TIME-OF-FLIGHT STUDIES OF MUON CATALYZED FUSION
WITH A MUONIC TRITIUM BEAM

by

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Abstract

In this thesis, we establish a new approach in muon catalyzed fusion studies, the time-of-flight method with an atomic beam of muonic tritium, and report results for muonic tritium scattering and epithermal $d\mu t$ resonant formation, providing the first quantitative measurements on these reactions.

Emission of muonic tritium from a solid hydrogen thin film into vacuum was observed via imaging of muon decay electrons, and the measurement of the position and the time of muon decay provided spectroscopic evidence for the Ramsauer-Townsend effect in the $\mu t + p$ interaction. The RT minimum energy was determined to be $13.6 \pm 1.0$ eV, in fair agreement with quantum three body calculations.

Using this $\mu t$ beam, we have confirmed theoretical $\mu t + d$ scattering cross sections to an accuracy of 10% by measuring the attenuation of $\mu t$ through a deuterium layer. The importance of $p$-wave scattering in the $\mu t + d$ interaction, as suggested by the theory, was also confirmed by our data via comparisons with Monte Carlo calculations assuming different scattering angular distributions.

The existence of a predicted resonance for $d\mu t$ formation in $\mu t + D_2$ collisions was directly confirmed for the first time. Our results correspond to a peak resonance rate of $(8.7 \pm 2.1) \times 10^9$ s$^{-1}$ in Faifman's model, more than an order of magnitude larger than the room temperature rates, and indicate a resonance energy of $0.42 \pm 0.04$ eV for the F=1 resonance peak in ortho deuterium.

Assuming the theoretical $[(d\mu t)\text{dee}]$ energy spectrum, these results imply sensitivity to the binding energy of the loosely bound state of the $d\mu t$ molecule, with an accuracy approaching the magnitude of the relativistic and QED effects.
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Chapter 1

Introduction

1.1 Muon Physics

Since its discovery in cosmic rays in 1937, the muon has played an important role in our understanding of nature. The muon has been extensively studied to determine its own properties and interactions and it has also been used as a probe to reveal the nature of other particles and systems. Indeed, it was the muon which gave us the first indication that nature replicates particles in a similar pattern, now known as generations. The existence of generations, and the muon itself for that matter, persists as a big mystery after over 50 years. With its 2.2 $\mu$s mean life, one of the longest of all elementary particles unstable under weak decay, it also provides a rich variety of applications in diverse areas of science, including condensed matter physics and chemistry.

From my somewhat biased point of view, currently there is renewed interest in basic muon physics, mainly on two fronts. High energy physicists are seriously considering the design and construction of a muon collider (the First Muon Collider), which if realized, has a potential to become a “Higgs Factory,” as well as achieving a much higher collision energy than the existing electron colliders. At low energy, or the precision front as it is sometimes called, muonic processes forbidden (or highly suppressed) by the Standard Model, such as the decays $\mu \rightarrow e\gamma$, and $\mu \rightarrow eee$, have drawn continuing and perhaps recently more intensified attention as a probe of the physics beyond the Standard Model. In particular, many Supersymmetric and/or Grand-Unified extensions of the Standard
Chapter 1. Introduction

Model, the current theoretical favorite, naturally require these processes to occur, which, if observed, would have a considerable impact on the way we see our Universe.

Although its fundamental properties and interactions are of great interest, for the purpose of this thesis, which focuses on atomic and molecular aspects of muon physics, much of the behavior of the negative muon can be described by that of a heavy electron. Nevertheless, because of the muon's heavy mass, comparable to that of light nuclei, the muonic system exhibits many unique characteristics, including the catalysis of nuclear fusion among hydrogen isotopes. As we will see, the nuclear and particle physics aspects of muonic processes become important experimentally in understanding various systematic effects and background processes encountered in our experiment. In addition, because of the use of solid targets, condensed matter physics comes into play. Indeed, this experiment allows me an opportunity to learn about a rich variety of physics at energy scales spanning over more than twelve orders of magnitude, from the hydrogen Debye temperature of $10^{-2}$ eV to the electroweak scale of $10^{11}$ eV.

In the rest of this chapter, I will give a brief review of $\mu$CF and describe the concept of our experiment. Chapter 2 focuses on the theoretical aspect, while the experiment apparatus is described in Chapter 3. Chapter 4 gives a summary of our experimental runs. After explaining our simulation codes in Chapter 5, the details of the analysis will be given in Chapters 6-8, which be followed by a discussion and conclusion.

1.2 Overview of the muon catalyzed fusion cycle

Figure 1.1 illustrates a greatly simplified scheme of the muon catalyzed fusion ($\mu$CF) cycle in a D/T mixture\(^1\). The system has attracted the greatest interest because of

\(^1\)When referring to a mixture of hydrogen isotopes, a notation $X/Y$ is used in this thesis, where $X$, $Y$ are protium ($^1$H), deuterium or tritium. It is usually assumed that the mixture has an equilibrium molecular composition, i.e., $X_2:XY:Y_2 = c_X^2:2c_Xc_Y:c_Y^2$, where $c_X$ and $c_Y$ denote the relative
its most favorable efficiency for fusion catalysis (see for a review [1–4]). Note that a homogeneous mixture, of mostly gas or liquid, has been used traditionally, as opposed to the inhomogeneous targets used in this thesis.

A muon injected into the hydrogen target will slow down and form a small atomic system, muonic deuterium (\(\mu d\)) or muonic tritium (\(\mu t\)), by replacing the electron in the atom. If a \(\mu d\) is formed, the muon will be transferred to a triton forming a more tightly bound \(\mu t\). The \(\mu t\) will then collide with a deuterium molecule and form the muonic molecule\(^2\) \(d\mu t\). Molecular formation occurs predominantly via a resonant mechanism, in which the energy released from the formation of the \(d\mu t\) molecule is absorbed by the rotational and vibrational excitation of the molecular complex \([(d\mu t)dee]\) where the compact object \(d\mu t\) acts as a pseudonucleus. Muonic molecule formation can also occur by releasing the energy via the Auger process (non-resonant formation), but this rate is much

\(^2\)To be precise, this should be called a muonic molecular ion, but we shall simply denote it as a muonic molecule according to the convention in the field.
smaller than resonant formation. Because the size of the muonic molecule is smaller than ordinary molecules by its mass ratio \(m_\mu/m_e\) in zeroth order, the internuclear distance in \(d\mu t\) is small enough that fusion takes place within \(10^{-12}\) s. After fusion, the muon is released more than 99% of the time, but a small probability exists for a process known as sticking in which the muon becomes attached to the charged fusion product, in this case an \(\alpha\) particle. If sticking occurs, the muon is lost from the cycle, and this indeed limits the ultimate number of fusions that one muon can catalyze. In the following sections, we will discuss each process involved in some detail.

### 1.2.1 Muonic atom formation

When a muon stops in a hydrogen isotope target, it replaces the electron of an atom to form a muonic atom in an excited state. Understanding muonic atom formation and the cascade process to the ground state is important, since they affect muon transfer from excited states (the \(q_{1s}\) problem), as well as the creation of hot atoms via acceleration. The sequence is, however, one of the least studied processes in \(\mu\)CF [2], partly due to the lack of direct experimental information. Recent theoretical developments on the formation processes are reviewed in Refs. [5-7].

In a simple picture\(^3\), the muon is expected to be captured in an excited orbital state with the principal quantum number \(n \sim \sqrt{m_\mu/m_e} \sim 14\), whose wave function has similar energy and spatial size to that of the ground state electron\(^4\). In reality, the target is molecular rather than atomic hydrogen, and muonic atom formation is predicted to occur via formation of an excited molecule \([xy\mu^-e^-]^\ast\), and its decay, in semi-classical

---

\(^3\)Recall that for the principle quantum number \(n\), the orbital radius \(r_n\), the energy \(E_n\) and the orbital velocity \(v_n\) are: 
\[ r_n = \frac{n^2h^2}{m_\mu Z^2}, \quad E_n = -\frac{Ze^2}{2r_n} = -\frac{m_\mu Z^2e^4}{2n^2h^2} \quad \text{and} \quad v_n = \frac{Ze^2}{n^2}, \]
where \(m_\mu\sim m_\mu\) is the reduced mass of the muon and the nucleus with the charge \(Z\).

\(^4\)Note that the corresponding velocity of the electron differs from that of the muon by the factor \(v_e/v_\mu \sim \sqrt{m_\mu/m_e}\).
calculations by Fesenko and Korenman [6], results in a broad distribution over quantum states $n$. More complete, but quasi-classical five-body dynamical calculations, including rotational and vibrational degrees of freedom of the molecule are being carried out by Cohen [7, 8].

1.2.2 Cascade

Muonic atoms formed in highly excited states quickly de-excite to the ground state via several competing mechanisms [9, 10]:

1. Radiative: $(\mu x)_i \rightarrow (\mu x)_f + \gamma$

2. External Auger: $(\mu x)_i + YZ \rightarrow (\mu x)_f + YZ^+ + e^-$

3. Stark Mixing: $(\mu x)_n + Y \rightarrow (\mu x)_{n'} + Y$

4. Coulomb Collisions: $(\mu x)_i + y \rightarrow (\mu x)_f + y$, $n_f < n_i$

5. Elastic Scattering: $(\mu x)_n + YZ \rightarrow (\mu x)_n + YZ$

In addition, muon transfer can take place from an excited state before reaching the ground state, as will be discussed below. Each process has a different dependence on target density and collision energy, and competition between the processes dictates the energy distribution of muonic atoms in the ground state. Coulomb de-excitation (item 4 above) has received recent attention as a possible mechanism for accelerating muonic atoms to up to more than 100 eV. Calculations by Markushin predict as many as 50% of muonic atoms have energies above 1 eV at the instant of reaching the ground state at liquid hydrogen density [9].
1.2.3 Muonic atom collisions

In the traditional description of the $\mu$CF cycle, it was assumed that the muonic atom was always thermalized at the target temperature [11], and the energy dependence of the reaction rates, if considered at all, was averaged out by integrating over the Maxwellian distribution. This type of analysis using the constant rates has proven very successful in particular for the pure liquid and gaseous deuterium system [12].

The importance of going beyond the constant rate approach was recognized by the suggestion of epithermal (i.e., non-thermalized) transient phenomena by Kammel [13] and by Cohen and Leon [14], but it was not until the complete set of theoretical cross sections [15-21] became available that an energy dependent analysis could be performed in $\mu$CF [22].

It is absolutely essential for our measurement to take into account the energy dependence; or rather, our experiment is designed to take advantage of its sensitivity to the energy dependent cross sections.

Theoretically, collisional processes of muonic atoms present challenging few-body problems. Because the muon mass is comparable to that of nuclei, the system is highly non-adiabatic, i.e., nuclear and muonic motions cannot be separated, as we shall see in detail in Section 2.1.2. The main collisional processes of muonic hydrogen isotope atoms include elastic scattering, muon transfer (also called charge exchange), and hyperfine transitions (spin flip). Table 1.1 lists the main properties of muonic atoms, while Fig. 1.2 compares the various cross sections of $\mu$t collisions with hydrogen isotope nuclei.

Elastic scattering

Elastic scattering processes dictate the deceleration of muonic atoms in the target, hence they determine the energy distribution at the time of the subsequent reaction (such as
Table 1.1: Main properties of muonic atoms from Ref. [23]a.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Nucleus mass ((M_x))</th>
<th>Ground state (E_{\mu x}) (eV)</th>
<th>Hyperfine splitting (\Delta E_{\mu x}^{hfs}) (eV)</th>
<th>Isotope splitting (\Delta E_{xy}) (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>p</td>
<td>1836.1515</td>
<td>-2528.517</td>
<td>0.1820</td>
<td>(\Delta E_{pd} = 134.709)</td>
</tr>
<tr>
<td>d</td>
<td>3670.481</td>
<td>-2663.226</td>
<td>0.0485</td>
<td>(\Delta E_{dt} = 48.042)</td>
</tr>
<tr>
<td>t</td>
<td>5496.918</td>
<td>-2711.268</td>
<td>0.2373</td>
<td>(\Delta E_{pt} = 182.751)</td>
</tr>
</tbody>
</table>

aThe masses are given in units of the electron mass \(m_e = 0.5109991\) MeV/c\(^2\). The muon mass is \(m_\mu = 206.7686m_e\), the Rydberg energy, \(R_y = 13.605804\) eV. These values are used in the calculations of muonic molecular energy levels given in Table 2.1.

muon transfer or molecular formation) in the \(\mu\)CF cycle.

There is a relatively limited amount of experimental information on muonic atom scattering cross sections [24]. In particular, there is hardly any data on energy dependent cross sections.

The elastic scattering cross section can be written in terms of the sum of the contributions of the partial waves:

\[
\sigma = \sum_l \sigma_l = \frac{4\pi}{k^2} \sum_l (2l + 1) \sin^2 \delta_l, \tag{1.1}
\]

where the physics is contained in the phase shift \(\delta_l\). As one can see from Eq. 1.1, if for a given partial wave \(l\), \(\delta_l\) is approximately equal to \(n\pi\) \((n = 1, 2, 3 \cdots)\), then the partial cross section \(\sigma_l\) goes to zero. For low energy collisions, where \(s\) wave \((l = 0)\) usually dominates, the total cross section can become very small, a process known as the Ramsauer-Townsend effect. If, on the other hand, \(\delta_l \sim (n + \frac{1}{2})\pi\), then the cross section becomes large and reaches a maximum value \((4\pi/k^2)\) called the unitarity limit. It is interesting to note that all the phase shifts at the zero energy limit are fixed at \(\delta_l = B_l\pi\) by Levinson’s theorem [25, 26], where \(B_l\) is the number of three-body bound states of given \(l\), illustrating the intricate connection between bound states and scattering properties. As we shall see it is the Ramsauer-Townsend effect that makes it possible to produce a muonic atom beam for our experiments.
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Muon transfer

Due to the differences in the binding energies, the muon can be transferred to the heavier isotopes:

$$(\mu x)_{ni} + y \rightarrow x + (\mu y)_{ni} + \frac{\Delta E_{xy}}{n^2},$$

where $\Delta E_{xy}$ is the isotope splitting (Table 1.1) due to the reduced mass difference. The subscript $n$ denotes the possibility of transfer from excited states (Section 1.2.4). In the D/T mixture target, the rate of muon transfer from the ground state of the deuterium atom to the triton $\lambda_{dt}$ is relatively slow ($\sim 3 \times 10^8 \text{ s}^{-1}$) and becomes one of the bottle-necks at low tritium concentrations. It should be noted that the released energy, divided between two projectiles, gives an acceleration to the muonic atom in the lab frame corresponding to the energy

$$E(\mu y)_{Lab} = \frac{m_x}{m_x + m_{\mu y}} \frac{\Delta E_{xy}}{n^2}. \quad (1.3)$$

The energy dependent calculations predict that $\lambda_{dt}$ is significantly larger at higher energies ($\gtrsim 10 \text{ eV}$). This may be exploited, for example, by triple isotope mixture targets (H/D/T) [27], in which a faster reaction, muon transfer from a proton to a deuteron, creates an energetic $\mu d$, thus effectively speeding up the transfer to tritium.

Hyperfine transitions

Some muonic processes such as resonant molecular formation, fusion, and nuclear muon capture are known to depend on the hyperfine state of the muonic atom, though for different reasons. The most spectacular example is a drastic dependence of the $d\mu d$ formation rate at low temperature on the $\mu d$ hyperfine state [28]. Other cases where spin flip plays a crucial role include the Wolfenstein–Gershtein effect in $p\mu d$ [29–31] and $p\mu t$ fusion [32], and muon capture on a proton (Ref. [33] and references therein). Spin flip is
Figure 1.2: Scattering cross sections for $\mu t$ with a hydrogen isotope nuclei from the Nuclear Atlas [16,17], showing the Ramsauer-Townsend minimum at around 10 eV for $\mu t + p$. $\mu t(F) + t$ cross sections plotted include both elastic and spin exchange reactions, where $\mu t(0)$ is the singlet state and $\mu t(1)$ is the triplet state.

usually considered in symmetric collisions:

$$\mu x(F) + x \rightarrow \mu x(F') + x + \Delta E_{\mu x}^{hfs},$$

where the reaction is dominated by the exchange of the muon between two identical nuclei. There is also the possibility, though much smaller, of spin flip in asymmetric collisions:

$$\mu x(F) + y \rightarrow \mu x(F') + y + \Delta E_{\mu x}^{hfs}. $$
Obviously, unlike the symmetric case, this cannot be achieved by muon exchange, and a relativistic interaction is required to flip the spin so the cross sections are much lower. According to Cohen [21], who calculated these processes in the Improved Adiabatic approach (Section 2.1.3), non-symmetric spin flip cross sections are several orders of magnitude smaller than symmetric spin flip cross sections.

In addition, there is a prediction that spin flip in the pure deuterium system occurs via the back decay of the muonic molecular complex (resonant spin flip reaction) [34],

\[ \mu d(F = \frac{3}{2}) + D_2 \rightarrow [(d\mu d)dee]^* \rightarrow \mu d(F = \frac{1}{2}) + D_2, \]  

(1.6)

but the experimental data become rather inconsistent with theoretical predictions, if the resonant spin flip is included [12,35].

1.2.4 Processes of excited muonic atoms

One of the primary questions in \( \mu \)CF is the so called \( q_{ls} \) problem. In a D/T mixture, some fraction of the muons captured in the excited state of muonic deuterium can be transferred to the triton before reaching the ground state with the probability \((1 - q_{ls})\), where the probability for the muon to reach the ground \( 1s \) state is denoted by \( q_{ls} \). Since the transfer rate from excited states is very fast\(^5\), \( q_{ls} \) can affect the cycling rate (the number of fusions catalyzed by the same muon per second). Conversely, the extraction of \( \mu \)CF parameters from the measurement of the cycling rates depends on the \( q_{ls} \) value (see Section 1.3.1).

There has been a longstanding discrepancy between theoretical predictions, based on semi-classical calculations, and experimental data derived indirectly (see for example,

\(^5\)This is expected from a naive argument that the muonic atom with higher \( n \) is much more extended in size, hence has larger cross sections, but the bigger effect presumably is the long range behavior of the potential for the excited states.
Chapter 1. Introduction

Ref. [1, 36]). Recent experimental developments [37, 38] using state-of-the-art X ray detection technologies are producing more direct information about \( q_{1s}^{pd} \) in the case of \( pd \) transfer in H/D mixtures, and it is hoped that they will give some insight into the more important D/T case.

Processes involving excited states, \( n > 1 \), of the muonic atom have gained increasing theoretical attention. In addition to semi-classical calculations of excited transfer reactions [39, 40], full quantal calculations are emerging [41] using the hyperspherical approach (Section 2.1.4). The transport cross sections of excited atoms including Stark transitions are being investigated mainly in semi-classical approaches [42, 43].

Recently, a new process has been suggested by Froelich and Wallenius [44–46]. They predict a high rate of formation of a muonically excited metastable three-body state \( \mu t^* \) (associated with the adiabatic 3\( \sigma \) potential [47]) in the collision of excited \( \mu t(2s) \) with \( D_2 \), which will then decay into \( \mu d(1s) \), effectively reversing the transfer reaction:

\[
\mu t(2s) + DX \rightarrow [(\mu t^*)^*dee] \rightarrow \mu d(1s) + TX,
\]

(1.7)

where \( X = D \) or \( T \). The excited molecule \( \mu t^* \) can decay into \( \mu t(1s) + d \) as well, but the decay width ratio is about 9 to 1 favoring the decay into \( \mu d(1s) + t \), because the wave function of the latter channel is less orthogonal to, hence has a larger overlap with, the state \( \mu t^* \) [48]. The proposed side path model appears to give a better agreement with measurement of the cycling rates [49], but clearly more studies are desirable. Understanding of excited muonic atom processes is important also for QED and weak interaction experiments utilizing the possible meta-stable population of 2s muonic atoms [50].

We note that the side path model is unlikely to impact our measurements using \( \mu t \) emitted from layers, since the Ramsauer-Townsend effect is not expected for excited muonic atoms.
1.2.5 Formation of muonic molecules

Non-resonant formation

A straightforward process for muonic molecular formation is via the emission of an Auger electron:

\[ \mu x + YZ \rightarrow [(x\mu y)^+ze] + e^- \]  \hspace{1cm} (1.8)

The rate for this non-resonant process is typically of the order of \(10^6\) s\(^{-1}\), hence one muon could catalyze not many more than one fusion, if this were the only process for formation.

The most recent calculations of the non-resonant molecular formation rates were performed by Faifman for all the combinations of collisions [51]. Using the molecular bound state energy levels and wave functions derived from the improved two level approximation in the Adiabatic Representation (Section 2.1.3), the rates were calculated taking into account monopole E0 and dipole E1 Auger transitions to all the bound states (earlier calculations considered only selected states). Note that the existence of weakly bound states, which is of paramount importance for resonant formation, also affects non-resonant formation.

Resonant formation

Observations were made in Dubna in the early 1960s for the temperature dependence of fusion yields in muon catalyzed \(dd\) fusion [52]; this was a strange phenomenon at the time since nuclear reactions of the MeV scale appeared to be affected by the target temperature which is of the meV scale. An Estonian graduate student, Vesman, proposed a resonant process for muonic molecular formation to explain the observations [53]. According to
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<table>
<thead>
<tr>
<th>Reaction</th>
<th>Collision energy (cms) [eV]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.003</td>
</tr>
<tr>
<td>$\mu p + p \rightarrow p\mu p$</td>
<td>1.81</td>
</tr>
<tr>
<td>$\mu d + d \rightarrow d\mu d$</td>
<td>0.03</td>
</tr>
<tr>
<td>$\mu t + t \rightarrow t\mu t$</td>
<td>2.71</td>
</tr>
<tr>
<td>$\mu d + p \rightarrow p\mu d$</td>
<td>5.69</td>
</tr>
<tr>
<td>$\mu p + d \rightarrow p\mu d$</td>
<td>0.17</td>
</tr>
<tr>
<td>$\mu t + d \rightarrow d\mu t$</td>
<td>0.56</td>
</tr>
<tr>
<td>$\mu d + t \rightarrow d\mu t$</td>
<td>2.26</td>
</tr>
<tr>
<td>$\mu t + p \rightarrow p\mu t$</td>
<td>6.47</td>
</tr>
<tr>
<td>$\mu p + t \rightarrow p\mu t$</td>
<td>0.03</td>
</tr>
</tbody>
</table>

Table 1.2: Non-resonant muonic molecular formation rates in $10^6$ s$^{-1}$ by Faifman [51].

The Vesman mechanism,

$$\mu x + Y Z_{\nu_i,K_i} \rightarrow [(x\mu y)^S_{J_v,\nu_v} z_{ee}]_{\nu_f,K_f},$$

(1.9)

where the kinetic energy of $\mu x$ and the energy released upon formation of a loosely bound $(x\mu y)_{J_v}$, with $(J, v)$ the ro-vibrational quantum numbers, is absorbed in the rotational $(K)$ and vibrational $(\nu)$ excitation of the molecular complex $[(x\mu y) z_{ee}]^*$, a hydrogen like molecule with $x\mu y$ playing a role of one of the nuclei. Since the final states have discrete spectra corresponding to ro-vibrational levels $(\nu_f, K_f)$, the process takes place only when the collision energy matches the resonant condition. The temperature dependence is derived from the varying degree of overlap between the Maxwellian distribution (assuming $\mu x$ is thermalized) of $\mu x$ atoms with the resonance energy profile at different temperatures. For the Vesman mechanism to occur, the muonic molecule has to have a level with the binding energy smaller than the dissociation energy ($\sim 4.5$ eV) of the host molecule. This is extremely loosely bound in comparison with the typical muonic energy scale of a few keV. Indeed such states have been shown to exist theoretically by Ponomarev and his co-workers for $d\mu d$ and $d\mu t$ molecules, with quantum numbers $(J, v) = (1, 1)$, where
Table 1.3: Non-relativistic Coulomb molecular binding energies -$\epsilon_{Jv}^0$ (eV).

<table>
<thead>
<tr>
<th>$J, v$</th>
<th>$\mu \nu$</th>
<th>$\mu \delta$</th>
<th>$\mu \tau$</th>
<th>$\delta \mu$</th>
<th>$\delta \tau$</th>
<th>$\tau \mu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0,0</td>
<td>253.15</td>
<td>221.55</td>
<td>213.84</td>
<td>325.07</td>
<td>319.14</td>
<td>362.91</td>
</tr>
<tr>
<td>0,1</td>
<td>35.84</td>
<td>34.83</td>
<td>83.77</td>
<td>226.68</td>
<td>232.47</td>
<td>289.14</td>
</tr>
<tr>
<td>1,0</td>
<td>107.27</td>
<td>97.50</td>
<td>99.13</td>
<td>226.68</td>
<td>232.47</td>
<td>289.14</td>
</tr>
<tr>
<td>1,1</td>
<td>1.97</td>
<td>0.66</td>
<td>45.21</td>
<td>226.68</td>
<td>232.47</td>
<td>289.14</td>
</tr>
<tr>
<td>2,0</td>
<td>86.45</td>
<td>102.65</td>
<td>172.65</td>
<td>226.68</td>
<td>232.47</td>
<td>289.14</td>
</tr>
<tr>
<td>3,0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>48.70</td>
</tr>
</tbody>
</table>

$J$ and $v$ denote rotational and vibrational states, respectively.

Muonic molecular ions are three-body systems interacting with both electromagnetic and strong forces, but to a good approximation ($\lesssim 10^{-4}$ eV), direct effects of the strong force can be neglected in calculating energy levels of loosely bound states, although other nuclear properties such as the charge form factors and polarizabilities play non-negligible roles. Now modern calculations have achieved amazing accuracies, as discussed in Section 2.1. Table 1.3 shows the nonrelativistic Coulomb bound state energy levels of muonic molecules.

The molecular complex [$(x\mu y)dee]^*$ is metastable, and can decay into several channels. An Auger transition of the $x\mu y$ molecule is an important step, which stabilizes the molecule, leading to fusion. This competes with the back decay process, which returns the excited molecular complex to $\mu x$ and $YZ$. The details of the formation and back decay processes are discussed further in Section 2.2 and in Appendix B.

### 1.2.6 Nuclear fusion

The rate of nuclear fusion in the $d\mu t$ molecule is of the order of $10^{12}$ s$^{-1}$, hence it is too fast to measure experimentally. The reaction is strongly dominated by the resonance $^{5}$He$^d, \pi=\frac{3}{2}^+$, which lies about 55 keV above $\mu t + d$ threshold. Because of the centrifugal
barrier of the $J = 1$ states of the molecule, fusion is expected to take place mainly from $J = 0$ states, i.e., $(J,v) = (0,1)$ and $(0,0)$, into which the initial state $(1,1)$ is quickly converted via fast $E1$ Auger transitions.

For $d\mu d$, on the other hand, the transition from $J = 1 \rightarrow 0$ is strongly suppressed (since it requires spin flip, analogous to the ortho-para transitions in the homonuclear hydrogen molecule), and fusion takes place mainly from the $J = 1$ states, if the molecule is resonantly formed in the $(1,1)$ state. This in fact offers a unique opportunity to study the $p$ wave fusion reaction at very low energy, which is difficult to do in a beam experiment. An interesting feature of this reaction is that the fusion width ratio of the $(n + {^3}\text{He})$ channel to the $(p + t)$ channel is about $1.4$, apparently violating charge symmetry.

In a simple approach, the fusion rate is calculated by treating three-body Coulomb physics (molecular wave functions) and nuclear physics separately [54], a method known as factorization. For $d\mu t$, we have

\[ \lambda_{d\mu t}^f = A \lim_{R_{dt} \to 0} \int S|\Psi_{d\mu t}(r, R_{dt})|^2 d^3r, \]

where $A$ is related to the zero energy limit of the astrophysical $S$ factor,

\[ A = \frac{\hbar}{\pi e^2 \overline{M}_{dt}} \lim_{E \to 0} S(E), \]

with $\overline{M}_{dt}$ being the reduced mass of $d$ and $t$. $S$ is obtained from the fusion cross section with Coulomb repulsion factored out, hence it contains all the nuclear physics. Thus in this approach, the overlap of the $d - t$ wave functions in the united-atom limit is multiplied by a single number describing the strong interaction.

More accurate approaches treat the nuclear force dynamically by directly incorporating it as a complex potential in the three-body Hamiltonian (optical potential method) [55,56], or as complex boundary conditions at the nuclear surface ($R$-matrix
method) [57–59]. These elaborate approaches agree rather well with each other, as well as with the simple factorization approach.

In $\mu x + y$ collisions, fusion “in flight”, i.e., without forming a bound $x\mu y$ molecule, is also possible, and its rates have been calculated by various methods including, most recently, Faddeev equations (see [26] and references therein). The reactions are enhanced, compared to bare nuclear collisions, especially if virtual states exist, due to the increased overlap of the nuclear wave functions.

1.2.7 Sticking and stripping

Sticking is a process in which the muon becomes attached to a positively charged fusion product such as an $\alpha$ particle. The probability of the process, denoted by $\omega_s$, imposes a more stringent limit on the efficiency of $\mu$CF than the short lifetime of the muon itself, hence it has attracted much attention, but discrepancy between experiments and theory persists, motivating further investigation.

Sticking takes place in two distinct steps:

\[
\begin{align*}
\text{sticking:} & \quad \mu^- + \alpha^* + n \\
\text{stripping:} & \quad \mu^- + \alpha + n
\end{align*}
\]

where $\mu\alpha^*$ refers to the muonic helium ion in a bound state $n, l$ recoiling with kinetic energy 3.46 MeV. The initial sticking $\omega_s^0$ is the intrinsic branching ratio of the sticking reaction channel immediately after the fusion, while stripping can occur during the slowing down of the $\mu\alpha$, that is the muon is detached from the $\alpha$ with probability $R$, in collisions.
with the neighbouring atoms. Hence the final sticking $\omega_s$ is:

$$\omega_s = \omega_s^0 (1 - R).$$  \hspace{1cm} (1.12)

The experimental results on sticking have been in disagreement with theory and, sometimes, with each other. The challenge lies in its low branching ratio and in the handling of tritium. Figure 1.3 summarizes the previous measurements of final sticking plotted against target density $\phi$. Apart from a longstanding discrepancy on the density dependence between two major experimental groups at PSI and LAMPF, in which the analysis of the latter now appears to be unreliable at low densities, most experimental values are systematically and substantially smaller than the theoretical prediction, even though different methods have been used. For a recent review, see Ref. [60].
1.3 Measurement of resonant molecular formation

In this section, we shall describe the concept of our experiment using an atomic $\mu t$ beam and discuss its expected advantages. But first, let us make a few remarks on the conventional experiments in D/T mixtures.

1.3.1 Cycling measurements in D/T mixture

In conventional experiments [61–69], the molecular formation rates are determined from the measurements of the cycling rate $\lambda_c$, which is $1/\tau_c$, where $\tau_c$ is the mean time between the 14 MeV neutrons from fusions catalyzed by the same muon.

In this method, the cycling rates, measured at various target conditions (such as the tritium concentration $c_t$, temperature $T$, and the density $\phi$), are fitted to an expression describing the kinetics model. Molecular formation rates $\lambda_{d\mu t}^F$, together with quantities such as the $d \rightarrow t$ transfer rate $\lambda_{dt}$ and $q_{1s}$, are the fitting parameters. A simplified expression for $\lambda_c$ reads [67]:

\[
\frac{1}{\lambda_c} \approx \frac{c_d q_{1s}}{c_t \lambda_{dt}} + \frac{3/4}{c_t \lambda_{10} + c_d \lambda_{d\mu t}^1} + \frac{1/4 + 3/4 \chi_1}{c_d \lambda_{d\mu t}^0 \chi_2}, \quad \text{with}
\]

\[
\chi_1 = \frac{c_t \lambda_{10}}{c_t \lambda_{10} + c_d \lambda_{d\mu t}^1},
\]

\[
\chi_2 = \frac{\lambda_{t\mu t}^F}{c_t \phi \lambda_{t\mu t} + \lambda_{t\mu t}^F},
\]

where $\chi_1$ is the fraction of $\mu t$, $F = 1 \rightarrow 0$ transition with the spin flip rate $\lambda_{10}$, and $\chi_2$ is a correction factor taking into account the $t\mu t$ fusion time, with $\lambda_{t\mu t}$ and $\lambda_{t\mu t}^F$ being $t\mu t$ formation and fusion rates, respectively (note $t\mu t$ formation is non-resonant, hence assumed to be independent of $F$). An improved formula would include a further correction term due to the muon recycling in $d\mu d$ and $t\mu t$ branches without producing the 14 MeV neutrons, which depends on parameters like $d\mu d$ formation and spin flip.
rates.

The molecular formation rate depends not only on the $\mu t$ hyperfine state $F$, but also on the target molecular species. Thus, $\lambda^F_{\text{dust}}$ consists of different components

$$c_d \lambda^F_{\text{dust}} = c_D \lambda^F_{\text{dust} - d} + \frac{C_D T}{2} \lambda^F_{\text{dust} - t} + c_d \lambda^r_{\text{dust}}$$

for $F=0,1$, (1.14)

which have to be disentangled in the fit. Often, assumptions have to be made in actual fits with guidance from theory. For example, at low temperatures all the terms in $\lambda^F_{\text{dust}}$ except $\lambda^0_{\text{dust} - d}$ are assumed to be zero [22,67]. We shall come back to the implications of these assumptions later in this thesis.

As Cohen points out [4], further complication in the conventional method comes from the lack of precise characterization of the parameter $q_{1s}$ (the probability that the muon reaches the ground state of $d\mu$ before transferring to a triton). Its behavior suggested by the cycling fit is in large disagreement with theoretical expectations of its $c_t$ and $\phi$ dependence. Independent measurements of $q^{pd}_{1s}$ for $pd$ transfer are emerging [37], but no direct information is yet available for the $dt$ case.

Furthermore, the recent suggestion by Froelich and Wallenius of a resonant side-path in D/T cycling [44,49] (see Section 1.2.4) could have significant impact in the extraction of the $d\mu t$ formation rate from the cycling rate.

Finally, it should be noted that in these analyses using single rates, the energy dependencies of the processes are averaged out.

### 1.3.2 Epithermal molecular formation

Muonic molecule formation with epithermal (non-thermalized) $\mu t$ atoms has attracted much recent interest because of the very large rate predicted by theory [70]. With the maximum rate approaching $10^{10} \text{ s}^{-1}$, this is nearly two orders of magnitude larger than
formation rates with $\mu t$ in thermal equilibrium at room temperature ($\sim 10^8 \text{ s}^{-1}$). Figure 1.4 shows the recently calculated epithermal resonant molecular formation rate in $\mu t + D_2$ collisions [71, 72], normalized to liquid hydrogen density (LHD). Also plotted is the elastic scattering rate of $\mu t$ on the $d$ nucleus [17].

Epithermal molecular formation, in fact, was first suggested by Cohen and Leon [14], and independently by Kammel [13], to explain the fusion time spectra observed in the PSI experiment with low density gas [64], in which an unexpected prompt peak was initially misinterpreted as due to the hyperfine effect$^6$. Later Jeitler et al. [22, 74] carried

$^6$A qualitative remark of the possibility of epithermal formation was given earlier by Rafelski [73].
out Monte Carlo studies in the *homogeneous* mixture of D/T\textsuperscript{7}. They obtained qualitative agreement between the experimental data and the simulations assuming strong epithermal resonances, but neither the strength nor the position the resonances could be determined by their analysis, because (a) the neutron time spectra are essentially insensitive to the position of epithermal resonances, and (b) the magnitude of the prompt peak is dependent both on the formation rate strength and the poorly known initial population of epithermal \( \mu t \), hence with the lack of knowledge of the latter, the former cannot be extracted reliably.

In order to access these resonances with thermal energies in a conventional target, one would require a temperature of several thousand degrees. The Dubna group has recently developed a target which can operate at up to 800 K, but this is still substantially smaller than the required temperature for the main resonances. Efforts are being made by the PSI/Vienna group to develop an ambitious target system for 2000 K, but the technical difficulties in handling tritium at such a high temperature have so far prevented the realization of such a system.

In this thesis, we will develop an alternative approach [76–79] for investigating the epithermal molecular formation, as illustrated in Fig. 1.5. Taking advantage of the Ramsauser-Townsend effect, a neutral atomic beam of muonic tritium is obtained [80]. By placing a second interaction layer, separated by a drift distance in vacuum, reaction measurements are possible in an event-by-event fashion. This is in sharp contrast to conventional methods where a muon is stopped in a bulk gas or liquid target in which complex, and inevitably interconnected chains of reactions take place, as seen in Section 1.3.1. The use of the muonic atom beam would thus help us to isolate the process of interest from the rest of the "mess." Furthermore, the time of flight of \( \mu t \) across the drift distance provides information on the \( \mu t \) kinetic energy, hence the energy dependence of

\textsuperscript{7}Markushin et al. also performed a Monte Carlo analysis of the triple mixture H/D/T [27,75]
the formation rates can be tested.

![Diagram of time-of-flight measurement of resonant molecular formation rates using the $\mu t$ atomic beam.](image)

Figure 1.5: Conceptual drawing of the time-of-flight measurement of resonant molecular formation rates using the $\mu t$ atomic beam.

The Basic processes involved in our experiments are as follows [81,82]:

**Production:** Muonic atoms are produced by stopping low-momentum muons ($\sim 27$ MeV/c) in a thin layer ($\sim 0.4$ mm) of hydrogen with a small tritium concentration. The knowledge of the fraction of muons which stops in the hydrogen layer is of great importance for our measurements, and will be discussed in Section 6.3.

**Acceleration:** The muon transfer reaction $\mu p + t \rightarrow p + \mu t$, taking place in $\sim 100$ ns [83], is used to create a fast $\mu t$ with a kinetic energy of $\sim 45$ eV, which results from the reduced mass difference in the binding energy.
Chapter 1. Introduction

**Extraction:** $\mu t$ loses its energy via scattering mainly with protons, until it reaches around 10 eV, where due to the Ramsauer-Townsend (RT) effect, the $\mu t + p$ elastic cross section drops by a few orders of magnitude, and the rest of the target becomes nearly transparent. Since there is no RT effect in the $\mu p + p$ scattering\(^8\), $\mu t$ is selectively extracted from the layer with the characteristic energy of $\sim 10$ eV. The understanding of the $\mu t$ emission mechanism is essential to our measurements, and will be discussed in Chapter 7.

**Moderation:** The $\mu t$ beam energy can be degraded, when necessary, by placing a moderation layer on top of the production layer. For the epithermal formation rate measurement, where the largest peak lies at around 0.5 eV, the beam energy of $\sim 10$ eV is indeed too high, hence a D\(_2\) overlayer was used as a moderator (not shown in Fig. 1.5). A thin layer ($\sim 5$ $\mu$m) of D\(_2\) is sufficient to slow down the $\mu t$, due to the large elastic cross section(see Fig. 1.2). Fusion events also occur at the upstream (US) moderation layer, which become background to the fusion at the downstream (DS) reaction layer, but because of their mostly prompt time structure, the US events can be separated from the DS signal.

**Reaction:** The $\mu t$, after travelling across the drift distance, collides with a target D\(_2\) molecule. Resonant $d\mu t$ formation can take place in two different manners: (a) the direct process, where $d\mu t$ is formed directly before $\mu t$ is scattered, and (b) the indirect process, in which $\mu t$ first loses energy in collision with another D\(_2\) molecule, before molecular formation (see Fig. 1.4 for a comparison of the rates). The process (a) is most interesting because it preserves the correlation between the $\mu t$ time of flight (which is roughly equal to the time between the muon stop and the detection of a fusion product, due to short emission and fusion times) and the energy at which

\(^8\)This is due in part to the existence of a virtual (nearly bound) state near $\mu p + p$ threshold.
molecular formation takes place. On the other hand, in the process (b), while the
correlation is obscured (e.g., a fast $\mu t$ with a short time-of-flight can slow down
significantly in the reaction layer and form $d\mu t$ at much lower energy), it can still
give us useful information, such as the magnitude of the resonance. The use of a
thin layer, together with a moderation of the $\mu t$ energy mentioned above, reduces
the fraction of the indirect process, thereby enhancing our sensitivity.

There are, of course, difficulties and disadvantages involved in our approach. In
addition to the obvious technical challenges in dealing with cryogenic and ultra-high
vacuum targets with the potential hazard of tritium, there are difficulties and limitations
which we did not foresee in advance (which are reflected in the volume of this thesis!).
They will be discussed in due course.

Before describing the details of our measurements and the analysis, we shall discuss
some theoretical aspects of muon catalyzed fusion in the following chapter, with partic­
ular emphasis on the muonic three body problem and molecular formation, in order to
highlight the importance of the physics involved in our experiment.
Chapter 2

Theoretical aspects of $\mu$CF

2.1 Muonic molecule and the three body problem

2.1.1 The challenge

Accurate descriptions of muonic molecule properties are essential to an understanding of $\mu$CF processes. The energy levels of the loosely bound state $(J,v) = (1,1)$ affect the temperature dependence of molecular formation rates; since 1 meV $\sim$ 10 K, it is desirable to achieve an accuracy of better than 1 meV for the energy level, which should be compared to the three body break up energy of muonic molecular ions of $\sim$ 3 keV. In addition to the energy levels, the molecular formation matrix elements are sensitive to details of the $d\mu t$ wave function, particularly in its asymptotic region, the formation rates being proportional to the square of a wave function parameter $C$ (see Section 2.2). Furthermore, the accuracy of the three-body wave function is important for $\mu\alpha$ sticking as well. The convergence in variational calculations of the sticking probability $\omega_s$ is much slower than that of the energy biding energy $\epsilon$; the general trend is that when $\epsilon$ is converged to $n_\epsilon$ digits, $\omega_s$ is accurate only to $n_s/3$ significant figures [3,84]. Thus, understanding two of the most important processes in $\mu$CF, i.e., molecular formation and sticking, demands the solution of the three-body problem to an accuracy of better than $10^{-7}$, a challenging task to the theorists. Indeed, in 1993, Szalewicz, one of the experts in the field, asserted these calculations to be "some of the most demanding few-body
calculations, taxing the most powerful supercomputers" [85].

Because the three body problem is central to processes involved in this thesis, we shall review its theoretical framework in some detail in the sections that follow, focusing on understanding the underlying physical concepts.

### 2.1.2 Three body coulomb problem

It is well known that the three body problem does not have a general analytical solution, and it has been the subject of considerable academic efforts since the 17th century. The history as well as recent progress on the oldest three body problem, the Moon-Earth-Sun system have been reviewed by Gutzwiller [87].

The Coulomb three body problem, in particular, has been a difficult one to solve accurately, due in part to the long range nature of the interaction, in comparison to few nucleon systems in which the interaction is short-ranged. It is an active area of research, as indicated by a number of recent developments and refinements of theoretical techniques with the help of ever-increasing computing resources. Efforts are being made to extend the calculations to full four-body muonic problems [92,93].

In general, the three body Hamiltonian can be written as

$$\mathcal{H} = T + V = \sum_{i=1}^{N=3} \frac{1}{2m_i} \Delta_{\rho_i} + \sum_{i<j}^{N=3} V_{ij}(|\rho_i - \rho_j|),$$  \hspace{1cm} (2.1)

where $\rho_i$ is the particle position vector [3]. The center of mass motion can be separated out, leaving a six dimensional problem in $\mathbf{r}_1$, $\mathbf{r}_2$ space.

The exact form of the Hamiltonian depends on the choice of the co-ordinates $\mathbf{r}_1$, $\mathbf{r}_2$. In terms of kinematics, there is no unique choice of the co-ordinates, hence choosing suitable ones, which will give accurate results, is one of the challenges that theorists face.

---

1. It is said that there were over 800 papers published on the subject between 1750 and 1920s [86].
2. See recent calculations of muonic and other three-body systems, [36,88–91].
2.1.3 Adiabatic approaches

Born-Oppenheimer approximation

In the conventional method for a molecular system, known as the Born-Oppenheimer approximation, nuclear motions are decoupled from electronic (or muonic) motions, *i.e.*, the total wave functions are assumed to have the form

\[ \Psi(r, R) = \chi(R)\psi(r, R), \]  

(2.2)

where \( \psi(r, R) \), which is obtained from

\[ H^0 \psi(r, R) = E^0(R)\psi(r, R), \]  

(2.3)

are the eigenfunctions for the Hamiltonian for fixed nuclei, \( H^0 \), with \( E^0 \) its eigenvalues. Here, the internuclear distance \( R \) is just a parameter. The nuclear wave function, \( \chi(R) \), is solved using \( E^0(R) \) as the effective potential for the nuclear motion;

\[ \left[ -\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} + E^0(R) - E \right] \chi(R) = 0. \]  

(2.4)

**Born's Adiabatic method**

The Born method [94] extends the Born-Oppenheimer approximation by including corrections to the effective potential \( E^0(R) \) in Eq. 2.4. The full Hamiltonian, separated in two parts, includes the term \( H' \) describing the relative motion of the two nuclei as well as the coupling between the electronic (muonic) and the nuclear motions, in addition to the term \( H^0 \) for the fixed nuclear problem;

\[ H = H^0 + H'. \]  

(2.5)

Note that \( H^0 \) includes nuclear repulsion in addition to the electronic interaction with the nuclei. The total wave function is expanded in the complete set of \( \psi_n(r, R) \):

\[ \Psi(r, R) = \sum_n \chi_n(R)\psi_n(r, R), \]  

(2.6)
where $\psi_n(r, R)$ are the eigenstates of the fixed nuclei problem Eq. 2.3. The solution to the problem

$$\left(\mathcal{H}^0 + \mathcal{H}'\right)\Psi(r, R) = E\Psi(r, R)$$

(2.7)

is obtained by substituting (2.6) into (2.5), multiplying the result by $\psi^*(r, R)$, and integrating over $r$, which yields

$$X_n(R) = \int \frac{\hbar}{2\mu} \Delta R + E_n^0(R) + \mathcal{H}'_{nn} - E \chi_n(R) = - \sum_{m \neq n} \mathcal{H}'_{mn}(R)\chi_m(R)$$

(2.8)

where

$$\mathcal{H}'_{mn} = \int \psi_m(r, R)\mathcal{H}'\psi_n(r, R)dr.$$  

(2.9)

Note that in (2.8), the diagonal term $\mathcal{H}_{nn}$ is moved to the left-hand side, hence the summation is for off-diagonal terms ($m \neq n$). In the Born adiabatic approximation, the right hand side of (2.8), i.e. the coupling of different electronic states $m$, is neglected, leaving only the first term in the expansion in Eq. (2.6) $\Psi = \chi_n\phi_n$. Thus, the equation for nuclear motion reads

$$\left[ - \frac{\hbar}{2\mu} \Delta R + U'_n(R) - E \right] \chi_n = 0,$$

(2.10)

with the effective potential for a given electronic state $n$ given by

$$U'_n(R) = E_n^0(R) + \mathcal{H}'_{nn}(R),$$

(2.11)

which includes the diagonal correction to the zero order potential, taking partly into account the coupling between the nuclear and electronic (muonic) motion. The Born-Oppenheimer approximation, on the other hand, neglects this coupling, and assumes $U'_n(R) = E_n^0(R)$, as was seen in Eq. 2.4.

The validity of the Born-Oppenheimer approximation is dependent on the smallness of the expansion parameter $\kappa \sim (m/M)^{1/4}$ where $M$ is the reduced mass of the two nuclei,
Chapter 2. Theoretical aspects of $\mu$CF

Figure 2.1: Co-ordinate system for muonic molecular ion $ab\mu$ used in the Adiabatic Representation method.

and $m$ the mass of electron or muon. The parameter $\kappa$ reflects the ratio of the amplitudes of the nuclear motion in comparison to the equilibrium internuclear distance, hence the assumption that $\kappa \ll 1$ allows one to neglect the term $\mathcal{H}'$ in (2.8), which contains derivatives of $\phi(r, R)$ with respect to $R$ [25]. Dependence of the Born approximation on $(m/M)$ is less clear and somewhat qualitative, and the accuracy depends on specific characteristics of the problem including the strength of off-diagonal coupling in Eq. 2.8.

In the case of the muonic molecule, $m/M$ is not small. For its loosely bound states, which are our main interest, the vibrational amplitudes are not small compared to the internuclear distance. Hence both the Born-Oppenheimer and the Born Adiabatic approximation fail to describe even qualitative characteristics of the muonic molecule, such as the number of the bound states [3]. As for loosely bound states, the former method gives a state bound much too deeply, while with the latter method the state is not bound at all [4].

Adiabatic Representation method

The Dubna group, led by Ponomarev, undertook extensive efforts in the 1970s to solve
the muonic three body problem with high accuracy. Their method, known as the Adiabatic Representation method, is based on the expansion of the three-body wave function (Eq. 2.12) in the basis of solutions to the two-centre problem (Eq. 2.13) describing the muonic motion in the field of two fixed nuclei:

\[ \Psi(r, R) = \sum_j \phi_j(r; R) \frac{1}{R} \chi_j(R), \] (2.12)

where \( \sum_j \) indicates summation over the discrete states and integration over continuum states, and \( r \) and \( R \) are given in the co-ordinates shown in Fig. 2.1\(^3\). The basis \( \phi_j \) is given by solving (in the units \( e = \hbar = 1 \))

\[ \left[ -\frac{1}{2m_a} \Delta_r - \frac{1}{r_a} - \frac{1}{r_b} \right] = \epsilon_j \phi_j(r; R), \] (2.13)

where the reduced mass is defined with respect to one of the nuclei \( M_a (M_a \geq M_b) \):

\[ \frac{1}{m_a} = \frac{1}{m_\mu} + \frac{1}{M_a}, \] (2.14)

such that the energy \( \epsilon \) is measured from the ground state of an isolated atom \( \mu a \). The right hand side of Eq. 2.13 is replaced by \( \frac{1}{2} k^2 \phi(R; k, R) \) for the continuum spectrum. The equation for the nuclear wave functions \( \chi_j(R) \) is given by an infinite set of coupled integro-differential\(^4\) equations:

\[ \left[ \frac{d^2}{dR^2} + \lambda - U_{ii}(R) \right] \chi_i(R) = \sum_{j \neq i} U_{ij}(R) \chi_j(R). \] (2.15)

where \( U_{ij} \) are the effective potentials due to the kinetic energy of the nuclear motion and its coupling to the muon motion, and \( \lambda \) are the eigenvalues, from which the binding energies are derived. Note that simplified notations given in Ref. [95] are shown here; for

\(^3\)Taking out the factor \( 1/R \) from the definition of \( \chi(R) \) is a standard procedure which simplifies the form of the operator.

\(^4\)Integration comes from the operator \( U_{ij} \).
the complete formulae, see a detailed review [96]. Equation 2.15 is similar to the starting point of the Born approximation derivation (Eq. 2.8), but contrary to the approximation made in Eq. 2.10, the non-adiabacity is fully taken into account by the off-diagonal coupling terms $U_{ij}$.

Ponomarev and his colleagues overcame the computational challenges (and limited computer resources at the time) and solved Eq. 2.15 via systematic expansion in the finite set of the basis functions where the continuum spectra are discretized via:

$$\sum_{j'} \sum_{k} \int dk \rightarrow \sum_{j'} \sum_{\alpha} \Delta k_{\alpha} = \sum_{n=1}^{N},$$  

(2.16)

which gives a truncated system of coupled integro-differential equations with finite number $N$. In contrast to the Born Adiabatic method, where accuracy was not well determined, the convergence of the expansion can be tested, and the solution was obtained with a controlled accuracy [96]. With a few hundred basis functions, the solution converged, giving an accuracy of $10^{-1}$ eV [97]. Thus, they showed for the first time the existence of the loosely bound state for the $dpd$ and $dpt$ molecules. In fact, for the loosely bound states, contributions from off-diagonal terms from both discrete and continuum states are significant [97], and this is why earlier adiabatic calculations failed to see such states.

**Scattering calculations with the Adiabatic Representation**

The Adiabatic Representation method can be applied to three body scattering problems. For scattering cross sections, the need for accuracy, from the point of view of $\mu$CF physics, is much less strict; the few percent level in cross sections is often sufficient (compared to the $10^{-7}$ level for the energy levels).
Effective two level approximation  Bubak and Faifman performed the first comprehensive calculations for all combinations (except for the asymmetric spin flip) of muonic atom scattering in a broad energy range [15]. They used an improved form of the two level approximation of the Adiabatic Representation method in which the first two states are used in the expansion of the three-body wave function in Eq. 2.12 \( (i.e., N=2 \) in Eq. 2.16), hence Eq. 2.15 reduced to the system of two equations coupled with off-diagonal effective potentials \( U_{12} \) and \( U_{21} \). In order to overcome the shortcomings of the Adiabatic Representation method with a small \( N \) (see Section 2.1.3), a correction was made to the reduced masses, giving an appropriate asymptotic behavior of the solutions [98]. A considerable reduction of computational complexity of using only two levels allowed crude, but very useful, estimates of various cross sections. They quoted accuracies of typically 10-20% for most cases. It should be noted that the effective two level approximation, when applied to the muonic three body bound state problem, correctly predicts the existence of loosely bound states, which conventional two level approaches have failed to find.

Multi-level calculations  An extensive effort was put together by Ponomarev and his colleagues to perform the task of truly multi-level calculations in the Adiabatic Representation [99]. The results for scattering on a bare nucleus have been published in an atlas of cross sections, referred in this thesis as the “Nuclear Atlas” [16–18]. The accuracy for the purely three-body problems \( (i.e., \) atomic and molecular effects ignored) is claimed to be \( \sim 3\% \) [24]. For a recent review of multi-level calculations and a comparison with other methods, see [100,101].

Improved Adiabatic methods

The difficulties associated with the Adiabatic Representation method have been discussed by many authors (for example, see Ref. [102]). Ponomarev summarizes its disadvantages
in treating boundary conditions [103,104]:

1. Incorrect dissociation limit of the system \((a, b, \mu)\), when it decays into \((a, \mu) + b\) and \(a + (b, \mu)\).

2. Incorrect value of momentum in the reaction channels.

3. Infinite and long-range behavior of the non-diagonal potential \(U_{ij}(R)\) as \(R \to \infty\).

Cohen and his co-workers at Los Alamos used what they call the “Improved Adiabatic” approach [105] to address some of the above limitations. In the standard Adiabatic Representation method, the Hamiltonian was separated into two parts (Eq. 2.5); the zero order term describing the two centre Coulomb problem \(\mathcal{H}_{TC}^0\), and the perturbative term for the relative motions of the nuclei as well as nuclear-muonic motion coupling \(\mathcal{H}_{AR}'\). In the asymmetric nuclear system (e.g., \(M_a > M_b\)), since infinite mass for both the nuclei is assumed in \(\mathcal{H}_{TC}^0\), the zeroth order solution cannot distinguish the channel \(a\mu\) from \(b\mu\), into the former of which the ground muonic state \((1s\sigma)\) should dissociate in the limit \(R \to \infty\) (recall the \(a\mu\) is more deeply bound than the \(b\mu\) due to the reduced mass difference). The Improved Adiabatic approach instead partitions the Hamiltonian into

\[
\mathcal{H} = \mathcal{H}_{IA}^0 + \mathcal{H}_{IA}'
\]

\[
= \{\mathcal{H}_{TC}^0 + \mathcal{H}_{mp} + \mathcal{H}_{ang}\} + \{\mathcal{H}_{sep}\},
\]

(2.17)

where the mass polarization (i.e. nuclear-muonic motion coupling) term \(\mathcal{H}_{mp}\) and the internuclear orientation term \(\mathcal{H}_{ang}\), which were part of the perturbation \(\mathcal{H}_{AR}'\) in the Adiabatic Representation, are now included in the zero order Hamiltonian. The term \(\mathcal{H}_{ang} \propto L_R^2\) acting in the nuclear centre of mass frame (as opposed to the geometric centre of nuclei frame) breaks the symmetry, and causes the zero order muon wave function to
move to the heavier nuclei as $R \to 0$ [105]. Therefore, the Improved Adiabatic method is expected to give more accurate results for the asymmetric nuclear case, compared to the standard Adiabatic Representation approximation, for the same number of basis functions $N$, hence achieving faster convergence. Of course, in the limit $N \to \infty$, both approaches should give the same results. The two level approximation of the Improved Adiabatic method has been applied by the Los Alamos group to the bound state calculations of HD$^+$ [105] and $d\mu t$ [57], as well as in the scattering calculations [19–21]. The latter cover all the possible combinations of muonic hydrogen isotope scattering, including the only calculation to date of the hyperfine transitions in asymmetric collisions.

Despite the criticisms of the Adiabatic Representation, it should be noted that the effective two level approximation used by Bubak and Faifman (p. 31) also deals with the dissociation problem within the framework of the Adiabatic Representation, and appears to give a reasonable accuracy for only $N = 2$.

### 2.1.4 Variational approaches

More accurate results for the bound state energy levels of muonic molecules have been obtained with variational calculations. Szalewicz reviewed recent progress on the variational approaches [85]. In these approaches, the exact wave function $\Psi$ for the Hamiltonian $\mathcal{H}$ is approximated by an expansion with a finite set of suitable basis functions

$$\Psi(r_1, r_2) \sim \tilde{\Psi} = \sum_{i=1}^{N} c_i \varphi_i(r_1, r_2).$$  \hspace{1cm} (2.18)

The approximate energy levels $\tilde{E}_n$, and approximate eigenfunctions $c_i \varphi_i$ are found by diagonalizing an $N \times N$ matrix. The variational principle states

$$\frac{\langle \tilde{\Psi} | \mathcal{H} | \tilde{\Psi} \rangle}{\langle \tilde{\Psi} | \tilde{\Psi} \rangle} \geq E_0, \hspace{1cm} (2.19)$$
where $E_0$ is the ground state energy, hence, all the approximate energies $\tilde{E}_n$ are upper bounds to the exact energies. The basis functions, also called trial functions, contain some non-linear parameters, which are to be optimized by repeating the diagonalization and finding $\tilde{E}_n$ with a systematic variation of the parameters.

The choice of the basis functions is a major factor which determines the accuracy and convergence of the calculations. Physical intuition and computational experience in choosing the functions can be rewarded with faster convergence and better accuracy.

**Hylleraas type basis expansion**

A popular choice are Hylleraas type functions [106-109], which for $J = 0$ states read

$$\varphi_n = (r_{\mu \nu})^{k_n}(r_{\mu \delta})^{l_n}(r_{\mu \ell})^{m_n}\exp(-\alpha_n r_{\mu \nu} - \beta_n r_{\mu \delta} - \gamma_n r_{\mu \ell}), \quad (2.20)$$

and are expressed in the interparticle co-ordinates, where $r_{xy}$ denotes the distance between the particle $x$ and $y$. The exponential parameters $\alpha_n, \beta_n, \gamma_n$ are often taken to be the same for all $n$ to reduce the number of parameters to be optimized. For loosely bound states $(1,1)$ of $d\mu d$ and $d\mu t$, similar functions with $k_i = l_i = m_i$, known as Slater gemials were found to be more useful in representing the large physical size of the states [84,110].

The disadvantage with Slater gemials is that one has to optimize a large number of exponential parameters, which is very time consuming. Another disadvantage is its "linear dependencies" problem. This is due to the fact that the basis in this set is nearly linearly dependent, i.e. rather non-orthogonal. Since the functions differ only by their exponents, an optimized basis set sometimes has several functions with close values of the exponents [85]. Thus, the use of extended arithmetic precision ($\sim 30$ decimal digits) is necessary.
Coupled rearrangement channel method

A very appealing method in calculating three body problems, known as the non-adiabatic coupled rearrangement channel method [111], was developed by Kamimura. The method has been applied to many muonic and other three-body problems [44–48, 112–115]. In his approach, the total three body-wave function is expanded in basis functions spanned over three rearrangement channels in the Jacobian co-ordinate system, which is illustrated in Fig. 2.2 for the case of the $d\mu t$ system. The method treats the muon on an equal footing with the other two nuclei. All three channels are explicitly employed with each having its own importance; since the $(J,\nu)=(1,1)$ state lies only $\sim 0.66$ eV below the $[\mu t]_{1s}−d$ ($c = 1$ in Fig. 2.2) break up threshold, and $\sim 48$ eV below the $[\mu d]_{1s}−t$ ($c = 2$) threshold\(^\text{5}\), the large components of the three body wave functions are expected to be in the $c = 1$ and $c = 2$ configurations. The $[dt]−\mu$ component ($c = 3$) is important for calculations of fusion and sticking where the details of the wave functions at small $r_3$ are needed.

\(^5\)These should be compared to, for example, the $[\mu t]_{2s,2p}−d$ threshold which is $\sim 2$ keV higher.
wave function. For each channel $c$, the basis has the form

$$\xi_c = \phi_i(r_c)\chi_I(R_c)$$

$$= r_c^{l_c} \exp \left[ -\left( \frac{r_c}{\bar{r}_{ic}} \right)^2 \right] R_c^{L_c} \exp \left[ -\left( \frac{R_c}{\bar{R}_{lc}} \right)^2 \right] \quad i = 1, n \quad I = 1, N \quad (2.21)$$

where $\bar{r}_{ic}$ and $\bar{R}_{lc}$ are the nonlinear parameters to be optimized, and $l_c, L_c$ stand for the relative angular momentum associated with co-ordinates $r_c, R_c$ respectively. The use of a Gaussian basis allowed straightforward analytical integrations in calculations of kinetic and potential energy matrix elements. Some detailed examples are given in Ref. [46]. A “faster damping” of the Gaussian tail, compared to the Hylleraas type functions with exponential tails, is not a problem even when representing a diffuse $(1,1)$ state, since the parameters $\bar{r}_{ic}$ and $\bar{R}_{lc}$ can be made much larger than the muonic molecular size.

In addition to the philosophical appeal of treating the muon on the same footing as the nuclei, as mentioned above, this method possesses some practical advantages, which include the following. First, because the basis spans three channels, linear dependencies among the basis functions is smaller, i.e. non-orthogonality is not too severe, compared to the single-channel basis function. This permitted calculations in double precision (64 bits, 14~16 decimal digits), which resulted in a very short computation time. Second, due to the explicit use of Jacobian co-ordinates with the rearrangement channels, the method is suitable for scattering calculations, where correct boundary conditions are satisfied (c.f. recall the problems with the Adiabatic Representation). This feature is also useful in describing the $d\mu t$ wave function in the calculation of molecular formation rates (see Section 2.2).

The relative contributions of each channel can be tested with this method by removing one channel from the calculations, which proves the relative importance of the channels, in the order $(c=1) > (c=2) > (c=3)$, as expected above, but also shows that to achieve the $10^{-3}$ eV level of accuracy in $\epsilon_{11}$, it is necessary to include $c = 3$. With the basis set
of \( N_b \sim 2600 \), the final accuracy of about \( 2 \times 10^{-5} \) eV was achieved, which includes the estimated uncertainty in extrapolation to \( N_b \to \infty \) [111].

Among other approaches for the three-body Coulomb problem, two of them, which have received recent attention in \( \mu \)CF theory, are briefly discussed below.

**Hyperspherical approach** Hyperspherical co-ordinates in the three body system are chosen in terms of one “hyperradius,” which measures the size of the system, and the rest of the five variables as angles. In the simplest case, two independent vectors \( \mathbf{r}_1, \mathbf{r}_2 \) (see Section 2.1.2) are replaced by

\[
\rho = \left[r_1^2 + r_2^2\right]^{1/2}, \quad \alpha = \tan^{-1} \frac{r_2}{r_1},
\]

where \( \rho \) is the hyperradius and the \( \alpha \) hyperangle. Thus we have \( (\mathbf{r}_1, \mathbf{r}_2) \to (\rho, \Omega) \), where \( \Omega = (\alpha, \theta_1, \phi_1, \theta_2, \phi_2) \) denotes five angles with \( \theta_i, \phi_i \) being the spherical angles of the two vectors. A thorough review by Lin exists for this approach [116]. He argues that the hyperspherical approach provides a unified view of the three body problem regardless of the masses of the particles involved\(^6\). When applied with the adiabatic approximations, this approach gives faster convergence than the Adiabatic Representation [39, 117, 118]. Abramov et al. discuss the physical relationship between the two approaches [119]. The hyperspherical approaches have been recently applied to excited muon transfer [41], sticking in \( d\mu t \) [120], properties of \( d\mu^3 \)He molecules [90], and the calculation of \( d\mu t \) formation rates [121].

**Faddeev equations** Faddeev equations, which provide a rigorous approach to three-body problems, have been popularly used in nuclear few-body calculations [86], yet for

\(^6\)We note that an alternative term Hyperradius approach has been suggested by Alexander Matveenko as a more general name.
purely Coulombic systems, the method had been considered impractical until recently especially for scattering problems, due to the long-range nature of the interaction [122]. Kvitsinsky, Hu and their co-workers have solved some of the difficulties, improving upon a previously proposed modification to the Faddeev equations [123], and have calculated the bound state properties, e.g., \( p\mu p \) [124] and \( d\mu t \) [125], as well as scattering and fusion in flight cross sections [26,122].

The calculations for the non-relativistic point-like three body problem have reached impressive accuracies. Some selections from the results are summarized in Table 2.1. At this level of accuracy, the results are sensitive to the variations in the physical constants used in the calculation, such as the masses of the particles, particularly that of the triton [126]. Even a relatively small change in the masses can influence the binding energies as much as 0.5 meV. Table 2.1 is compiled using the constants given in Table 1.1 on page 7, in which I have made corrections when a different mass set was used in the calculations.

### 2.1.5 Non-Coulombic corrections

At the level of accuracy required (\( \lesssim 1 \) meV), the effects beyond the Coulombic, nonrelativistic point-like description of the muonic molecules are important. Because of the heavy mass of the muon, muonic hydrogen atoms and molecules are sensitive to effects such as QED vacuum polarization and the nuclear charge form factor as well as hyperfine effects. However, it is the difference between the corrections to the atomic and molecular energy levels which affects the binding energy \( \varepsilon_{Jv} \),

\[
\Delta \varepsilon_{Jv} = \Delta E_{Jv}^{mol} - \Delta E_{1s}^{atom}. \tag{2.23}
\]
Table 2.1: Comparison of the binding energy for the \((J,v) = (1,1)\) state of \(d\mu t\), \(-\epsilon_{11}^{nt}\) (in meV).

<table>
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<tr>
<th>Authors</th>
<th>Year</th>
<th>Method</th>
<th>(N)</th>
<th>(-\epsilon_{11}^{nt})</th>
</tr>
</thead>
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<tr>
<td>Vinitskii et al. [97, 127]</td>
<td>1978-80</td>
<td>Adiabatic</td>
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<td>640</td>
</tr>
<tr>
<td>Gocheva et al. [128]</td>
<td>1985</td>
<td>Adiabatic</td>
<td>884</td>
<td>656</td>
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<td>Bhatia and Drachman [106]</td>
<td>1984</td>
<td>Hylleraas</td>
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<td>224</td>
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<tr>
<td>Frolov and Efros [129, 130]</td>
<td>1984-85</td>
<td>Slater</td>
<td>400</td>
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<tr>
<td>Hu [107]</td>
<td>1985</td>
<td>Hylleraas</td>
<td>500</td>
<td>628</td>
</tr>
<tr>
<td>Hu [108]</td>
<td>1987</td>
<td>Hylleraas</td>
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<tr>
<td>Korobov et al. [131]</td>
<td>1987</td>
<td>Elliptic</td>
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<td>Szalewicz et al. [109]</td>
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<td>Hylleraas</td>
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<tr>
<td>Kamimura et al. [111]</td>
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<td>Jacobian</td>
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<td>660.104 (30)</td>
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<tr>
<td>Haywood et al. [84]</td>
<td>1991</td>
<td>Slater</td>
<td>2600</td>
<td>660.1778(^b) (10)</td>
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</table>

\(^a\)Taken from Ref. [85], which is the value normalized to the set of masses given in Table 1.1.

\(^b\)Published value shifted by 0.0252 meV to account for the different masses used.

Because the loosely bound \((d\mu t)_{11}\) states resemble a \(\mu t\) atom and a weakly bound deuteron orbiting around it, the corrections \(\Delta E_{J\nu}^{\text{mol}}\) and \(\Delta E_{1s}^{\text{atom}}\) are similar and partly cancel each other, hence the corresponding corrections \(\Delta \epsilon_{J\nu}\) are suppressed by a factor of about \(\sim 10^2\) [132]. These corrections (without hyperfine effects) amount to about a \(-30\) meV shift (towards deeper binding) in the \(d\mu t\) binding energy \(\epsilon_{11}\) [4].

Hyperfine structure of the \(\mu t\) and \(d\mu t\) further introduces corrections for \(F = 0\) of \(+35.2\) meV (\(S = 0\)), and for \(F = 1\) of \(-8.3\) meV (\(S = 0\)), \(-14.9\) meV (\(S = 1\)), and \(-8.5\) meV (\(S = 2\)), where \(F\) is the total spin of \(\mu t\) and \(S\), that of \(d\mu t\).

### 2.2 Resonant molecular formation

In this section we shall discuss the theory of resonant muonic molecular formation. Focusing on the \(\mu t + D_2\) case, which is the subject of this thesis, the standard theory based on the Vesman model is treated first with a particular emphasis on the work by
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Faifman et al.[70-72, 133, 134], followed by the extensions to the standard theory.

2.2.1 The standard Vesman model

As mentioned earlier, resonant molecular formation refers to the process:

$$(\mu t)_F + [D_2]_{\nu_i,K_i} \rightarrow [(d\mu t)^S_{j\nu,dee}]_{\nu_f,K_f},$$  \hspace{1cm} (2.24)

where the kinetic energy of $\mu t$ and the energy released upon formation of $d\mu t$ is absorbed in the rotational ($K$) and vibrational ($\nu$) excitation of the molecular complex $[(d\mu t)dee]^*$, a hydrogen like molecule with $d\mu t$ playing the role of one of the nuclei. The process is resonant in nature because of the discrete spectra in the final states, corresponding mainly to the ro-vibrational levels $(\nu_f, K_f)^7$, and the collision energy has to match the resonant condition. The resonance condition can be written, when considering purely two body collisions (i.e., neglecting three-body or phonon effects):

$$\epsilon_{\text{res}}[\mu t + D_2] = \epsilon_{11}^{FS}[d\mu t] + E_{\nu_f,K_f}[(d\mu t)dee] - E_{\nu_i,K_i}[D_2].$$  \hspace{1cm} (2.25)

Figure 2.3 gives a schematic energy level diagram for the resonant $d\mu t$ formation process. Plotted on the left is the potential curve of $d\mu t$ showing the “shallow” (in the muonic scale) bound state, while on the right the molecular complex energy levels are plotted. Note that due to the reduced mass difference, the energy levels of $D_2$ and $[(d\mu t)dee]$ are different (by 33.7 meV for the ground states according to Faifman et al. [138]). In fact it is this difference in reduced mass which introduces the dependence of the $d\mu t$ formation rates on the target molecular species, i.e., $\mu t + D_2$ versus $\mu t + HD$ or $\mu t + DT$.

\footnote{There are recent studies on the $[(d\mu t)dee]$ energy level splitting (of a few meV level) due to the $d\mu t$ interactions with the host molecular complex including the quadrupole finite-size corrections [135-137]. This would greatly increase the number of levels in the final state spectra (hence further complicating the notation!), but it is neglected here. Due to the Doppler effect the resonance profiles in the $\mu t$ lab frame are already broad (see the discussion later in this section), so presumably these splittings would not have much effect in our measurement of epithermal molecular formation, though more care may be necessary at low energy.}
Figure 2.3: Schematic energy diagram for resonant molecular formation $\mu t + [D_2]_{K_i=0,\nu_i=0} \rightarrow [(d\mu)]_{j=1,\nu=1}^{F=0,S=0}dee]_{K_f,\nu_f}$, from Ref. [4]. The energy released in the formation of $d\mu t$ is absorbed by the ro-vibrational excitation of the host molecular complex.

Under ordinary target conditions, only $\nu_i = 0$ is populated. On the other hand, the initial rotational population depends on the target preparation procedure as well as on temperature. At 3 K, an equilibrated target would have nearly 100% $K_i = 0$ population (ortho deuterium). Our targets, however, were prepared by rapidly freezing the statistically populated deuterium (67% $K_i = 0$ and 33% $K_f = 1$) from a hot palladium filter, and because the rotational relaxation is very slow in the absence of a catalyst, we expect this population will last for the entire measurement time. The initial ortho-para
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population is relevant, because angular momentum conservation requires:

$$L + K_i = J + K_f,$$

(2.26)

where $L$ is the relative angular momentum in the reaction (2.24). Since the $d\mu t$ rotational angular momentum $J$ is 1, we have $K_f = K_i \pm 1$ for low energy collisions with $L = 0$ [139]. However, at epithermal energies $L > 0$ becomes important, allowing various $K_f$ states. This is one of the reasons why we expect such a high rate at epithermal energies.

Formulation of the formation process

Calculation of the resonant molecular formation rates is difficult since it is a rearrangement process involving six bodies. Several different groups have published the calculations [45, 70, 71, 127, 133, 139–146], but each work is often criticized by other groups, which creates for the experimentalists a situation that is confusing (and sometimes frustrating!).

For the conventional homogeneous target experiments, the resonant molecular formation rate $\lambda^{m_f}$, as a function of the target temperature, is written as [4, 147]:

$$\lambda^{m_f}(T) = N \sum_i \int de2\pi |i|H'|f > |^2 f(\epsilon, T)I(\epsilon - \epsilon_{if}, T),$$

(2.27)

where $N$ is the target density, $|i|H'|f >$ is the matrix element for molecular formation, $f(\epsilon, T)$ is the kinetic energy distribution in the center of mass for the collision at the temperature $T$ (e.g., the Maxwellian distribution if $\mu t$ is thermalized), $\epsilon_{if}$ is the resonant energy, and $I(\Delta, T)$ is the resonance intensity profile.

The popular choice of the interaction operator has been:

$$H' = d \cdot E,$$

(2.28)

where $d$ is the dipole operator of $d\mu t$ and $E$ is the electric field at the center of mass of the $d\mu t$ due both to the spectator nucleus and electrons in the molecular complex.
The importance of including the field of the electrons was pointed out by Cohen and Martin [142] and independently by Menshikov and Faifman [143]; it screens the nuclear field, substantially reducing the matrix elements.

The use of the dipole operator in $H'$ was criticized by Petrov et al. [144–146] and by Scrinzi [148], who each proposed alternative forms of the operators, but the accuracies of simplifications adopted were in turn questioned by Faifman et al. [71,134]. Faifman and his colleagues recently included the quadrupole corrections in the interaction operator in Ref. [71], and those are what we used in our analysis of the experimental data. We note that the perturbative formulation used by Faifman as well as by Petrov and by Scrinzi were criticized by Lane [141] and Wallenius [45,46]. Lane [141] also criticized the formulation of earlier work by Cohen and Leon [14]. In fact, Wallenius in his thesis goes as far as calling Menshikov and Faifman’s justification for the Born approximation “nonsense.” Wallenius concludes, however, that different “working formulae” in the literature actually end up to be equivalent (presumably due to cancellation of what he calls mistakes), and would yield the same cross sections, if no further approximations were made [46].

Thus theorists seem to agree, though maybe for different reasons, at least on the form of the starting formula they use to calculate the matrix elements. For actual evaluations, however, further approximations are necessary in the interaction operators, such as the multipole expansion as mentioned above, or in the wave functions as discussed next.

**Wave functions**

The initial and final wave functions $\psi_i, \psi_f$ in the molecular formation matrix element calculation are usually approximated as:

$$\psi_i = \phi_{1i}(r) \, D_{\alpha \beta}^{ij}(r_1, r_2, \rho_2) \, e^{ip \cdot r}, \tag{2.29}$$
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where $\psi_{13}^{\mu}(r)$ is the $\mu$ ground state wave function with $r$ being the interparticle vector from $t$ to $\mu$, and $\Psi_{\nu_{t},J_{t}}^{D_{2}}(r_{1},r_{2},\rho_{2})$ is the initial $D_{2}$ wave function with internuclear vector $\rho_{2}$ and electron vectors $r_{1},r_{2}$. The factor $e^{ip\cdot r}$ describes the relative motion of the centre of mass of the $\mu t$ and $D_{2}$.

The final wave function is written:

$$
\Psi_{f} = \psi_{11}^{d\mu}(r,R) \Psi_{\nu_{f},K_{f}}^{N\text{dec}}(r_{1},r_{2},\rho),
$$

(2.30)

where $\psi_{11}^{d\mu}(r,R)$ is the $(d\mu)_{11}$ wave function, and $\Psi_{\nu_{f},K_{f}}^{N\text{dec}}(r_{1},r_{2},\rho)$ is the final state molecular complex wave function where $d\mu$ is replaced by a fictitious particle of mass and charge equal to the sum of $d\mu$ and located at the centre of mass of $d\mu$.

Following Menshikov [149], most authors use an analytical approximation of $\psi_{11}^{d\mu}(r,R)$ with its asymptotic form valid in the limit of infinite $\mu t + d$ separation, since the main contribution to the matrix element integral comes from the region where the $\mu t + d$ separation is large. The asymptotic wave function is characterized by a normalization constant $C$ [149,150]. Recently Kino et al. [114] obtained a new value of $C$ using Kamimura’s variational wave function (Section 2.1.4, page 36), calculated with the Gaussian basis functions in the Jacobi co-ordinate. As we have discussed, Kamimura’s wave function treats the break up channel into $\mu t + d$ directly, hence it gives an accurate description of asymptotic behaviour, while overcoming the difficulty of the Gaussian basis ("fast dumping") by using a sufficiently large number of basis functions. The formation rates are proportional to the square of the constant $C$, and its new value decreased the predicted formation rates by 14% compared to an earlier calculation in Ref. [149] using the Adiabatic Representation, but increased them by 33% compared to Ref. [150] using a variational wave function with the Slater-type basis functions [110]. It is interesting to note that the Slater-type basis method, which was used so successfully in the energy level calculations (see Table 2.1), gives a less accurate description (assuming Kino et al. are
correct) of the asymptotic wave function compared even to the Adiabatic Representa-
tion method, perhaps illustrating the difficulty in achieving a unified description of the
non-adiabatic three body problem. In our analysis, the new value of $C$ by Kino et al. is
used.

Decay of the molecular complex

Once the $[(d\mu t)\text{dee}]$ complex is formed, it can either stabilize itself, which leads to fusion,
or go through a back decay [151] into the entrance channel $\mu t + D_2$:

\begin{equation}
[(d\mu t)_{J_F}^{S_F}\text{dee}]_{\nu f K_f} \\
\quad \rightarrow (\mu t)_{F} + [D_2]_{\nu f'K'_f} (\Gamma^{SF}_{\nu f',K_f';\nu f'K'_f}) (2.31)
\end{equation}

\begin{equation}
\quad \rightarrow \text{stabilization } (\tilde{\lambda}_f) (2.32)
\end{equation}

The back decay width $\Gamma^{SF}_{\nu f,K_f;\nu f'K'_f}$ is determined by the same matrix element as the forma-
tion, though it should be noted that, in general, $\nu f'K'_f \neq \nu fK_f$ (see footnote\(^8\)).

The rate for stabilization, often called the effective fusion rate $\tilde{\lambda}_f$, includes contributions from: (1) fusion from the $(d\mu t)_{11}$ state, (2) radiative decay of the $(d\mu t)_{11}$ into a
lower state, (3) Auger deexcitation of $(d\mu t)_{11}$, and (4) collisional deexcitation of the com-
plex due to interaction with the surrounding environment. Because of the centrifugal
barrier, fusion from $J = 1$ states is relatively slow ($< 10^8$ s\(^{-1}\); see Ref. [3] for a recent
summary), and Lane estimates the radiative decay of $d\mu t$ to be even slower ($\leq 10^6$ s\(^{-1}\)
[151]). Hence $\tilde{\lambda}_f$ is dominated by the Auger deexcitation rate, at least at low densities.
This rate has been calculated by several authors with an increasing degrees of sophistica-
tion, most recently by Armour et al. [152], who took into account the molecular nature of

\(^8\)This possibility of what I call the "resonant excitation" of a $D_2$ molecule by $\mu t$ is not apparent in the
papers of Faifman et al. [71,133,134]. In fact, we point out that for kinematic reasons, this occurs only
for $\nu f \geq 4$ for most $K_f$ (for all $K_f$, if the $K_f$ distribution is relaxed to low temperature equilibrium).
the host complex, and estimated the rates for \((J,v) = (1,1) \rightarrow (0,1)\) to be in the range \((6.9-10.3) \times 10^{11} \text{ s}^{-1}\), depending on the model used.

The collisional rotational de-excitation of the complex was first calculated by Ostrovskii and Ustimov [153], and later by Padial, Cohen and Walker [154]. The latter, who claim better accuracy but still neglect the ro-vibrational transitions in the target (i.e., the surrounding) molecule, found the rates substantially smaller than the former (by a factor of two to ten depending on the transition).

To our knowledge, there is no accurate calculation of collisional vibrational quenching, except a rough estimate by Lane of \(\sim 10^7 \text{ s}^{-1}\) at room temperature [151,155], which is much slower than other processes. However, he claims this rate increases drastically with target temperature (for equilibrated targets) and increasing \(\nu\) [155], rising to the order of \(10^{10} \text{ s}^{-1}\) at 2000 K, therefore it may compete with Auger decay for the molecular formation at epithermal energies with high \(\nu_f\) (see footnote\(^9\)).

In Faifman's calculations used in our analysis [71,72], the rotational relaxation rate of \(\lambda_K K_f = 10^{13} \phi \text{ s}^{-1}\), with \(\phi\) being the target density in units of LHD (liquid hydrogen density), and the effective fusion rate of \(\lambda_f = 1.27 \times 10^{12} \text{ s}^{-1}\) [156], were used. The latter is the sum of the dipole E1 Auger transition from the \((J,v) = (1,1)\) state to \((0,1), (2,0),\) or \((0,0)\) with each rate being 11.4, 1.3 and 0.02 (\(\times 10^{11} \text{ s}^{-1}\)), respectively.

**Total and effective formation rates**

For our analysis, it is convenient to express the formation rates as a function of the \(\mu t\) laboratory (lab) energy, as opposed to the target temperature given in Eq. 2.27.  

\(^9\)Padial, Cohen and Walker promised to extend their calculations to vibrational transitions [154], and we await their results eagerly.
Specifying the initial and final states, we then have\(^{10}\):

\[
\lambda_{dut}^{F}(E_{\mu t}^{lab}) = \sum_{\nu_fSK_f} \lambda_{\nu_f,K_f}^{FS},
\]

where

\[
\lambda_{\nu_f,K_f}^{FS} = \sum_{K_i} \omega_K(K_i) \lambda_{\nu_i=0,K_i,\nu_f}^{SF}
\]

\[
\lambda_{\nu_iK_i,\nu_fK_f}^{SF} = 2\pi N \gamma(E_{\mu t}^{lab},\epsilon_{res}) A_{i:f} |<i|H'|f>|^2
\]

where \(\omega_K(K_i)\) is the initial rotational population distribution, which for equilibrated targets is the standard Boltzmann distribution, and \(A_{i:f}\) is a coefficient which depends on initial and final quantum numbers. The factor \(\gamma(E,\epsilon_{res})\) is the Doppler broadening profile due to target motion and recoil, whose exact form is derived in Ref. [133], but for \(E_{\mu t}^{lab} \gg kT\) can be approximated [157] as a Gaussian distribution with the width of:

\[
\sigma_D = \sqrt{\frac{4E_{\mu t}^{lab} kT M_{\mu t}}{M_{D_2}}},
\]

In Eq. 2.35, a \(\delta\) function resonance profile was assumed (the classical Vesman model), but even with that, the formation rate in the lab frame has a distribution with non-zero width due to the above-mentioned Doppler broadening.

At epithermal energies, the transition formation matrix elements become very large, which means both the formation rate and back decay width are large, resulting in a significant probability for \(d\mu t\) not fusing but returning to the entrance channel \(\mu t + D_2\). The effective formation rate is a renormalized rate taking into account the fusion probability, as defined in Ref. [134]:

\[
\tilde{\lambda}_{dut}^{F} = \sum_{\nu_f,S,K_f} W_{\nu_f}^{SF}(K_f) \lambda_{\nu_f}^{SF},
\]

\(^{10}\)We sometimes refer to these rates as the total formation rates, as opposed to the effective rates described below.
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where

$$W_{\nu_f}^{SF} (K_f) = \frac{\tilde{\lambda}_f}{\tilde{\lambda}_f + \Gamma_{\nu_f K_f}^{SF}},$$  \hspace{1cm} (2.38)$$

$$\Gamma_{\nu_f K_f}^{SF} = \sum_{\nu'_f K'_f} \Gamma_{\nu'_f K'_f}^{SF},$$  \hspace{1cm} (2.39)$$

For a high density ($\phi \gtrsim 0.1$) target such as ours, Faifman assumes complete rotational relaxation of the $K_f$ levels, hence dropping the $K_f$ dependence,

$$\lambda_f = \sum_{\nu_f, S} W_{\nu_f}^{SF} \lambda_{\nu_f K_f}^{SF},$$  \hspace{1cm} (2.40)$$

$$W_{\nu_f}^{SF} = \frac{\tilde{\lambda}_f}{\tilde{\lambda}_f + \Gamma_{\nu_f}^{SF}},$$  \hspace{1cm} (2.41)$$

$$\Gamma_{\nu_f}^{SF} = \sum_{K_f} \omega_B (K_f) \Gamma_{\nu_f K_f}^{SF},$$  \hspace{1cm} (2.42)$$

where $\omega_B (K_f)$ is the Boltzmann distribution of the $K_f$ states$^{11}$.

The effective fusion probability for the molecular complex is defined as [133]:

$$W^F = \frac{\tilde{\lambda}_f^{F} + \lambda_{\nu_f}^{nr}}{\lambda_{dut}^{F} + \lambda_{\nu_f}^{nr}},$$  \hspace{1cm} (2.43)$$

where $\lambda_{\nu_f}^{nr}$ is the rate for non-resonant formation, which does not back-decay. For $\tilde{\lambda}_f^{dut} \gg \lambda_{\nu_f}^{nr}$, $W^F$ can be written:

$$W^F = \frac{\sum_{\nu_f, S, K_f} W_{\nu_f}^{SF} (K_f) \lambda_{\nu_f K_f}^{SF}}{\sum_{\nu_f, S, K_f} \lambda_{\nu_f K_f}^{SF}},$$  \hspace{1cm} (2.44)$$

which can be understood as the average of fusion probabilities from each state weighted by the formation rate of that state.

Figure 2.4 shows the molecular formation rates $\lambda_{dut}^{F} (E)$ and effective fusion probabilities $W^F$ for $K_i = 0$ (ortho) and $K_i = 1$ (para) cases, calculated by Faifman et al. [70–72]

$^{11}$In Ref. [71,134], the dependence on $S$ in the fusion probabilities $W_{\nu_f}^{SF} (K_f)$ (Eq. 2.38) and $W_{\nu_f}^{SF}$ (Eq. 2.41) is not explicitly indicated, but they do indeed depend on $S$. 
Figure 2.4: Formation rates $\lambda_{d_{\mu t}}^F$ for $\mu t + D_2 \rightarrow [(d\mu t)dee]^*$ (top) and the fusion probability $W^F$ (bottom), calculated by Faifman [70–72] for 3 K. Also shown in dashed lines are the effective rate $\tilde{\lambda}_{d_{\mu t}}^F \sim \lambda_{d_{\mu t}}^F W^F$. The rates are normalized to liquid hydrogen density.

for a 3 K target. Also shown are the effective rates $\tilde{\lambda}_{d_{\mu t}}^F \approx W^F \lambda_{d_{\mu t}}^F$. Note that $\lambda_{d_{\mu t}}^{nr} \lesssim 10^7$ s$^{-1}$ is invisible in the scale plotted.
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2.2.2 Subthreshold resonances

Rather constant behaviour of the formation rate $\lambda_m(T)$ is observed by experiments, especially at low temperature [62,65], together with an unexpected density dependence of the formation rate [63]. This led theorists to consider extensions of the classical Vesman mechanism of resonant molecular formation which assumed an isolated two-body collision and the $\delta$ function resonance profile, as adopted in Refs. [71,72,133]. Recall that the Vesman model was very successful in the $d\mu d$ case [12]. In the $d\mu t$ case, however, strong resonance levels are expected to exist just below the $\mu t + d$ threshold, e.g., $K_i \rightarrow K_f = 0 \rightarrow 1$ at $-14.0$ meV, $0 \rightarrow 2$ at $-4.3$ meV, and $1 \rightarrow 2$ at $-11.7$ meV for $F = 0$ [4]. These transitions have large matrix elements because of the strong overlap of the wave functions.

Two main mechanisms to access these subthreshold (i.e., negative energy) resonances are: (1) intrinsic resonance width due to the finite lifetime of the molecular complex (mainly the Auger decay width), and (2) three-body or many-body collisions in which the other body (or bodies) absorbs the excess energy. The mechanism (1) is density independent, but (2) depends on the surrounding environment.

There have been many attempts to treat the subthreshold resonances [144,158–163], however, complete understanding has not yet been achieved. For example, the use of the Breit-Wigner profile adopted in Ref. [158] was criticized in Ref. [160,161] at least for the high density situation. Despite these criticisms, Petrov's calculation for the formation rate for $\lambda_{d\mu t}^{F=0}$ at low temperature and low density [145,146] seems to agree with the value suggested by the PSI measurement ($130 \pm 20$ $\mu$s) [22]. Armour recently proposed a new approach beyond the Born approximation [164,165], which also predicts non-zero width resonance profiles, but its application is still limited [121] and comparison with experiment is not yet possible.
We note, however, that for epithermal molecular formation, the resonant widths considered in these models are smaller than the Doppler broadening; thus the effects are negligible.

2.2.3 Condensed matter effects

An important advance was the observation of a striking condensed matter effect in solid D\(_2\) [77,166], where we measured an unexpectedly high \(d\mu d\) formation rate. This stimulated the theoretical efforts in \(\mu CF\) to be extended to solid state physics. In addition to the study of thermalization processes [167], investigation of molecular formation in solids has been started by several authors [167–170].

Condensed matter effects, important mainly at collision energies comparable to or below the Debye temperature of hydrogen \((k\Theta_B \approx 10\) meV\)), include the reduced mass effect which shifts the resonance energies in the lab frame, and phonon assisted resonant molecular formation. The latter is similar to the three-body collision mentioned in the previous section, but in this case it is one or more phonons which carry away the excess energy.

Fukushima made the first and so far only calculation of \(d\mu t\) formation in solid hydrogen [168], but he considered only metallic hydrogen targets at extremely high pressure in order to avoid the quantum crystal nature (involving large non-harmonic lattice vibrations) of solid hydrogen at zero pressure. Therefore its applicability to our experiments is questionable even at an order of magnitude level. Menshikov and Filchenkov claimed that our measured \(d\mu d\) formation rate [77,166] could be explained by the phonon assisted resonant formation alone, but again because of the lack of proper treatment of the phonon spectrum, the reliability of their calculation is rather unclear\(^{12}\). Adamczak

\(^{12}\)Note that their conclusion contradicts at least partly Adamczak’s slow thermalization model [167], so they cannot both be right.
has recently reported the first realistic calculations of resonant molecular formation in a solid for the $d\mu d$ case [170], extending his work on muonic atom thermalization in solid targets [167]. Unfortunately, there are no realistic calculations for $d\mu t$ formation in the solid state available to date. We further note that the role of condensed matter effects in the ro-vibrational relaxation of the molecular complex (perhaps involving rotons or vibrons), which affects the fusion probability $W^F$, is an open question.

Finally, let us note that the use of renormalized effective rates $\tilde{\lambda}_{d\mu t}^F$ (Eq. 2.37–2.42), which takes into account the fusion probability, needs very careful consideration as to its applicability. In the case of epithermal molecular formation, the use of the renormalized rates in the Monte Carlo calculations, as was done in Refs. [27, 82] would result in a significant overestimate of the fusion yield [171]. We shall give a detailed discussion on the use of effective rates in Appendix B (Section B.1).

I shall come back to some of the theoretical details when we discuss our results. In the chapters that follow, we shall see what we can contribute to the understanding of the rich physics described in this chapter, involving resonant molecular formation as well as the muonic few body problem. We shall start in the next chapter with a description of our experimental apparatus, which makes these measurements possible.
Chapter 3

Experimental Apparatus

The experiment described here has been conducted at the M20B channel of TRIUMF (Canada's national meson facility) located on the University of British Columbia campus in Vancouver, Canada. A novel target system for films of solid hydrogen isotopes and other gases was developed, and detectors which take advantage of the thin film targets have been utilized. In this chapter, we describe the muon beam, the cryogenic target system including target characterization, and detection systems.

3.1 Muon beam

TRIUMF is one of the two remaining meson factories in the world, providing intense continuous beams of protons, pions and muons. The TRIUMF accelerator is a six-sector focusing cyclotron, which accelerates H⁻ ions. By stripping the two electrons with a thin foil, protons are extracted at energies between 183 and 520 MeV with an intensity up to 140 μA. The primary proton beam usually has an RF time structure of 23 MHz¹. The cyclotron and the beam lines are shown in Fig. 3.1. Two main beam lines are BL4, primarily for proton experiments, and BL1, mainly for pion and muon experiments. In addition, BL2A will be used for the ISAC radioactive nuclear beam facility now under construction.

¹It is, however, possible to change this structure. For a measurement of pion lifetime, for example, the beam cycle was reduced by a factor of five [172].
Figure 3.1: Schematic site drawing of TRIUMF facilities and beam lines.
The muon beam at the M20B channel is obtained from primary protons striking the meson production target 1AT2, typically water cooled beryllium 10 cm thickness and 5 mm × 15 mm cross sectional area (Fig. 3.2). A cloud of pions produced near the surface of 1AT2 decays into muons and neutrinos.

For our experiment, negative muons of momentum about 27 MeV/c were selected by the momentum analyzing magnet (B1). The electron separator, a crossed electric and magnetic field, selected muons, improving the muon to electron ratio in the beam by more than two orders of magnitude. Final focusing of the beam was done by a set of quadrupole magnets, resulting in a beam size of about 30 mm (FWHM) with a rate of order $5 \times 10^3$ s$^{-1}$.

Figure 3.2: Layout of the M20B muon channel.
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3.2 Cryogenic target system

A cryogenic target system for solid hydrogen thin films, developed and improved over several years at TRIUMF, was used in our Muonic Hydrogen research program. It is this target system which allows us to perform unique experiments including the one described in this thesis.

The TRIUMF target system is capable of making two separate solid layers of hydrogen isotopes (protium $^1$H$_2$, deuterium, tritium and their mixtures), and other gases such as neon, which face each other. The thicknesses range from a few $\mu$g-cm$^{-2}$ ($\sim$1 $\mu$m for protium) to a few mg-cm$^{-2}$, and can be controlled to better than 1% relative accuracy.

The system consisted roughly of a gas mixing system, a gas deposition system, a cryogenic system, a vacuum system and a tritium safety system. The details of the target system are given in Refs. [173,174]. A general description of each part is given below.

3.2.1 Gas handling and mixing

Hydrogen isotope mixtures and other gases were prepared by a gas handling system, and introduced to the experimental vacuum space (EVS). The topology of the EVS and gas handling systems is shown in Fig. 3.3.

Highly isotopically pure protium, generated by electro-decomposition of deuterium depleted water, was passed through a palladium filter heated to a few hundred degrees Celsius, providing protium gas with an impurity level of a few ppm or less, necessary because of possible muon transfer to heavier nuclei. Similarly, research grade deuterium gas (purity $>$ 99.99%) was passed through another palladium filter to remove non-hydrogenic impurities. A small amount of protium contamination in deuterium could be tolerated,
Figure 3.3: Topology of important parts of the experimental vacuum and the gas handling systems. The gases were purified and mixed in the gas handling system on the right, then transferred to the experimental vacuum system where they were condensed. After measurements were completed, tritiated targets were pumped into a uranium storage bed, while subsequent pumping was provided by a turbo molecular pump (TMP), backed by a spiral pump (SBP) and a bellows pump (MDP). Important valves (VAs) were air-controlled, and various gauges (CGs and IG) monitored pressures.

since muon transfer to protons from other nuclei is kinematically forbidden at the energies at which we are working. A total inventory of 200 \(Ci\) (22.5 mg) of tritium gas\(^2\) was supplied by Ontario Hydro, and stored in a 5 g uranium tritium getter bed. By heating the tritium bed to the temperature 200–400° C, the gas was released into a buffer volume.

\(^2\)1 cm\(^3\) of tritium at 1 bar and 273 K contains 2.38 \(Ci\) of activity.
When a mixture of gases was used, its isotopic concentrations were determined by
the volume of the buffer and its pressure. The gases were mixed in a volume which was
connected to the experimental vacuum region via gas transfer lines. The gas mixing took
place after each gas was passed through the palladium filter, hence a protium-tritium
mixture, for example, is expected to be a mixture of H\textsubscript{2} and T\textsubscript{2}, but not HT molecules.
Before starting a gas deposition, we waited typically for a few hours so that gas mixing
was complete. Gas composition was monitored also by a residual gas analyzer (RGA),
sometimes called a quadrupole mass spectrometer.

3.2.2 Gas deposition

In the experimental vacuum space, the gases were injected onto a cold substrate via a gas
diffusion mechanism called a diffuser. The details of the diffuser are shown in Fig. 3.4.
By independently diffusing gas on each side of the diffuser, the system was capable of
making two films which faced each other.

Different gas deposition systems have been developed to incorporate experimental
demands, and to improve the performance [173,174]. For system 1, the original design,
the gas was released through a diffuser made of a thin stainless steel foil perforated with
many holes (~0.2 mm diameter). System 2 used the same diffuser foil, but as a result of
gas line modifications to incorporate tritium compatibility, it had gas inlet tubing with a
higher volume, which acted as an unwanted buffer volume as we shall see in section 3.3.2.
System 3 is the latest version with the gas inlet lines replaced with low volume, high
conductance tubing, and a sintered metal diffuser (2 \mu m porosity) employed to ensure
microscopic homogeneity of deposition. For the present work, systems 2 and 3 were used
for Run Series I and II, respectively.

The target substrates were made of 50 \mu m thick gold foil, and cooled to about 3 K
with a helium flow cryostat. The cryostat, designed by a company specializing in custom refrigerators\(^3\), was based on a standard unit\(^4\). The helium flow line was extended to suppress the thermal conductivity between the mounting flange and the cold stage, and was surrounded by a buffer space (Tritium Barrier Space, TBS) for safety purposes as discussed below.

The heat shield was cooled below 100 K, and the diffuser kept below 150 K to reduce radiation heating of the target. Two flexible copper braids (detail K in Fig. 3.4) connected the diffuser and the radiation shield to provide a flexible thermal conduction path, allowing the diffuser to move vertically.

The temperatures of different parts of the cryogenic system were monitored. Constant current silicon diode sensors permitted wide ranges of temperature readings at heat shields and diffuser. The coldest part of the cryostat, on the other hand, was measured by carbon glass resistors which provided a sensitivity necessary for fine control for helium flow optimization and depositions.

### 3.2.3 Experimental vacuum space

Figure 3.5 illustrates the central part of experimental vacuum space (EVS). The diffuser (detail L) moved vertically into the space between the upstream (US) and downstream (DS) target supports foils (detail F), and was retracted during the muon beam measurements. The muon beam, defined by a 250 \(\mu m\) plastic scintillator (T1), entered from the front port through a thin (25 \(\mu m\)) stainless steel window, which isolated the high-vacuum target region from the lower vacuum of the beam line and muon production target area. This also prevented tritium contamination of the rest of the beam lines. The beam then went through the upstream heat shield (consisting of a pair of 6 \(\mu m\) gold plated copper

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\(^3\)Quantum Technology Co., 1370, Alpha Lake Road, Unit 15, Whistler, B.C. Canada.
\(^4\)SuperTran, Janis Research Company, Inc. Wilmington MA, 01887-0090.
foil) and the US target support (50 μm gold foil), before stopping in the hydrogen layers. Almost all muons which penetrated the hydrogen layers stopped in the DS target support foil (also 50 μm gold), and did not reach the downstream heat shield (1.6 mm gold plated copper plate).

Different kinds of detectors were placed at the remaining ports of the EVS (see Section 3.4). When the MWPCs were used, a thin side window (50 μm) was necessary to reduce multiple scattering of electrons. For neutron and gamma detection, thicker side windows could be tolerated. Silicon detectors were placed in vacuum, held on the heat shield, viewing directly the target without any window in between. This turned out to be a powerful feature of our experiment, allowing the detection of low energy charged particles with a high resolution. The distance between the US and DS gold foils (detail F) was adjustable, and was 42.5 mm for Run Series I, and 17.9 mm for Run series II.

3.2.4 Vacuum system

The experimental vacuum space was pumped with a turbo-molecular pump, backed by a spiral pump and a bellows pump, all of which were free of oil and elastomers. Most of the other vacuum components were made also with elastomer free materials, in order to reduce activation by the tritium. The system typically achieved a $10^{-7}$ torr level of vacuum at room temperature, with the main residual being water vapor and hydrogen; when cooled to 3 K, cryopumping reduced the pressure to about $10^{-9}$ torr. Figure 3.6 illustrates the vacuum system.

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5 TurboVac 340M, Leybold Vacuum Products Inc. D5000 Cologne 51, Germany.
6 The contamination takes place through exchange reactions of tritium with any compound containing hydrogen.
Figure 3.4: A view of the diffuser system showing the central diffuser support (L), gas supply lines (M), diffuser chamber (N), mechanical support (O), bellows and vertical motion guide shafts (P, Q), and diffuser cooling copper braid connected the heat shield (K). The figure only shows one side of the diffuser; the gas can be deposited from the other side as well.
Figure 3.5: A perspective view of the central part of the experimental vacuum system (EVS) with a side window and a panel open (side panels normally hold Si detectors; see Fig. 3.18). Shown are the thermal heat shield (I), gold target support foils (F), the retractable diffuser (L) and its guide rails (J).
Figure 3.6: A pictorial view of vacuum system, roughly to scale. For detail of the experimental vacuum space, see Fig. 3.5. The description of the cryostat is given in Section 3.2.2.
3.2.5 Tritium safety system

Of the total tritium inventory of 200 Ci, no more than about 10 Ci was present in the target vacuum at any time. This is considerably smaller than other dt fusion experiments, which typically use up to 100 kCi of tritium. Also because of low permeability at low temperature, tritium permeation through target walls and windows is less of concern in our normal operating conditions\(^7\). Thus, the use of the muonic atom emission mechanism from the cryogenic target allowed an experiment with less stringent safety requirements. Nevertheless, various precautions have been implemented in order to ensure the safety of the personnel and to avoid incidents.

The entire target system was contained in safety enclosures, from which the air was continuously exhausted to a roof vent through a fan at a rate of 50 m\(^3\)/min. This ensured that tritium was diluted with air, and exhausted from the enclosures in case of any tritium leak from the system. Necessary access to the inside of the enclosures, such as valve operations, were done through glove panels. The tritium level was monitored with tritium monitors (TM) at different places, i.e., in the exhaust stack, in the room air in the experimental area, and at specific places in the enclosures during particular operations. See Fig. 3.7 for the schematic representation of the safety enclosure and the positions of the tritium monitors.

The helium flow line for the cryostat was doubly isolated from the EVS and tritium target region with an intermediate space (Tritium Barrier Space, or TBS) in between, reducing the possibility of contaminating the helium recovery system\(^8\). The TBS was evacuated and its pressure monitored, separately from the main vacuum system.

The readings of the tritium monitors and the target vacuum ionization gauge were

\(^7\)Tritium permeation through target walls and windows can be significant sometimes even at room temperature [175].

\(^8\)At TRIUMF, helium gas is collected for recycling after use.
Figure 3.7: Schematic representation of the safety enclosure, showing position of tritium air monitors (TM), air inlets, glove panels, and the air flow direction.
taken into a programmable logic controller to provide an interlock system which con­
trolled important pneumatic valves and tritium supply systems. It not only prevented
operational errors by not allowing certain operations under particular conditions, but also
automatically implemented basic protective measures in case of emergency. In addition,
various alarm signals, such as target temperature, ventilation air flow and air pressure to
the pneumatic valves, were sent to the experimental counting room to give early warning
of unusual conditions.

3.2.6 Operation

The amount of gas injected into the system was measured in units of Torr-litre (abbrevi­
ated T·l), where one T·l corresponds to the number of molecules in one litre of gas
at a pressure of 1 Torr and ambient temperature (~295 K). This unit was operationally
convenient since the number of molecules can be compared, independent of the isotopic
composition.

Depositing targets required some experience in fine handling of the metering valve
which controlled the gas release onto the cold foil. The target was deposited typically
at a rate of the order of a few (T·l) · s⁻¹ or less in the steady condition, the limit
imposed by the cryostat cooling power, as well as by the requirement to avoid significant
intermolecular interactions which would lead to heat conduction from the relatively hot
(~100 K) diffuser to the cold plate (~3 K). Close monitoring of the temperature and
the pressure and quick response to any condition change during possibly several hours of
target deposition was necessary in order to avoid losing the target.

The cryogenic system consumed less than 3 l/hour of liquid helium in normal condi­
tions, hence 6 days of beam experiments were possible with a 500 l helium dewar,
conveniently fitting the beam schedule at TRIUMF where beam stops once a week for
accelerator maintenance. Since changing the dewar requires warming up of the system to nearly room temperature, it would cost many hours of beam time, if it could not have been done during the maintenance day.

The tritium-containing target film, once measurements were finished, was removed by stopping the cryostat helium flow, and the evaporated gas was mostly captured by the recovery getter bed, similar to the supply one in function, but significantly larger in volume. After the pressure went down to a few mTorr level, the remaining gas was pumped via the turbo pump into a 100 l waste volume to provide a closed cycle pumping system. After beam experiments were finished at the end of several weeks of running, the content of the waste volume was very slowly released into the air exhaust, to be diluted greatly with the air, so that the monitored tritium level would always remain much below the allowed limit.

Modifications and changes to the target system, such as installation of the calibration setup described below, were performed in a separate enclosure, where personnel safety was ensured. The weekly urine samples of the operators working in a possibly contaminated environment were tested by the TRIUMF Safety Group during the period of the operations, but no sign of the contamination has been found to date. To prepare for a potential contaminating incident, a dehydrating substance was stored in the experimental counting room during experimental periods in order to facilitate timely discharge of the radioactivity.

3.3 Target characterization

In our experiments using thin solidified gas films as targets, characterization of the target films is often important in the analysis of the experimental data. For cross section measurements, in particular, the accurate knowledge of the thickness and uniformity of
the target films is essential, since the uncertainty in the thickness directly propagates to the final results. This section gives a description of the thickness calibration method and the results under various conditions, together with comparisons to Monte Carlo simulations of the gas deposition mechanism.

3.3.1 Characterization method

Due to the spatial and cryogenic limitations, conventional methods for thin film thickness measurements, such as optical interferometry, cannot be used with our system.

Thickness measurements of condensed gases have been reported by several authors. Sørensen et al. used a quartz crystal oscillator to measure solid hydrogen film thicknesses [176]. However, the method suffered from a severe non-linearity, which was attributed to the low density of hydrogen. Rutherford Backscattering (RBS) [177] which was used, for example, by Chu et al. [178] to measure the thickness of solid argon, oxygen, and CO₂, is kinematically impossible for protium targets.

We have used the energy loss of α particles traversing the film to measure the thickness [179–181]. Uniformity was determined by measuring the thickness at different positions with an array of sources.

Characterization Setup

Figure 3.8 shows a schematic view of the experimental setup for the target characterization. A linear array of five alpha spot sources was custom-manufactured⁹ by electrodeposition of ²⁴¹Am onto a gold-plated oxygen-free copper plate. The spot sources had nominal diameters of 3 mm, a center-to-center separation of 10 mm, and were covered by a thin gold layer (∼200 μg·cm⁻²) for safety and ease of handling.

⁹Isotope Products Laboratories, 1800 N. Keystone St., Burbank, CA, USA.
Figure 3.8: Schematic view of the target characterization setup. The upstream target support foil (detail F in Fig. 3.5) was replaced by a gold plated copper plate onto which $^{241}$Am sources were implanted (shown as Cold Plate in this figure). A Si detector was mounted on top of the diffuser for this measurement to directly view the target film (normally Si detectors were mounted in the side panels. See Fig. 3.18).
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As in the beam experiments, the target support plate, enclosed in an evacuated chamber, was cooled to approximately 3 K, and hydrogen gas solidified onto it when introduced through the diffuser. All of the different gas deposition systems described in Section 3.2.2 were tested for the calibration measurements, including System 1 not actually used in the experiments for this thesis, but the results are given here for completeness.

The adjustable distance between the diffuser surface and the target plate was set to about 14 mm for System 1 and 2, and about 8 mm for System 3 for the present calibration measurements.

Alpha particles penetrating the hydrogen film were detected by a passivated, implanted planar silicon detector\textsuperscript{10} of active thickness 150 $\mu$m and area 600 mm$^2$. The detector was mounted on the diffuser frame which was part of a mechanism that allowed the diffuser to be inserted and retracted (Fig. 3.8). The detector thus moved vertically to allow a measurement of the thickness at different positions by detecting the $\alpha$ particles from each of the five spot sources. A collimating device which consisted of an array of small holes (diameter $\sim$1 mm) restricted the angular path of the $\alpha$ particles to accept alphas from only one spot source at a time. The signal from the detector was recorded with a standard spectroscopy system consisting of a charge sensitive pre-amplifier, linear amplifier, and analog-to-digital converter, together with a CAMAC/VAXstation data acquisition system, a system similar to the one described in Section 3.4.4. The energy scale of the alpha detection system was calibrated using a separate $^{241}\text{Am}$ source. Calibration was frequently required since temperature variations in the detector could cause significant shifts in the gain. The system achieved a typical resolution of 0.4% (FWHM) at a detector temperature near 100 K. The profile of alpha counts versus the vertical position of the collimated detector for a bare target (i.e. no solid hydrogen layer) is shown in Fig. 3.9. The plot confirms that we detected $\alpha$ particles from only one source spot at a time.

\textsuperscript{10}Canberra, model FD/S-600-29-150-RM.
Figure 3.9: Counts versus vertical position of the silicon detector with respect to the center. Each peak corresponds to one of the five source spots. The detector is collimated such that it sees only one spot at a time.

**Thickness Determination**

Figure 3.10 shows an example of the energy spectra of $\alpha$ particles penetrating hydrogen films with different amounts of gas injected, namely 0, 150, and 300 T\cdot l. The shift of the peaks to lower energy with increased injected gas is clearly visible. Note also the peak broadening (due mainly to straggling) and the asymmetric peak shape which is due in part to the energy loss in the protective gold layer on the source. We determine the mean energy value $< E >$ from the centroid of the energy distribution $f(E)$ in the spectrum
Figure 3.10: Alpha particle energy spectra for the central source spot with different thicknesses of hydrogen film, where the peaks are normalized to the same number of counts. The numbers above each peak indicate the amount of hydrogen gas injected.

via

\[
\langle E \rangle = \frac{\int_{\langle E \rangle - \epsilon}^{\langle E \rangle + \epsilon} f(E)E\,dE}{\int_{\langle E \rangle - \epsilon}^{\langle E \rangle + \epsilon} f(E)\,dE},
\]

where \( \epsilon \) is a finite cutoff value. The use of the centroid in the analysis achieves sufficient accuracy while avoiding the difficulties in fitting the irregular peak shapes, which vary depending on the source spot and target thickness.

In the approximation that the angular dispersion of \( \alpha \) particles is avoided due to the use of a collimator, the thickness of the target \( T \) can be obtained from \( R(\langle E \rangle) \), the alpha range as a function of energy:

\[
T = R(\langle E_{\text{init}} \rangle) - R(\langle E_{\text{fin}} \rangle),
\]
where $<E_{\text{init}}>$ is the initial energy of the $\alpha$ particles and $<E_{\text{fin}}>$, the energy after traversing the target.

For our analysis, the stopping power or the range in the solid state of hydrogen for $\alpha$ particles in the energy range of $\sim$2–5 MeV was needed, but no experimental data is available for solid hydrogen at these energies. The detailed discussion of the effect of physical phase on stopping power for heavy charged particles, which is reported for keV projectiles in hydrogen [182] and in nitrogen [183], as well as for MeV ions in organic and other materials [184–186], can be found in Ref. [179] (see also reviews [187–189]). After a critical survey of the literature [179], a recent compilation for gaseous hydrogen by the International Commission of Radiation Units and Measurements (ICRU) [190] was used for the conversion.

**Uncertainties**

The systematic uncertainties considered here include knowledge of stopping power, the effect of energy cuts, and energy calibration of the detector. The uncertainty in the stopping power is difficult to estimate, since neither experimental data nor satisfactory theory is available for our case. The ICRU table [190] claims an accuracy of $\sim$1-2% at 4 MeV and $\sim$2-5% at 1 MeV for gaseous hydrogen.

Ziegler et al., in another commonly used compilation [191], give an estimate of stopping power of solids for which there are no data available, by interpolating (or, for hydrogen, extrapolating) the data from other elements under certain assumptions. They quote a 5% average accuracy for $\alpha$ particle stopping powers in solids, which is simply the average of deviations taken from the collections of experimental data in the literature for many elements.

The two tables ([190] and [191]) agree with each other within 3% at $\sim$ 2 MeV, and the thicknesses derived using both tables agree within 2% (the differences in stopping powers
partly cancel with one another upon integration over energy). We shall conservatively quote 5% of the derived thickness as the uncertainty due to the stopping power, including a possible physical phase effect. This was the limit on our accuracy in most cases.

The effect of the finite cutoff values (\(\epsilon\) in Eq. 3.1) in determining the centroid of the energy spectra was investigated by changing the cuts and its uncertainty was found to be small compared to that from the stopping powers in most cases. The uncertainty in the detector energy calibration is estimated to be less than 2 keV, except for a few measurements during which a temperature variation in the silicon detector resulted in a gain shift.

These systematic uncertainties, as well as the statistical uncertainties, were added in quadrature to obtain the total uncertainty in the thickness. Except for very thin layers, whose thicknesses had to be determined from a small difference in the initial and final alpha energies, the knowledge of the stopping power dominated the uncertainty.

### 3.3.2 Film thickness and uniformity

**Linearity of film deposition**

The linearity of the deposition was tested by measuring the thickness of films made with different amounts of gas input under different conditions. The data shown in Fig. 3.11 were taken at the central source position (0 mm) for films made by diffuser system 1. Some of the films (40, 300 T-l) were made by depositing gas on top of existing films, whereas others (20, 150, 400 T-l) were made with a single deposition. The solid line represents a weighted least-squares fit to the data (plotted with error bars\(^{11}\)). The slope represents the conversion factor from gas input to the film thickness at this source position (0 mm). A possible non-linear component in deposition was investigated by allowing a

\(^{11}\)Error bars presented in this section represent total uncertainties including that from stopping power, unless otherwise stated.
Figure 3.11: Test of the linearity of deposition. The solid line represents a least-squares fit to the data points plotted with error bars.

quadratic term in the fitting function. The maximum allowable non-linear contribution, when extrapolated to a 1000 T·l target, was similar in size to the uncertainty in the thickness due to the stopping power. Similarly, the data from other source positions on the plate, and with diffuser systems 2 and 3, showed good linearity for these moderate thicknesses.

The extrapolation of the results to very thin films requires some caution; if a small amount of the gas remained in the gas transfer tubes, it would be lost from the layer, giving a small offset in gas deposition. This gas loss is negligible for thick layers, but can be important for thin layers. The effect was examined for systems 2 and 3 by comparing two series of measurements; (1) thick films made by a single deposition of a large amount of input gas, in which the gas loss is negligible, and (2) thin films made by sequential depositions of small amounts of gas, where the gas loss from each deposition, if it exists,
is multiplied to give a measurable effect after several depositions.

Due to the small energy loss, measurement of very thin films was difficult, and we have assumed that the same cutoff value, $\epsilon$ in Eq. 3.1, can be used to determine both the initial and the final energies (this is not generally valid in the thick film measurement due to the peak broadening). Provided that the same value is used for both initial and final spectra, the choice of $\epsilon$ did not affect the resulting thickness values.

Figures 12(a) and 12(b) show the results of the comparisons for systems 2 and 3 respectively. For system 2 (Fig. 12(a)), sequential deposition of small amounts of gas (measurement (2), filled squares) resulted in a smaller thickness per unit gas input than thick films made with a single deposition (measurement (1), open squares), indicating that when making very thin films a non-negligible amount of the gas remains in the gas transfer tubes without being deposited. On the other hand, System 3 (Fig. 12(b)), which was in fact designed to remove the effect, shows no evidence of such gas loss.

**Thickness Profile**

Shown in Fig. 3.13 are the thickness profiles from different deposition systems. An asymmetric non-uniformity with respect to the center can be observed. The error bars do not include the uncertainty from the stopping power, since it is not relevant when considering relative uniformity.

One may assume that the thickness of a film at a particular position depends only on the relative distance from the diffuser, which is the case if, for example, the molecules stick onto the cold plate at the first contact as suggested from the measurements described in Section 3.3.2.

A series of measurements were made using system 3 with films that were deposited with the diffuser displaced by 0 mm, +5 mm, +10 mm and -20 mm from the nominal
Figure 3.12: Sequential deposition of very thin films shown as filled squares is compared to the thick film deposition shown as open squares for different systems: (a) system 2, where the difference in slopes between the two series of depositions indicates the gas loss effect, and (b) system 3, showing no evidence for such an effect. Inserted boxes in the figures illustrate details at small thicknesses.
Figure 3.13: Thickness profiles for different diffuser systems. Thickness per unit gas input is plotted against the source positions. Because these are relative measurements, the error bars do not include the uncertainty from the stopping power.

standard position, the positive direction being upward. In Fig. 3.14, the resulting thicknesses are plotted against diffuser coordinates, i.e., the relative vertical distance from the center of the diffuser, unlike Fig. 3.13 which was plotted against the distance from the center of the source plate. The data from different films, thus plotted, are consistent, indicating the translational invariance of the film profile under the diffuser displacement. One exception is the filled star point at distance 0 mm in the diffuser coordinates, which represents the thickness at the center of the diffuser for a film deposited with the diffuser retracted 20 mm downwards. At this position the diffuser, which has an active diameter of 60 mm, was aligned with the bottom source spot (see Fig. 3.8) and a significant fraction of emitted gas molecules missed the cold plate, hence the molecules may bounce
around inside the vacuum system, eventually sticking nearer to the bottom rather than the top of the cold plate. Nevertheless, validity of the translational invariance indicated above suggests that it is possible to measure a thickness at an arbitrary point along the vertical axis with the present technique, despite the fact that α particles are emitted only from discrete source spots. Figure 3.14 can thus be considered as a good representation of the thickness profile of the film made with system 3. Based on these results, the calibration factors (thickness per unit gas input) for system 3 at each position with respect to the diffuser center, including the 5% uncertainty from the stopping power, are given in Table 3.1. When two or more measurements exist for the same position, an average was taken, except that some points from the diffuser position of -20 mm, which may be
Table 3.1: Protium film thicknesses at various distance from the center of the film for system 3. The distance between the diffuser surface and cold plate surface was 8 mm.

<table>
<thead>
<tr>
<th>Position with respect to film center (mm)</th>
<th>Thickness per unit gas input ((\mu g \cdot cm^{-2} \cdot (T/l)^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>40</td>
<td>0.21 ± 0.08</td>
</tr>
<tr>
<td>30</td>
<td>1.52 ± 0.11</td>
</tr>
<tr>
<td>20</td>
<td>2.92 ± 0.17</td>
</tr>
<tr>
<td>15</td>
<td>3.10 ± 0.18</td>
</tr>
<tr>
<td>10</td>
<td>3.21 ± 0.16</td>
</tr>
<tr>
<td>5</td>
<td>3.33 ± 0.19</td>
</tr>
<tr>
<td>0</td>
<td>3.46 ± 0.17</td>
</tr>
<tr>
<td>-5</td>
<td>3.64 ± 0.18</td>
</tr>
<tr>
<td>-10</td>
<td>3.76 ± 0.19</td>
</tr>
<tr>
<td>-15</td>
<td>3.92 ± 0.21</td>
</tr>
<tr>
<td>-20</td>
<td>3.77 ± 0.19</td>
</tr>
<tr>
<td>-25</td>
<td>3.06 ± 0.17</td>
</tr>
<tr>
<td>-30</td>
<td>1.68 ± 0.09</td>
</tr>
</tbody>
</table>

in error as discussed, were not included in the final average.

It should be noted that, in general, the calibration factor depends on the distance between the diffuser surface and the cold plate surface which, in the present case, was 8 mm. For system 1, which had a 14 mm separation between the diffuser and cold plate, the calibration factors at the standard vertical diffuser position are given in Table 3.2. The values for system 2 are similar and agree with system 1 within the uncertainty (see Fig. 3.13).

Thicknesses of deuterium and tritium in units of \(\mu g \cdot cm^{-2}\) are factors of 2 and 3, respectively, larger than for a protium film with the same number of molecules due to the isotopic mass difference. The measurement of a deuterium film, when corrected by this factor, showed good agreement with protium films.
Table 3.2: Thickness of the protium film for system 1, deposited with standard diffuser position.

<table>
<thead>
<tr>
<th>Source position (mm)</th>
<th>Thickness per unit gas input ($\mu g\cdot cm^{-2} \cdot (T\cdot l)^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>2.37 ± 0.13</td>
</tr>
<tr>
<td>10</td>
<td>2.94 ± 0.15</td>
</tr>
<tr>
<td>0</td>
<td>3.32 ± 0.17</td>
</tr>
<tr>
<td>-10</td>
<td>3.60 ± 0.19</td>
</tr>
<tr>
<td>-20</td>
<td>3.31 ± 0.17</td>
</tr>
</tbody>
</table>

Other Measurements

Measurements were made with films deposited under different conditions to see the effects on thickness and uniformity. No deviation was found for films made with and without pumping the target vacuum during deposition within the relative uncertainty of about 1%. Reducing the deposition rate by an order of magnitude also did not noticeably affect either thickness or uniformity.

As described in Section 3.2, the gas deposition system is capable of making a second film on a separate cold surface through the opposite side of the diffuser. The apparatus was designed to minimize unwanted cross deposition from one side to the other by shielding with cold surfaces. By intentionally releasing a large amount of gas through the opposite side, cross deposition on the spot source target was checked. The measurement, made with similar assumptions on the cutoff value $\varepsilon$ to the measurement of very thin films described in Section 3.3.2, indicated that less than one part in a thousand of the injected gas arrived at the central spot. This result, together with the fact that pumping the system did not affect deposition, suggests that the gas molecules are likely to stick to the cold surface on first contact, which would explain the observed translational invariance of film profiles (Section 3.3.2).
Table 3.3: Thickness calibration factors for a neon film at source positions -20 mm, 0 mm and 20 mm.

<table>
<thead>
<tr>
<th>Source position (mm)</th>
<th>Thickness per unit gas input ($\mu g/cm^2 \cdot (T \cdot l)^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>24.7 ± 1.7</td>
</tr>
<tr>
<td>0</td>
<td>32.5 ± 2.0</td>
</tr>
<tr>
<td>-20</td>
<td>31.5 ± 1.9</td>
</tr>
</tbody>
</table>

Beam experiments using the same solid hydrogen target sometimes last for a few days, so it is important to check the effect of target aging. The results of measurements, made 8 hours apart, of the same film were consistent with each other, giving an upper limit in thickness variation $\Delta T/T \leq 0.5\%$ over 8 hours. In the analysis, a similar assumption in the cutoff value was made.

Films of other gases can also be deposited with the target system (for example, neon films have been used in the experiments by our group [192,193]). Thickness measurements of neon films were made using two different gas inputs at three of the five source positions. The stopping power in gaseous neon taken from the ICRU tables [190] was used to convert energy loss to thickness. The same 5% uncertainty in stopping power was assumed. Calibration factors for the three spots are given in Table 3.3.

**Monte Carlo Simulations of Gas Deposition**

In order to better understand the mechanism of gas deposition, Monte Carlo simulations were performed with the following assumptions: (1) molecules are emitted uniformly from the gas diffuser surface, (2) the molecules stick to the cold surface at the position of first contact, and (3) there is no interaction between the molecules. The last assumption is justified since the requirement to keep the target film temperature cold demands...
an insignificant intermolecular interaction, minimizing heat conduction from the warmer diffuser to the target plate. Pressure must be maintained low enough during deposition to keep the mean free path of the emitted molecules comparable to or larger than the diffuser-to-plate separation. Three different models for the angular distribution of molecules emitted from the diffuser surface were used in simulations: (a) the (unrealistic) forward emission model assuming $\theta = 0$, where $\theta$ is the angle with respect to the normal to the diffuser surface, (b) the isotropic emission model assuming that the number of molecules $dw$ emitted into unit solid angle $d\Omega = \sin \theta d\theta d\phi$ is constant, independent of $\theta$, i.e. $dw/d(\cos \theta) = \text{const.}$, and (c) the $\cos \theta$ emission model using a diffusion-like angular distribution $dw/d(\cos \theta) \sim \cos \theta$. The diffuser diameter of 60 mm and the distance of 8 mm between the diffuser and cold plate surface were used in simulations to compare with the measurements for system 3. There are no free parameters in the simulations other than the emission angular distribution. Figure 3.15 compares the simulation results with the film thickness profile for system 3 as deduced above, which is plotted with error bars that now include the uncertainty from the stopping power. The agreement with the $\cos \theta$ model (c) is quite good, except for the asymmetry in the shape.

The asymmetry, which obviously cannot be reproduced with our simulations, is unlikely to be due to an effect of gravity on the target, since the relative non-uniformity remains constant over a wide range of the thickness. The shape may be partly explained by the fact that the gas is introduced from the bottom of the diffuser system, hence gas molecules may have a larger probability of diffusing out at the bottom rather than the top of the diffuser, indicating a breakdown of assumption (1).

Similar simulations with the diffuser-to-plate distance taken to be 14 mm were compared to data for system 1. The $\cos \theta$ model best describes the data, but comparison indicates a slightly more forward peaked emission angular distribution. It should be recalled, however, that system 1 had a different perforation structure from system 3.
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Figure 3.15: The data, with error bars which include the uncertainty from the stopping power, denote the averaged thickness profile for system 3 plotted against the relative vertical distance from the diffuser center. The histograms are the the simulated thickness profiles from Monte Carlo calculations with different assumptions for the angular distribution of molecules emitted from the diffuser surface.

Reasonable success in the simulation with the $\cos \theta$ model gives us some confidence in scaling the thickness as a function of distance between the diffuser surface and the target support foil surface. Figure 3.16 shows a comparison of the simulated thickness profiles with different distances.

Effective Thickness

The results given in Tables 3.1, 3.2, and 3.3 should be used with caution when the films are used as a target for beam experiments. Since the present calibration measures
Figure 3.16: Monte Carlo simulations comparing different distances between the diffuser surface and the cold foil surface.

the profile only in the vertical dimension, the profile in the horizontal dimension has to be assumed when estimating an average target thickness. For a non-uniform layer, the average thickness depends on the width and profile of the beam which stops in the target. The angular divergence of the beam also contributes to the effective thickness.

Thus the method of the averaging depends on the type of measurement. For example, when the target is used to stop particle beams from an accelerator as in Ref. [194], a Gaussian distribution with a certain width may be justified. However, when muonic hydrogen atoms emitted from a solid hydrogen layer are stopped in a deuterium film, as in Ref. [83], the atomic beam is divergent with an angular distribution close to $\frac{d\omega}{d(\cos \theta)} \sim \cos \theta$, and depends on scattering cross sections. In this case the detailed averaging has
to be done with a Monte Carlo simulation which includes differential scattering cross sections. This will be done in a following chapter.

3.3.3 Summary of target characterization

Deposited films of solid hydrogen isotopes and neon have been characterized via the energy loss of $\alpha$ particles. The method can be applied to a relatively wide range of film thicknesses, e.g. for protium, from $\sim 1\ \mu g\cdot cm^{-2}$ to $\sim 1\ mg\cdot cm^{-2}$. The accuracy of the measurement is limited by the uncertainty in the stopping powers, but the relative accuracy can reach better than 1%. The uniformity can be measured by sampling the thickness at different positions with an array of alpha sources. Furthermore, it was possible to determine the thickness at an arbitrary vertical position by measuring films deposited with different diffuser positions. We note that muon catalyzed fusion reactions could also be used as a mono-energetic alpha source for the thickness measurement [179] (see also [195]).

As for our target system, the linearity of deposition was confirmed, with the exception of very thin film deposition with system 2. An asymmetric non-uniformity was observed in all films, while deposition conditions (such as pumping of the high vacuum region surrounding the target, or change in gas deposition rate) did not affect the thickness or uniformity. Deposition from one side of the diffuser did not contaminate the cold plate on the other side to a limit of one part in a thousand, enabling experiments using two separated solid hydrogen targets described in this thesis. No evidence was seen for the change in the film thickness over time. The comparison with Monte Carlo simulations indicates that the angular distribution of gas emission is close to a $\cos \theta$ model. The thickness calibration as well as film profile for the target is presented in Tables 3.1, 3.2, and 3.3.
3.4 Detection System

3.4.1 Overview

As illustrated in Sec 1.2, muonic processes in hydrogen isotopes produce a rich variety of complex reactions, leading to various kinds of radiations. Obviously, detecting as many different types of radiation as possible is advantageous in identifying and understanding the processes. The TRIUMF system was designed to allow this versatility.

Figure 3.17 illustrates a top view of a detector arrangement used in Run Series 1. Muons entering the system were detected by a thin plastic muon beam counter (T1). On the beam right (i.e., on the right hand side when facing the same direction as the beam flux) was placed a set of MWPCs, which tracked trajectories of muon-decay electrons, enabling measurement of the positions and the time of muon decay. A silicon detector and a neutron detector detected fusion products, while a germanium detector monitored target impurities via muonic X-rays. Si detector was placed in vacuum, and held on the side of the heat shield, viewing the targets without any window in between. Electron, Neutron and Ge detectors were placed outside the safety enclosure (Section 3.2.5), which contained the vacuum system.

For Run Series 2 (Fig. 3.18), the distance between the two gold foils was reduced to 17.9 mm from 42.5 mm in order to enhance the number of lower energy $\mu t$ atoms surviving to reach the downstream reaction layer. Because of the smaller drift distance, the MWPCs, which had a position resolution of several mm, were not useful and they were replaced by a silicon detector and neutron detector to gain better efficiency for fusion product detection.
Figure 3.17: Schematic top view of the detector arrangement for Run Series 1. A MWPC system reconstructed the paths of the muon decay electrons, whose time was measured with plastic scintillators, and the energy with a NaI crystal. A silicon detector and a neutron detector detected fusion products, while a Ge detector monitored target impurities via muonic X-rays. See also Fig. 3.18.
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Figure 3.18: Top view of the detector arrangement for Run Series 2. Compared to Run Series 1, the target spacing was reduced and the MWPCs were replaced by another Si detector and neutron detector.
Detector signals were processed via a NIM/CAMAC electronics system. The signals were converted with Analog-to-Digital Converters (ADCs) or Time-to-Digital Converters (TDCs) modules and read into a workstation (DEC Vax Station) with a TRIUMF developed VDACS data acquisition system [196]. The VDACS uses a PDP-11 microprocessor (CES 2180 Starburst, hereafter called a Starburst) as a front-end-processor which handles the data acquisition according to a procedure specified by the user using a high level language called TWOTRAN [197]. The data were logged onto a magnetic tape on an event by event basis for off-line analysis, while an analysis program MOLLI [198] was used for on-line analysis to provide a rapid diagnosis of the experiment.

3.4.2 Trigger

The data acquisition system was “triggered” to read in the data, when appropriate conditions were met. This decision making was done in a hardware logic circuit illustrated in Fig. 3.19. Our basic strategy for triggering can be summarized as:

1. Provide a gate for $\sim 10\mu$s after muon arrival, which is sufficiently long compared to the muon lifetime.

2. Inhibit the triggering if a muon had been already accepted and the gate was opened ($BUSY$), or the previous data was being processed by the computer ($INH$). 

3. Identify a pile up, if another muon arrived after the gate was opened.

4. Collect the signals from all of the detectors, if any one of the detectors fired during the gate.

5. Clear the TDC and ADC buffers, if no detector fired.

6. Record various scaler values for data normalization and diagnosis.
Figure 3.19: Schematic diagram for the trigger electronics, illustrating the key components. Pulses from the beam counter $T_1$, beam busy signal $BUSY$, and computer busy $INH$ were combined to provide an event gate $EVG$, which allowed individual sub-detector systems to accept events and give a sub-trigger $TRG_n$. The circuit diagrams are for illustrative purposes only, and not all of the details are given. 

\[\text{aElectronics diagrams shown in this section are taken from or based on those in Ref. [199], for which I thank Paul Knowles.}\]
The timing diagram is given in Fig. 3.20. A muon, defined by a sufficient energy deposit in T1, opened a pile up gate (PUG), which supplied a BUSY signal for 10 $\mu$s. The BUSY gate, together with the general inhibit signal (INH) due to computer delay\(^\text{12}\), was used to ensure (2.) above. Only when no muon had arrived in the previous 10 $\mu$s, and the computer was ready, was the coincidence $T1 \cdot \overline{B} \cdot \overline{I}$ satisfied, opening the event gate (EVG) as well as giving a common start to TDCs. The EVG was sent to all the detector signal processing logic allowing them to accept events.

If any of $n$ sub-detection systems described below gave a trigger $DET_n$ during EVG, a trigger gate $TRG_n$ was opened, which was subsequently closed with the end of event gate pulse (EEVG). The master event trigger $EVTR$ was provided as a coincidence of $TRG$ and $EEVG$, requesting the Starburst to collect the ADC and TDC values. On the other hand, when there was no detector giving a trigger, $EVCL$ was given from $EVCL = EEVG \cdot TRG$, clearing the ADCs and TDCs so that they could accept new values in the following event.

Once the master trigger was given, the Starburst provided a computer busy signal (CINH) in a CAMAC output register while processing the signal ($\sim 1$ ms). However, since it took a few hundred microseconds for CINH to turn on, an inhibit had to be provided by hardware (HINH) to prevent a pile up during this time. Furthermore, an extended inhibit (IEX) was provided while clearing the ADCs and TDCs, whose duration was determined empirically by monitoring the number of lost events\(^\text{13}\).

If more than one muon arrived during the gating period, the time correlation between the muon stop and the detected reaction would be lost, hence the event had to be discarded. The rejection of pile up events, which amounted to about 5% of total events at our typical beam rate of 5000 s$^{-1}$, was achieved in software using a pile-up bit pattern.

\(^{12}\text{When MWPCs were used, INH was also activated by a high voltage supply failure WCINH.}\)

\(^{13}\text{This was derived from comparing the number of triggers going into the Starburst (EVTR), and the number it actually accepted (STAR, a scaler counting Starburs events).}\)
Figure 3.20: Schematic timing diagram for trigger electronics. Lower lines indicate “on,” and higher lines “off,” corresponding to 0 V and -0.8 V, respectively, in fast-negative NIM logic standard. The horizontal axis represents approximate time flow. The second pulse in T1 illustrates a pile up event, which extends B, and delays EB. The EVCL pulse, not shown in the figure, was given when there was no trigger, and was generated at the same timing as EVTR.

given by the PUG module. Also in the case of pile up, the module extended its BUSY signal for 10 $\mu$s from the time of the second muon entrance, which in turn delayed the end-of-busy pulse (EB) (see dotted lines in the figure).
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An important scaler \textit{GMU (good muons)} was derived as a coincidence between \textit{EB} and \textit{EEVG (end of event gate)}. Since \textit{EEVG} always occurred 10 \( \mu s \) after \textit{EVG} was turned on, regardless of whether another muon came in or not, the coincidence was satisfied only if there was no pile up. Recalling that \textit{EVG} (\( = T1 \cdot B \cdot I \)) takes into account the computer dead time, \textit{GMU} provided the number of incident muons which satisfied all the trigger conditions except detector triggering, hence is to be used for absolute normalization.

Various scalers including \textit{GMU} were recorded for normalization and system diagnosis purposes. Their values were read into the Starburst typically every 5 s, independent of the muon trigger. Occasionally, some of the scalers were lost due to module malfunction, but there was enough redundancy to recover important scalers. For example, an alternative way of deriving the number of "good" incident muons is:

\[
GMU = T1 \cdot B \cdot I \times \frac{T1 \cdot B \cdot I}{T1} \times \frac{MON}{MON \cdot I},
\]

where \textit{MON} is a scaler for a pulser signal\(^{14}\), hence \( MON \cdot I/MON \) is the measure of live time fraction. The factor, \((T1 \cdot B \cdot I/T1) \times MON/(MON \cdot I)\) gives the fraction of events which do not have pile up \( f(pile) \), and \textit{GMU} is derived by \( T1 \cdot B \cdot I \times f(pile) \).

A further diagnosis and normalization tool was provided by a trigger called \( 1/N \), which was activated every 1024 hits in \( T1 \) scintillator, regardless of its deposited energy (i.e., including beam electron hits), and independent of the sub-detector triggers. The latter feature of this trigger allowed yet another way of checking the GMU scaler.

3.4.3 Multi-wire proportional chambers

Multi-Wire Proportional Chambers (MWPCs) provided information of the position of muon decay electron hits. The electronics diagram for the MWPC imaging system is

\(^{14}\) For the purpose here, this can be any signal uncorrelated to the muon trigger.
given in Fig. 3.21. When an electron passed through the chamber, it ionized a gas mixture of argon, isobutane, freon, and methylal vapor, which filled the chamber. The ionized electrons drifted toward the closest anode wire(s), and near the anode where the electric field became very strong, electrons were increased from the consecutive series of ionization of the gas molecules, leading to a multiplication process. This process induced a current in the nearest cathode wires in each of two planes in orthogonal directions (y and z in our case). Each wire in a plane was connected to a delay line, and the signal reached the end of the line with a time delay (with respect to the electron hit, measured by EM1 and EM2) that was proportional to the distance of the hit position from the edge of the plane, or the end of the delay line. The delay time was measured at both ends of the delay line, and the difference between them was proportional to the distance from the center of the chamber.

For instance, for z wires, the distance from the center $Z_{WC}$ is given by

$$Z_{WC}(mm) = (ZL - ZH) \times dispZ + offsetZ$$  \hspace{1cm} (3.4)$$

where $ZL$ and $ZH$ are the delay times at the beam upstream and downstream ends of the delay line, respectively, and $dispZ$ is a dispersion constant, determined from calibrations using an electron source, where the physical displacement of the source by a known distance was mapped to the shift in the delay time. The parameter, $offsetZ$ was adjusted such that the center of the distribution of electron hits corresponded to zero in the z scale. The position in the y-dimension was measured in an identical manner. With three sets of MWPCs providing hit positions, the trajectories of muon decay electrons were determined from a least-squares fit to three points on a straight line. The fitted line was extrapolated back to a perpendicular plane bisecting the target, providing positional information of the muon at the time of muon decay.

The energy of electrons was measured by a large NaI crystal (MINA). The cuts in
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Figure 3.21: Electronics diagram for the Multi-Wire Proportional Chamber imaging system. Only one of three identical chambers is shown. The time of the electron is measured by a pair of plastic scintillators, while its energy is given by the NaI crystal MINA.

MINA energy improved position resolution of the imaging system, by favoring higher energy electrons which scatter through smaller angles in window materials. More details of the data treatment will be given in Sec. 7.1.

3.4.4 Silicon detector

A silicon detector, placed in vacuum, detected low energy charged particles such as 3.5 MeV α particles from dt fusion and 3 MeV protons from dd fusion. For Run Series 1,
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Figure 3.22: Electronics diagram for a silicon detector. The detector output was pre-amplified to give a fast timing signal and a slow energy signal. The timing filter amplifier, with the use of an external delay, optimized the pulse shape which gave a sub-trigger $SITR$ if in coincidence with $EVG$, and opened the ADC gate for the amplified energy signal. In Run Series 2, where two silicon detectors were used, both functioned in an identical manner. A 4-input 8000-channel ADC was shared between silicon detector(s) and a germanium detector.

A passivated, ion-implanted planar silicon (PIPS) detector\textsuperscript{15} of active thickness 150 $\mu$m and area 600 mm\textsuperscript{2} was operated at a bias voltage of 30 V. For Run Series 2, two similar PIPS detectors\textsuperscript{16}, but of active thickness 300 $\mu$m and area 2000 mm\textsuperscript{2} (diameter 50 mm) were used. Silicon detectors, such as PIPS detectors, and more commonly used silicon surface barrier (SSB) detectors, are in principle reverse-biased diodes. Charged particles slowing down in a carrier-depleted region of the silicon crystal creates electron-hole pairs, which are swept toward and collected at the electrodes by an applied bias field. PIPS detectors offer advantages over SSB detectors, such as thinner yet more robust entrance windows, and lower leakage currents, which improves energy resolution.

Figure 3.22 shows the electronics diagram for silicon detectors. Pre-amplifiers were placed immediately next to the feed-through connectors of the vacuum system, in order to reduce the total capacitance. Amplification of energy and timing output of the preamps

\textsuperscript{15}Canberra, model FD/S-600-29-150-RM
\textsuperscript{16}Canberra, model FD/CY-2000-37-300-RM
was done with linear amplifiers and timing filter amplifiers, respectively, before the signals were sent to the counting room.

Considerable efforts were made to improve energy resolution in the tritium environment. This included shielding the pre-amplifier and the cables leading to it with a copper foil and copper braid material respectively, grounding the detectors and electronics system to the target vacuum chamber, and using a noise-filtered power supply.

Mounted on the cryogenic thermal shields, the detectors were kept at about 90 K, which reduced the leakage currents and thereby improved the energy resolution.

For Series 2 the horizontal dimension of the detectors was collimated to 13.9 mm to view only the inner sides of the gold foils, where the reactions of interest took place. Furthermore the collimation restricted the angular path of the alphas and protons entering the detectors, preventing the ones with grazing angles which suffer large energy loss in the layer. This reduced variations in the detection efficiencies when layers with different thicknesses were used. The energy scale of the silicon detectors was calibrated with an $^{241}$Am source.

### 3.4.5 Neutron detector

Detection of neutrons is among the standard techniques in conventional studies of $\mu$CF, since they easily exit from the target containers which typically consist of thick walls.

Detectors using the liquid scintillator NE213 were used with pulse shape discrimination (PSD) provided by hardware modules (Link System PSD-5010 and PSD-5020). Taking advantage of the difference in the liquid scintillator light output time-structure between neutrons and gammas, the PSD module could discriminate the two at the trigger level.

The electronics diagram for neutron detection is given in Fig. 3.23. A pair of plastic
Figure 3.23: Electronics diagram for a neutron detector. Charged particle anti-coincidence was required for the neutron/gamma detection. The pulse shape discriminator LINK 5010 (5020 for the second detector) provided a trigger level discrimination of neutrons from gammas, though the latter were also recorded, pre-scaled by factor of 30. For the sub-trigger $N1TRG$ to fire, the event has be in coincidence with EVG.

scintillators placed in front of the detector was used in anti-coincidence to reject charged particles. Together with neutron triggers, gammas were recorded but were pre-scaled by a factor of 30.
3.4.6 Germanium X-ray detector

Detection of muonic X-rays emitted upon muon transfer from hydrogen isotopes to heavier elements was used mainly for two purposes in our experiments. One was to detect arrivals of muonic hydrogen at a specific spatial position, while the other was to measure the level of impurities in the target layers. For the work of this thesis, the latter was the main use, and because of the anomalously long diffusing path length of muonic tritium in protium, the detection of the transfer X-rays provided a sensitive measure of impurities such as nitrogen and neon in the layer. Examples of the former use of muonic X-rays are given in Ref. [193, 200, 201].

Figure 3.24 shows the circuit for the germanium detector with a pair of plastic scintillators used for charged particle anti-coincidence. The detector was placed in a lead collimator to reduce neutron induced background [202].
Figure 3.25: Electronics diagram for the electron detection by plastic scintillators and the dynode output of the neutron detectors. Only scintillators in front of the neutron detector are shown here, but all other ones were also connected to the logical OR module to give an electron trigger DELE. The neutron dynode output provided not only the timing of the electron, but also its energy.

3.4.7 Electron detection system

Plastic scintillators, in addition to providing a charged particle veto for neutron and photon detection, were used to detect the electrons with an independent trigger, recording the electron time spectrum (Fig 3.25). Since statistics for the electron measurement were not a problem, the trigger was pre-scaled by factor of 4, in order to reduce the load on the data acquisition system.

This circuit also provided a tool called delayed electron (Del) coincidence for reduction of muon capture related background, where one demands that the muon-decay electron be detected after the reactions of interest, hence ensuring the muon was alive, (i.e., had not decayed nor been captured by) then. In case of nuclear muon capture, the muon was converted to its neutrino, and no electron would be observed in the delayed time
window, which was typically from 0.2 to 5 $\mu$s after the reaction. For the Del coincidence, the master trigger was always given by the detector of interest, therefore no pre-scaling of the signal was necessary.

The neutron detectors could also be used for electron detection by utilizing a fast signal from the dynode of the photo-multiplier. A separate trigger, called telescope or simply Tel trigger, was implemented which required a triple coincidence of two plastic scintillators and the dynode signal, with the latter energy also recorded.
Chapter 4

Experimental Runs

Muonic hydrogen research at TRIUMF has evolved for over a decade, making continual progress in improving the measurements and understanding the physics. Of nearly a dozen series of experimental runs performed up to now by the Collaboration, some of which have or will have resulted in Master’s, as well as Ph.D. theses [30,179,199,203], the runs analyzed for this thesis are the first two with tritium used in the target, which are summarized in Table 4.1.

<table>
<thead>
<tr>
<th>Run (date)</th>
<th>Au foil spacing</th>
<th>Gas diffuser(^a)</th>
<th>Si Detector Active area</th>
<th>Thickness</th>
<th>MWPC imaging</th>
<th>Main goals Physics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Series I (Nov–Dec 93)</td>
<td>39.0 mm</td>
<td>System 2</td>
<td>600 mm(^2)</td>
<td>150 (\mu m)</td>
<td>yes</td>
<td>Commission (\mu t) emission</td>
</tr>
<tr>
<td>Series II (Jul–Aug 94)</td>
<td>17.9 mm</td>
<td>System 3</td>
<td>2(\times2000) mm(^2)</td>
<td>300 (\mu m)</td>
<td>no</td>
<td>Production TOF fusion</td>
</tr>
</tbody>
</table>

\(^a\)See Section 3.2.2 the for description of diffusers.

\(^b\)Each detector had a Cu collimator with an opening of 13.9 mm\(\times\)50 mm.

Table 4.1: Comparison of Run Series

4.1 Run Series I

Run Series I was performed for three weeks in November and December 1993. After many months of work spent on target system modification to incorporate tritium compatibility, observing a clear peak built up within minutes after starting the first \(dt\) fusion
Table 4.2: Run summary for Series 1. For thin deuterium overlayers shown with \( \oplus \), the nominal gas input values are given, but the actually thickness has some uncertainties (see Section 3.3.2). GMU, the number of “good muons,” is derived from Eq. 3.3. Data was collected typically at the rate of \( 3.5 \times 10^3 \) GMU/s, i.e., \( 12-15 \times 10^6 \) GMU/hour.

<table>
<thead>
<tr>
<th>ID</th>
<th>Run</th>
<th>Target Upstream</th>
<th>GMU (( \times 10^6 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>I-1</td>
<td>1526-1529</td>
<td>SET1 (1000 T·l ( \text{H}_2 ) with 0.1% ( \text{T}_2 ))</td>
<td>9.179, 47.377, 77.682, 102.109</td>
</tr>
<tr>
<td>I-2</td>
<td>1536</td>
<td>SET1 ( \oplus ) 3 T·l</td>
<td>43.726</td>
</tr>
<tr>
<td>I-3</td>
<td>1538-1541</td>
<td>SET1 ( \oplus ) 6 (3+3) T·l</td>
<td>13.471, 34.918, 39.038, 88.066</td>
</tr>
<tr>
<td>I-4</td>
<td>1542-1545</td>
<td>SET1 ( \oplus ) 12 (3+3+6) T·l</td>
<td>12.533, 9.027, 24.896, 49.753</td>
</tr>
<tr>
<td>I-5</td>
<td>1546</td>
<td>SET1 ( \oplus ) 24 (3+3+6+12) T·l</td>
<td>21.302</td>
</tr>
<tr>
<td>I-6</td>
<td>1547</td>
<td>SET1 ( \oplus ) 36 (3+3+6+12+12) T·l</td>
<td>22.423</td>
</tr>
<tr>
<td>I-7</td>
<td>1555-1556</td>
<td>SET3 (1000 T·l ( \text{H}_2 ) with 0.3% ( \text{T}_2 ))</td>
<td>6.671, 33.033</td>
</tr>
<tr>
<td>I-8</td>
<td>1572</td>
<td>1000 T·l ( \text{H}_2 )</td>
<td>26.844</td>
</tr>
</tbody>
</table>
Run was quite exciting indeed.

Run Series I was a commissioning run for the new tritium target system, and much of the time was devoted to testing of various systems and establishing safe procedures of target handling. It was the first time that we observed an $\alpha$ particle from muon catalyzed $dt$ fusion with high resolution. In addition, the emission of muonic tritium into vacuum was observed for the first time.

For this thesis, MWPC imaging data was analyzed to study muonic tritium emission, on which the atomic beam method is based. Shown in Table 4.2 is a summary of runs analyzed from this Series. The standard emission target, denoted SET1 (or SET3) consisted of a mixture of 1000 T·l pure protium with 0.1% (or 0.3%) of tritium. An additional layer of thin deuterium, called an overlayer, was sometimes deposited on top of the emission layer. Its thickness for this Series, however, had some uncertainty due to the effects of gas remaining in the transfer tube, as described in Section 3.3.2 (see Fig. 12(a)). Other than the thin overlayers, the average conversion factor for T·l is $3.2 \mu g \cdot cm^{-2}$ per T·l. With a T1 beam counter rate of $\sim 5 \times 10^3/s$ GMU events were accumulated at the rate of $10^{-12}\times 10^6$/hour. The scaler GMU (Fig. 3.19) suffered an occasional malfunction during this run, therefore GMU derived from Eq. 3.3 was used for normalization.

4.2 Run Series II

Run Series II was our production run, where the measurement of $d\mu t$ molecular formation via time of flight was attempted. Improvements in the target system had been made after Series I. The gas diffuser was replaced with the one with sintered metal to ensure microscopic uniformity of the target film. The gas transfer tubing was modified to remove the problem of gas remaining in the transfer tube which had caused uncertainties in thin layer thicknesses. Target support foil spacing was reduced from 39 mm to 17.9
## Table 4.3: Run summary for Series 2 (part 1)

<table>
<thead>
<tr>
<th>ID</th>
<th>Run</th>
<th>Target Upstream</th>
<th>Target Downstream</th>
<th>GMU (×10^6)</th>
</tr>
</thead>
<tbody>
<tr>
<td>II-1</td>
<td>1625</td>
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<tr>
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<td>1631</td>
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<td>20 T·l</td>
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<tr>
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<td>subtotal 1530–31</td>
<td>48.323</td>
</tr>
<tr>
<td>II-2</td>
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<td>20 T·l</td>
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<td>33.483</td>
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<td>1654</td>
<td>1000 T·l H₂</td>
<td>–</td>
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<td>SET1</td>
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<td>500 T·l H₂ ⊕ 3 T·l</td>
<td>38.143</td>
</tr>
<tr>
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<td>500 T·l H₂ ⊕ 3 T·l</td>
<td>81.813</td>
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<tr>
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<td>500 T·l H₂ ⊕ 3 T·l</td>
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<tr>
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<td>1684</td>
<td>SET1 ⊕ 14 T·l</td>
<td>500 T·l H₂ ⊕ 3 T·l</td>
<td>41.567*</td>
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<td>1685</td>
<td>SET1 ⊕ 14 T·l</td>
<td>500 T·l H₂ ⊕ 3 T·l</td>
<td>29.243*</td>
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<td></td>
<td>1686</td>
<td>SET1 ⊕ 14 T·l</td>
<td>500 T·l H₂ ⊕ 3 T·l</td>
<td>30.421*</td>
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<td>subtotal 1671–86</td>
<td>469.808*</td>
</tr>
<tr>
<td>II-9</td>
<td>1688</td>
<td>SET1 ⊕ 14 T·l</td>
<td>500 T·l H₂ ⊕ 6 (3+3) T·l</td>
<td>50.370*</td>
</tr>
<tr>
<td>II-10</td>
<td>1690</td>
<td>SET1 ⊕ 14 T·l</td>
<td>500 T·l H₂ ⊕ 20 (3+3+8) T·l</td>
<td>54.109*</td>
</tr>
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</table>
## Table 4.4: Run summary for Series 2 (part 2)

<table>
<thead>
<tr>
<th>ID</th>
<th>Run</th>
<th>US Target</th>
<th>DS Target</th>
<th>GMU ($\times 10^6$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>II-12</td>
<td>1708</td>
<td>SET2 (1000 T·l H$_2$ with 0.2% T$_2$)</td>
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<td>46.441</td>
</tr>
<tr>
<td>II-13</td>
<td>1709</td>
<td>SET2 $\oplus$ 14 T·l</td>
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<td>52.875</td>
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<td>1711</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td></td>
<td>11.266</td>
</tr>
<tr>
<td></td>
<td>1712</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td></td>
<td>40.259</td>
</tr>
<tr>
<td></td>
<td>1713</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td></td>
<td>14.359</td>
</tr>
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<td></td>
<td>1714</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td></td>
<td>48.087</td>
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<tr>
<td>subtotal 1708–14</td>
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</tr>
<tr>
<td>II-14</td>
<td>1719</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td>3 T·l</td>
<td>58.499</td>
</tr>
<tr>
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<td>1723</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td>3 T·l</td>
<td>51.871</td>
</tr>
<tr>
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<td>1728</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td>3 T·l</td>
<td>37.135</td>
</tr>
<tr>
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<td>1729</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td>3 T·l</td>
<td>38.114</td>
</tr>
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<td>1730</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td>3 T·l</td>
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<tr>
<td>subtotal 1719–30</td>
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</tr>
<tr>
<td>II-15</td>
<td>1741</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td>23 (3+20) T·l</td>
<td>51.847</td>
</tr>
<tr>
<td></td>
<td>1742</td>
<td>SET2 $\oplus$ 14 T·l</td>
<td>23 (3+20) T·l</td>
<td>7.385</td>
</tr>
<tr>
<td>subtotal 1741–42</td>
<td></td>
<td></td>
<td></td>
<td>59.232</td>
</tr>
<tr>
<td>II-16</td>
<td>1693</td>
<td>SET0.5 $\oplus$ 20 T·l</td>
<td></td>
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</tr>
<tr>
<td></td>
<td>1694</td>
<td>SET0.5 $\oplus$ 20 T·l</td>
<td></td>
<td>70.664</td>
</tr>
<tr>
<td>subtotal 1694–95</td>
<td></td>
<td></td>
<td></td>
<td>93.266</td>
</tr>
</tbody>
</table>
mm, in order to increase at the downstream reaction layer the number of $\mu t$ for energies 0.5–2 eV, at which the largest resonances were predicted. The MWPC imaging system, which would not have been as effective due to the smaller foil spacing, was replaced by another silicon detector. The z-dimension (beam direction) of both of 2000 mm$^2$ silicon detectors were collimated to 13.9 mm, to view only the inner side of the Au foils, where target films exist, with reasonable (i.e., non-grazing) angles.

Shown in Tables 4.3 and 4.4 is the run summary for Series 2. The standard emission targets with tritium concentration 0.1% (SET1) and 0.2% (SET2) were used with deuterium overlayers shown with $\oplus$ in the table. The GMUs were taken from the GMU scalers, but the runs with (*) in the table (Run 1684-90) had a problem with an ADC affecting silicon detector normalization, which will be corrected in Section 8.1.2.

Apart from the data for TOF measurements of molecular formation, test data for new experiments were taken during this run, which resulted in new proposals. These include precision measurement of muonic atom scattering cross sections and direct measurement of the $\mu$CF efficiency–limiting sticking probability.

### 4.3 Histogramming and analysis tools

Data collected during the experimental runs, which were recorded on tape on an event-by-event basis, were analyzed using several different histogramming and analysis packages. A TRIUMF program, MOLLI [198] with the FIOWA histogramming package [204], which was used on the TRIUMF Data Analysis Vax cluster, was the main mode of the histogram collection. CERN packages PAW and HBOOK, featuring NTUPLE analysis, were used on DEC Alpha Unix servers, when correlations of many different variables were important. Graphic drawing and data manipulation including fitting were done with REPLAY [205] and PHYSICA [206] both TRIUMF programs, as well as PAW, on
both VAX and UNIX (Alpha/OSF, Pentium/Linux) platforms.
Chapter 5

Monte Carlo simulation codes

In order to gain quantitative understanding of the spectra observed by our experiments, comparison with detailed Monte Carlo simulations is essential. Several simulation codes have been developed by different authors. In particular, the prime tool for the quantitative analysis for the present thesis is SMC (Super Monte Carlo) developed by Tom Huber at Gustavus Adolphus College in Minnesota, USA. In the following section, SMC is described in detail, followed by a brief discussion of other simulation codes used in this thesis.

5.1 SMC Monte Carlo code

Developed over several years by Huber, SMC simulates muonic processes in full three-dimensional geometry with energy-dependent cross sections and rates as inputs. The code also simulates the MWPC imaging process, and was a prime tool for quantitative analysis for this thesis. This section explains the physical processes modelled in the simulation.

5.1.1 Muonic atom generation

The simulation was started by creation of a muonic atom in the emission layer. The spatial distributions, both in the beam direction $z$ and in the radial direction $r$, of the muonic atom generation could be controlled by the input file. A uniform distribution
in $z$ and a Gaussian distribution with a flat top in $r$ were used as nominal input. For determination of the radial beam distribution, see Section 6.1. The fraction of muonic protium $\mu p$ to muonic tritium $\mu t$ was proportional to that of protium to tritium in the target\textsuperscript{1}. The initial energy of the muonic atom was taken to be a Gaussian distribution with mean 1 eV and standard deviation 1 eV. Hyperfine states of muonic atoms were populated with the statistical weights, i.e., 25% F=0 and 75% F=1 for both $\mu p$ and $\mu t$, which are composites of two spin one half particles.

\section*{5.1.2 Reaction selection}

Once a muonic atom is created, various reactions can take place, which include muon decay, elastic scattering, inelastic scattering, muon transfer, muonic molecular formation followed by fusion or back decay, and muon capture (after transfer to a heavy element). Some of the theoretical input for density dependent reactions is given in terms of cross sections, but it is convenient to convert them to rates. For the process $i$:

$$
\Lambda_i = \sigma_i \cdot v_{rel} \cdot n_i,
$$

where $v_{rel}$ is the relative velocity between the projectile and the target, and $n_i$ is the number density of the relevant target species. Inputs for other reactions are given by rates, some such as molecular formation, dependent on the density and/or collision velocity, while others such as muon decay independent of the above.

The simulation was done in an energy dependent manner, and for a given state of the projectile with a particular energy, the probability for $i$th reaction $P_i$, out of $m$ possible

\textsuperscript{1}Deviations of the capture ratio from the concentration ratio were reported for pionic hydrogen and deuterium (for a review, see [207]), but this does not affect our simulations, because of very small tritium concentrations used in the measurements.
different reactions, is given by:

\[ P_i = \frac{\Lambda_i}{\sum_i \Lambda_i}, \quad (5.2) \]

where \( \Lambda_i \) is the reduced rate for each reaction, given by Eq. 5.1 for cross section inputs, or by

\[ \Lambda_i = C_i \cdot \phi \cdot \lambda_i, \quad (5.3) \]

for input by rates, where \( C_i \) is the relative concentration of relevant species, \( \phi = n/n_0 \) is the target number density in the unit of liquid hydrogen atomic density \( n_0 = 4.25 \cdot 10^{22} \) cm\(^{-3} \), and \( \lambda_i \) is the rate normalized to \( n_0 \). The processes such as muon decay and nuclear muon capture are independent of the target concentration \( C_i \). For muon decay, for example, \( \Lambda_i = \lambda_0 Q_k \), where \( \lambda_0 \) is the free muon decay rate and \( Q_k \) the Huff factor\(^2 \) in the \( k \)th element.

### 5.1.3 Time and position evolution

The time between reactions \( \tau_r \) is given by

\[ \tau_r = \frac{-\log(\text{RAN})}{\sum_i \Lambda_i}, \quad (5.4) \]

where \( \text{RAN} \) is a random number between 0 and 1. This follows from the requirement that

\[ d \left( \exp \left[ -\frac{\tau_r}{\tau_{tot}} \right] \right) = \text{const}. \quad (5.5) \]

where \( \tau_{tot} = \left( \sum_i \Lambda_i \right)^{-1} \) is the total reaction time constant.

\(^2\text{See Section 6.4 for the description of the Huff factor.}\)
The position $\vec{X}$ of the reaction is calculated from $\vec{X} = \vec{v}_{lab} \times r$. If $\vec{X}$ lays outside the boundary of the current medium, the projectile is advanced to the edge of the new medium, from which a new random number is generated for Eq. 5.4.

### 5.1.4 Muonic processes

In this section, we discuss the details of physical processes modelled in the simulation.

**Elastic scattering**

\[
\begin{align*}
\mu p^F + p & \rightarrow \mu p^F + p, \quad F = 0, 1 \\
\mu d^F + d & \rightarrow \mu d^F + d, \quad F = \frac{1}{2}, \frac{3}{2} \\
\mu p^F + t & \rightarrow \mu p^F + t, \quad F = 0, 1 \\
\end{align*}
\]

(Eq. 5.6)

\[
\begin{align*}
\mu p^F + d & \rightarrow \mu p^F + d, \quad F = 0, 1 \\
\mu p^F + t & \rightarrow \mu p^F + t, \quad F = 0, 1 \\
\mu d^F + p & \rightarrow \mu d^F + p, \quad F = \frac{1}{2}, \frac{3}{2} \\
\mu d^F + t & \rightarrow \mu d^F + t, \quad F = \frac{1}{2}, \frac{3}{2} \\
\mu t^F + p & \rightarrow \mu t^F + p, \quad F = 0, 1 \\
\mu t^F + d & \rightarrow \mu t^F + d, \quad F = 0, 1 \\
\end{align*}
\]

(Eq. 5.7)

Elastic scattering cross sections for the symmetric (Eq. 5.6) and asymmetric (Eq. 5.7) cases were taken from Bracci et al. [16], and Chiccoli et al. [17], respectively. These are known as "Nuclear Atlas cross sections," since the calculations assume scattering with the bare nucleus, as opposed to the atom or molecule. For the symmetric case, differential cross sections published by Melezhik and Wozniak [18] were used to give the final angular
distributions. Similar differential cross sections for asymmetric collisions have not been published yet, but were calculated by Wozniak [208] and used as inputs to the simulation.

There are more recent calculations which take into account the molecular effects [23] as well as solid state effects [167]. However, not all these cross sections are available in differential forms as of the writing of this thesis. Because we are interested in the transport of muonic atoms, it is essential to use differential cross sections, and therefore we chose to use the above “nuclear” cross sections which are available in differential form. For energies above about 0.2 eV, the molecular and solid state effects are small, therefore for the resonant molecular formation at $\mu t$ energies of 0.5 – 2 eV, the use of nuclear cross sections should be a good approximation. At lower energies, however, solid state processes become increasingly important.

**Spin flip**

\[
\begin{align*}
\mu p^{F=0} + p & \rightarrow \mu p^{F=1} + p -0.1820 \text{ eV} \\
\mu p^{F=1} + p & \rightarrow \mu p^{F=0} + p +0.1820 \text{ eV} \\
\mu d^{F=0} + d & \rightarrow \mu d^{F=1} + d -0.0485 \text{ eV} \\
\mu d^{F=1} + d & \rightarrow \mu d^{F=0} + d +0.0485 \text{ eV} \\
\mu t^{F=0} + t & \rightarrow \mu t^{F=1} + t -0.2373 \text{ eV} \\
\mu t^{F=1} + t & \rightarrow \mu t^{F=0} + t +0.2373 \text{ eV}
\end{align*}
\] (5.8)

Spin flip cross sections are also taken from the Nuclear Atlas by Bracci et al. [16] with differential cross sections provided by Melezhik and Wozniak [18]. Spin flip in asymmetric collisions such as $\mu t + p$ requires relativistic interactions, hence is much suppressed. Cohen calculated those cross sections and found that they are smaller by 6 to 8 orders of magnitude compared to their elastic scattering counterparts [21]. Therefore
we do not consider the asymmetric spin flip in our simulations.

\textbf{Charge exchange}

\begin{align*}
\mu p + d &\rightarrow \mu d + p + 134.709 \text{ eV} \\
\mu p + t &\rightarrow \mu t + p + 182.751 \text{ eV} \\
\mu d + t &\rightarrow \mu t + d + 48.042 \text{ eV} \\
\mu p + Z &\rightarrow \mu Z + p \\
\mu d + Z &\rightarrow \mu Z + d \\
\mu t + Z &\rightarrow \mu Z + t
\end{align*}

(5.9)

For isotopic charge exchange, or muon transfer reactions (5.9), differential nuclear cross sections similarly calculated by Chiccoli \textit{et al.} [17] and Wozniak [209] are used, except for \( \mu p + t \rightarrow \mu t + p \), where the rate measured by our group, \( \lambda_{pt} = 5.86 \times 10^9 \text{ s}^{-1} \) (energy independent) [83], was used as a nominal input. Although the experimental \( \lambda_{pt} \), obtained in similar conditions to this thesis, appeared more reliable than the theory, simulations with the theoretical \( \lambda_{pt} \) were also performed, wherever possible, to check the systematic effects.

Muon transfer to heavy elements takes place when muonic hydrogen reaches target materials such as gold. The rate for these reactions is taken to be a high value \( 10^{12} \text{ s}^{-1} \) to ensure rapid transfer in the simulation.

\textbf{Nonresonant molecular formation}

\begin{align*}
\mu p + p &\rightarrow p\mu p \\
\mu d + p &\rightarrow p\mu d \\
\mu t + p &\rightarrow p\mu t
\end{align*}

(5.11)
Since Faifman's predictions of nonresonant molecular formation rates [51] depend only weakly on the energies, a constant rate from Ref. [51] was used: \( \lambda_{pud} = 5.6 \times 10^6 \text{ s}^{-1} \) for \( p\mu d \) and \( \lambda_{p\mu t} = 6.5 \times 10^6 \text{ s}^{-1} \) for \( p\mu t \) formation. For \( p\mu p \) formation, on the other hand, the measured rate by our group \( \lambda_{p\mu p} = 3.2 \times 10^6 \text{ s}^{-1} \) [83] was used in the simulations.

**Resonant molecular formation**

\[
\mu t^F + D^\text{ortho}_2 \rightarrow [(d\mu t)\text{dee}], \quad F = 0, 1 \tag{5.12}
\]

\[
\mu t^F + D^\text{para}_2 \rightarrow [(d\mu t)\text{dee}], \quad F = 0, 1 \tag{5.13}
\]

As discussed in detail in earlier chapters, these are indeed the key rates which we wish to test by our measurements. The nominal rates for the resonant formation were taken from the work of Faifman and his colleagues (Refs. [70–72, 133]), calculated for isolated target molecules at 3 K with the quadrupole interaction included, but correlations among target molecules not taken into account. These rates are calculated separately for different \( \mu t \) hyperfine states, and target molecule rotational states (ortho and para). Since our \( D_2 \) layers were made by rapid freezing of warm gas, it is expected that the ortho-para ratio is statistical (2 to 1), and is so assumed in the simulations. The predicted rates by Faifman are given Fig. 2.4. A peak value for \( F = 1 \) in ortho \( D_2 \) for example is \( 1.16 \times 10^{10} \text{ s}^{-1} \) with a resonance energy of 0.45 eV.

**Fusion and back decay**

\[ d\mu t \rightarrow n + \alpha + 17.6 \text{ MeV} \tag{5.14} \]

\[
[(d\mu t)\text{dee}] \rightarrow \mu t^F + D_2, \quad F = 0, 1 \tag{5.15}
\]

Once the molecule is formed, fusion can take place rapidly releasing the energy. If the formation took place resonantly, it can break up through a back decay. The branching
ratio for fusion ($W$) versus back decay ($1 - W$) is given by Faifman et al. [70–72, 133]).

The hyperfine state of $\mu t$ after the back decay was kept the same as before the molecular formation. The outgoing energy of $\mu t$ was one of the free parameters in the simulation.

### 5.1.5 Muon decay and imaging

The decay rate of a muon in muonic hydrogen is taken to be the same as that of a free muon ($\lambda_0 = 0.455 \times 10^6$ s$^{-1}$), ignoring the small nuclear capture rate ($\lambda_c^{H_2} = 531 \pm 33$ s$^{-1}$ [210]). When muon decay is selected as the reaction, an imaging routine is called to simulate the MWPC response.

Two MWPC planes (instead of three as in the actual experiment), as well as the copper thermal heat shield and stainless steel vacuum window, were defined in the program. The electron was generated isotropically (within a cone containing the MWPC planes) from the muon decay position, and passed through the shield and the window, where multiple scattering deflected the electron angle according to a Gaussian distribution with a width given by

$$\theta = \frac{13.6 \text{MeV}}{\beta c_p} \sqrt{x/X_0[1 + 0.038 \ln(x/X_0)]}$$  \hspace{1cm} (5.16)

where $p, \beta c$ are the momentum and velocity of the electron, and $x/X_0$ is the medium thickness in radiation lengths [211]. The electron energy spectrum $\mathcal{F}_{\text{dec}}(E)$ is approximated by

$$\mathcal{F}_{\text{dec}}(E) \sim \begin{cases} 
\frac{1}{\sqrt{2\pi} \sigma_{\text{dec}}} \exp \left( -\frac{(E - 53)^2}{2\sigma_{\text{dec}}^2} \right) & \text{if } 0 < E < 53 \text{ MeV} \\
0 & \text{otherwise}
\end{cases} \hspace{1cm} (5.17)$$

with $\sigma_{\text{dec}} = 50/(2\sqrt{2 \ln 2})$ MeV (normalization arbitrary). This is half of a Gaussian with FWHM of 50 MeV, displaced by 53 MeV, which is sufficiently close to the real spectrum for our purposes. When the electron track intersected with the $i$th MWPC at $[x_i, y_i, z_i]$,
the position was smeared in $y$-$z$ plane by a Gaussian with a distribution of standard deviation $\sigma_y^{WC}, \sigma_z^{WC}$, characterizing the finite resolution of MWPCs. The new positions $[x_i, y_i', z_i']$ were fitted with a straight line, and extrapolated back to the perpendicular plane bisecting the target, similar to the way the real data was analyzed.

5.2 Other simulation codes

5.2.1 Full muonic processes

MCKIN, developed by Valeri Markushin$^3$ of Russian Research Center, Moscow, was used in the early stages of the experiment, providing guidance for planning the measurements.

As well, a program developed for TRIUMF Experiment 742 by Jan Wozniak of Institute of Physics and Nuclear Techniques, Cracow, Poland, was used to test some of the systematics of the measurements such as solid state effects in muonic atom scattering.

5.2.2 Specific processes

In addition to the above codes for full simulation of muonic processes, various smaller programs were used to study specific parts of the experiment. A GEANT-based code was developed by Marshall and used to understand the energy deposited in the liquid scintillators by the muon-decay electrons. A charged-particle energy loss calculation program APEC-97[212] featuring a package PEPPER[213], which I originally developed for the planning of the muon-alpha sticking experiment, was used to study the energy spectra of silicon detectors as well as for determining detector solid angles and energy cut efficiencies. The detail of these codes will be discussed in the analysis sections.

$^3$Now at Paul Scherrer Institute, Switzerland.
Chapter 6

Analysis I – Absolute normalization

6.1 Effective target thickness

Because of the non-uniformity of the target observed in Section 3.3, the average layer thickness depends on the width and profile of the beam which stops in the target. Also recall that we have measured only the profile in the $Y$ (vertical) dimension, hence the horizontal profile has to be assumed. The effective thickness can be defined via

$$T_{\text{eff}} = \frac{\sum w_i T_i}{\sum w_i},$$  \hspace{1cm} (6.1)

where $T_i$ is the thickness at the $i$th measured spot, and $w_i$ the weighting factor. A weighted root–mean–square deviation of thickness is defined via

$$\Delta T_{\text{rms}}^{\text{eff}} = \sqrt{\frac{\sum w_i (T_i - T_{\text{eff}})^2}{\sum w_i}}.$$  \hspace{1cm} (6.2)

This is a quantitative measure of the non-uniformity and is useful when optimizing the vertical position of the diffuser for deposition.

Figure 6.1 illustrates the dependence of the effective thickness on the beam parameters. The average was calculated assuming rotational symmetry of the thickness profile, and weighted with two different beam parameterizations (i) Gaussian beam, and (ii) a flat top Gaussian, for which the radial intensity at the distance $r$, $f(r)$ is defined by:

$$f(r) \propto \begin{cases} 
1 & \text{if } 0 < r < R_{\text{flat}} \\
\exp \left[ -\frac{(r - R_{\text{flat}})^2}{2\sigma^2} \right] & \text{otherwise}.
\end{cases} \hspace{1cm} (6.3)$$
Figure 6.1: Dependence of the effective thickness (above) and the effective deviation (below) on the beam parameters and the radial cut-off values. (a) (b) (c): Flat-top Gaussian beam (FG, defined in Eq. 6.3) with $R_{\text{flat}} = \alpha$ and FWHM$_g = 2.355 \times \sigma_g = \alpha/2$. (d) (e) (f): Gaussian beam (G) with FWHM$_g = \alpha$. The points are slightly shifted horizontally for visual ease. The error bars represent the 5% uncertainty from the stopping power.
Figure 6.2: The Y-distribution of decay electrons in the upstream layer imaged by the MWPC system plotted with error bars is compared with the Monte Carlo simulations, in the histograms, assuming Gaussian distributions of FWHM 20, 25, and 30 mm (from inside out) as the initial muon beam distribution in the XY plane.

The figure also shows the dependence on the radial cut-off values which reflect the physical limit of the beam radius. For the upstream layer, the cut-off value can be considered to be 32.5 mm which is the radius of the thin gold target support frame (beyond this radius the muon would have to go through more than a millimeter of copper). As for the downstream layer, which is directly facing the upstream layer, there is no collimation of the $\mu t$ beam due to the target support frame, hence an R cut-off of 35 mm in the $X$ direction (from the external rectangular size of the gold plated copper frame) and slightly larger in the $Y$ dimension can be expected. As can be seen from Fig. 6.1, the average thickness depends on beam profile, especially if the beam width is large.

Since the knowledge of the thickness is very important, all the available information
Figure 6.3: Similar to Fig. 6.2 except simulations are with the initial muon distribution being Gaussian, with flat top. The histograms, from inside out, show the beam distribution of $8\oplus8$, $10\oplus10$, $12\oplus12$, $15\oplus15$, where first number is the flat top radius $R_{\text{flat}}$ and the second is the full width at half maximum of the Gaussian part $\text{FWHM}_g \equiv 2.355 \times \sigma_g$ (see Eq. 6.3).

needs to be combined to determine the effective thickness. We attempted this in the following manner: (1) first parameterize the muon beam distribution (in the $XY$ plane) from the image of decay electrons obtained by the MWPC system, (2) then use that as an input to the Monte Carlo simulation to calculate the distribution of the $\mu t$ beam reaching the downstream layer, and (3) finally take a weighted average for each of the upstream and downstream layers, using the assumed beam profiles. Note that depending on the thickness of the upstream moderator, the $\mu t$ beam profile at the downstream layer, hence the effective thickness, could be different.

The knowledge of the beam profile is also necessary for the determination of the silicon
Table 6.1: The effective upstream (US) layer thickness with different beam parameters. Also shown is the corresponding acceptance $\Omega_{Si}$, for the silicon detector.

detector acceptance, which is tabulated in Tables 6.1-6.3, together with the effective thicknesses, but will be discussed in the following section.

Figures 6.2 and 6.3 compare the experimental data and simulations, with different input parameters, of the $Y$ distribution of the decay electrons image in the upstream layer. The data were obtained via the MWPC imaging system, while the Monte Carlo code, SMC, simulated the imaging process in the detector with the initial muon beam stopping distribution and the wire chamber resolutions as input parameters. In Fig. 6.2, Gaussian distributions with varying FWHM were assumed for the initial beam distribution in the $XY$ plane, while flat-top Gaussian distributions, defined by Eq. 6.3, with varying $R_{flat}$ and $\sigma_g$ were used for Fig. 6.3. The Gaussian beam of FWHM $20 \sim 25$ mm, and the flat-top Gaussian with the flat top radius $R_{flat}$ of $10 \sim 12$ mm and Gaussian FWHM $g$ of $10 \sim 12$ mm seem to reproduce the experimental data rather well. The resulting effective thicknesses are summarized in Table 6.1. The variation in the thicknesses with these values of beam parameters is less than 3%. The wire chamber resolution, $\sigma_{y}^{WC}$, of 1 mm is used for this analysis, but variation of the wire chamber resolutions between 0.2 mm and 4 mm did not affect this conclusion.

\[1\] The subscript $g$ is given here to stress it is the width only of the Gaussian part of the beam, as opposed to that of the entire beam.
The profile of the $\mu t$ atomic beam reaching the downstream layer generally depends on, but differs from, that of muon beam stopping in the upstream layer. The SMC, with all the physics in it, was used to simulate the former, using the latter as SMC input. The resulting profiles were parameterized similarly to the upstream case.

Figure 6.4 illustrates an example of the simulated radial profiles of the $\mu t$ beam reaching the downstream layer (plotted with error bars), for which the input to the simulation of a flat-top Gaussian beam, with $R_{\text{flat}} = 12$ mm and $\text{FWHM}_g = 12$ mm, was assumed for the upstream beam profile. Shown as a histogram is a parameterization of that profile using a flat-top Gaussian function. The flat radius of 4 mm and Gaussian FWHM of 28 mm give a reasonable $\chi^2$ per degree of freedom (DOF) of 1.05. If a Gaussian
distribution is assumed, a FWHM of 32.4 mm with $\chi^2$/DOF of 1.80 was obtained in the fit.

Table 6.2 summarizes the effective thickness for the downstream layer with the different US beam distribution. For the US beam of 10×10 mm, a single Gaussian function fits the simulated $\mu t$ distribution entering the DS target reasonably well, but for larger US widths, the fitted FWHM values depend on the region of the fit (e.g. [-29;29] mm vs. [-35;35] mm), and a flat-top Gaussian function gave better $\chi^2$ as well as a more stable fit. The different DS beam parameters for the same US parameter in the table reflect variations due to the fitting region size. The deuterium overlayer of 48.3 ($\sim 14$ T·l) was assumed for the simulations.

As discussed in Section 3.3.2 in Chapter 3 (see Fig. 3.16 in page 86), the layer thickness depends on the distance between the diffuser surface and the target foil surface according to our film deposition simulations. Although the distance for the upstream layer was similar to the calibration measurements in Section 3.3 ($\sim 8$ mm), that for the downstream foil was closer, i.e. $\sim 2.8$ mm. Therefore the calibration factors have to scaled by a factor 1.106±0.034. This factor was determined by comparing the areas between the simulations for the foil distance 8 mm and 2.8 mm in Fig. 3.16. About 3% uncertainty in the scaling is due to the choice of the interval within which the ratio was taken; [-15, 15] mm and [-30, 30] mm are the two extreme intervals considered, and the average between them was used as the scaling factor.

In general, the $\mu t$ beam profile at the downstream layer depends on the thickness of the overlayer in the upstream layer. This is partly due to the more forward peaked angular divergence of $\mu t$ going through the overlayer, and also because the moderated $\mu t$ with larger angles are less likely to survive to reach the downstream layer due to the longer flight path. This effect was investigated in Table 6.3 with the US beam of 10×10 mm assumed, and found to have about 6% effect in the effective thickness.
Chapter 6. Analysis I – Absolute normalization

Table 6.2: The effective thickness and the silicon detector acceptance ($\Omega_{Si}$) for the downstream (DS) layer, evaluated with different parameterizations of the $\mu$ beam profile at DS, which in turn were simulated using different US profiles as input to the Monte Carlo. The upstream moderating overlayer of 14 T-l is assumed for the simulation. See the text for the details. The summary of effective thicknesses and Si solid acceptance will be given in Table 6.5.

<table>
<thead>
<tr>
<th>US beam (mm) $R_{flat}$ FWHM$_g$</th>
<th>DS beam (mm) $R_{flat}$ FWHM$_g$</th>
<th>Effective thickness ($\mu$g-cm$^{-2}$ (T-l)$^{-1}$)</th>
<th>$\Omega_{Si}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 10 0 28.8 3.60</td>
<td>2.44</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12 12 0 32.1 3.50</td>
<td>2.47</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0 33.8 3.45</td>
<td>2.48</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4 28 3.53</td>
<td>2.46</td>
<td></td>
<td></td>
</tr>
<tr>
<td>15 15 0 42.1 3.25</td>
<td>2.50</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0 37.2 3.36</td>
<td>2.53</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7.5 30 3.36</td>
<td>2.51</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0 20 0 28.0 3.63</td>
<td>2.43</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0 25 0 31.8 3.51</td>
<td>2.47</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 6.3: The effective thickness of the downstream (DS) layer with different upstream overlayer thickness, and the corresponding silicon detector acceptance. The US beam parameter of the flat-top Gaussian 10+10 mm was used as input for simulations.

<table>
<thead>
<tr>
<th>US overlayer thickness (T-l)</th>
<th>DS beam (mm) $R_{flat}$ FWHM$_g$</th>
<th>Effective thickness ($\mu$g-cm$^{-2}$ (T-l)$^{-1}$)</th>
<th>$\Omega_{Si}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 0 35.8 3.40</td>
<td>2.49</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3 0 32.1 3.50</td>
<td>2.47</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6 0 30.6 3.55</td>
<td>2.46</td>
<td></td>
<td></td>
</tr>
<tr>
<td>14 0 28.7 3.60</td>
<td>2.44</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

6.2 Silicon detector acceptance

The silicon detector acceptance depends both on the geometry and the distribution of the beam (which determines the distribution of the particle source). The Monte Carlo method was used to determine the acceptance. We define our Si acceptance $\Omega_{Si}$ as the
Table 6.4: Variations in Si acceptance due to uncertainties in the geometry.

<table>
<thead>
<tr>
<th>Source</th>
<th>Uncertainty</th>
<th>Relative change in Si Acceptance</th>
</tr>
</thead>
<tbody>
<tr>
<td>beam X position shift</td>
<td>±5 mm</td>
<td>+2.7 %</td>
</tr>
<tr>
<td>beam Y position shift</td>
<td>±5 mm</td>
<td>-0.9 %</td>
</tr>
<tr>
<td>Si distance</td>
<td>-0.5 mm</td>
<td>+2.1 %</td>
</tr>
<tr>
<td>Si distance</td>
<td>+0.5 mm</td>
<td>-1.9 %</td>
</tr>
<tr>
<td>Si Z position shift</td>
<td>-1 mm</td>
<td>-2.4 %</td>
</tr>
<tr>
<td>Si Z position shift</td>
<td>+1 mm</td>
<td>+1.7 %</td>
</tr>
<tr>
<td>collimator width</td>
<td>-0.2 mm</td>
<td>-1.4 %</td>
</tr>
<tr>
<td>collimator width</td>
<td>+0.2 mm</td>
<td>+1.5 %</td>
</tr>
<tr>
<td>Total uncertainty</td>
<td></td>
<td>+4.1 %</td>
</tr>
</tbody>
</table>

The position of the silicon detectors is rather well defined, since they are mounted on the thermal shield box made of gold plated copper. A copper collimator sheet (aperture of 13.9 ± 0.2 mm × 50 ± 0.2 mm at room temperature) in front of the detector, further defined the acceptance. Thermal contraction, for both the thermal shield box and the collimator sheet, is taken into account using the values given in Ref. [214] and is estimated to give a combined relative correction of about 1.3% to the Si acceptance. Uncertainties due to the possible variations in the geometry are summarized in Table 6.4, where the total uncertainty is given as a quadratic sum of the all the uncertainties. Note that shifting the beam with respect to the target centre in the X direction always increases the solid angle (the two detectors are symmetric to the beam on the X axis), while shifting in the Y direction always reduces it. The variations of the silicon detector acceptance due to the beam distribution and its parameterizations were already given in Tables 6.1 and 6.2, for the upstream layers and the downstream layers, respectively.
Chapter 6. Analysis I – Absolute normalization

<table>
<thead>
<tr>
<th>Target</th>
<th>Effective thickness ((\mu g \cdot cm^{-2} \cdot (T \cdot l)^{-1}))</th>
<th>(\Omega_{Si}) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>US</td>
<td>3.43 ± 0.18</td>
<td>2.375 ± 0.086</td>
</tr>
<tr>
<td>DS (0 T \cdot l US)</td>
<td>3.34 ± 0.22</td>
<td>2.512 ± 0.105</td>
</tr>
<tr>
<td>DS (3 T \cdot l US)</td>
<td>3.44 ± 0.23</td>
<td>2.486 ± 0.089</td>
</tr>
<tr>
<td>DS (6 T \cdot l US)</td>
<td>3.49 ± 0.23</td>
<td>2.474 ± 0.103</td>
</tr>
<tr>
<td>DS (14 T \cdot l US)</td>
<td>3.54 ± 0.23</td>
<td>2.461 ± 0.088</td>
</tr>
</tbody>
</table>

Table 6.5: Summary of effective average thickness and Si acceptance for all the target arrangements used in this thesis.

Except for the case of the US beam of size 15×15 mm which appears inconsistent with the electron imaging data (Fig. 6.3), the variations of the Si acceptance due to the beam parameters are found to be less than 1%.

Given in Table 6.3 are the changes in the Si acceptance due to varying the upstream overlayer thicknesses, in which about 2% relative difference is found between 14 T \cdot l overlayer and no overlayer.

Table 6.5 summarizes the final values and their uncertainties for the effective thickness and Si acceptance for all the target arrangements used in this thesis. The uncertainties for the thickness include the ones due to the beam parameterization, the scaling factor for the diffuser distance (DS only) and the stopping power, which were added quadratically. Uncertainties for \(\Omega_{Si}\) are due to the geometry and the beam parameterization, also added in quadrature.
6.3 Muon stopping fraction determination

In order to obtain the normalization, we need to know the fraction of muons which stop in the hydrogen target with respect to the number of entering muons counted by the beam counter T1. Since our layer is rather thin, some of the muons stop in the Au target foils or possibly in the Cu thermal shield. We attempted to determine this fraction by two different methods, both of which use the decay electron time spectrum: (i) the amplitude ratio of different lifetime components in the electron time spectrum (Section 6.4: amplitude ratio method), and (ii) an absolute measurement of the number of electrons decaying with the hydrogen lifetime, with the detection efficiency determined using fusion signals in the silicon detectors (Section 6.5: absolute amplitude method). These methods are largely independent, but we initially observed an apparent discrepancy of some 20% between the two methods. This is why we go into great detail for the stopping fraction determination in the sections that follow.

6.4 Amplitude ratio method

The first method takes advantage of the fact that the lifetime of the muon is different in hydrogen and gold, being about 2.2 $\mu$s and 70 ns, respectively. The electron time spectrum is measured with a pair of electron scintillation counters surrounding the target chamber (En1, En2, and Ege), and with NE213 neutron detectors (N1 and N2) with their fast dynode output (Tel1 and Tel2) in coincidence with the scintillator pair hit in front of them.

The time of the first hit among En1, En2, or Ege (first E), as well as the first hit of Tel1 and Tel2 (first Tel), are also histogrammed, which if we accept only one muon at a time, and if the effect of background is small, should equal to the sum of En1, En2 and Ege, or Tel1 and Tel2 respectively.
The recorded time spectra were fitted to exponential functions with a constant background\textsuperscript{2}. The general form of the fitting function, taking into account capture and other muon losses, as well as the finite size of the time bin, is derived in Eq. 6.4 in Appendix C:

\[
\mathcal{F}^{fit}(t_i) = \sum_{k}^{M} \epsilon_k \frac{Q_k \lambda_0}{\lambda_k} N_k^0 \times \left\{ \exp\left[ -\lambda_k \left( t_i - \frac{1}{2} \Delta t \right) \right] - \exp\left[ -\lambda_k \left( t_i + \frac{1}{2} \Delta t \right) \right] \right\} + \text{bkgd}, \tag{6.4}
\]

where for kth element (\(k = 1 \ldots M\)), \(\epsilon_k\) is the electron detection efficiency, \(Q_k\) the Huff factor, \(N_k^0\) the initial muon population, and \(\lambda_k\) the muon disappearance rate in element \(k\), with \(\lambda_0\) being that in free space. The above \(\mathcal{F}^{fit}\) is a function of a discrete time variable \(t_i\) with interval \(\Delta t\).

Let us consider the specific case of \(M = 2\) (two exponentials), where we assume all observed muon decay takes place in one of the two materials. Also we take \(k = 1\) for a heavy element such as gold present in the system, and \(k = 2\) for hydrogen\textsuperscript{3}. We define the fraction of muon stopping in hydrogen \(S_H\), hence \(N_1^0 = N^0(1 - S_H)\) and \(N_2^0 = N^0 S_H\). In actual fits, the time histogram is normalized to GMU (number of “good muons”), and thus the fit function is defined to give, together with the decay rates \(\lambda_k\), the normalized amplitudes \(A_k \equiv \epsilon_k Q_k N_k^0 \text{/GMU}\). Hence, the stopping fraction can be determined from the ratio

\[
S_H^{AR} = \frac{N_2^0}{N_1^0 + N_2^0} = \frac{A_2}{\frac{A_1}{\epsilon_1 Q_1} + \frac{A_2}{\epsilon_2 Q_2}}, \tag{6.5}
\]

the superscript “AR” indicating the stopping fraction being derived from the amplitude

\textsuperscript{2}The validity of assuming a constant background will be discussed later.

\textsuperscript{3}A fit with \(M = 3\) was also performed to investigate a possible existence of the copper component in the time spectrum, with two of the three lifetimes fixed to those of Au and Cu from Ref. [215], but this did not affect the derived hydrogen stopping fraction. Fixing of two lifetimes was necessary to avoid a fitting problem due to a strong correlation between them.
ratio. We define a reduced stopping fraction $S_H^{AR}$ as,

$$S_H^{AR} = \frac{A_2}{A_1/Q_1 + A_2},$$

(6.6)

which would equal $S_H^{AR}$, if we assume (a) $Q = 1$ for hydrogen, and further (b) $\epsilon_1 = \epsilon_2$. The assumption (a) is valid since the binding energy of muonic hydrogen ($\sim 2.5$ keV) is much smaller than the average electron energy ($\sim 35$ MeV), but (b) needs careful consideration, which I shall revisit later.

### 6.4.1 Electron scintillator measurements

Shown in Fig. 6.5 is an example of the electron time spectrum and fit with two exponential functions with a constant background. Plotted with error bars are the first electrons detected by electron pair scintillators for a target of 1000 $T\cdot l$ pure hydrogen, while the solid curve is a fit in the time interval of [0.02, 6] $\mu$s.

The resulting fit amplitudes ($A_1/Q_1$, $A_2$, and bkgd), normalized to GMU, and the lifetimes ($1/\lambda_1$ and $1/\lambda_2$) are given in Table 6.6, for the fit from each detector (Ege, En1, and En2) as well as that from the first hit in the three detectors (1st). Also shown is the reduced muon stopping fraction in hydrogen ($S_H^{AR}$) derived from $A_2/(A_1/Q_1 + A_2)$ with the Huff factor for Au of $Q_1 = 0.85$. The data of runs 1650 and 1654, taken with the same conditions, were analyzed both separately and together.

Fits are mostly of satisfactory quality, but in some cases we observed rather low confidence levels. This could be in part due to beam related background which is correlated to the 23 MHz (43 ns) cyclotron RF cycle. This periodic background, when fitted with a straight line, would increase the total $\chi^2$, but should not affect the fit parameters for the signals, since it is the average over the relevant time scale that affects the fit results$^4$.

$^4$The time scales for the RF cycle and the gold signal are rather similar, but the background amplitude is several orders of magnitude smaller, so the effect should be negligible.
Figure 6.5: An example of electron time spectrum and fit with two exponential functions with a constant background. Plotted with error bars are the first electrons detected by electron pair scintillators, while the solid curve is a fit in the time interval of [0.02, 6] µs.

Our fitted short lifetime $1/\lambda_1$ is reasonably consistent with $74.3\pm1.5$ ns for gold given by Suzuki, Measday and Roalsvig [215], but the long lifetime $1/\lambda_2$ is somewhat shorter than $2.195$ µs for hydrogen [215].

In the single hit mode\(^5\), if the detection efficiency is high, there could be a distortion in the recorded accidental background, since the detector is more likely active at earlier time. But the individual detector fits and the fit to the first hit among them ("1st") are consistent with each other, indicating that the effect of accidental background in distorting the fit is negligible ("1st" can be considered as one single-hit detector with an efficiency three times as large as a single detector, hence if an efficiency dependent

\(^5\)In this mode, the detector can accept only one event per incident muon, as opposed to the multi hit mode in which multiple events per incident muon can be recorded.
Chapter 6. Analysis I – Absolute normalization

Table 6.6: The result of a fit to the electron time spectrum with two exponential functions with a constant background, and the muon stopping fraction to hydrogen ($S_H^{AR}$) derived from the amplitude ratio, $A_2/(A_1/Q_1 + A_2)$ where $Q_1 = 0.85$ is the Huff factor for Au. The fit amplitudes ($A_1/Q_i$, $A_2$, bkgd) are normalized to GMU. Fits to the spectrum from each detector (Ege, En1, En2) as well as that for the first hit in the three detectors (1st) are listed.

<table>
<thead>
<tr>
<th>Run</th>
<th>Det.</th>
<th>$A_1/Q_1$ (10^{-2})</th>
<th>$A_2$ (10^{-2})</th>
<th>bkgd (10^{-4})</th>
<th>$1/\lambda_1$ (ns)</th>
<th>$1/\lambda_2$ (µs)</th>
<th>$S_H^{AR}$ (%)</th>
<th>$\chi^2$/dof(cl)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1650</td>
<td>Ege</td>
<td>5.06(13)</td>
<td>1.760(9)</td>
<td>2.4(4)</td>
<td>76.9(20)</td>
<td>2.030(23)</td>
<td>25.81(50)</td>
<td>0.99 (71%)</td>
</tr>
<tr>
<td></td>
<td>En1</td>
<td>4.86(13)</td>
<td>1.734(9)</td>
<td>2.3(4)</td>
<td>76.0(21)</td>
<td>2.027(24)</td>
<td>26.28(52)</td>
<td>1.02 (18%)</td>
</tr>
<tr>
<td></td>
<td>En2</td>
<td>4.86(12)</td>
<td>1.563(9)</td>
<td>2.9(4)</td>
<td>79.0(21)</td>
<td>2.041(26)</td>
<td>24.34(48)</td>
<td>1.05 (3.8%)</td>
</tr>
<tr>
<td>1st</td>
<td></td>
<td>14.71(22)</td>
<td>5.061(17)</td>
<td>6.9(7)</td>
<td>77.1(12)</td>
<td>2.029(15)</td>
<td>25.59(30)</td>
<td>1.09 (.03%)</td>
</tr>
<tr>
<td>1654</td>
<td>Ege</td>
<td>4.72(12)</td>
<td>1.770(10)</td>
<td>2.5(4)</td>
<td>80.7(22)</td>
<td>2.003(23)</td>
<td>27.28(51)</td>
<td>1.01 (32%)</td>
</tr>
<tr>
<td></td>
<td>En1</td>
<td>4.71(12)</td>
<td>1.722(9)</td>
<td>2.1(4)</td>
<td>77.8(21)</td>
<td>2.043(24)</td>
<td>26.75(51)</td>
<td>0.99 (69%)</td>
</tr>
<tr>
<td></td>
<td>En2</td>
<td>4.95(13)</td>
<td>1.584(9)</td>
<td>3.0(4)</td>
<td>74.9(20)</td>
<td>2.012(24)</td>
<td>24.26(48)</td>
<td>1.04 (7.0%)</td>
</tr>
<tr>
<td>1st</td>
<td></td>
<td>14.30(21)</td>
<td>5.081(16)</td>
<td>6.8(7)</td>
<td>77.6(12)</td>
<td>2.017(13)</td>
<td>26.22(29)</td>
<td>1.02 (26%)</td>
</tr>
<tr>
<td>1650</td>
<td>Ege</td>
<td>4.88(9)</td>
<td>1.765(7)</td>
<td>2.4(3)</td>
<td>78.8(15)</td>
<td>2.016(16)</td>
<td>26.56(36)</td>
<td>0.99 (62%)</td>
</tr>
<tr>
<td></td>
<td>En1</td>
<td>4.77(9)</td>
<td>1.728(7)</td>
<td>2.2(3)</td>
<td>76.9(15)</td>
<td>2.035(17)</td>
<td>26.52(37)</td>
<td>1.04 (8.6%)</td>
</tr>
<tr>
<td></td>
<td>En2</td>
<td>4.90(9)</td>
<td>1.574(7)</td>
<td>2.9(3)</td>
<td>76.9(15)</td>
<td>2.025(19)</td>
<td>24.30(36)</td>
<td>1.13 (.01%)</td>
</tr>
<tr>
<td>1st</td>
<td></td>
<td>14.50(16)</td>
<td>5.071(12)</td>
<td>6.8(5)</td>
<td>77.4(9)</td>
<td>2.023(10)</td>
<td>25.91(21)</td>
<td>1.09 (.04%)</td>
</tr>
</tbody>
</table>

effect was important in the fit, it would show up as the difference between the 1st and other detectors). In fact, fits with explicitly non-constant backgrounds were tried, but the stopping fraction was found rather insensitive to the background model.

The dependence of the derived stopping fraction $S_H^{AR}$ was tested by changing the fit region from [0.02;6] µs to [0.02; 9.5] µs. While the H$_2$ component lifetime was slightly increased (~ 2%), the variation in stopping fraction was negligible.

We have also fitted the later time $t > 1$ µs, at which the gold signal is negligible, to a single exponential and a background. In these fits, in which 100 ns bin size was used (averaging out the RF structure), a lifetime of 2.11(1) µs was obtained with a confidence level of 8%, a lifetime closer to, yet still significantly smaller than, the literature value for muons in hydrogen. An exponential background was also tried, giving $1/\lambda_2 = 2.07(3)$ µs.
with a background lifetime of 0.0829 $\mu$s with a confidence level of 11%\textsuperscript{6}. The amplitude for the hydrogen component thus derived is somewhat smaller than the ones in Table 6.6, but by no more than 3%.

Alternatively, fits were tried with the lifetime fixed to 2.195 $\mu$s and with a constant background (varying the amplitude) or an exponential background (varying the lifetime and the amplitude). Neither background gave a satisfactory fit, suggesting that the deviation of the lifetime is unlikely due to a trivial error in modeling the background, thus pointing to the existence of one or more muon loss mechanisms. This could include a small amount of non-hydrogenic contamination in the target, which would not affect the extraction of the stopping fraction, as long as it is well approximated with an exponential function. Emission of muonic protium from the solid layer, recently observed by our collaboration for the first time \cite{216}, could be partly responsible for the discrepancy also. According to Wozniak \cite{217}, several percent of muonic protium atoms may be emitted back to the gold foil, whereby the muon transfers and is captured by the gold nucleus.

\textsuperscript{6}One could conceive the background to have a form $\text{exp}(-\epsilon \lambda t)$, where $\epsilon$ is the detection efficiency and $1/\lambda$ the signal lifetime, if the background is an accidental one. In fact, $0.0829/\lambda_2 = 0.172$, derived from the above, is close to the combined efficiency for the 1st electron obtained in Table 6.17 in page 157.

<table>
<thead>
<tr>
<th>$\delta t_0$ (ns)</th>
<th>$\overline{S}_H^{AR}$ (%)</th>
<th>$(\delta \overline{S}<em>H^{AR})</em>{rel}$</th>
<th>$\chi^2/dof$</th>
<th>$\text{cl}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>-4</td>
<td>27.07 (23)</td>
<td>4.4%</td>
<td>1.09</td>
<td>0.053</td>
</tr>
<tr>
<td>-2</td>
<td>26.45 (22)</td>
<td>2.1%</td>
<td>1.09</td>
<td>0.045</td>
</tr>
<tr>
<td>-0</td>
<td>25.91 (21)</td>
<td>0 %</td>
<td>1.09</td>
<td>0.043</td>
</tr>
<tr>
<td>+2</td>
<td>25.38 (21)</td>
<td>-2.0%</td>
<td>1.09</td>
<td>0.041</td>
</tr>
<tr>
<td>+4</td>
<td>24.85 (20)</td>
<td>-4.1%</td>
<td>1.09</td>
<td>0.039</td>
</tr>
</tbody>
</table>

Table 6.7: Sensitivity of the reduced stopping fraction $\overline{S}_H^{AR}$ to the shift of time zero ($\delta t_0$) for the first electron time spectrum for the sum of runs 1650 and 1654. $(\delta \overline{S}_H^{AR})_{rel}$ is the relative shift with respect to the $\delta t_0 = 0$ value. The variation in $\chi^2/dof$ is smaller than the last digit shown.
Table 6.8: The results of fit to the electron telescope time spectrum for the sum of Runs 1650 and 1654, to be compared with the scintillator fit in Table 6.6. The fits for Telescope 1 (Tell), located on the beam left, and Telescope 2 (Tel2) downstream, together with first hit of the two (1st) are listed.

The sensitivity of $S_H^{AR}$ to the shift of the time zero definition ($\delta t_0$) was also investigated. An example for the first electron spectrum is given in Table 6.7. The fit to the individual detectors as well as to individual runs gives similar trends. The error $\delta t_0 = \pm 4$ ns is a rather conservative estimate of the shift, and it is more likely to be less than $\pm 2$ ns. The variation in the amplitude for the hydrogen component in this time scale is, nevertheless, completely negligible, hence the change in $S_H^{AR}$ comes from that in the gold component. In any case, this alone cannot explain the 20% difference between method 1 and method 2, as will be discussed later.

While $S_H^{AR}$ for Ege and En1 are consistent with each other, there is a systematic difference between En2 and the others, with En2 being relatively lower by nearly 10%. This discrepancy will be addressed later in relation to the acceptance difference for the upstream and downstream targets.

We note that agreement between Ege and En1, which were symmetrically located across the target (beam-left and beam-right, respectively), exclude the possible effects of polarized muon precession about the vertical axis.

### 6.4.2 Electron telescope measurements
Chapter 6. Analysis I – Absolute normalization

The liquid NE213 scintillation detectors, normally used for fusion neutron detection, can be used as charged particle detectors by reading out a fast dynode signal. We define a telescope event as the event in which the dynode fires in coincidence with the pair of plastic scintillators in front of it. Telescope 1 (Tell1) is in the beam right position (as is Neutron 1 or N1), while Telescope 2 (Tel2) was in the downstream position; both used an independent data acquisition trigger.

Given in Table 6.8 are the results of fits to the telescope spectra and derived reduced stopping fractions \(S_H^{AR}\). The fits were performed in the time region of [0.02; 9.5] \(\mu\)s, and the time spectra were obtained with a “nominal” energy cut on the dynode output, which was \(200 \text{ ch} < E_{dy} < 1200 \text{ ch}\). Fits in this section are done to the sum of runs 1650 and 1654. As can be observed in a comparison of Tables 6.6 and 6.8, the stopping fractions given by the telescopes were systematically higher than those from the electron scintillators. We shall look into possible effects due to the difference in the energy sensitivity of the detectors in the following section.

Energy dependence of the stopping fraction

We plot in the top of Fig. 6.6 the telescope energy spectra, e.g., the neutron detector dynode output, recorded in coincidence with the corresponding scintillator pair. The time cut of \(t > 0.02 \mu\)s was applied to remove the prompt beam signal (which would otherwise slightly increase counts at energies less than 200 ch). With the minimum ionizing energy loss \(dE/dx \sim 2 \text{ MeV/}(\text{g/cm}^2)\) for scintillator materials, and our detector depth being about 10 cm (4 inches), the minimum ionizing electrons deposit energy of order 20 MeV. The muon decay electrons (in free space) range from 0 to about 53 MeV in energy, hence some would stop in the detector, while others would go through. Thus, the peak in Fig. 6.6 (above) near channel 700 should be due to the minimum ionizing electrons corresponding to some 20 MeV. Detailed GEANT calculations indeed confirm
Figure 6.6: (Top) The energy of the dynode output for the telescope events. The peak near 700 is due to minimum ionizing electrons going through the detector, depositing nearly 20 MeV. (Bottom) Energy dependence of $\frac{S_H^{AR}}{A} (=A_2/(A_1/Q_1 + A_2))$, the muon stopping fraction in hydrogen obtained from the ratio of amplitudes, in a two-exponential lifetime fit to the telescope time spectrum. The width of the histogram bins roughly corresponds to the intervals of the energy cuts to which each fit was performed.
Chapter 6. Analysis I – Absolute normalization

this picture [218].

The bottom figure in Fig. 6.6 illustrates the reduced muon stopping fraction in hydrogen determined from the amplitude ratio of a two-exponential lifetime fit, $S_{H}^{AR}$ ($= A_2/(A_1/Q_1 + A_2)$), as a function of the energy deposit in the NE213 detector. The fits were performed with the various energy cuts, with their intervals indicated roughly by the position and width of the histogram bars. This clearly demonstrates that the stopping fraction determined from the amplitude ratio is dependent on the energies to which the detector is sensitive. Possible mechanisms for this energy dependence are considered in the following sections, and will lead to the corrections to $S_{H}^{AR}$.

6.4.3 Corrections to $S_{H}^{AR}$ in the amplitude ratio method

In this section, we investigate the possible corrections to the reduced muon stopping fraction in hydrogen $S_{H}^{AR}$ derived from the amplitude ratio, in an attempt to explain the observed energy dependence of $S_{H}^{AR}$, as well as to resolve the discrepancy with the absolute amplitude method (Section 6.5).

Charged particles from muon capture

Muon capture on a proton,

$$\mu^- + p \rightarrow \nu_\mu + n,$$  \hfill (6.7)

is an elementary process (at least at the nucleon level), but when the muon is captured on a nucleus the process is rather complex. Muon capture in a heavy nucleus often leads to the emission of particles, normally neutrons but sometimes charged particles. These have the same time constants as the decay electrons, hence if detected in either the scintillator pairs or the telescope, they would bias the determination of the stopping fraction from the amplitude ratio.
The detection of charged particles would modify the branching ratio of muon decay versus total muon disappearance $B_k$ (Eq. C.4) into an effective branching ratio $\tilde{B}_k$, 

$$\tilde{B}_k = \frac{Q_k \lambda_0 + \lambda^C_k \sum m W_{mp}}{Q_k \lambda_0 + \lambda^C_k + \lambda^X_k}, \quad (6.8)$$

where $W_{mp}$ are fractional probabilities per muon capture for emission of $m$ charged particles. Alternatively, the effect can be incorporated as the efficiency for the electron detection $\varepsilon_k$ in Eq. 6.5 

$$\tilde{\varepsilon}_k = \varepsilon_k \left( 1 + \frac{\lambda^C_k \sum m W_{mp}}{Q_k \lambda_0} \right) \quad (6.9)$$

$$= \varepsilon_k \beta_k \quad (6.10)$$

with $\beta_k \equiv \tilde{\varepsilon}_k/\varepsilon_k$ defined as a correction factor.

Since the ratio $\lambda^C_k/Q_k \lambda_0$ is $\sim 34$ for the case of $k = \text{gold}$, a relatively small fraction of charged particle emission can affect the stopping fraction determination, hence this effect needs to be considered in some detail.

The charged particle emission following muon capture has been traditionally less studied compared to neutral emission, but there is some recent interest in relation to nucleon pairing in nuclei. A few reviews exist on this subject [219–221]. Following muon capture, the emission of neutrons (and gammas) dominates over that of charged particles ($p, d, \alpha$ etc.), as the latter requires a multi-step reaction or capture on correlated nucleons, since the process (6.7) only gives outgoing neutrons. The total probability of the charged particle emission decreases with increasing $Z$ of the nucleus, due to the increasing Coulomb barrier [219,220], but there is some evidence from recent measurements that the yield of the highest energy protons is independent of $Z$ [221].

Table 6.9 lists measured and calculated probabilities for charged particle emission reactions ($\mu^-\tilde{p}xn$), where $x$ is the neutron multiplicity, and $\tilde{p}$ includes deuterons and
Chapter 6. Analysis I – Absolute normalization

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Probability per muon capture $W_p$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{63}\text{Cu}$</td>
<td>&gt;1.7±0.3</td>
</tr>
<tr>
<td>$^{181}\text{Ta}$</td>
<td>&gt;0.07±0.01</td>
</tr>
<tr>
<td>$^{208}\text{Pb}$</td>
<td>&gt;0.30±0.08</td>
</tr>
</tbody>
</table>

Table 6.9: Measured and calculated fractional probabilities (per captured muon) $W_p$ for inclusive charged particle emission reactions ($\mu^-, p\pi n$). (a) experimental lower limit for observed channels, (b) estimated total inclusive probabilities derived from (a) using approximate ratio of various channel, (c) and (d) theoretical calculations. (a), (b) and (c) are from Ref. [220], and (d) from Ref. [219].

We ignore the charged particle multiplicity in the following analysis, i.e., we let $\sum_m mW_{mp} = W_p$. Note that emission of $\alpha$ particles is suppressed by at least several times [220]. Assuming $W_p = 0.4\%$ for gold, we obtain $\varepsilon_{Au} = 1.14 \cdot \varepsilon_{Au}$ from Eq. 6.9, which in turn gives an upward correction to the apparent hydrogen stopping fraction $S_H^{AR}$. The magnitude of the correction varies from 0 to 14% (relative) as a decreasing function of $S_H^{AR}$, but for $S_H^{AR} = 25\%$, the corrected value will be $S_H^{AR} = 27.5\%$, a relative shift of 10%.

For copper which is also present in parts of the target system, the emission probability $W_p$ is nearly an order of magnitude higher than gold (Table 6.9). However, the fit to decay electron data with three exponential functions, two of which were fixed to the lifetimes corresponding to Au and Cu measured by Suzuki, Measday and Roalsvig [215], indicated that the apparent Cu component cannot be more than 10% (in most cases less
Table 6.10: Integrated yields of proton emission following muon capture, based on the compilation in Ref. [221]. $E_{th}$ is the energy threshold above which the yield is integrated. $E_0$ is derived from a fit of the energy spectrum to the form $\exp(-E/\tau_0)$.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Authors</th>
<th>$E_{th}$ (MeV)</th>
<th>$\Gamma_{E_{th}}$ ($10^{-4}$)</th>
<th>$E_0$ (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S</td>
<td>Baladin et al. (1979) [222]</td>
<td>40.5</td>
<td>2.07±0.15</td>
<td>6.4±0.1</td>
</tr>
<tr>
<td>Ca</td>
<td>Martoff et al. (1991) [223]</td>
<td>40</td>
<td>2.32±0.26</td>
<td>8.0±1.5</td>
</tr>
<tr>
<td>Cu</td>
<td>Budyashov et al. (1971) [224]</td>
<td>42.5</td>
<td>3.8±1.4</td>
<td>13.1±1.6</td>
</tr>
<tr>
<td>Cu</td>
<td>Krane et al. (1979) [225]</td>
<td>40</td>
<td>19.6±1.2</td>
<td>8.3±0.5</td>
</tr>
<tr>
<td>Y</td>
<td>Cummings (1991) [221]</td>
<td>40</td>
<td>0.72±0.07</td>
<td>7.2±1.0</td>
</tr>
<tr>
<td>Pb</td>
<td>Krane et al. (1979) [225]</td>
<td>40</td>
<td>1.71±0.28</td>
<td>9.9±1.1</td>
</tr>
</tbody>
</table>

than a few percent) of gold. Furthermore, the capture rate for copper is less than half that of gold, while the Huff factor for copper is larger. All this suggests that the relative contribution of charged particles from copper is significantly smaller than that from gold and thus is neglected.

Nevertheless, the estimate of 14% effect on $\epsilon_{Au}$ is a worst possible case of detecting all the charged particles produced. In reality, the emitted particles can be ranged out in the walls and other materials, hence the effect is expected to be smaller. This of course depends on the energy spectrum of the emitted charged particles, which we discuss in the following.

Table 6.10, based on a compilation by Cummings [221], lists previously measured yields $\Gamma_{E_{th}}$ of high energy protons following muon capture, integrated above a threshold energy $E_{th}$. According to Cummings [221], some of the older results by Budyashov et al. and by Krane et al. may be unreliable, and excluding those would suggest high energy proton yields of $1-2\times10^{-4}$, independent of the atomic number.

For the En2 counter located downstream of the target, there is substantial material
between the gold target support and the detector, including 1.6 mm gold-plated copper, 2.5 mm stainless steel and 3 mm Plexiglas. According to the ICRU range table [190], the copper and stainless steel alone can stop protons up to ~ 50 MeV. Therefore, the effect of charged particles should be less than of the order of $10^{-3}$, which is completely negligible for the $S_H$ determination.

On the other hand, the amount of material between the gold and electron counters on the sides of the target (Ege, Enl) is considerably less, in particular, for the paths which go through the silicon detector. A rough estimate based on the range table [190] suggests that protons of energies above 20 to 25 MeV may reach the second scintillator E2ge (providing a coincidence of the scintillator pair) after going through the silicon detector (300 µm), a copper thermal shield foil (13 µm), a stainless steel window (76 µm), and the first scintillator E1ge (3.2 mm) with three layers of mylar wrappings (3 × 285 µm) (recall that $E_{ge} = E_{1ge} \cdot E_{2ge}$).

Assuming the proton spectrum from the gold is similar to that from yttrium given by Cummings [221] and using the partial yield above 40 MeV and the exponential decay rate in Table 6.10, the yield above 20 MeV, $\Gamma_{20}$ can be derived to be $2.3 \times 10^{-3}$. This would give $\beta_{Au} = \bar{\epsilon}_{Au}/\epsilon_{Au} = 1.08$ (see Eq. 6.9), which in turn would give a relative 6% correction, if $\Sigma_{AR}^{Eg} = 25\%$, to 26.5% from Eq. 6.5.

However, if the charged particle did not go through a Si detector, but hit the Cu collimator around the active Si area, it would be less likely to reach the scintillators. The relative acceptance of Ege (or Enl) compared to that of Si detector, which was assumed to be unity above, is actually less than a half, hence the effect of charged particles is expected to be smaller than the above, and of a few percent level for the side counters Ege and Enl.
Figure 6.7: The energy spectra of muon decay electrons, for the muon bound in lead and iron, calculated by Huff [226]. Also plotted is the electron spectrum for the muon decay in free space (without radiative corrections), $R(E) \propto 16E^2[3(1-2E) + \frac{2}{3}\rho(8E-3)]$, with the Michel parameter $\rho = 0.75$, and the maximum energy of 52.8 MeV. All spectra are normalized to a free muon decay rate of unity.

**Electron energy spectrum**

As was seen in the previous section, the effect of capture induced charged particles alone cannot explain the energy dependence of $S_H^\Delta$. Indeed, for the downstream counters, it was found very unlikely to cause any measurable effect. We therefore turn our attention to other effects in these two sections, i.e., electron energy spectra, and upstream-downstream acceptance difference.

The energy spectrum of decay electrons from muons bound in a nucleus is known to differ from that in free space, due to effects such as the final state Coulomb interaction and reduced phase space [226–228] (the same effects are responsible for the Huff factor
describing the reduction in decay rates). Shown in Fig. 6.7 are the electron energy spectra calculated by Huff [226], for muons bound in lead and iron, together with that for free muon decay. As all the spectra are normalized to a free muon decay rate of unity, the integral of the spectra gives the Huff factor \( Q = 0.844 \) for lead, 0.975 for iron. More recent and more elaborate calculations on the energy spectra [227,228] essentially agree with Huff's.

Figure 6.7 illustrates that the strength of the spectra above \( \sim 20 \text{ MeV} \), energies which would give a peak in the telescope spectrum (Fig. 6.6), is significantly larger for free decay than for lead (which are similar to decay for hydrogen and for gold, respectively). This would cause the amplitude ratio for hydrogen \( \overline{S}_{H}^{AR} \) to increase at higher energies. At lower energies, on the other hand, the spectrum is stronger for lead than for free decay, which would decrease \( \overline{S}_{H}^{AR} \). Thus the difference in the electron energy spectra shown in Fig. 6.7 provides an explanation, at least in part, for the energy dependence of the amplitude ratio \( \overline{S}_{H}^{AR} \) observed in Fig. 6.6.

Relative acceptance

In our stopping fraction measurements, most or all of the hydrogen was placed on the upstream target, while the muon can stop in gold at both upstream and downstream targets. The relative geometrical acceptance for the electrons from hydrogen and gold is expected to be similar for the counters at the sides (Ege, En1), but can be different for the downstream counters (En2). Taking this into account, the expression for the stopping fraction Eq. 6.5 should read:

\[
S_{H} = \frac{\frac{A_{2}}{\varepsilon_{2} \Omega_{2} Q_{2}}}{\frac{A_{1}}{\varepsilon_{1} \Omega_{1} (F_{u} + \kappa_{\Omega} F_{d}) Q_{1}} + \frac{A_{2}}{\varepsilon_{2} \Omega_{2} Q_{2}}}, \tag{6.11}
\]
where $F_u, F_d$ are relative fractions of muons which stops in the upstream and downstream gold foil, respectively ($F_u + F_d = 1$), and $\kappa_\Omega = \Omega_d/\Omega_u$ is the acceptance ratio. Obviously, when $F_d \ll 1$ or $\kappa_\Omega \sim 1$, the expression reduces to Eq. 6.5.

If the beam momentum width is larger than the corresponding thickness of gold foils and the hydrogen layer, one would very roughly expect $F_u \sim F_d$ for the beam momentum optimized for the maximum stopping in hydrogen. $\kappa_\Omega$ can be roughly estimated to be about 1.3 for En2 from the fact that target spacing between the upstream and the downstream layers is some 12% of the detector distance. Assuming a point source, this would make about a 10% upward correction to $S_H^{AR}$ (from 25% to 27.4%) for the downstream counter pair En2.

**Combined corrections**

We have looked above at the possible individual influence on the stopping fraction determination by charged particles following muon capture, decay electron energy spectra, and upstream-downstream acceptance variations. Here we combine all the factors to give a general expression. When the muons stop in $M$ different materials, located in $L$ different locations (such as upstream foil, downstream foil and so on), the fraction of the muon which stops in the $k$-th material, $S_k^{AR}$, determined from the ratio of the amplitudes $A_k$ for the electron spectrum lifetime fit, is

$$S_k^{AR} = \frac{\sum_l F_l^k \xi_l^k}{\sum_{k'} \left( \sum_l \frac{A_{k'}}{F_l^{k'} \xi_l^{k'}} \right)}$$

(6.12)
where \( F_l^k \) is the fraction of muons stopping at the location \( l \) for the specific material \( k \) (\( \sum_l F_l^k = 1 \)), and \( \xi_l^k \) is the effective efficiency for electron detection from muons stopped on material \( k \) at location \( l \),

\[
\xi_l^k = Q^k \varepsilon \Omega_l^k \rho_l^k \gamma_l^k,
\]

with \( Q^k \) the Huff factor for the material \( k \), \( \varepsilon \) the intrinsic detector efficiency, \( \Omega_l^k \) the detector acceptance for material \( k \) at location \( l \), and \( \beta_l^k \) and \( \gamma_l^k \) the corrections to the efficiencies due to charged particle emission and electron energy spectrum, respectively.

In our specific case where we assume \( M = 2 \) and hydrogen is present only at the upstream foil \( (F_{us}^H = 1, F_{\neq us}^H = 0) \), while the muons stop in gold both at the upstream and the downstream layer with the relative fraction \( F_{Au}^u, F_{Au}^d \) but nowhere else \( (F_{Au}^{\neq u,d} = 0) \), we can simplify the expression to

\[
S_{H}^{AR} = \frac{A^H}{A_{Au}} \frac{A_{Au}}{\eta (F_{us}^H + \kappa F_{ds}^H)} + A^H,
\]

where \( \eta \) is the ratio of the effective efficiencies for gold and hydrogen both at the upstream location, and \( \kappa \) that for the upstream and the downstream gold:

\[
\eta = \frac{\xi_{us}^{Au}}{\xi_{us}^{H}} = \frac{Q^{Au} \Omega_{us}^{Au} \beta_{us}^{Au} \gamma_{us}^{Au}}{Q^{H} \Omega_{us}^{H} \beta_{us}^{H} \gamma_{us}^{H}},
\]

\[
\kappa = \frac{\kappa_{us}^{Au}}{\kappa_{us}^{H}} = \frac{\Omega_{ds}^{Au} \beta_{ds}^{Au} \gamma_{ds}^{Au}}{\Omega_{ds}^{H} \beta_{ds}^{H} \gamma_{ds}^{H}}.
\]

The expressions above reduce to Eq. 6.11, if \( \eta \sim Q^{Au} \) (i.e., gold and hydrogen have the same efficiencies for the upstream except the Huff factor), and further to Eq. 6.6 if \( \kappa \sim 1 \) (the same efficiencies for upstream and downstream gold) or \( F_{ds}^{Au} \ll F_{us}^{Au} \) (negligible stopping in the downstream gold).
We consider the case for counters at the sides (Ege, En1) and downstream (En2) separately. For En2, if we assume $\beta^k = 1 \ (k = Au, H$ and $l = us, ds)$, and $\Omega^Au \sim \Omega^H_{us}$, we approximately have

\[
\eta(En2) \sim Q^Au \frac{\gamma^Au_{us}}{\gamma^H_{us}}, \quad (6.18)
\]

\[
\kappa(En2) \sim \frac{\Omega^{Au}_{ds}}{\Omega^{Au}_{us}}, \quad (6.19)
\]

On the other hand, for Ege, En1, assuming $\beta^A_{ds} \sim \beta^A_{us}, \Omega^A_{ds} \sim \Omega^A_{us}$, we have

\[
\eta(Ege, En1) \sim Q^Au \frac{\Omega^Au_{us} \beta^A_{us} \gamma^Au_{us}}{\Omega^H_{us} \beta^H_{us} \gamma^H_{us}}, \quad (6.20)
\]

\[
\kappa(Ege, En1) \sim 1, \quad (6.21)
\]

where there is some uncertainty in $\eta(Ege, En1)$ due to the charged particle emission effect $\beta^Au_{us} / \beta^H_{us}$. In addition, there is potentially a large error in the factor $\Omega^Au_{us} / \Omega^H_{us}$ due to the possible non-flatness of the gold foil which could cause significant shadowing of the electrons either from gold or hydrogen\(^7\). Thus, the use of the downstream counters En2, in which the shadowing effect will not be present and the charged particle emission effect is negligible, appears more reliable for the determination of the stopping fraction.

The factors $F^Au_{us}, F^Au_{ds}, \eta, \kappa$ in Eq. 6.15 were determined from detailed GEANT simulations [218] taking into account the full geometry. Simulation for the muon beam assuming a momentum of 27.0 MeV/c with $\Delta p/p$ of 5.7% yielded the gold stopping upstream and downstream to be $F_u = 0.48$ and $F_d = 0.52$. A separate series of simulations for the decay electrons using the energy spectrum for lead (Fig. 6.7) to represent that for gold, and the free muon to represent hydrogen gave the absolute detection efficiencies $\varepsilon \Omega^k_{l} \gamma^k_{l}$ ($k = H, Au; l = US, DS$), presented in Table 6.11. Note that only the relative efficiencies are relevant in this context.

\(^7\)The frame for the foil is rather thick, so the foils could be bent such that the view from the side is blocked by the foil frame. We have observed evidence for such effects in the November 1993 runs with the MWPC imaging system.
Table 6.11: The absolute efficiencies, $\varepsilon \Omega_k^k \gamma_k^k (k = \text{H, Au}; l = \text{US, DS})$ for electron detection, calculated by GEANT simulations [218]. Uncertainties given are statistical only.

<table>
<thead>
<tr>
<th>Efficiency</th>
<th>US</th>
<th>DS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ege</td>
<td>6.03(8)</td>
<td>4.88(7)</td>
</tr>
<tr>
<td>En1</td>
<td>6.14(8)</td>
<td>5.02(7)</td>
</tr>
<tr>
<td>En2</td>
<td>5.11(7)</td>
<td>4.06(7)</td>
</tr>
</tbody>
</table>

Table 6.12: The correction factors $\eta$, $\kappa$ in Eq. 6.15, and assumed quantities for the derivation by Eqs. 6.16, 6.17. The first row is the average of Ege and En1. Values with * are taken from the ratio of the efficiencies calculated by a GEANT simulation given in Table 6.11.

<table>
<thead>
<tr>
<th>Detector</th>
<th>$Q^\text{Au}$</th>
<th>$\Omega_{\text{us}}^\text{Au} / \Omega_{\text{us}}^\text{H}$</th>
<th>$\beta_{\text{us}}^\text{Au} / \beta_{\text{us}}^\text{H}$</th>
<th>$\beta_{\text{ds}}^\text{Au} / \beta_{\text{us}}^\text{Au}$</th>
<th>$\eta$</th>
<th>$\kappa$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ege+En1</td>
<td>0.85</td>
<td>0.813*</td>
<td>1.04</td>
<td>1</td>
<td>0.719</td>
<td>1.007*</td>
</tr>
<tr>
<td>En2</td>
<td>0.85</td>
<td>0.795*</td>
<td>1</td>
<td>1</td>
<td>0.676</td>
<td>1.375*</td>
</tr>
</tbody>
</table>

The correction factors, which take into account the differences in solid angle for upstream and downstream, electron energy spectra between gold and hydrogen, and effects of charged particle emission, can now be determined. Together with the assumed quantities, the values of $\eta$ and $\kappa$ are given in Table 6.12. Note that assuming $\beta_{\text{ds}}^\text{Au} / \beta_{\text{us}}^\text{Au} = 1$, $\kappa$ is the direct ratio of the electron detection efficiencies $\Omega_{\text{ds}}^\text{Au} / \Omega_{\text{us}}^\text{Au}$. As mentioned before, $\eta(Ege, En1)$ can have relatively large uncertainties, due to possible errors in $\Omega_{\text{us}}^\text{Au}$ and $\beta_{\text{ds}}^\text{Au}$.

The effect of these corrections on the stopping fraction is summarized in Table 6.13. The uncertainty in the estimated value of $\beta^\text{Au}$ (1.04) for Ege, En1 is assumed to be ±0.04, which is included quadratically in the uncertainties presented in the table. Possible errors in $\Omega_{\text{us}}^\text{Au}$ for Ege and En1 are not included. The error for En2 is statistical only.
Table 6.13: The uncorrected stopping fraction $S_{\text{HN}}^{AR}$ from Eq. 6.6 and the corrected one $S_{\text{HR}}^{AR}$ from Eq.6.15 for the runs 1650, 1654. The errors on $S_{\text{HR}}^{AR}$ for Ege, En1 include estimated 4% (relative) uncertainties due to the charge particle emission. The other errors are statistical only.

The total of about $-12\%$ relative correction to $S_{\text{HN}}^{AR}$ for Ege, En1, is dominated by the $-14\%$ correction due to the difference in the relative electron detection efficiency $\Omega_{\text{ds}}\gamma_{\text{ds}}/\Omega_{\text{us}}\gamma_{\text{us}}$ (which in turn is dominated presumably by the electron energy spectrum effect), partly offset by a $+3\%$ effect due to charged particle emission.

For En2, the correction to $S_{\text{HR}}^{AR}$ is rather small as a result of cancellation between the $-16\%$ relative correction, due to the difference in the energy spectrum, and the $+14\%$ correction, due to the difference in the upstream–downstream relative acceptance. The latter correction in $S_{\text{HR}}^{AR}$ is reasonably close to the approximate $+10\%$ given on page 146, estimated with simplified assumptions without detailed simulations.

### 6.5 Absolute amplitude method

In this section, we shall discuss another method for the determination of the stopping fraction, referred to earlier in this chapter as the absolute amplitude method.

In this method, the same fit is performed to the decay electron time spectra as in Fig. 6.5, but we use only the amplitude information for the hydrogen component, and do not rely on the gold component in the fit. With the knowledge of the absolute electron detection efficiency, the number of muon stops in $\text{H}_2$ can be directly measured.
Chapter 6. Analysis I – Absolute normalization

The absolute efficiencies for electron detection are determined by taking advantage of a condition called delayed electron coincidence, or Del-e, and delayed telescope coincidence, Del-t (together, generally called Del cuts). These cuts were implemented originally to suppress the muon-capture related background, particularly useful for detection of low energy fusion neutrons from \( dd \) fusion [166]. In the Del cut, it is demanded that the electron be observed in the scintillator (telescope) within a certain time window after a hit in a detector of interest (e.g., neutron or Si). This ensures with a high probability that the event in the detector comes from a \( \mu \text{CF} \) related process, as opposed to the capture process, since in capture the muon would be converted into a neutrino, and would not decay into an electron.

The suppression of background by more than two orders of magnitude in neutron detection was essential in order to overcome the poor signal-to-noise ratio in the \( dd \) fusion neutron measurements, but in the Si detectors, \( dt \)-fusion \( \alpha \) particle signals can be identified without Del-e (Del-t) cuts, thanks to the good resolution of the detector. This leaves us with the ability to measure with precision the efficiency of Del-e (Del-t) cuts, from which the absolute efficiency of the electron scintillators (telescopes) can be determined.

The delayed electron (telescope) coincidence cut efficiency \( \epsilon_{\text{del}} \) is a product of the electron scintillator or telescope detection efficiency (including the solid angle) \( \epsilon_e \Omega_e \), the branching ratio for electron emission \( B_e \), and the time cut efficiency \( \epsilon_{\text{time}} \) (assuming the Huff factor is 1 for hydrogen),

\[
\epsilon_{\text{del}} = \epsilon_e \Omega_e \cdot B_e \cdot \epsilon_{\text{time}}
\]

\[
= \epsilon_e \Omega_e B_e \int_{t_1}^{t_2} \lambda_{\text{del}} \exp \left[ -\lambda_{\text{del}} (t_{\text{del}} - t_{\text{Si}}) \right] dt
\]

(6.22)  \hspace{1cm}  (6.23)

where the \( \epsilon_{\text{time}} \) integral is over the time difference between the Si event time \( (t_{\text{Si}}) \) and the delayed electron time \( (t_{\text{del}}) \), and the total electron disappearance rate \( \lambda_{\text{del}} = \lambda_0 + \lambda_X \)
is the sum of the free decay rate $\lambda_0$ and muon loss rate $\lambda_X$ accounting for possible muon loss, as observed in the electron lifetime. The branching ratio $B_e$ is defined as

$$B_e = \frac{\lambda_0}{\lambda_0 + \lambda_X}. \quad (6.24)$$

It should be noted that in our earlier paper [166], an incorrect expression for the delayed electron efficiencies (Eq. (4)) was used without the factor $B_e$. That would be correct only when there is no muon loss (i.e. $\lambda_{del} = \lambda_0$), which is generally not the case either in Ref. [166] or in this thesis. For Ref. [166], however, $\epsilon_{del}$ was used only for relatively minor corrections related to background subtraction, hence the conclusion there should not be significantly affected. For our case, on the other hand, $B_e$ enters directly into the stopping fraction, as we shall see, so this distinction is rather important.

The Del cut efficiencies, $\epsilon_{del}$, were determined experimentally from the ratio of the yield $Y_{\alpha\text{f}}$ for fusion $\alpha$, to that for the $\alpha$ with a Del-e condition demanded, $Y_{\alpha\text{del}}$. The electron disappearance rate was obtained by fitting the time spectrum of the electrons detected after the fusion $\alpha$ signal in the Si detectors.

Thus, the electron detection efficiency can be derived via:

$$\epsilon_e \Omega_e = \frac{\epsilon_{del}}{B_e \epsilon_{\text{time}}} = \frac{Y_{\alpha\text{del}}}{Y_{\alpha\text{f}}} \left( \frac{\lambda_0}{\lambda_{del}} \right)^{-1} \left[ \exp(-\lambda_{del} t_1) - \exp(-\lambda_{del} t_2) \right]^{-1} \quad (6.25)$$

The fit of decay electrons to exponential functions, discussed in the previous section, had given us the amplitude $A_2$ corresponding to the hydrogen component (see Table 6.6). With $A_2$ normalized to the number of incident muons (GMU), in was the case in Table 6.6, we have $A_2 = \epsilon_e \Omega_e N_2^0$/GMU, where $N_2^0$ is the number of muons stopping in the hydrogen. Hence, the muon stopping fraction in hydrogen $S_{H}^{\text{ABS}} (\equiv N_2^0$/GMU), using the knowledge of the absolute efficiency of electron detection $\epsilon_e \Omega_e$ obtained from the Del analysis, can
be given by:

\[ S_H^{ABS} = \frac{A_2}{\epsilon_e \Omega e} = A_2 \frac{B_e \epsilon_{time}}{\epsilon_{del}}. \]  \hspace{1cm} (6.26)

We shall discuss each factor in Eq. 6.26 in the following subsections.

### 6.5.1 Delayed electron lifetime

The disappearance rate of delayed electrons \( \lambda_{del} \) enters in the determination of stopping fraction (6.26), directly in the branching ratio \( B_e \) and indirectly in the time cut efficiency \( \epsilon_{time} \). The rate \( \lambda_{del} \) was determined by fitting the time \( t_{del} - t_{Si} \), i.e., the time between the silicon \( \alpha \) event in Si1 or Si2 and the first electron (telescope) event following the Si event, with a single exponential function. In order to ensure accurate determination of \( \lambda_{del} \), cuts were made on both energy and time of the Si events.

A cut on \( t_{Si} \) (Si time with respect to muon entrance time \( t_0 \)) of \( 0.02 < t_{Si} < 0.5 \mu s \) was applied to select prompt fusion events from the upstream D\(_2\) moderator overlayer (where the signal-to-background ratio is most preferable), and to ensure a uniform time efficiency of the Del cuts. Since the event gate was open for a finite width (~ 10\( \mu \)s) after the muon entrance, the efficiency for the Del cuts for the Si events occurring late (with respect to \( t_0 \)) is reduced, due to the smaller time window for the detection of delayed electrons\(^8\).

Two different energy cuts were applied. The nominal [2000, 3700] ch\(^9\) cut (noted as Energy cut "a" in Tables 6.14, 6.15) covered a good portion of the fusion \( \alpha \) peak,

---

\(^8\)Recall that any event, including the delayed electron event, can be collected only when the Event Gate is open. Thus, if for example, a Si event occurred at 9 \( \mu \)s, there is only about a 1 \( \mu \)s window for the delayed electrons to be detected to fulfill the Del cut, as opposed to the nominal Del window \( (t_2 - t_1) \) of \( \sim 5 \mu s \). The Del time efficiency is constant as long as \( t_{Si} \leq T_{EV} - (t_2 - t_1) \), where \( T_{EV} \) is the event gate width.

\(^9\)Recall that 1 ch \( \sim 1 \) keV.
Chapter 6. Analysis I – Absolute normalization

Table 6.14: The lifetime of the first electrons (1st E) after the Si signal, fitted to a single exponential with a constant background. Energy cut $\alpha$ represents Si energy of [2000, 3700] ch, selecting fusion $\alpha$ events, while the cut $l$ is for [2000, 3000] ch, avoiding fusion events occurring at the surface.

While the lower energy cut [2000, 3000] ch (noted as "l" in Tables 6.14, 6.15) avoided the fusion events which occurred near the surface of the $D_2$ overlayer. The latter cut was implemented to test a possible systematic effect which depends on the depth of the fusion event in the layer, such as $\mu^-$ or $\mu d$ escaping from the layer. The energy of the $\alpha$ is related to the event depth thanks to $\alpha$ energy loss in the layer.

Tables 6.14 and 6.15 give the fitted results of $\lambda_{\text{del}}$ for Del-e and Del-t time spectra, respectively. Two different series of runs, (A) Runs 1671-83$^{10}$, and (B) Runs 1709-30$^{11}$, were used for the fit. While a thick hydrogen layer (500 T-l) was present in the downstream target for Run A, there was no (Target II-13) or only a very thin (II-14) layer in the downstream target for Run B.

For the delayed electron (Table 6.14), the use of a constant background term was necessary to obtain reasonable fits. This was not the case for the delayed telescope (Table 6.15) where the background was smaller, and fits both with and without the constant term were tried to check the consistency.

As shown in Tables 6.14 and 6.15, for Si1, Runs A and B give a consistent value of

---

Table 6.14:

<table>
<thead>
<tr>
<th>Run</th>
<th>Energy cut</th>
<th>1st E after Si1</th>
<th>1st E after Si2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$1/\lambda_{\text{del}}$ ($\mu$s)</td>
<td>$\chi^2$/dof</td>
</tr>
<tr>
<td>A</td>
<td>$\alpha$</td>
<td>2.025(31)</td>
<td>1.05</td>
</tr>
<tr>
<td></td>
<td>$l$</td>
<td>2.095(56)</td>
<td>0.99</td>
</tr>
<tr>
<td>B</td>
<td>$\alpha$</td>
<td>2.027(32)</td>
<td>1.03</td>
</tr>
<tr>
<td></td>
<td>$l$</td>
<td>2.150(66)</td>
<td>1.09</td>
</tr>
</tbody>
</table>

$^{10}$Target ID = II-9.

$^{11}$Target ID = II-13, II-14.
Table 6.15: The lifetime of the first telescope (1st T) after Si signal, fitted to a single exponential with (bkgd yes) and without (bkgd no) a constant background term.

\[
\begin{array}{|c|c|c|c|c|c|c|}
\hline
\text{Run} & \text{Energy cut} & 1/\lambda_{\text{del}} (\mu s) & \chi^2/\text{dof} & \text{cl} (\%) & 1/\lambda_{\text{del}} (\mu s) & \chi^2/\text{dof} & \text{cl} (\%) & \text{bkgd} \\
\hline
A & \alpha & 1.978(48) & 1.20 & 9.7 & 2.102(46) & 0.95 & 62 & yes \\
 & l & 2.054(89) & 1.21 & 9.0 & 2.113(78) & 0.92 & 69 & \\
B & \alpha & 2.034(51) & 1.13 & 19 & 1.952(44) & 0.99 & 50 & \\
 & l & 2.141(91) & 0.94 & 64 & 2.077(94) & 1.18 & 12 & \\
A & \alpha & 2.116(31) & 1.38 & 1.0 & 2.096(26) & 0.94 & 84 & \\
 & l & 2.102(52) & 1.21 & 8.4 & 2.102(44) & 0.91 & 73 & no \\
B & \alpha & 2.090(30) & 1.15 & 16 & 2.013(28) & 1.04 & 37 & \\
 & l & 2.187(51) & 0.94 & 65 & 2.103(54) & 1.17 & 13 & \\
\hline
\end{array}
\]

\( \lambda_{\text{del}} \) for both Del-e and Del-t, while for Si2, Run B gives a smaller value than Run A by 2 to 3 \( \sigma \). If Runs A and B are averaged, however, Si1 and Si2 give consistent \( \lambda_{\text{del}} \). The averages over Si1 and Si2 as well as over Runs A and B were thus taken and are presented in Table 6.16. We note the following. First, Del-e and Del-t are for the most part consistent with each other. Second, not including the constant background term increases the value of fitted \( \lambda_{\text{del}} \). Though not shown in the tables, this holds true for the Del-e fits as well. Third, the energy cut \( l \) gives a \( \lambda_{\text{del}} \) that is 2–4 \( \sigma \) lower than the cut \( \alpha \) in all cases in Table 6.16. Our determination of \( \lambda_{\text{del}} \) is thus limited by systematic effects, which are possibly due to the finite thickness of our layer. Taking the average of the two extreme values in Table 6.16 we assign \( \lambda_{\text{del}} = 2.081 \pm 0.064 \) \( \mu s \) with the error covering the two extremes. Thus we have the time cut efficiency, \( \epsilon_{\text{time}} = 82.0 \pm 0.4\% \), and the electron branching ratio, \( B_e = 94.7 \pm 2.9\% \), which combine to give the factor \( B_e \cdot \epsilon_{\text{time}} = 77.7 \pm 2.8\% \) for Eq. 6.26. Note that the errors are correlated.
6.5.2 Delayed electron cuts efficiency

The efficiencies for the delayed electron and telescope cuts were determined from the ratio of the $\alpha$ signal yields with and without the Del cuts. From each spectrum the background was subtracted, usually using a pure H$_2$ target in which no fusion takes place$^{12}$. The $\alpha$ events were selected with an energy cut of [2000, 3700] ch, and a time cut of [0.02, 0.4] $\mu$s was applied to the Si signals.

Columns 3 to 5 in Table 6.17 show the Del cut efficiency, $\epsilon_{del}$ for Si1, Si2, and their average. Run A is the same as that in Tables 6.14 and 6.15. Run C was similar to A but had no downstream layers (500 T-l H$_2$ and 3 T-l D$_2$ were present for Run A). Run D had a 6 T-l upstream overlayer (as opposed to 14 T-l as in others), testing possible effects of the layer thickness and the reaction depth. Given in brackets for $\epsilon_{del}$ are statistical errors. Run A had sufficient statistics, while Runs C and D, as well as Del-t cuts (Tel1, Tel2) had relatively poor statistical precision. Some variations of the Del cut efficiencies are observed in Table 6.17: Run C and D have a lower $\epsilon_{del}$ than A by 2 to 3 $\sigma$, possibly pointing to a systematic effect due to the target conditions.

The interpretation of the $\epsilon_{del}$ results requires some caution; recall that in the present analysis, detection of the fusion reactions is used to tag the muon as a well-defined

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$^{12}$For Run D below, the standard emission target without overlayer (target ID=II-7) was used.
Table 6.17: The Del cut efficiency, $\epsilon_{\text{del}}$, the absolute electron detection efficiency, $\epsilon_{e}\Omega_{e}$, and the muon stopping fraction, $SF_{H}^{\text{ABS}}$, determined from the absolute yield method. Run A is the same as that in Table 6.14, and Run C is similar to A but without the downstream 500 T-l H$_2$ and 3 T-l D$_2$ (target ID=II-7). Run D had a 6 T-l upstream overlayer (target ID=II-3) instead of 14 T-l as in others. The errors (in brackets) for $\epsilon_{\text{del}}$ and $\epsilon_{e}\Omega_{e}$ are statistical only, while those for $SF_{H}^{\text{ABS}}$ include the $\sim$ 3.6% systematic uncertainty due to $B_{e}$. The values in bold face highlight the downstream detectors (En2 and Tel2) which are less susceptible to the Si solid angle effect.

<table>
<thead>
<tr>
<th>Run</th>
<th>Det.</th>
<th>$\epsilon_{\text{del}}$(%)</th>
<th>$\epsilon_{e}\Omega_{e}$(%)</th>
<th>$SF_{H}^{\text{ABS}}$(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Si1</td>
<td>Si2</td>
<td>Average</td>
</tr>
<tr>
<td>A</td>
<td>Ege</td>
<td>3.99(5)</td>
<td>5.26(5)</td>
<td>4.63(4)</td>
</tr>
<tr>
<td></td>
<td>En1</td>
<td>5.05(6)</td>
<td>3.87(7)</td>
<td>4.46(5)</td>
</tr>
<tr>
<td></td>
<td>En2</td>
<td>4.13(6)</td>
<td>4.25(5)</td>
<td>4.19(4)</td>
</tr>
<tr>
<td></td>
<td>1st E</td>
<td>13.1(1)</td>
<td>13.3(1)</td>
<td>13.2(1)</td>
</tr>
<tr>
<td></td>
<td>Tel1</td>
<td>2.35(4)</td>
<td>1.99(4)</td>
<td>2.17(3)</td>
</tr>
<tr>
<td></td>
<td>Tel2</td>
<td>1.90(4)</td>
<td>1.98(4)</td>
<td>1.94(3)</td>
</tr>
<tr>
<td></td>
<td>1st T</td>
<td>4.24(6)</td>
<td>3.96(6)</td>
<td>4.10(4)</td>
</tr>
<tr>
<td>C</td>
<td>Ege</td>
<td>3.67(13)</td>
<td>5.12(11)</td>
<td>4.40(9)</td>
</tr>
<tr>
<td></td>
<td>En1</td>
<td>4.85(15)</td>
<td>3.78(16)</td>
<td>4.32(11)</td>
</tr>
<tr>
<td></td>
<td>En2</td>
<td>4.07(13)</td>
<td>3.93(13)</td>
<td>4.00(9)</td>
</tr>
<tr>
<td></td>
<td>1st E</td>
<td>12.54(25)</td>
<td>12.76(24)</td>
<td>12.65(17)</td>
</tr>
<tr>
<td></td>
<td>Tel1</td>
<td>2.23(10)</td>
<td>2.07(10)</td>
<td>2.15(7)</td>
</tr>
<tr>
<td></td>
<td>Tel2</td>
<td>1.78(9)</td>
<td>1.83(9)</td>
<td>1.81(6)</td>
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<tr>
<td></td>
<td>1st T</td>
<td>4.01(13)</td>
<td>3.89(13)</td>
<td>3.95(9)</td>
</tr>
<tr>
<td>D</td>
<td>Ege</td>
<td>4.17(23)</td>
<td>4.44(22)</td>
<td>4.31(16)</td>
</tr>
<tr>
<td></td>
<td>En1</td>
<td>4.63(25)</td>
<td>3.80(27)</td>
<td>4.22(18)</td>
</tr>
<tr>
<td></td>
<td>En2</td>
<td>4.11(23)</td>
<td>3.81(24)</td>
<td>3.96(17)</td>
</tr>
<tr>
<td></td>
<td>1st E</td>
<td>12.86(44)</td>
<td>11.93(44)</td>
<td>12.40(31)</td>
</tr>
</tbody>
</table>

source for the electron, into which it decays. Detecting the fusion $\alpha$, however, biases the electron source to preferentially concentrate towards the region where the acceptance for the particular Si detector is higher, e.g., the beam-left edge of the target for Si1 (see Fig. 3.18, page 90). The overall efficiency of the Del cuts, which is a convolution of the $\alpha$ particle acceptance in the Si detector and the electron acceptance in the scintillators
(telescope), is thus higher in the case of the Del coincidence in the same side of the target than that in the opposite side (Fig 3.18); e.g., for Si1, $\epsilon_{\text{det}}$ is higher for Ege at the beam left than En1 at the beam right, as can be seen in Table 6.17. This acceptance bias effect can be largely avoided by taking the average of Si1 and Si2, yet the effect persists to increase $\epsilon_{\text{det}}$ (unless the target is infinitely small), and is at the several percent level in our case.

On the other hand, the downstream electron detectors (En2, Tel2), which were on the beam (Z) axis (as opposed to the perpendicular axis (X)), are less susceptible to the bias effect, since they have relatively uniform acceptance over the entire target. This can be seen in the consistency between Si1 and Si2 in $\epsilon_{\text{det}}$ for En2 and Tel2 in Table 6.17. En2 and Tel2, which have lower values of $\epsilon_{\text{det}}$, are thus more reliable compared to other detectors.

With all the factors in Eq. 6.26 determined, the stopping fraction with the absolute yield method can now be computed and the results are given in the last column of Table 6.17. The error includes 3.6% systematic uncertainty due to the delayed electron lifetime, $\tau_{\text{det}}$, which dominates the total error for Run A, but is comparable to the statistical error for Runs C and D. The values in bold face are for the detectors downstream (En2, Tel2), which are more reliable than other detectors, as discussed above. Among these $SF_{H}^{ABS}$ values for En2 and Tel2, there are about ±3.3% variations depending on the target conditions or the choice of the detector (i.e., En2 versus Tel2), which indicates a measure of further systematic effects. Note that unlike the case for the relative amplitude method, in which there was some 30% discrepancy in $SF_{H}^{AR}$ between the scintillators and telescopes (see Table 6.6 in page 134 and 6.8 in page 136), the results in the two detectors are nearly consistent in the absolute yield method here. Taking the average of the two extremes of the downstream $SF_{H}^{ABS}$, we quote $SF_{H}^{ABS} = (29.9 \pm 1.5)\%$, where with the assumption that the uncertainty is dominated by the systematics, the 5% error is given
by a quadratic addition of the 3.6% systematic error from each measurement and 3.3% due to run variations.

6.6 Discussion of stopping fraction

6.6.1 The discrepancy

As we have seen in the previous sections, the results of two methods for stopping fraction determination are in disagreement. For the downstream detector En2, we have:

\[ S^{AR}_{H} = 23.4 \pm 0.7\% \quad \text{(Relative Amplitude Method)} \]
\[ S^{ABS}_{H} = 29.9 \pm 1.5\% \quad \text{(Absolute Amplitude Method)} \]

(6.27)

where the disagreement is about 25% (at the 4 \( \sigma \) level). Although the absolute amplitude method has about twice as large a quoted error, it is not necessarily less reliable than the relative amplitude method. The advantages of the former include that it: (1) does not rely on the Au component fit in electron spectrum, which can be rather difficult due to the fast time slope, (2) is much less sensitive to the time zero, \( t_0 \), position, (3) does not require the corrections via Monte Carlo simulations for the electron detection threshold and decay energy spectrum effects, as well as the relative (upstream versus downstream) solid angle effect, and (4) is not susceptible to the effects of particle emission after muon capture.

On the other hand the disadvantages of the absolute amplitude method include: (1) lack of a complete understanding of the muon loss mechanism in a thin layer, which might affect the results via the branching ratio factor, \( B_c \), (2) possible bias due to the different acceptance functions for electron detection in scintillators (telescope) and \( \alpha \) detection in Si. It should be stressed, however, that these effects have been estimated rather reliably to be less than a few percent by using our data, and are thus reflected in our quoted
errors for this method.

6.6.2 A solution

In an attempt to resolve this discrepancy, we revisit the relative amplitudes in the telescope measurements. As discussed in Section 6.4.2 (p. 136), the telescope data gave some 30% higher value of the reduced stopping fraction $S_{HR}^{AR}$, compared to the electron scintillator data. We suspect that this might be due to the energy sensitivity of the detectors to the stopping fraction, as the telescope events required a certain energy deposit in the liquid scintillator. This was experimentally demonstrated in Fig. 6.6 (page 138), and qualitatively explained to be due in part to the muon decay electron energy spectra (p. 144).

Here we postulate the possibility of muon-capture related processes still not fully accounted for by various corrections which we have made in Section 6.4.3. One such example is gamma induced reactions in the target.

We make use of the minimum ionizing peak ($\sim 15 - 20$ MeV) in the telescope energy spectra, and make a comparison with the GEANT simulations of full electronic processes, to extract the relative amplitude. The possible advantages of using the energy cuts at this peak, as opposed to no energy cuts, include: (a) energy deposited from a muon-capture related process, if it exists, should be less important at high energies, and (b) with the lack of an energy calibration for the Tel spectra, the minimum ionizing peak itself can provide a calibration. Nevertheless, in order to account for some ambiguity\textsuperscript{13} in the exact cut positions corresponding to the experimental energy cut at the peak ($590 < E < 790$ ch for both Tel1 and Tel2; see Fig. 6.6), we applied two different sets of cuts in the Monte

\textsuperscript{13}In addition to lack of independent energy calibration, we note that the simulation does not \textit{explicitly} include the resolution of the detector, though the simulated physics processes in the detector partly account for it.
Figure 6.8: Energy spectra for Tel1 (top) and Tel2 (bottom), obtained by GEANT simulations. Vertical lines indicate the cuts used to determine the relative electron detection efficiencies, corresponding to the cuts in the experimental data (590<E<790 GeV in Fig. 6.6). Narrower cuts (cn1, cn2) and wider cuts (cw1, cw2) were tried to check the systematic effects.
Table 6.18: The correction factors $\eta$, $\kappa$ in Eq. 6.15, and assumed quantities for the derivation by Eqs. 6.16, 6.17. Values with ‘*’ are taken from the ratio of the efficiencies calculated by GEANT simulation with the cuts given in Fig 6.8. The first parentheses are the Monte Carlo statistical errors, while the second indicate systematic uncertainties due to the different cuts.

Carlo spectra. Shown in Fig. 6.8 are the simulated spectra and applied cuts. With these cuts, we determined the relative efficiencies of electron detection necessary to derive the stopping fraction, as given in Table 6.18. The first parentheses in the tables are the Monte Carlo statistical errors, while the second indicate systematic uncertainties due to the different cuts in the Monte Carlo energy spectra. As can be seen, the resulting relative efficiencies are not very sensitive to the choice of the cuts. Note that the charged particle effect ratio $\beta_{us}^{Au} / \beta_{us}^{H}$ of 1 was used for Tel1 (bold face in the table), as opposed to 1.04 in the scintillator measurement, because of the higher energy cuts applied here. The stopping fractions $S_{H}^{AR}$ determined with the telescope thus are given in Table 6.19, together with uncorrected values.

Table 6.19: The uncorrected stopping fraction $S_{H}^{AR}$ and the corrected one $S_{H}^{AR}$ using the minimum ionizing peak of the telescope detectors in the relative amplitude method. Uncorrected values with nominal energy cuts (200<$E$<1200 ch) are also given for comparison.
The results for $S_H^{AR}$ from the telescope are in good agreement with the absolute amplitude method, while in disagreement with the relative method with the scintillators. The stopping fractions determined from various methods are summarized in Table 6.20. Also listed is the result of a GEANT based beam stopping simulation performed by Marshall assuming the peak momentum of 27.0 MeV/c with $\Delta p/p = 5.7\%$ (these values were carefully tuned to match the relative hydrogen stopping fractions for a range of beam momentum). Our results suggest that the relative amplitude method, using electron scintillators, may be unreliable for a stopping fraction determination, even with elaborate corrections applied as in the preceding sections, due perhaps to further capture related background such as nuclear gammas which are not accounted for. In fact, a preliminary study by Art Olin using an evaporation model for particle emission following nuclear excitation from muon capture suggests this may be the case. Excluding the scintillator relative amplitude values, we obtain consistent results on the stopping fraction. The absolute amplitude method with its advantages discussed above, appears to be more reliable, hence I shall use the value $S_H = 29.9 \pm 1.5\%$ for the present work.

<table>
<thead>
<tr>
<th>Method</th>
<th>Detector</th>
<th>$S_H$ (%)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative amplitude</td>
<td>En2</td>
<td>23.4(7)</td>
<td>$\mu$ capture related background?</td>
</tr>
<tr>
<td>Relative amplitude</td>
<td>Tel1, 2</td>
<td>30.5(14)</td>
<td>Energy cut $\sim$16–20 MeV</td>
</tr>
<tr>
<td>(min. ionizing peak)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Absolute amplitude</td>
<td>En2, Tel2</td>
<td>29.9(15)</td>
<td>No MC correction necessary</td>
</tr>
<tr>
<td>Beam simulation</td>
<td>(GEANT)</td>
<td>31.1</td>
<td>$p = 27.0$ MeV/c, $\Delta p/p = 5.7%$</td>
</tr>
</tbody>
</table>

Table 6.20: Summary of stopping fractions determined via various methods. The absolute amplitude result (bold face) will be used for this work.

Finally, I point out that since many of the previous experiments in the field used the simple relative amplitude method, often without any efficiency corrections, their results may require some re-interpretation, particularly when muon stopping in non-hydrogenic
materials is significant, as in a low density gas target.
Chapter 7

Analysis II – Emission of muonic tritium

In this chapter, we present the analysis of Run Series I, in which the emission of muonic tritium is studied. The first section deals with the treatment of the MWPC imaging data, and in the second section we present a quantitative analysis using detailed Monte Carlo calculations.

7.1 MWPC imaging data

7.1.1 Cuts

As described in Section 3.4.3, the MWPC system determines the position of the electron hit from the delay times, the time it takes the signal to reach both ends of the delay line (see Eq. 3.4). In addition to a hardware discrimination (via pre-amplifier threshold) against noise in the delay signal lines, the following off-line software cut procedure was applied in order to ensure the quality of imaging.

Since the delay lines have a fixed length, the sum of the delay times is expected to be constant, i.e., for the Z-direction, \( SUMZ \equiv (ZL + ZH) \times dispZ = Const. \), where \( ZL \) and \( ZH \) are the delay time measured at beam upstream and downstream ends of the delay lines, respectively, and the sum is multiplied by the dispersion constant, \( dispZ \) with a length dimension for convenience. In reality, however, a small but finite amount of time is required between the ionizing electron and the current being induced in the
cathode wires, mainly due to the drift time of electrons to the anode, which creates a
distribution in \( \text{SUM}Z \) (and \( \text{SUM}Y \equiv (\text{YL} + \text{YH}) \times \text{dispY} \)) reflecting the proximity
of the initial hit to the anode. There is a strong correlation between distributions of
\( \text{SUM}Z \) and \( \text{SUM}Y \), since they are due to the same physical process for both \( z \) and \( y \)
wires, \( i.e. \), electron drift to the anode (\( e.g. \) if ionization occurs very close to the anode,
both \( \text{SUM}Z \) and \( \text{SUM}Y \) are small due to a small drift time).

Because of this correlation, the plot of \( \text{SUM}Z \) vs. \( \text{SUM}Y \) for good events occupies only a small area of two dimensional space (a long rectangular box oriented in a
diagonal direction), so making independent cuts on each parameter \( \text{SUM}Z \) and \( \text{SUM}Y \)
(corresponding to accepting a large square box on the plane) is rather inefficient in terms
of noise elimination. We therefore performed a linear transformation of the coordinate
system, introducing new uncorrelated variables, \( \text{SUMSUM} \equiv \text{SUM}Z + \text{SUM}Y \) and
\( \text{DIFSUM} \equiv \text{SUM}Z - \text{SUM}Y \). With these coordinates, applying the cuts becomes
straightforward. \( \text{DIFSUM} \), the difference in the sums, typically had a smaller disper-
sion compared to that of \( \text{SUM}Z \) or \( \text{SUM}Y \) and \( \text{SUMSUM} \) larger. Therefore a small
interval for \( \text{DIFSUM} \) cut (±8 or ±10 mm for this analysis) and a large interval for
\( \text{SUMSUM} \) cut (±50 mm), cover the entire parameter regions for good events, while
efficiently eliminating the noise.

Good hits in each of three MWPCs, thus passing the cut, were fitted to a straight line
with the least-squares method. Cuts in the chi-square (<4) and the fit-residual for the
second of three chambers (<10 mm) for both \( z \) and \( y \) variables, were applied to ensure
a reasonable fit.

For this analysis, all the above cut values described in this section were taken to
be relatively non-restrictive, the largest effect being about 20% rejection due to the
chi-square cut, in order to prevent biasing of the data set and to allow for maximum
statistics.
Chapter 7. Analysis II – Emission of muonic tritium

7.1.2 Muonic tritium emission spectra

The trajectories of muon-decay electrons, reconstructed from the least-squares fit of three wire chamber positions, were extrapolated back to a perpendicular plane bisecting the target, providing an estimate of the position of the muons at the time of decay. The time of electrons was measured by the fast output signal of plastic scintillators. Recalling that the number of muon decays in unit time is proportional to the muon population \((-dN_\mu/dt = \lambda N_\mu)\), the detection of decay electrons provides the measurement of the muon population at a particular position and time.

The evidence for \(\mu t\) emission is clearly seen in Fig. 7.1. The time and the extrapolated position of muon decay in the \(z\)-direction (along the beam) are shown in contour plots.
for the targets of (a) H$_2$ with 0.1% tritium, and (b) pure H$_2$. The intense region near $z = -20$ mm in both figures is from muons decaying inside the hydrogen layer and upstream target support, whereas the events near $z = 20$ mm are muons stopping in the bare downstream gold foil where they disappear quickly via nuclear capture. While the pure H$_2$ target (b) shows very few events in the vacuum region between two gold foils (around $z = [-10,10]$ mm), a strong signal is observed in this region for the H/T target (a), indicating emission of the muonic system into vacuum. The emitted muonic system, when allowed to collide with a separate D$_2$ layer, produced $dt$ fusion providing unambiguous identification as $\mu t$.

The information on the velocity distribution of the emitted $\mu t$ can be obtained from the correlation between the time and position of muon decay. Since the time for emission is relatively short ($\sim$100 ns), the slope $z/t$ in the plot roughly corresponds to the longitudinal component of the $\mu t$ velocity. Shown in Fig. 7.1 (c) is another measurement, where the standard H/T target was covered by an additional very thin layer of D$_2$ (overlayer). A significant shift in average slope, hence velocity, is observed.

Figure 7.2 compares a series of measurements with varying overlayer thickness of D$_2$ as well as with no overlayer, where the time spectra of muon decay in the vacuum region ($z = [-10,10]$ mm) are plotted. The events were normalized to GMU, and the pure H$_2$ run (Fig. 2 (b)) was used for background subtraction. The change in the mean of the time distribution is clear, demonstrating that we can control the velocity of the emitted $\mu t$, an important feature for optimizing the $\mu t$ beam energy for the time-of-flight measurements.

Further quantitative analysis requires a comparison with Monte Carlo calculations, which will be discussed in the following section (7.2).
Figure 7.2: Time spectra of muon decay in the region $z=\pm[10,10]$ mm with varying thickness of $D_2$ overlayer. The thicknesses given are approximate.
7.2 Measurement of the Ramsauer-Townsend effect

In this section, we shall quantitatively analyze the muonic tritium emission spectrum obtained in Section 7.1, by comparing with Monte Carlo calculations based on theoretical cross sections. This will constitute the first spectroscopic measurement of the Ramsauer-Townsend effect in the muonic system.

7.2.1 Monte Carlo parameter determination

In order to make a comparison with data, some parameters in the SMC code need to be determined, including (a) muon beam distribution, and (b) wire chamber position resolution. This was achieved using a pure $\text{H}_2$ layer. We have already discussed (a) in the context of determining the effective thicknesses (Section 6.1), where the beam distribution was determined from the MWPC image of the target region in the $Y$ direction. For the present analysis, a flat-top Gaussian beam distribution with a flat top radius $R_{\text{flat}}$ of 10 mm and Gaussian FWHM$_y$ of 10 mm (noted $10\pm10$ mm) was used.

Because the target is extended in the $Y$ direction, the $Y$ image is rather insensitive to the wire chamber resolution parameter $\sigma_{WC}^y$. We therefore used the $Z$ image to determine $\sigma_{WC}^z$ and assumed that $\sigma_{WC}^y$ had a comparable value. Note that we need not know $\sigma_{WC}^y$ too precisely because the $Y$ distribution is already broad and the $Y$ image is not affected much by the choice of $\sigma_{WC}^y$.

We have performed iterative fits of the Monte Carlo calculations to the data to find the best value for $\sigma_{WC}^z$ (Fig. 7.3). Plotted with error bars in Fig. 7.3 (a) is a $Z$ image of MWPC system for a pure $\text{H}_2$ target$^1$, from which one expects no muonic atom emission except at very low energies [216]. A time cut of $t - t_0 \geq 2\mu$s ($t_0$ being muon entrance time) is applied to eliminate the background from muon stopping in heavy elements in

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$^1$Target ID 1-8 in Table 4.2 in page 105.
the target frames\(^2\). The data was fitted in the interval shown in the figure with a Monte Carlo assuming \(\sigma^W_C = 1.3\) mm with a beam parameter of \(10 \pm 10\) mm (histogram). The normalization factor being the only free parameter, the fit gave \(\chi^2/\text{DOF}\) of 1.62 (10.3% confidence level).

Figure 7.3 (b) shows the result of an iterative fit, varying \(\sigma_z\) for two different beam parameters\(^3\). Thus we conclude that the best value for the MWPC resolution parameters is \(1.3 \pm 0.1\) mm with only a small dependence of the beam parameters.

### 7.2.2 The Ramsauer-Townsend minimum energy

With these Monte Carlo parameters fixed, we can now compare the time of flight spectrum of the emitted muonic tritium with Monte Carlo simulations to test theoretical cross sections based on few-body theory. We shall focus on the measurement of the position of the Ramsauer-Townsend minimum \(E_R\), since this is important both as a source of the \(\mu t\) beam and as a test of the quantum three-body calculations.

The Nuclear Atlas cross sections [16, 17] for the muonic processes were used as a nominal input to the Monte Carlo. Molecular and condensed matter effects [167, 170, 216] are expected to play negligible roles in the transport properties of muonic atoms at the energies above a few eV. Iterative calculations were performed by multiplying the energy scale of the \(\mu t + p\) elastic scattering cross section by a constant factor \(\kappa\), thus shifting the RT minimum as \(E_R \rightarrow \kappa E_R\). The resulting simulated time spectrum is fitted to the experimental data with one free parameter (relative normalization), and \(\chi^2\) was calculated for each value of \(\kappa\). For both the data and the MC, a longitudinal spatial cut of \(z = [-10, 10]\) mm was applied to select the vacuum region.

\(^2\)Recall that muons bound in the atomic states of heavy elements disappear quickly (within a few 100 ns) due to a high rate of nuclear muon capture via the semi-leptonic weak interaction.

\(^3\)See Section 6.1 for the notation of beam parameters.
Figure 7.3: Determination of MWPC resolution parameters.
Figure 7.4: Above: Plotted with error bars is a background-subtracted time spectrum of μt emitted from the standard emission target (protium with 0.1% tritium). A spatial cut of $z = [-10, 10]$ mm was applied. The histogram shows a fit to a Monte Carlo calculation, with an energy scaling factor $\kappa = 1.1$. Two different intervals for the fit are also shown. Below: residuals from the fit in the region A.
Figure 7.4 shows an example of such a fit (top) and its residuals (bottom). Plotted with error bars is the $\mu t$ time spectrum from the standard emission target\footnote{Target ID = I-1 in Table 4.2 in page 105.} (1000 T-l H$_2$ with 0.1% T$_2$) from which a background run\footnote{Target ID = I-8.} of pure 1000 T-l H$_2$, normalized to GMU, was subtracted, while the histogram shows a simulated time spectrum assuming the energy scaling factor of $\kappa = 1.1$, which turned out to give the best $\chi^2$/DOF of 1.06 for fit interval $t = [0,6] \mu$s (region A) with DOF = 59. Plotted in the bottom of Fig. 7.4 is the fit residual, i.e. the difference between the Monte Carlo and the data, normalized to one standard deviation.

Illustrated in Fig. 7.5 is the global trend of total $\chi^2$ versus the energy scaling factor $\kappa$, while Fig. 7.6 shows the details near the minimum. The horizontal axis is plotted against the inverse of square root of $\kappa$ to reflect our sensitivity to the time of flight, rather than the energy\footnote{Recall that $t = l \times \sqrt{M_{\mu t}/2E}$, where $t$ is the time of flight over the drift distance $l$.}.

Dependences of the various parameters were investigated in detail for the potential systematic effects. Figs. 7.5 and 7.6 show some examples of such investigations including:

1. Muon transfer rate from protons to tritons, $\lambda_{pt}$, as input to the simulation. Our standard input uses an experimental rate measured by our collaboration, $\lambda_{pt} = 5.86 \times 10^9$ s$^{-1}$\footnote{Target ID = 1-1 in Table 4.2 in page 105.} for this process, but fits using the theoretical energy-dependent value of $\lambda_{pt}$ from the Nuclear Atlas cross section\footnote{Target ID = 1-8.} [17] were also tried.

2. Time interval of the fit region. Region A: $t = [0,6] \mu$s, Region B: $t = [0.1,3] \mu$s (see Fig. 7.4). If our fit model is correct, the fit results should be independent of fit region except to add a constant value in total $\chi^2$, which was confirmed in Figs. 7.5, 7.6.

3. MWPC resolution parameter $\sigma_y^{WC}$, $\sigma_z^{WC}$. Our nominal value 1.3 mm was varied...
Figure 7.5: Total $\chi^2$ versus $\kappa^{-1/2}$ in the fit of $\mu t$ emission data with Monte Carlo calculations, where $\kappa$ is a scaling parameter shifting the Ramsauer-Townsend energy minimum. The inverse of square root $\kappa$ is plotted to reflect our sensitivity to the time of flight, rather than the energy. Effects of the muon transfer rate from protons to tritons, $\lambda_{pt}$, and the time interval of fit regions are also shown. $\lambda_{pt}(\text{exp}) = 5.86 \times 10^9 \text{ s}^{-1}$ is taken from Ref. [83], and $\lambda_{pt}(\text{th})$ from Ref. [17]. Fit region A is the time interval $t = [0, 6] \mu s$ with DOF = 59, and region B, $t = [0.1, 3] \mu s$ with DOF = 29 (see Fig. 7.4).

from 1.0 mm to 1.5 mm (only some selections are shown in Fig. 7.6.)

4. Beam width parameters characterizing a flat top Gaussian distribution in the $XY$ plane. $10 \oplus 10 \text{ mm (nominal)}$ and $15 \oplus 15 \text{ mm}$ were plotted.

5. Muon beam stopping distribution in the $z$ direction of the emission layer.

$\alpha$: Gaussian with the peak at the surface of the layer with the standard deviation width of half the layer thickness.

$\beta$: Gaussian with the peak at the centre of the layer with the standard deviation
width of 0.4 times layer thickness.

No symbol: uniform $Z$ stopping distribution.

For the nominal values of parameters, we obtained the best fit with $\kappa = 1.1$ ($\kappa^{-1/2} \sim 0.95$ in the figures). The value of $\kappa$ shifted between 1.05 to 1.15 depending on the parameters (see Fig. 7.6). In addition, we estimate that the uncertainty in the drift distance scale can give rise to a shift in $\kappa$ of order $\pm 0.05$. With the two major systematic errors (due to the parameters (1)-(5) and the distance scale) added in quadrature, and the statistical errors much smaller, as can be observed in Fig. 7.6, our measurement indicates
a scaling factor of $\kappa = 1.10 \pm 0.07$, i.e., the Ramsauer-Townsend minimum energy of $E_{RT}^{\text{exp}} = 13.6 \pm 1.0$ eV (c.f. the theoretical minimum, $E_{RT}^{\text{th}} = 12.4$ eV). The results of a similar analysis for a measurement using an emission target with a tritium concentration of 0.3%\(^7\) were consistent with $\kappa = 1.1$.

In conclusion, we have reported in this chapter, (1) the first observation of $\mu t$ in vacuum, (2) the first quantitative spectroscopic evidence of the Ramsauer-Townsend effect in an exotic system, confirming the theoretical RT minimum energy in the $\mu t + p$ elastic scattering at the 10% level, an accuracy sufficient for our goal of molecular formation rate measurements.

\(^7\)Target ID = I-7.
8.1 Si detector data

Detection of the alpha particles provided the signature of $d\mu t$ molecule formation. From the data, information on two somewhat independent aspects can be extracted, namely, time spectra and absolute yields. As is the case in many experiments, the absolute measurement of the data was more difficult compared to the time spectrum measurement. In this section, the procedure for the analysis of the $\alpha$ particle data from Run Series 2 is presented.

8.1.1 Si detector energy calibration

The energy scales of the two silicon detectors were calibrated with $^{241}$Am sources, which were attached to the diffuser and hence could be removed during the muon beam measurements.

Figure 8.1 illustrates the calibration spectra for one of the silicon detector (Si2) taken with the source. Measurement with no target present in the system is shown in Fig. 1(a) which had a full width half maximum resolution of about 40 keV. This is partly due to the resolution of the source itself, and the detector resolution may be better. The spectrum in Fig. 1(b), on the other hand, was taken with 2 T.l, or about 6.8 Ci of tritium$^1$ present (Target II-14 in Table 4.4 in page 108). The effect of tritium caused the peak width to

\footnote{The specific activity of tritium is 2.58 Ci/cm$^3$ at STP, hence 1 T.l corresponds to about 3.4 Ci.}
Figure 8.1: Silicon (Si2) energy spectra taken with $^{242}$Am source (peak energy 5.486 MeV). One ADC channel is close to 1 keV. Si1 had similar spectra.
be about 65 keV, due to the $\beta$ decay background.

Since energy spectroscopy is not the purpose of our experiments, the absolute energy scale was not very crucial. However, as we found in the thickness measurements in Section 3.3, the change in the detector temperature could cause significant variation in the energy gain, hence frequent calibration was performed to ensure the relative stability of the energy scale. In addition, the fusion signal itself, whose energy is well known, could provide the calibration information.

Figure 8.2 shows the position of the centroid of the peak corresponding to $^{241}$Am for different calibration runs. The centroid value was determined by Eq. 3.1 (page 73) in a similar manner to the thickness measurements described in Section 3.3 with the lower and upper cut off values 5300 ch and 5600 ch, respectively. The error bars in the figure
indicate statistical uncertainties in the centroid. Changing the lower cut off value to 5200 ch caused less than 3 ch change in the centroid, indicating the systematic uncertainty in the calibration. The average of the measured centroid values is 5486.9 ch (4.7 ch) for Si1 and 5473.5 ch (3.4 ch) for Si2, respectively with the values in parentheses being the standard deviation of 14 measurements. These should be compared to the weighted average of 3 alpha lines of $^{241}$Am, 5491 keV (note that the peak energy for the strongest line is 5486 keV). Assuming there is no significant offset, which was confirmed by a test using a pulse generator, the calibration is very close to 1 ch $= 1$ keV for both detectors.

### 8.1.2 ADC dead time correction

Various off-line systematic checks of scaler values, as well as comparison of the fusion yield amongst different runs were done to make sure that the system was functioning properly. During one such check, it was discovered that some runs had fewer counts in the Si spectra compared to other runs. After some investigation, it was realized that a change made to the germanium detector circuit had caused the problem in the Si detectors, and a method was devised to correct for the loss.

The runs in questions are Runs 1683 – 1690 taken with Target II-9 (Table 4.3, page 107). A few initial observations included:

- During Runs 1683 – 1690, both fusion signal and background counts were reduced by the same factor, indicating a normalization problem rather than target related real physical effects.

- There were very many zeros in the silicon energy spectra for Runs 1683 –1690, which went unnoticed during the measurements, since on-line spectra had a cut to remove zeros.
• These zero energy events had a similar time structure as the other non-zero events.

• Other detector yields appeared unaffected.

Attention then was drawn to a circuit modification which took place during Run 1683. Before this run, a germanium detector had been disabled simply by unplugging the fast timing signal from the circuit, because its high trigger rate was causing a significant dead time for the data acquisition. (Removing the germanium detector from the trigger reduced the dead time from about 30% to 20%). In the middle of Run 1683, the circuit was modified with intention of allowing the germanium data to be recorded when other sub-detector systems fired while keeping the germanium sub-trigger out of the master trigger so that there would be no trigger if only the germanium fired. This was implemented by reviving the germanium sub-detector circuit, but with its sub-trigger (c.f. TRGn in Fig. 3.19 in page 92) not connected into logical OR for the master trigger (TRGF in Fig. 3.19).

The problem was, as it turned out, the spectroscopic ADC (AD413A, Ortec), which was shared among the germanium detector and two silicon detectors, and which was being "blocked" when only the germanium sub-trigger fired, but nothing else. AD413A, unlike other camac ADCs and TDCs, had no “fast clear” function which would ensure in the event of no master trigger, the ADC/TDC buffer was cleared by a fast pulse signal in order to be ready for the next event. Of course, when there was a master trigger, the ADC buffer was cleared (by a CAMAC command) after the data was read into the computer. Even when no sub-trigger but germanium was activated, however, the ADC gate\(^2\) was opened by the sub-trigger, hence the germanium energy data was still accepted by the ADC. The problem is that AD413A worked by design in such a way that when the master gate closes, no pulses were accepted until a CLEAR command was given, even

\(^2\)Here I am referring to the master gate for the ADC; the use of a gate for individual channels was redundant in our case.
if the master gate opened again. Therefore, if the germanium fired without any other triggers, and then either one of the Si detectors triggered on the following event, the Si signal was not accepted by the ADC, hence only zero was recorded. Yields of Si events were thus affected by the change in the germanium circuit.

In order to obtain a correct normalization, we derived a procedure to determine the ratio of “good” ADC events to “bad” ones. We first define the following conditions in order to identify the different events:

1. Firing of Si sub-trigger (Sil or Si2)
2. Recorded Si energy = 0
3. Recorded Ge energy > 0 (i.e., not 0)
4. Recorded Ge time is an over flow

We used a combination of above conditions to select the events. Several different types of events can give a similar signature, and they are summarized in Table 8.1.

<table>
<thead>
<tr>
<th>Type</th>
<th>Event ((i - 1))</th>
<th>Event (i)</th>
<th>Condition</th>
<th>ADC blocking</th>
</tr>
</thead>
<tbody>
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<td>Si</td>
<td>Ge</td>
<td>Si</td>
<td></td>
</tr>
<tr>
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<td>yes</td>
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<td>no</td>
<td>yes</td>
</tr>
<tr>
<td>II</td>
<td>yes</td>
<td>no</td>
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<td>yes</td>
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<td>no</td>
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<tr>
<td>IV</td>
<td>no</td>
<td>no</td>
<td>yes</td>
<td>LLD</td>
</tr>
</tbody>
</table>

Table 8.1: Comparison of different types of events and their signatures. Event \((i - 1)\) is the event preceding a silicon detector event (Event \(i\)). LLD stands for low level discrimination. The conditions are defined in the text.

From the discussion above, the conditions 1 and 2 select obvious candidates for the “blocked” events. These candidate events could have zero silicon energy for two reasons; [a] the ADC blocking which is our concern (Event type I, II in Table 8.1), or [b] a real
Chapter 8. Analysis III – Molecular formation

silicon event, but with its energy lower than the low level discrimination\(^3\) (LLD) of the ADC (Event type III, IV)\(^4\).

Ignoring for the moment the small chance of coincidence between an LLD event and germanium trigger in Event \(i\) (Type IV in Table 8.1), the above \([a]\) and \([b]\) can be distinguished by Condition 3. That is, if the event was due to blocking (case \([a]\)), there should be non-zero germanium energy in the ADC which came from the previous Ge event (Type I, II). On the other hand, if it was due to LLD (case \([b]\)), there is no germanium energy information left over from the previous event, hence the germanium energy is zero (Type III), unless it is a Ge-LLD coincidence event (Type IV).

The condition 4 provides information as to whether there was a germanium hit in Event \(i\). If there was no germanium trigger, then the TDC clock would not be stopped and an overflow would be recorded. This could be used to discriminate between event type I and II, as well as between III and IV. However, in the end only Condition (1-2-3) was used to identify the blocking events. This condition inevitably included event type IV as well, which was not a blocking event, but the analysis of normal runs suggested that this type of event occurs in less than 0.5% of the total Si events. The alternative Condition (1-2-3-4) would have been much worse, since it would miss selecting event type II which has a higher probability than type IV. The problem here was that when there was Ge hit in Event \(i\), there is no way of knowing where a Ge hit also occurred (and blocked the ADC) in Event \((i - 1)\).

An independent check of the ADC problem was conducted using a quite different effect. It took advantage of an unwanted background for our main measurements, namely, the signal from the scattered beam muon directly stopping in the Si detectors\(^5\). When

\(^3\)LLD was specified by a CAMAC command, and set a threshold, below which energy only zero was recorded in the ADC. This could save read out time.

\(^4\)For our purpose, an event where the silicon detector was triggered due to noise on the logic signal line can be included in \([b]\).

\(^5\)This background is removed in future runs by placing a thin Cu foil to shield the Si from the beam.
looking at the fusion signals, this background was reduced to a negligible level by rejecting
the signals at prompt times, but here we used the intensity of this prompt beam peak to
check to see if the silicon detector system was properly functioning. When the ADC was
blocked, not only fusion events but also the beam stopping background events would be
lost. Hence, by normalizing the beam background intensity to GMU, we could effectively
measure the ADC live-to-dead time ratio. The limitation was that we had to assume that
the beam was stable; a small change in the position of the beam could change the number
of muons that could go through a tiny gap in the cryostat to reach the Si detectors. The
ADC dead times derived from this method and the above method using scalars agree
reasonably well, giving us some confidence in correcting for the ADC blocking events.

8.1.3 Energy spectrum features

Before we go into the detailed quantitative analysis, it may be instructive to take a look
at gross qualitative features of our data. Therefore we shall spend this section and the
next for that purpose.

Characteristics of the Si energy spectra, for different time cuts, are shown in Figs. 8.3
and 8.4. Figure 8.3 is for our standard time-of-flight measurement arrangement, i.e.,
1000 T·l of H₂ with 0.1 % tritium and 14 T·l of D₂ overlayer in the upstream target,
plus 3 T·l D₂ downstream (target ID = II-9 in Table 4.3 in page 107). Three spectra
are plotted with the time cuts of (1) \( t > 0 \), (2) \( t > 0.02 \) µs, and (3) \( t > 1.5 \) µs. The
histogram (1) shows a signal from direct beam muon stops in the Si detector, in addition
to a broad fusion signal near 3.5 MeV, predominantly from the US moderating overlayer,
and a strong low energy background peak. The beam signal is very prompt in time, and
a 20 ns delay cut eliminates this very efficiently (histogram (2)). The histogram (3) with
a time cut of \( t > 1.5 \) µs, on the other hand, shows a much narrower fusion peak mostly
Figure 8.3: Energy spectrum of Si detector (Si1) for standard TOF target with different time cuts, showing important characteristics as indicated in the figure. The energy scale is close to 1 ch = 1 keV. Si2 has a similar energy spectrum.

coming from the DS reaction layer. The US fusion takes place typically in the first few 100 ns, while fusion in DS occurs after $\mu t$ travelled a separation distance of 18 mm, hence appropriate time cuts can separate these two events. Furthermore, the US $D_2$ layer had 14 T-l thickness while the DS only 3 T-l, resulting in the difference in the peak width. There is indication that we are seeing 3 MeV protons from $d\mu d$ fusion, which contributes to the background. This will be treated in detail in Section 8.2.4.

Comparison of Fig. 8.3 with Fig. 8.4, the latter for pure $H_2$ target with no $D_2$, confirms that the peaks near 3000 – 3500 ch in the former come from fusion in the $D_2$ layers. On the other hand, the peak near 2 MeV persists in Fig. 8.4 (also in the bare target runs), and in fact the peak energy shifted as beam momentum was changed, providing the
evidence that this peak is due to muons stopping in the detector.

Other sources of background include: (a) muon decay electrons, (b) charged particle (proton, deuteron etc.) emission following nuclear muon capture on heavy elements, (c) muon induced nuclear break up, and (d) scattered beam electrons. Also the neutrals, such as neutrons and gammas following muon capture, muonic X rays, or bremsstrahlung from tritium $\beta$ decay, could cause background signals in the Si detectors, but probabilities for these are expected to be small since the detectors had a thickness of only 300 $\mu$m. Among other possible background processes for targets containing tritium are conversion muons from muon catalyzed $pt$ fusion: $\mu pt \rightarrow ^4\text{He} + \mu$ (19 MeV), and muon capture on Si from emitted $\mu t$ hitting Si detectors.
8.1.4 Time spectrum features

Gross features of Si time spectra, with different energy cuts, are illustrated in Figs. 8.5 and 8.6. Shown in Fig. 8.5 is the Si1 time spectrum for the standard time-of-flight arrangement, whereas that for pure H$_2$ is given in Fig. 8.6. All the histograms have a sharp spike at time zero, which, at least in part, comes from direct muon stops in the Si detector. The low energy part ($E < 2000$ ch) of the spectra, which we saw was dominated by a large background signal (Figs. 8.3, 8.4), has two exponential components, a fast one with the order of 100 ns and a slow one about 2 $\mu$s. This is consistent with muon disappearance rates in heavy elements and hydrogen, respectively, suggesting the signals in this energy region come from muon decay electrons and charged particles from muon capture. Conversion muons from \textit{p}+\textit{t} fusion (19 MeV) could also contribute to the
Figure 8.6: Time spectrum of Si detector (Si1) for pure H$_2$ target with different energy cuts. The fusion peak is absent, while muon beam peak persists.

long lifetime.

The time spectrum with an energy cut 2001 < $E$ < 4000 ch in Fig. 8.5 exhibits fusion time signals; exponentially decaying in early time ($t < 1\mu s$) is fusion from the upstream target, while events in $\sim 2 - 4\mu s$ are mostly due to fusion from $\mu t$ flying across the drift distance to reach the downstream layer (though the signal is not so clear from the figure due to the unrestrictive energy cut).

Whereas these are obviously absent from the same energy region in Fig. 8.6, comparison between the two figures of the higher energy part (4001 < $E$ < 8000 ch) of the spectra indicates excess events in Fig. 8.5. These events, unlikely due to $d\mu t$ fusion since the maximum $\alpha$ energy is about 3.5 MeV and the probability of pile up is very small, are attributed to emitted $\mu t$ reaching the Si detector where the muon is transferred to Si and...
then captured, emitting charged products. In fact, the signal in this region is enhanced when there is no moderating overlayer because of the higher yield of $\mu t$ emission into vacuum.

8.2 The $d\mu t$ fusion measurements

In this section we shall extract, taking into account the background and various other systematic effects, the time-of-flight fusion spectra as well as the fusion yields. While the former contains the main physics we are seeking, i.e., the resonant molecular formation at epithermal energies, the latter can provide us with valuable information, as we shall see.

Our fusion measurements can be divided into four distinct categories, three for the fusion yield and one for the time spectrum measurement: (a) US fusion yield with varying US layer thickness (denoted USY measurements), (b) DS fusion yield with varying US thickness but DS thickness fixed (Moderation yield or MODY measurements$^6$), (c) DS fusion yield with varying DS thickness but US thickness fixed (Time-of-flight yield or TOFY measurements), and (d) Time spectrum of the DS fusion for various DS thickness (Time-of-flight spectrum or TOFS measurement). Note that (a) and (b), as well as (c) and (d) use the data from mostly the same run series, respectively, but they each look at different aspects of the data. Let us first discuss the background in general before going into the specifics of the each measurements.

$^6$The notation MOD is given because of its sensitivity to the $\mu t$ moderation process in the US $D_2$ layer.
8.2.1 Background subtraction

Since the Si data contains counts from non-fusion events as seen in the previous sections, subtracting appropriate background is important in the yield and spectrum determination. It is one of the advantages of our multi-layer film targets that the process of interest can be controlled, i.e., "turned on" and "turned off" without much affecting other processes (e.g., by simply depositing a thin layer), hence providing rather reliable means of background subtraction. Thus our strategy generally is to use the data from the target, in which the particular processes is turned off, for the background subtraction whenever possible (with both the signal and background data normalized to the number of muons GMU). For a given measurement, nonetheless, there were sometimes more than one possibility for the choice of the background runs, or no one best choice, in which case different methods were compared to see the systematic effects. Summaries of background runs used are given in Table 8.2. As we will see, in some cases further corrections were necessary to account for a residual background.

<table>
<thead>
<tr>
<th>Label</th>
<th>ID</th>
<th>(c_t) (%)</th>
<th>US (T·t)</th>
<th>DS (T·t)</th>
<th>Use</th>
<th>GMU (10^6)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BG1</td>
<td>II-5</td>
<td>pure (H_2)</td>
<td>1000</td>
<td>0</td>
<td>USY, MODY</td>
<td>84.9</td>
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<td>BG2</td>
<td>II-1</td>
<td>0.1</td>
<td>1000</td>
<td>20 (D_2)</td>
<td>USY</td>
<td>51.9</td>
</tr>
<tr>
<td>BG3</td>
<td>II-6</td>
<td>0.1</td>
<td>1000</td>
<td>0</td>
<td>USY, MODY</td>
<td>62.2</td>
</tr>
<tr>
<td>BG4</td>
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<td>1000</td>
<td>0</td>
<td>USY</td>
<td>46.4</td>
</tr>
<tr>
<td>BG5</td>
<td>II-7</td>
<td>0.1</td>
<td>1000(\oplus14) (D_2)</td>
<td>0</td>
<td>MOD, TOFY, TOFS</td>
<td>60.7</td>
</tr>
<tr>
<td>BG6</td>
<td>II-8</td>
<td>0.1</td>
<td>1000(\oplus14) (D_2)</td>
<td>500 (H_2)</td>
<td>TOFY, TOFS</td>
<td>54.8</td>
</tr>
<tr>
<td>BG7</td>
<td>II-13</td>
<td>0.2</td>
<td>1000(\oplus14) (D_2)</td>
<td>0</td>
<td>TOFY, TOFS</td>
<td>166.8</td>
</tr>
</tbody>
</table>

Table 8.2: Summary of runs used for background subtraction.
8.2.2 Yield measurements

Cuts

<table>
<thead>
<tr>
<th>Yield measurement</th>
<th>US layer (T·l)</th>
<th>DS layer (T·l)</th>
<th>Time cut</th>
</tr>
</thead>
<tbody>
<tr>
<td>USY</td>
<td>3, 6, 14, 14†, 20‡</td>
<td>20/0</td>
<td>0.02 &lt; t &lt; 0.4 μs</td>
</tr>
<tr>
<td>MODY</td>
<td>0, 3, 6, 14</td>
<td>20</td>
<td>0.3 to 0.8 &lt; t &lt; 6 μs</td>
</tr>
<tr>
<td>TOFY Series A</td>
<td>14</td>
<td>3, 6, 20</td>
<td>1 &lt; t &lt; 6 μs</td>
</tr>
<tr>
<td>TOFY Series B†</td>
<td>14†</td>
<td>3†, 23†</td>
<td>1 &lt; t &lt; 6 μs</td>
</tr>
</tbody>
</table>

Table 8.3: The target combinations for fusion yield measurements. Tritium concentration of $c_t = 0.1\%$ was used, except the ones marked † ($c_t = 0.2\%$) and ‡ ($c_t = 0.05\%$). The layers with bold face were kept fixed during the measurements. Some of the US yield measurements were done with no DS layer.

The time cuts for the yield measurements have been chosen to maximize the fusion signal of interest, while avoiding the background. For example, the US and DS fusions, when they coexist, can largely be separated by appropriate time cuts, since most US fusion takes place nearly promptly after the muon stop and disappears with typical time constant of $\sim 100$ ns (depending on the tritium concentration), while DS events occurs after $\mu t$ time-of-flight across the drift distance, typically of order $\mu s$. The standard time cuts of $0.02 < t < 0.4 \mu s$, and $1 < t < 6 \mu s$ were used for the USY and TOFY measurements, respectively, while the MODY time cuts were slightly varied to maximize the signal-to-background ratio (e.g., with a thin US moderator, the $\mu t$ energy is rather high, hence the time-of-flight short, which could overlap with US fusion). Table 8.3 summarizes the main conditions for the yield measurements.

Together with the partial yields for given time cuts, we give the total yield with the acceptance for the time cut estimated by SMC simulations. These corrections are typically 10% or less. The comparisons with Monte Carlo calculation results, which will be discussed later, will be done using the same time cuts for both the data and MC, hence
the time cut efficiencies will cancel in those analyses (assuming the time distribution is predicted correctly by the MC).

The effects of the energy cuts on the yield were carefully studied by using different cuts, as well as estimating the efficiency using a dedicated Monte Carlo. These and other corrections will be discussed later.
USY: Upstream Yield

The time cut of $0.02 < t < 0.4$ was used for the US measurements to select the fusion events in the upstream layer. The first 20 ns was avoided due to the prompt beam muon background, while the long time was excluded to remove the fusion events in the downstream layer.
For these measurements, we had two main choices of background run in most cases: BG1, a pure H\textsubscript{2} target, and BG2, a standard emission target with 20 T·l D\textsubscript{2} DS (see Table 8.2). Fortunately, thanks to a good signal-to-background ratio and the high yielding US fusion, the effect of background choice is small.

Examples of the US fusion data with the background are illustrated in Figs. 8.7 and 8.8 for 3 T·l and 14 T·l cases, respectively. Notice the differences in the peak width and the extent of the lower energy tail, both due to α particle energy loss in the D\textsubscript{2} layer.

To determine the α yield, we typically used at least two different energy cuts. The energy cut efficiency corrections were done using a dedicated Monte Carlo code, which will be discussed later. Si yield for the US fusion measurements with different backgrounds are given in Table 8.4. When the effects due to different background subtractions are larger than the statistical errors, we considered the former as a measure of the systematic uncertainty. These cases are indicated with * in the table.

Runs with target ID=II-2, II-3, and II-4, were performed consecutively in identical conditions (except US thickness), hence the relative yields are more reliable than other separate runs.

**MOD: Moderation Yield**

Similar to the US measurements, runs BG1 and BG2 were used for background subtraction for most runs. Because of the lower signal-to-background ratio, the choice of background had larger effects on the yield determination for the MOD measurements. Since B1 has no tritium or deuterium, it may underestimate the background, while unmoderated μt emission from B2 may contribute to the over-subtraction of the background. Some events in B2 appear to come from μt reaching Si detectors (μ capture on Si produces charged products), which have time structure similar to the real data with a characteristic time-of-flight. Table 8.5 gives the yield with different backgrounds.
### Table 8.4: Si yield per GMU for the US fusion measurements with different background subtraction and energy cuts. A time cut of $0.02 < t < 0.4 \mu s$ was applied to select the US events. The error values with * include systematic effects due to the different background subtraction. Tritium concentration of $c_t = 0.1\%$ was used, unless otherwise stated.

<table>
<thead>
<tr>
<th>US (DS)</th>
<th>ID</th>
<th>E cut $(\times 10^3 \text{ ch})$</th>
<th>BG</th>
<th>Yield $(10^{-4}/\text{GMU})$</th>
<th>BG avg.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Si1</td>
<td>Si2</td>
<td>Si avg.</td>
</tr>
<tr>
<td>3 T-(l) (20)</td>
<td>II-2</td>
<td>2.7; 3.7</td>
<td>BG1</td>
<td>0.580(15)</td>
<td>0.572(15)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG2</td>
<td>0.582(15)</td>
<td>0.575(16)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.1; 3.7</td>
<td>BG1</td>
<td>0.537(13)</td>
<td>0.541(13)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG2</td>
<td>0.543(13)</td>
<td>0.545(14)</td>
</tr>
<tr>
<td>6 T-(l) (20)</td>
<td>II-3</td>
<td>2.0; 3.7</td>
<td>BG1</td>
<td>1.64(2)</td>
<td>1.67(2)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG2</td>
<td>1.64(2)</td>
<td>1.68(2)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.7; 3.7</td>
<td>BG2</td>
<td>1.55(2)</td>
<td>1.60(2)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG2</td>
<td>1.55(2)</td>
<td>1.61(2)</td>
</tr>
<tr>
<td>14 T-(l) (20)</td>
<td>II-4</td>
<td>1.0; 3.7</td>
<td>BG1</td>
<td>4.26(4)</td>
<td>4.27(4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG2</td>
<td>4.28(4)</td>
<td>4.28(4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>BG1</td>
<td>3.97(4)</td>
<td>3.99(4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG2</td>
<td>3.97(4)</td>
<td>3.99(4)</td>
</tr>
<tr>
<td>14 T-(l) (0)</td>
<td>II-7</td>
<td>1.0; 3.7</td>
<td>BG1</td>
<td>4.24(3)</td>
<td>4.41(3)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG3</td>
<td>4.24(3)</td>
<td>4.48(3)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>BG1</td>
<td>4.01(3)</td>
<td>4.15(3)</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>BG3</td>
<td>4.02(3)</td>
<td>4.21(3)</td>
</tr>
<tr>
<td>14 T-(l) (0) $c_t = .2%$</td>
<td>II-13</td>
<td>1.0; 3.7</td>
<td>BG1</td>
<td>4.55(2)</td>
<td>4.54(2)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG3</td>
<td>4.58(2)</td>
<td>4.65(3)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>BG1</td>
<td>4.28(2)</td>
<td>4.28(2)</td>
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<tr>
<td></td>
<td></td>
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<td>BG3</td>
<td>4.30(2)</td>
<td>4.34(2)</td>
</tr>
<tr>
<td>20 T-(l) (0) $c_t = .05%$</td>
<td>II-16</td>
<td>1.0; 3.7</td>
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<td>4.41(3)</td>
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<td></td>
<td></td>
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<td>BG2</td>
<td>4.41(3)</td>
<td>4.60(3)</td>
</tr>
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<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>BG1</td>
<td>3.99(2)</td>
<td>4.10(2)</td>
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<td>BG2</td>
<td>4.00(2)</td>
<td>4.16(2)</td>
</tr>
</tbody>
</table>

Similarly to the US measurements, the background subtraction effects are included in the error (noted with *), if it is larger than the statistical error. When one background run is clearly more applicable than the other, that alone was used for the yield determination, which was the case for 0 and 14 T-\(l\) measurements.
Table 8.5: Si yield per GMU for the MOD measurements (DS fusion with varying US moderator thickness) with different background subtractions and energy cuts. The error values with * include a systematic effect due to the different background subtraction. When one BG method is clearly more applicable than the other, we quote that value in the final column. Tritium concentration of $c_t = 0.1\%$ was used.

**TOFY: Time-of-flight Yield**

Two separate series of TOF measurements were performed with tritium concentration $c_t = 0.1\%$ (Series A) and $c_t = 0.2\%$ (Series B). Due to the small counting rates, the TOF measurements, especially the ones with thin DS layers, were most difficult among the fusion measurements. We give in Table 8.6 the Si detector yield per GMU for TOFY measurements with different background and energy cut conditions. The information

<table>
<thead>
<tr>
<th>US (ID)</th>
<th>T cut (μs)</th>
<th>E cut ($\times10^3$ ch)</th>
<th>BG</th>
<th>Yield ($10^{-5}$/GMU)</th>
<th>BG avg.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Si1</td>
<td>Si2</td>
</tr>
<tr>
<td>0 T-1 (II-1)</td>
<td>0.3; 6</td>
<td>1.0; 3.7</td>
<td>BG1</td>
<td>27.3(3)</td>
<td>26.6(3)</td>
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<tr>
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<td>BG3</td>
<td>25.7(3)</td>
<td>25.0(3)</td>
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<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>BG1</td>
<td>25.4(2)</td>
<td>24.6(2)</td>
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<td>24.5(2)</td>
<td>23.6(2)</td>
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<td>0.5; 6</td>
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<td>9.38(15)</td>
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<td>8.34(16)</td>
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<td></td>
<td></td>
<td>BG5</td>
<td>1.79(15)</td>
<td>2.07(15)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>BG1</td>
<td>1.92(9)</td>
<td>2.06(10)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5</td>
<td>1.76(10)</td>
<td>1.91(10)</td>
</tr>
<tr>
<td>14 T-1a (II-11)</td>
<td>0.8; 6</td>
<td>2.0; 3.7</td>
<td>BG5</td>
<td>1.99(10)</td>
<td>1.89(10)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>1.96(10)</td>
<td>1.76(10)</td>
</tr>
</tbody>
</table>

*With 500 T-1 H$_2$ layer at DS under 20 T-1 D$_2$ layer.
we obtain in this section, such as the background and the fusion yield, will be directly applicable to the time-of-flight spectrum measurements which are our main physics data. Hence we give some detailed discussion here of the background subtraction.

**TOF series A**

The measurement for a 3 T·l layer in Series A \((c_t = 0.1\%)\) had an additional difficulty in the background subtraction, which was overcome by using different methods of background estimation and comparing them.

The most suitable background run for the \(c_t = 0.1\%\) series (BG6 in Table 8.2, Page 191) had rather limited statistics with about 1/9 of GMUs of the production run. Performing our standard background subtraction procedure using BG6, illustrated in Fig. 8.9, exhibited two potential systematic effects: (a) a Si1 to Si2 detector ratio of up to about 30% was observed in the yield using BG6 for 3 T·l (see values with † in Table 8.6), which appear to be due mostly to asymmetry in the background BG6, not the signal, and (b) there appears to be low energy tails in the background subtracted spectra down to 2 MeV (2000 ch) (see Fig. 8.9, and Table 8.6).

Using instead BG5, which does not have DS 500 T·l H\(_2\) under the DS deuterium layer, makes the Si1/Si2 ratio consistent with 1. However, the lack of the H\(_2\) layer in BG5 could cause an underestimate of the background, since muon decay electrons are a major source of background especially at lower energies. In fact, yields with BG5 are higher than those with BG6 in all cases, but clearly with a lower energy cut (see values with †† in Table 8.6).

Another possible run to be used for background subtraction is BG7, which had an emission target with \(c_t = 0.2\%\), and no H\(_2\) DS substrate layer. Despite the difference in \(c_t\), which might possibly introduce further systematic error, this run had much better statistics (about 3 times that of BG6).
### Table 8.6: Si yield per GMU for the TOF measurements (DS fusion with US moderator fixed at 14 T-l) with different background subtractions and energy cuts. Two separate series of runs were performed with tritium concentration $c_t = 0.1\%$ and 0.2%.

<table>
<thead>
<tr>
<th>US (ID)</th>
<th>$c_t$ (%)</th>
<th>E cut (x10^3 ch)</th>
<th>BG</th>
<th>Yield (10^{-6}/GMU)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Si1</td>
</tr>
<tr>
<td>3 T-l (II-9)</td>
<td>0.1</td>
<td>2.0; 3.7</td>
<td>BG5</td>
<td>5.74(40)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>5.64(42)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>5.66(24)</td>
</tr>
<tr>
<td></td>
<td>2.5; 3.7</td>
<td></td>
<td>BG5</td>
<td>4.95(33)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>5.20(34)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>4.88(21)</td>
</tr>
<tr>
<td></td>
<td>3.1; 3.7</td>
<td></td>
<td>BG5</td>
<td>4.04(25)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>4.35(25)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>4.12(16)</td>
</tr>
<tr>
<td>6 T-l (II-10)</td>
<td>0.1</td>
<td>2.0; 3.7</td>
<td>BG5</td>
<td>8.83(81)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>8.73(82)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>8.75(74)</td>
</tr>
<tr>
<td></td>
<td>2.7; 3.7</td>
<td></td>
<td>BG5</td>
<td>8.79(70)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>9.01(70)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>8.68(66)</td>
</tr>
<tr>
<td>20 T-l (II-11)</td>
<td>0.1</td>
<td>1.0; 3.7</td>
<td>BG5</td>
<td>30.2(15)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>22.4(16)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>29.1(14)</td>
</tr>
<tr>
<td></td>
<td>2.0; 3.7</td>
<td></td>
<td>BG5</td>
<td>19.3(10)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>19.2(10)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>19.2(9)</td>
</tr>
<tr>
<td></td>
<td>2.7; 3.7</td>
<td></td>
<td>BG5</td>
<td>16.3(8)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG6</td>
<td>16.5(8)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG5+6+7</td>
<td>16.2(8)</td>
</tr>
<tr>
<td>3 T-l (II-14)</td>
<td>0.2</td>
<td>2.0; 3.7</td>
<td>BG7</td>
<td>2.81(33)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG7</td>
<td>3.02(29)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG7</td>
<td>2.86(23)</td>
</tr>
<tr>
<td>23 T-l (II-15)</td>
<td>0.2</td>
<td>1.0; 3.7</td>
<td>BG7</td>
<td>23.6(10)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG7</td>
<td>21.1(7)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>BG7</td>
<td>18.6(7)</td>
</tr>
</tbody>
</table>
Given this situation for the 3 T-l measurement in the c_t = 0.1% series, we made several different background subtractions, and compared them to see the systematic effects. Method 1 is the standard procedure using BG6, but with a wide energy cut (2.5 < E < 3.7 MeV) to account for the possibility that the lower energy tails are due indeed to real $\alpha$ particle events, which gave the Si averaged yield of $Y_{\text{meth1}} = 4.43(26) \times 10^{-6}$/GMU.

The energy spectra obtained with Method 1 had nonzero counts even at energies
Figure 8.10: The energy spectra for the TOF 3 T-l ($c_t = 0.1\%$) measurement with the background as sum of BG5, 6, and 7, describing Method 2. The additional correction to the yield in the signal region SG was applied for the residual background present in the bottom figure, using the average of the background region B-LO and B-HI. See text for the details.

much higher than the fusion signal, where we expect an average of counts consistent with zero. In Method 1-b, taking into account the possibility that this is due to a potential error in the relative normalization (although the ADC blocking effect in Si detectors was corrected carefully in Section 8.1.2), we fitted the region between 4000 and 8000 ch with a constant base line, and subtracted that from the yield from Method 1 above to obtain
Figure 8.11: Background subtraction Method 2-c, where the background subtracted spectrum obtained using BG5+6+7 (bottom in Fig. 8.10) was fitted with an exponential function to remove residual background.

\[ Y_{\text{meth1-b}} = 4.06(27) \times 10^{-6} \]. Using a line with a slope or exponential curve in the fit gave a larger base line yield (hence smaller fusion yield), but we use the value \( Y_{\text{meth1-b}} \) as our upper limit in calculating the average later.

Method 2 used the entire background data set available (sum of BG5, BG6, and BG7) to make a first order background subtraction (see bottom in Fig. 8.10). But keeping in mind some differences in the conditions, we made an additional, second order correction from the above background. From the narrowly defined signal region (denoted SG, 3.1 < \( E < 3.7 \) MeV), the average of counts from the region below (shown as B-LO, 2.2 < \( E < 2.8 \) MeV) and above (B-HI, 3.8 < \( E < 4.6 \) MeV) were subtracted, i.e., \( Y_{\text{meth2}} = \text{SG} - 0.5(\text{B-LO} + \text{B-HI}) \). With SG=4.05(11) \( \times 10^{-6} \), B-LO=0.34(6) \( \times 10^{-6} \), and B-HI=0.73(10) \( \times 10^{-6} \), we have \( Y_{\text{meth2}} = 3.52(12) \times 10^{-6} \).

The systematics in Method 2 were checked for the manner in which the additional
correction was made; the background cut, B-LO, might contain the signal, which, to­
together with the narrow cut for SG, might lead to over-subtraction of background. We
present two more systematic checks of Method 2, in which the second order additional
correction was done in a different way, while keeping the first order the same as Method
2.

Method 2-b is simply to use a wider signal region of $2.7 < E < 3.7$ MeV, with B-L0
$2.1 < E < 2.6$ MeV and B-HI $3.8 < E < 4.3$ MeV. Note that because of the low energy
structure of background, we do not desire to use the spectrum much lower than 2 MeV.
Thus B-LO and B-HI now each have half the energy width of SG. The detector averaged
yield thus obtained with Method 2-b is: $Y_{meth2b} = 3.65(18) \times 10^{-6}$.

In Method 2-c, illustrated in Fig. 8.11, we have fitted the residual background obtained
in Fig. 8.10 with an exponential function. The fit was performed simultaneously in the
regions B-LO ($2.0 < E < 2.4$ MeV) and B-HI ($3.8 < E < 6.0$ MeV), and are plotted
with a solid line in Fig. 8.11. The yield was determined in the SG region ($2.5 < E < 3.7$
MeV), which was excluded from the fit, by subtracting the area under the fitted curve.
We obtain $Y_{meth2c} = 3.45(15) \times 10^{-6}$, where the error is due to the statistics in the SG
counts.

The Si yield for 3 T-l in TOF Series A ($c_t = 0.1\%$), using different background sub­
traction methods, are summarized in Table 8.7. Energy cut width dependent corrections,
*i.e.*, the energy cut efficiency and the $dd$ proton contribution, which will be discussed in
detail in Sections 8.2.4 and 8.2.3, are applied here.

As can be seen, Method 2, 2-b, 2-c, which all used combined background BG5+6+7
but had different methods of residual background subtraction, gave consistent results,
giving some confidence at least in the residual background subtraction procedure. In
comparison with Method 1, Method 1-b appears more reliable since it takes into account
the possible relative normalization error by subtracting a constant base line. Note that in
Table 8.7: Summary of the detector averaged Si yield for 3 T-1 TOF measurement 
\( (c_t = 0.1\%) \) with different background subtraction methods. The energy cut dependent 
corrections (cut efficiency and \( dd \) proton contribution) are made. Taking the average of 
two bold values we obtain \( Y_{3T1} = 3.51 \pm 0.25 \times 10^{-6}/\text{GMU} \), without the time cut and the 
nitrogen contamination corrections (see text for the details).

<table>
<thead>
<tr>
<th>Method</th>
<th>1</th>
<th>1-b</th>
<th>2</th>
<th>2-b</th>
<th>2-c</th>
</tr>
</thead>
<tbody>
<tr>
<td>BG run</td>
<td>BG6</td>
<td>BG6</td>
<td>BG5+6+7</td>
<td>BG5+6+7</td>
<td>BG5+6+7</td>
</tr>
<tr>
<td>SG cut (MeV)</td>
<td>2.5; 3.7</td>
<td>2.5; 3.7</td>
<td>3.1; 3.7</td>
<td>2.7; 3.7</td>
<td>2.5; 3.7</td>
</tr>
<tr>
<td>SG yield ((10^{-6}/\text{GMU}))</td>
<td>4.43(26)</td>
<td>4.43(26)</td>
<td>4.05(11)</td>
<td>4.64(14)</td>
<td>4.81(15)</td>
</tr>
<tr>
<td>B-LO cut (MeV)</td>
<td>–</td>
<td>–</td>
<td>2.2; 2.8</td>
<td>2.1; 2.6</td>
<td>2.0; 2.4</td>
</tr>
<tr>
<td>B-HI cut (MeV)</td>
<td>4.0; 8.0</td>
<td>3.8; 4.4</td>
<td>3.8; 4.3</td>
<td>3.8; 6.0</td>
<td>3.8; 6.0</td>
</tr>
<tr>
<td>Fit/sum for ( i )th bin</td>
<td>( y_i = \text{const.} )</td>
<td>( \sum_i y_i )</td>
<td>( \sum_i y_i )</td>
<td>( y_i = a e^{-\frac{x_i}{b}} )</td>
<td></td>
</tr>
<tr>
<td>B yield ((10^{-6}/\text{GMU}))</td>
<td>0.38(6)</td>
<td>0.54(6)</td>
<td>0.99(11)</td>
<td>1.36</td>
<td></td>
</tr>
<tr>
<td>TOF yield ((10^{-6}/\text{GMU}))</td>
<td>4.43(26)</td>
<td>4.06(27)</td>
<td>3.52(12)</td>
<td>3.65(18)</td>
<td>3.45(15)</td>
</tr>
<tr>
<td>( dd ) proton</td>
<td>2.4(9)%</td>
<td>2.4(9)%</td>
<td>0</td>
<td>2.4(9)%</td>
<td>2.4(9)%</td>
</tr>
<tr>
<td>E cut efficiency</td>
<td>1</td>
<td>1</td>
<td>0.980(3)</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>TOF corr ((10^{-6}/\text{GMU}))</td>
<td>4.32(26)</td>
<td><strong>3.96(27)</strong></td>
<td>3.59(12)</td>
<td>3.56(18)</td>
<td><strong>3.37(15)</strong></td>
</tr>
</tbody>
</table>

Series B, the constant term was consistent with zero, as expected. Discarding Method 1, 
we chose the two extreme values in (Method 1-b and Method 2-c) as representative of the 
deviations due to the background subtraction procedure. Since these measurements are 
not completely independent, determining the average and its error is somewhat tricky. 
Obviously, since the two values are incompatible within in the quoted error, simply taking 
a weighted mean and combining the error would underestimate the total uncertainty.

Here we adopt a procedure used by the Particle Data Group [229, 230], which scales 
the error bar to give \( \chi^2/\text{dof} = 1 \). In our case, with the numbers of points and the free 
parameter being 2 and 1 respectively (hence \( \text{dof} = 1 \)), we scale the error by a factor 1.9 
and obtain \( Y_{3T1} = (3.51 \pm 0.25) \times 10^{-6}/\text{GMU} \).

As for other TOF series A measurements (with \( c_t = 0.1\%) \), the statistics of the pro­
duction runs and the standard background run (BG6) are comparable, and the accuracy 
is not limited by the background statistics. Also because of a better signal-to-background
ratio, effects of the background are not as serious. Furthermore, the Si1/Si2 asymmetry in Si yields are at the 10% level for the most part. Thus, we shall use BG6 for our background subtraction. Using other backgrounds in fact gives consistent results for a reasonable range of the energy cuts (Table 8.6).

**TOF series B**

The TOF measurements with $c_t = 0.2\%$ (Series B) had a good background run (BG7) with high statistics, comparable to that of the production run. Neither of the runs had a $H_2$ substrate under the DS $D_2$. The yields with different energy cuts are given in Table 8.6.

In the 3 T-l run with $c_t = 0.2\%$ there is some 10% increase in the yield when the cut lower limit is extended from 3.1 MeV to 2.5 MeV (Method 3-nr and Method 3-wd). This is due to an increase in the Si2 counts (Si1 remains constant). Again, we test different background subtractions. In Method 3-b, the high energy ($> 4$ MeV) part of the background subtracted spectra (bottom of Fig. 8.12) were fitted with a constant line as in Method 1-b above. These gave the results consistent with zero base line, indicating there is no normalization problem. The spectra was then fitted with either exponential or linear functions in the region excluding the signal ($1500 < E < 2400$ ch and $3800 < E < 6000$ ch) to look for a potential residual background (Method 3-c). While the Si1 fit was consistent with zero background, the Si2 fit found $0.50(6) \times 10^6$/GMU background in the signal region ($2500 < E < 3700$). The results are summarized in Table 8.8. The shift between Method 3-wd and Method 3-c is due to the yield B (the area under the fitted curve in the signal region), which is well determined from the fit. Taking into account both possibilities that B is due to the background or due to the real signal, we take an average of two values and accept half the difference as our systematic uncertainty. Adding the statistical error in quadrature, we obtain the yield for Series B,
Figure 8.12: Si energy spectra for the TOF yield measurement with 3 T·l DS D₂ in Series B ($c_t = 0.2\%$), with the time cut of $1 < t < 6 \mu s$. Top figures show the fusion signal (filled circles) and a background run BG 6 (open circles), while bottom figures give background subtracted spectra.

$Y_{3\text{H}}^B = (3.05 \pm 0.24) \times 10^{-6}/\text{GMU}$. 
Table 8.8: The detector averaged Si yield for Series B \( (c_t = 0.2\%) \) 3 T-\( l \) TOF measurement with different energy cuts, with and without a residual background fit. The energy cut dependent corrections (cut efficiency and \( dd \) proton contamination) are included. Taking into account the systematic effect in the background we obtain \( Y_{\text{Tl}} = 3.05 \pm 0.24 \times 10^{-7}/\text{GMU} \).

### 8.2.3 Systematic effects

**Si1-Si2 symmetry**

Generally, higher count rates were recorded in Si2 compared to Si1. This is presumably due to the higher background rates, as Si2 was geometrically slightly closer to the beam axis. However, as was seen in Tables 8.4-8.6, the background subtracted yields are mostly consistent with the Si1-Si2 symmetry, except when the signal-to-noise ratio is low as in 3 T-\( l \) TOF measurements where we observed up to 30% asymmetry. We note, however, that while the individual detector yield can be sensitive to a slight shift in the beam position, this effect is cancelled in the averaged yield to first order. For example, in the limit of a small beam size, a 5 mm shift in the beam can cause some 40% asymmetry in the individual detector yields, yet the averaged yield is affected only by a few percent. Therefore it is crucial to have two detectors particularly for the absolute
yield determination.

**Time zero shifts**

We have observed some shift in the time zero over the course of the three week experimental run. The time shift between the signal runs and background could cause some errors in background subtraction. The uncertainty introduced by the few ns shift is obviously negligible for the TOF measurements which has a time scale of $\mu$s, but its effect on the US yield was investigated by intentionally introducing the time shift upon background subtraction, and found to be about 0.5% for a shift of 4 ns, which can also be safely neglected.

**Si detector double peaking**

Both Si1 and Si2 time spectra exhibited a small satellite peak for a prompt signal, which came 15 to 20 ns before the nominal time zero with an amplitude (averaged over two detectors) of about 5% relative to the prompt peak. The origin of the second peak is not clear, but if it contains real counts, the total yield would be underestimated if we only counted the events in the prompt peak. Again, it is negligible for the TOF and MOD measurements. The possible effect on the US yield was estimated assuming the fusion decay time of 100 ns to be less than 1%, hence we neglected the double peaking effect in our analysis.

**Cut efficiency correction**

Efficiencies for the time cuts can be determined by making similar cuts in the full Monte Carlo time spectra. For this purpose, we used the standard SMC input with modified low energy formation rate (the nominal input, see Section 8.3). For the comparison with
Figure 8.13: Energy cut efficiency determination using a dedicated Monte Carlo code APEC-97 [212] (plotted with histograms), featuring a package PEPPER [213] which incorporates a non-uniform fusion depth profile calculated with the full Monte Carlo SMC. Without PEPPER, a uniform fusion distribution is assumed. US 14 T-l Si1 data are plotted with error bars. The use of PEPPER in APEC-97 does not seem to be justified.

For energy cut efficiencies, we have used a dedicated Monte Carlo code, APEC-97 (Alpha Particle Energy Computation ver. 1997), featuring a package PEPPER (Profile Evaluation Package for Particle Energy and Range). An example of the APEC-97 outcome is illustrated with and without the use of PEPPER in Fig. 8.13. APEC-97 with
PEPPER assumes the fusion depth profile calculated by SMC, while APEC-97 without PEPPER uses a uniform fusion profile.

It is interesting to notice that the simple uniform fusion depth profile (without PEPPER) seem to describe the observed Si spectrum better than the profile derived from a detailed full simulation of the muonic processes leading to fusion (with PEPPER). This might be due to real physics, but from these data alone we cannot exclude other possibilities such as non-uniform target layers (which is only included as the effective average thicknesses, see Section 6.1) or an inaccurate assumption of beam radial distribution\(^7\).

Nonetheless we take the average of the cut efficiency derived from each depth profile, and take the difference as a measure of uncertainty. The derived energy cut efficiencies, together with various other corrections, will be given later in Table 8.13.

**Nitrogen contamination**

If there was a contamination of heavier elements in the emission layer, the yield of \(\mu t\) would be reduced due to muon transfer from the proton to the heavy element. In some of the runs, we observed muonic X rays from muonic nitrogen atomic transitions, indicating contamination of the target. From the analysis of the Lyman series X rays, the contamination level was estimated to be a few ppm level by Francoise Mulhauser \[83, 231\].

In the constant rate, infinite medium approximation, the probability of \(\mu t\) production \(P_{\mu t}^N\) can be expressed as:

\[
P_{\mu t}^N = \frac{\phi c_1 \lambda_{pt}}{\lambda_0 + \phi (c_p \lambda_{p\mu p} + c_t \lambda_{pt} + 2c_{N_2} \lambda_{pN})},
\]

where \(c_{N_2}\) is the concentration of \(N_2\) *molecules*, and \(\lambda_{pN}\) the muon transfer rate from a proton to a nitrogen *atom*. Using the proton to triton transfer rate \(\lambda_{pt} = 5.86 \times 10^3\ \mu s^{-1}\), and \(p\mu p\) formation rate \(\lambda_{p\mu p} = 3.21\ \mu s^{-1}\), both obtained from our earlier measurements.

\(^7\)Flat-top Gaussian 10 $\oplus$ 10 mm was assumed for the fusion radial profile in these simulations.
in solids [83], together with the transfer rate to nitrogen $\lambda_{PN} = 0.34 \times 10^{11} \text{ } \mu s$ from Ref. [232] (measured in a gas), we can estimate the reduction factor for the $\mu t$ production $\epsilon^N \equiv \mathcal{P}^N_{\mu t}/\mathcal{P}^0_{\mu t}$, which is normalized to the pure target yield $\mathcal{P}^0_{\mu t} = 0.621$. Table 8.12 presents the correction factor for two series of target sets which are of relevance in our analysis.

\begin{center}
\begin{tabular}{|c|c|c|c|c|}
\hline
Target ID & cN$_2$ (Ref. [231]) & $\mathcal{P}^N_{\mu t}$ & $\epsilon^N$
\hline
SETc$_t = 0.1\%$ (a) & II-1 to II-4 & 3.3$^{+5.5}_{-3.3} \text{ ppm}$ & 0.621$^{+0.14}_{-0.28}$ & 0.966$^{\pm 0.034}$

SETc$_t = 0.1\%$ (b) & II-6 to II-11 & 2.3$^{+4.6}_{-2.3} \text{ ppm}$ & 0.611$^{+0.11}_{-0.23}$ & 0.976$^{\pm 0.024}$
\hline
\end{tabular}
\end{center}

Table 8.9: The $\mu t$ production probability $\mathcal{P}^N_{\mu t}$ with possible nitrogen contamination, and the reduction factor $\epsilon^N$, normalized to a pure emission target $\mathcal{P}^0_{\mu t} = 0.621$.

We note that the estimate of nitrogen contamination by Mulhauser gives asymmetric errors as quoted in the table, but we take the average of two extreme values of 1$\sigma$ error bars for convenience in the data treatment. In our analysis, we assume that reduction in $\mu t$ emission is proportional to the factor $\epsilon^N$ given here, neglecting the effects of the $\mu t \rightarrow \mu N$ transfer, which is presumably much smaller.

### 8.2.4 $d\mu d$ fusion

Protons from $d\mu d$ fusion in the deuterium layer can cause a background in the $\alpha$ measurements depending on the energy cut (recall that the proton energy is 3 MeV). They can come from two different sources: (a) direct stopping of muons in the deuterium layer, or (b) recycled muons, i.e., the muons released after the fusion reaction (with the probability $1 - \omega_s$, where $\omega_s$ is the sticking probability). For both cases, because of solid state effects and finite thickness, estimating the proton yield is difficult, and there is yet no satisfactory theoretical model. The use of the Monte Carlo is untested in these
conditions, and its reliability is questionable without solid state effects included in the input.

Nonetheless, we first make an analytical estimate using the two node kinetics model which successfully, if accidentally, described the time evolution of \( \mu d \) fusion in a bulk solid deuterium [199]. According to this kinetics approximation, with an assumption of an infinite target and ignoring \( \mu d \) cycling, \( \mu d \) fusion yield per muon \( \mathcal{P}_{\mu d} \) can be obtained as:

\[
\mathcal{P}_{\mu d}^{\text{kin}} = f_\frac{2}{3} \left[ \frac{\phi \lambda_{\mu d}^{\frac{3}{2}}}{\phi(\lambda_{\mu d}^{\frac{3}{2}} + \lambda_{\mu d}^{\frac{1}{2}}) + \lambda_0} + \frac{\phi \lambda_{\mu d}^{\frac{3}{2}}}{\phi(\lambda_{\mu d}^{\frac{3}{2}} + \lambda_{\mu d}^{\frac{1}{2}}) + \lambda_0} \right] + f_\frac{1}{3} \frac{\phi \lambda_{\mu d}^{\frac{1}{2}}}{\phi \lambda_{\mu d}^{\frac{1}{2}} + \lambda_0},
\]

(8.2)

where \( \phi = 1.4 \) is the target density in units of liquid hydrogen atomic density, \( f_\frac{2}{3} = \frac{2}{3}, f_\frac{1}{3} = \frac{1}{3} \) are the initial hyperfine populations, and \( \lambda_0 = 0.456 \ \mu s^{-1} \) is the free muon decay rate. Using the effective formation rates from our measurements in thick solid deuterium targets [166, 199], i.e., molecular formation rates from each hyperfine state, \( \lambda_{\mu d}^{\frac{3}{2}} = 2.7 \ \mu s^{-1}, \lambda_{\mu d}^{\frac{1}{2}} = 0.044 \ \mu s^{-1} \), and the spin flip rate, \( \lambda_{\mu d}^{\frac{1}{2}} = 34 \ \mu s \), we obtain \( \mathcal{P}_{\mu d}^{\text{kin}} \sim 0.16 \). The fusion branching ratio into protons \( B_p \) is a somewhat complicated function of atomic hyperfine states and molecular angular momentum states, but for our analysis here, it suffices to let \( B_p \sim 0.5 \). Hence we have the proton yield per muon in the kinetics model, \( \mathcal{P}_p^{\text{kin}} \)

\[
\mathcal{P}_p^{\text{kin}} = \mathcal{P}_{\mu d}^{\text{kin}} \cdot B_p \sim 0.08,
\]

(8.3)

of which 30% comes from the fast \( (34 \ \mu s^{-1}) \) component, and the rest with the slower time slope.

This level of proton yield would give a non-negligible contribution in the \( \alpha \) yield measurements, when the direct stops and the recycling components are combined. However,
a proton yield at the 10% level relative to $\alpha$ can be excluded by comparing the expected peak shape, simulated with an energy loss Monte Carlo program [212], with experimental energy spectra. Recall that the $dd$ proton, with an energy of 3 MeV, has energy loss much smaller than a 3.5 MeV $\alpha$, by a factor of roughly $(Z_p/Z_\alpha)^2(M_p/M_\alpha) \sim 1/16$, hence a 10% proton yield would appear as a sharp peak in the Si energy spectra, which is absent from the data. Given the inconsistency with this simple analytical approach, we pay closer attention to our data set to extract the proton contribution in the sections which follow.

**Direct stop contribution at late time**

As we mentioned, the fusion yields in the upstream layers were measured with a time cut of $0.02 < t < 0.4 \mu s$, while the downstream measurements were done mainly with $1 < t < 6 \mu s$. The proton contributions at these two time ranges, as well as those from different sources (e.g., direct vs. cycling), need to be considered separately.

We first investigate the protons from direct muon stops in a $D_2$ layer at late time. This is important for the measurements using thick downstream layers, from which the DS direct proton contribution cannot be subtracted by using the standard background runs which had no downstream layer.

In order to estimate this contribution, we looked at the yield of protons coming from the 14 T-l upstream deuterium layer when only a very thin downstream layer was present. Given in Table 8.10 is the yield in the peak at 3 MeV ($2960 < E < 3040$ ch) with time cut of $1 < t < 6 \mu s$ for two different series of runs. The counts just above and below the peak in the energy spectra (with the same total bin width), were subtracted to account for the background.

As we will see later cycling from the US layer contributes very little at this late time, and assuming cycling from DS is negligible\(^8\) since the layer is very thin, we obtain $Y_{p\text{-late}}^{\text{dir}}$.

\( ^8 \)This approximation introduces an error of no more than 0.5% in the final correction.
Table 8.10: \( Y_{p\text{-late}}^{dir} \), the proton yield in late time (\( 1 < t < 6 \mu s \)).

The proton yield from direct stops in 17 T-l (14 US + 3 DS) of deuterium layers in the time region of \( 1 < t < 6 \mu s \). Averaging the above values, we have \( Y_{p\text{-late}}^{dir} = 1.68(32) \times 10^{-7} \) protons per GMU. Our downstream thicknesses ranged from 6 T-l to 23 T-l, hence this yield was scaled proportionally to the thickness in order to give the direct stop contribution at late time.

**Cycling contribution**

For the cycling contribution, we take advantage of the fact that we had two independent Si detectors, and study the correlations between them. With an energy cut of \( 2501 < E < 3700 \) ch and the time cut of \( 0.02 < t < 0.4 \mu s \) in one of the detectors, which selected alpha events with a high probability, we looked at the other detector.

With an energy cut in the second detector of \( 2701 < E < 3100 \) ch to select proton events, we obtained the coincidence rates with two different time cuts: (a) \(-0.01 < (T_p - T_\alpha) < 6 \mu s\), and (b) \(0.01 < (T_p - T_\alpha) < 6 \mu s\), where \( T_p - T_\alpha \) is the time difference between the proton and the \( \alpha \) candidate events. The yield with time cut (b) was corrected for the cut efficiency assuming the \( d\mu d \) fusion disappearance rate of \( 34 \mu s^{-1} \), given by the hyperfine transition rate in the kinetics model [166], as well as the average time-of-flight difference between the 3 MeV proton and 3.5 MeV \( \alpha \), while the cut (a) required no time corrections. The conditional solid angle for the protons given an \( \alpha \) is detected, could be different from the solid angle singles events, since the \( \alpha \) detection biases the proton.
distribution towards the first detector (i.e., away from the proton detector). This effect was estimated using a Monte Carlo [212] taking into account the geometrical correlation of the two detectors, and found to give about 10% (relative) reduction in the second detector. Using the coincidence solid angle, $\Omega_{\text{coin}} = 2.1\%$, the $\alpha$ yield with the energy cut $2501 < E < 3700$ ch, $Y_{\alpha} = 4 \times 10^{-3}/\text{GMU}$, and the time cut efficiency $\epsilon_t = 0.71$ for cut (a) ($\epsilon_t = 1$ for the cut (b)), we obtain the proton yield per $dt$ fusion from cycling, $P_{p}^{\text{cyc}}$ as

$$P_{p}^{\text{cyc}} = \frac{Y_{\text{coin}}}{Y_{\alpha} \Omega_{\text{coin}} \epsilon_t},$$

(8.4)

where $Y_{\text{coin}}$ is the coincidence yield per GMU, and we ignored the small sticking probability in $d\mu t$ fusion. The resulting $P_{p}^{\text{cyc}}$ from Target II-13 are given in Table 8.11.

<table>
<thead>
<tr>
<th>Time cut $T_{\alpha} - T_p$</th>
<th>Proton yield/dt fusion $(\times10^{-2})$</th>
<th>Si1(p) • Si2(α)</th>
<th>Si2(p) • Si1(α)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$[-0.1, 6] \mu s$</td>
<td>2.6 (4)</td>
<td>2.3 (4)</td>
<td></td>
</tr>
<tr>
<td>$[0.1, 6] \mu s$</td>
<td>0.9 (3)</td>
<td>0.9 (3)</td>
<td></td>
</tr>
</tbody>
</table>

Table 8.11: $P_{p}^{\text{cyc}}$, the probability that the proton is produced after $dt$ fusion is observed, is given for each coincidence pairs and the time cuts.

The inconsistency between the results from the two time cuts can be due either to the time cut efficiency or possible presence of a prompt background (such as $\mu$ decay electron). For example, if the $d\mu d$ fusion disappearance rate is different in a thin layer from 34 $\mu s^{-1}$ measured in bulk solid targets, the time correction made is not appropriate. As well, with limited statistics, it is difficult to estimate the prompt background contribution. Given the uncertainty, we take the average of the two cuts, and quote an error covering two extremes of the error bars. Thus we have $P_{p}^{\text{cyc}} = 1.6(9) \times 10^{-2}$ as the contribution from cycling. In our analysis below, we assume the cycling proton yield per $dt \alpha$ to be independent of the $D_2$ layer thickness, which is sufficient for the accuracy required here.
We note that at late time $1 < (T_p - T_a) < 6 \mu s$, the coincidence yield is more than an order of magnitude smaller than at early times, hence the slow lifetime component of $d\mu d$ (observed in bulk solid and other targets) appears nearly absent from cycled fusion in a thin layer.

The value of $P_p^{cyc}$ here should be compared with our earlier estimate with the kinetics model $P_p^{kin} \sim 0.08$ given by (Eq. 8.3), which is significantly higher. It is interesting to note, however, that our value is more or less consistent with the estimated fast component yield in the kinetics model $(0.3 \times P_p^{cyc} \sim 0.024)$. One possibility is that the muonic deuterium in the lower hyperfine state ($F = \frac{1}{2}$) escapes from the layer before fusion takes place, since the low rate of non-resonant fusion ($\lambda^F \sim 0.04 \mu s$), which is responsible for the slow component, implies a rather long interaction length. Another possibility, of course, is that the muon escapes from the layer before stopping to form muonic deuterium [233]. Physics of the muon cycling and the $\mu d$ transport in thin layers is a very interesting topic on its own, indeed.

**Direct stop contribution at early time**

The contributions from direct muon stops in the deuterium layer at early times are difficult to estimate experimentally from our data set, since at such times, an overwhelming $d\mu t$ signal is present in the spectrum. We therefore use the information from the cycling contribution and express the direct stop proton yield per GMU $Y_{p-fast}^{dir}$ as:

\[
Y_{p-fast}^{dir} = S_H T_{D_2} \frac{P_p^{cyc}}{\eta_{cyc}} \Omega_{Si},
\]

where $S_H = 0.299$ is the muon stopping fraction in the emission layer, $T_{D_2} = (14 \ T \cdot l)/(1000 \ T \cdot l)$ is the thickness fraction of the $D_2$ layer with respect to the emission layer, and $\eta_{cyc} \equiv P_p^{cyc}/P_p^{dir}$ is a phenomenological parameter which describes the ratio in the proton yields between $d\mu d$ fusion after $dt$ cycling and the direct stop. If we assume
the spatial distribution of cycled muons is the same as that of direct stop muons, the \( \eta_{cyc} \) parameter simply accounts for the muon escape from the layer before stopping to form \( \mu d \). Even if we allow a value as low as \( \eta_{cyc} = 0.5 \) (i.e., up to 50\% of muons escaping), the yield will be \( Y_p^{dir} \sim 1.8(1.0) \times 10^{-6} \), which is only 0.46(26)\% of the \( \alpha \) yield. Thus the uncertainty in \( \eta_{cyc} \) is not serious for our purpose here. For thicknesses other than 14 T-l, we estimate the yield by scaling to the thickness ratio. Table 8.12 summarizes the corrections due to protons from \( d\mu d \) fusion. We shall apply the correction for the DS (estimated here for the time cut \( 1 < t < 6 \mu s \)) to the MOD measurements as well, ignoring some differences in the time cut.

<table>
<thead>
<tr>
<th>D(_2) target</th>
<th>Proton yield ( Y_p^{d\mu d} ) per ( \alpha ) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cycling</td>
</tr>
<tr>
<td>US 20 T-l</td>
<td>1.64 (94)</td>
</tr>
<tr>
<td>14 T-l</td>
<td>ditto</td>
</tr>
<tr>
<td>6 T-l</td>
<td>ditto</td>
</tr>
<tr>
<td>3 T-l</td>
<td>ditto</td>
</tr>
<tr>
<td>DS 23 T-l</td>
<td>1.64 (94)</td>
</tr>
<tr>
<td>20 T-l</td>
<td>ditto</td>
</tr>
<tr>
<td>6 T-l</td>
<td>ditto</td>
</tr>
<tr>
<td>3 T-l</td>
<td>ditto</td>
</tr>
</tbody>
</table>

Table 8.12: Summary of the correction due to protons from \( d\mu d \) fusion. US is with the time cut of \( 0.02 < t < 0.4 \mu s \) and DS with \( 1 < t < 6 \mu s \).

### 8.2.5 The yield results

Table 8.13 summarizes our results for the yield measurements with the corrections discussed in this section and others. Those include the energy cut efficiency \( E_{cut}^E \), \( dd \) proton contribution \( Y_p^{d\mu d} \), the time cut efficiency \( T_{cut}^T \), and nitrogen contamination effects \( N \). The \( dt \) fusion yield \( Y_{dt} \) is normalized to stopped muons using the stopping fraction.
0.299(15) and the target thickness dependent Si solid angle (Table 6.5, p. 129). That is,

\[ Y_{dt} = \frac{Y_{Si}}{S_H \Omega_{Si} \prod_i \epsilon_i}, \]  

where \( Y_{Si} \) is the \( \alpha \) yield in the Si detector per GMU (averaged over Si1 and Si2), \( S_H \) is the stopping fraction, \( \Omega_{Si} \) is Si solid angle per detector, and \( \epsilon_i \) are the various corrections such as the cut efficiencies and \( N_2 \) contamination. The final uncertainties quoted in the table include quadratically added errors for these corrections. The results with different energy cuts are mostly consistent with each other\(^9\). In order to avoid the occasional presence of low energy background, bold-faced values with narrower energy cuts (when more than one is available) are chosen as our final results.

Figure 8.14 plots our final yield results for the US, MOD, and TOF measurements. The comparison of the data taken with different conditions, e.g. with different \( c_t \), requires some caution. For a quantitative understanding, the comparison with Monte Carlo calculations is essential, and will be dealt with in the sections that follow.

---

\(^9\)In a very crude statistical estimate, 6 cases out of 16 data point have the deviation between two energy cuts which is larger than the uncorrelated error (i.e., without normalization errors). This is perfectly consistent with statistical fluctuation.
### Chapter 8. Analysis III – Molecular formation

#### Table 8.13: The fusion yields $Y_{dt}$ per stopped $\mu$ in the US (top), MOD (middle) and TOF (bottom) measurements, with corrections due to the energy and the time cuts $E_{cut}$, $T_{cut}$, the effects of $dd$ proton $Y_{p}^{d,d}$, and of $N_2$ contamination $e^{N}$. The Si solid angles from Table 6.5 and the stopping fraction of 0.299(15) are used. Bold-faced values are chosen as our final results.

<table>
<thead>
<tr>
<th>US (DS)</th>
<th>ID (ct)</th>
<th>E cut $(10^3 \text{ ch})$</th>
<th>Si yield $(10^{-5} / \text{GMU})$</th>
<th>$E_{cut}$</th>
<th>$Y_{p}^{d,d}$ $(% / \alpha)$</th>
<th>$T_{cut}^d$</th>
<th>$e^{N}$</th>
<th>$Y_{dt}$ $(10^{-3} / \mu \text{ stop})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 T-l (20)</td>
<td>II-2</td>
<td>2.7; 3.7</td>
<td>5.77(9)</td>
<td>1.0</td>
<td>1.7(9)</td>
<td>.980(3)</td>
<td>.890</td>
<td>.966(34)</td>
</tr>
<tr>
<td>6 T-l (20)</td>
<td>II-3</td>
<td>2.0; 3.7</td>
<td>16.58(14)</td>
<td>1.0</td>
<td>1.8(9)</td>
<td>.975(5)</td>
<td>.886</td>
<td>.966(34)</td>
</tr>
<tr>
<td>14 T-l (20)</td>
<td>II-4</td>
<td>2.0; 3.7</td>
<td>42.73(24)</td>
<td>.970(4)</td>
<td>2.1(10)</td>
<td>.884</td>
<td>.966(34)</td>
<td>72.6(54)</td>
</tr>
<tr>
<td>14 T-l (0)</td>
<td>II-7</td>
<td>1.0; 3.7</td>
<td>43.43(19)</td>
<td>.970(4)</td>
<td>2.1(10)</td>
<td>.884</td>
<td>.976(24)</td>
<td>73.8(55)</td>
</tr>
<tr>
<td>14 T-l (0)</td>
<td>II-13</td>
<td>.5%</td>
<td>45.75(35)</td>
<td>.970(4)</td>
<td>2.1(10)</td>
<td>.884</td>
<td>1.0</td>
<td>75.2(50)</td>
</tr>
<tr>
<td>20 T-Z (0)</td>
<td>II-16</td>
<td>1.0; 3.7</td>
<td>44.90(16)</td>
<td>.914(16)</td>
<td>2.3(10)</td>
<td>.884</td>
<td>1.0</td>
<td>78.2(53)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>US (T cut)</th>
<th>ID (ct)</th>
<th>E cut $(10^3 \text{ ch})$</th>
<th>Si yield $(10^{-5} / \text{GMU})$</th>
<th>$E_{cut}$</th>
<th>$Y_{p}^{d,d}$ $(% / \alpha)$</th>
<th>$T_{cut}^d$</th>
<th>$e^{N}$</th>
<th>$Y_{dt}$ $(10^{-3} / \mu \text{ stop})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 T-l (.3; 6)</td>
<td>II-1</td>
<td>1; 3.7</td>
<td>25.35(20)</td>
<td>.954(18)</td>
<td>2.6(10)</td>
<td>1.0</td>
<td>.966(34)</td>
<td>36.6(28)</td>
</tr>
<tr>
<td>3 T-l (.5; 6)</td>
<td>II-2</td>
<td>1; 3.7</td>
<td>15.38(88)</td>
<td>.954(18)</td>
<td>2.6(10)</td>
<td>1.0</td>
<td>.966(34)</td>
<td>22.5(22)</td>
</tr>
<tr>
<td>6 T-l (.3; 6)</td>
<td>II-3</td>
<td>1; 3.7</td>
<td>8.82(90)</td>
<td>.954(18)</td>
<td>2.6(10)</td>
<td>.988</td>
<td>.966(34)</td>
<td>13.1(22)</td>
</tr>
<tr>
<td>14 T-l (.8; 6)</td>
<td>II-4</td>
<td>1; 3.7</td>
<td>1.93(11)</td>
<td>.954(18)</td>
<td>2.6(10)</td>
<td>.998</td>
<td>.966(34)</td>
<td>2.85(27)</td>
</tr>
<tr>
<td>14 T-l (.8; 6)</td>
<td>II-11</td>
<td>1; 3.7</td>
<td>2.15(12)</td>
<td>.954(18)</td>
<td>2.6(10)</td>
<td>.998</td>
<td>.976(24)</td>
<td>3.14(29)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>DS (ct)</th>
<th>ID</th>
<th>E cut $(10^3 \text{ ch})$</th>
<th>Si yield $(10^{-5} / \text{GMU})$</th>
<th>$E_{cut}$</th>
<th>$Y_{p}^{d,d}$ $(% / \alpha)$</th>
<th>$T_{cut}^d$</th>
<th>$e^{N}$</th>
<th>$Y_{dt}$ $(10^{-4} / \mu \text{ stop})$</th>
<th>Final Yield $(10^{-4} / \mu \text{ stop})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 T-l (.1%)</td>
<td>II-9</td>
<td>3.51(25)×$10^{-6}$ (see Table 8.7)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>.969</td>
</tr>
<tr>
<td>6 T-l (.1%)</td>
<td>II-10</td>
<td>2.0; 3.7</td>
<td>8.55(60)</td>
<td>1.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>.962</td>
</tr>
<tr>
<td>20 T-l (.1%)</td>
<td>II-11</td>
<td>1.0; 3.7</td>
<td>20.7(11)</td>
<td>.954(18)</td>
<td>2.6(10)</td>
<td>.948</td>
<td>.976(24)</td>
<td>31.9(29)</td>
<td></td>
</tr>
<tr>
<td>3 T-l (.2%)</td>
<td>II-14</td>
<td>3.05(24)×$10^{-6}$ (see Table 8.8)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>.971</td>
</tr>
<tr>
<td>23 T-l (.2%)</td>
<td>II-15</td>
<td>1.0; 3.7</td>
<td>23.16(73)</td>
<td>.936(29)</td>
<td>2.8(10)</td>
<td>.948</td>
<td>1.0</td>
<td>34.6(27)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>21.08(52)</td>
<td>.876(52)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 8.14: Fusion yield per stopped muon for the US (top), MOD (middle), and TOF (bottom) measurements. The error bars include the absolute normalization error.
8.2.6 Time-of-flight spectra

In this section, we shall discuss our time-of-flight spectrum (TOFS) measurements. Table 8.14 summarizes the runs for TOF time spectra measurements. In all of these runs, we had a 14 T-l D$_2$ layer US to slow down the $\mu t$ beam emitted from the emission layer. Recall that the RT minimum is around 10–15 eV in lab $\mu t$ energy, which is too high for the resonant molecular formation energies that we are interested in. While Series A had a 500 T-l H$_2$ substrate under the D$_2$ reaction layer DS, in Series B the deuterium was deposited directly on the Au foil. The H$_2$ substrate was initially used with the intention of reducing a background (especially in the neutron detectors) from muon capture on Au, which comes from $\mu t$ passing through the thin reaction layer and reaching the Au foil, hence having the time structure similar to that of a real signal. But as we saw in the yield measurements, the H$_2$ causes significant background due to muon decay, and as far as the background with the characteristic time-of-flight is concerned, it is dominated by the $\mu t$ directly hitting the Si detectors. Furthermore, it was learned in the course of the analysis that the H$_2$ substrate creates some ambiguity in the interpretation of the data due to possible $\mu t$ re-emission.

<table>
<thead>
<tr>
<th>TOF spect.</th>
<th>ID</th>
<th>US (T-l)</th>
<th>DS (T-l)</th>
<th>Energy cut (MeV)</th>
<th>BG</th>
</tr>
</thead>
<tbody>
<tr>
<td>Series A</td>
<td>II-9</td>
<td>14</td>
<td>H$_2$</td>
<td>3.1; 3.7</td>
<td>6+7+8 2</td>
</tr>
<tr>
<td></td>
<td>II-10</td>
<td></td>
<td>(500 T-l)</td>
<td>2.7; 3.7</td>
<td>6   standard</td>
</tr>
<tr>
<td></td>
<td>II-11</td>
<td></td>
<td></td>
<td>2.0; 3.7</td>
<td>6   standard</td>
</tr>
<tr>
<td></td>
<td>II-14</td>
<td>14</td>
<td>Au foil</td>
<td>3.1; 3.7</td>
<td>7   3-nr</td>
</tr>
<tr>
<td></td>
<td>II-15</td>
<td></td>
<td>(50 $\mu$m)</td>
<td>2.0; 3.7</td>
<td>7   standard</td>
</tr>
</tbody>
</table>

Table 8.14: Summary of the runs for the TOF time measurements. See Tables 8.7, 8.8 for the background subtraction methods.

Much of the details about background and various other corrections were already
given in the previous sections. It should be recalled that the background includes events from muon decay electrons, muon capture on Si, delayed $dt$ fusion from US, and $dd$ fusion protons (if the energy window extends lower than 3 MeV). The complexity in the background processes makes it virtually impossible to predict the time structure, and makes it unreliable to use a simple analytical function, hence we rely on bin-by-bin subtraction, which at the cost of statistical precision allows us better control of the
Figure 8.16: The TOF run with 3 T·l DS for Series B ($c_t = 0.2\%$) in filled circles, together with background run BG7 in open circles, both with an energy cut of $3101 < E < 3700$ ch (top). Background subtracted time-of-flight fusion spectrum (bottom).

We show some examples of the fusion time-of-flight spectra in Figs. 8.15 and 8.16. Figure 8.15 shows the TOF spectrum for Series A ($c_t = 0.1\%$) 3 T·l measurement, together with the background data from BG6, both with the energy cut of $3.1 < E < 3.7$ (see Table 8.2 for the background run information), while Figure 8.16 is for the 3 T·l measurement in Series B ($c_t = 0.2\%$). In the top figures, the non-exponential peak in the
fusion time spectra is noticeable at around 2-4 $\mu$s, which, together with the lack of such a peak in the runs without DS reaction layers, indicate the fusion is indeed taking place in the DS layer after a traversing the drift space. The fact that for the thin DS layer measurement, the energy width of these delayed events was so narrow in Fig. 8.9–8.12 in the previous section, corroborates that fusion is occurring at the downstream layer in which the $\alpha$ particle suffers less energy loss.

8.3 Monte Carlo analysis

In this section, we make use of the Monte Carlo simulations to understand our data and extract the physics. The details of our Monte Carlo code SMC has been already given in Section 5.1. The physics input into the simulations is mostly taken from the Nuclear Atlas [16,17] for scattering cross sections, and Faifman et al. for the resonant [70–72,133] and nonresonant [51] molecular formation rates, with exceptions that (a) the muon transfer rate in $\mu p \rightarrow \mu t$, $\lambda_{\mu t} = 5.86 \times 10^9$ s$^{-1}$, and $p\mu p$ formation rate (nonresonant) $\lambda_{p\mu p} = 3.2 \times 10^6$ s$^{-1}$ were taken from our measurements using the same data set [83], and (b) the low energy behavior of Faifman’s resonant $d\mu t$ formation rate was modified to reflect the recent measurement [22] and theory [146]; the rate in $\mu t^{F=0} + D_2^{ortho} \rightarrow [(d\mu t)dee]$ at $E_{\mu t} < 15$ meV was set to 130 $\mu$s$^{-1}$. This set of physics input parameters will be referred to as the nominal input to the simulations. Our experimental data will confront these physics cross sections and rates.

8.3.1 Fusion yield analysis

Before we test our main results on time-of-flight fusion spectra in the next section, let us first discuss the fusion yield measurements, which give us some confidence in our model as well as an indication of any new effects. Let us start with the analysis of the US yield
Chapter 8. Analysis III – Molecular formation

Figure 8.17: Monte Carlo simulations for US fusion yield (lines) compared to the experimental yield (open squares). MC(1) shows the nominal predictions with the uncertainties due to the layer thickness indicated with the dotted lines. In MC(3), a constant rate $\lambda_{\text{dpt}} = 5 \text{ ps}$ is added to the nominal $d\mu t$ formation rates, whereas MC(2) is with the original $d\mu t$ formation from Ref. [70–72, 133] without the low energy modification.

US yield

Figure 8.17 shows a comparison of the upstream fusion yields with the different Monte Carlo calculations. Only data from the same series of runs (ID=II-2, II-3, and II-4) are plotted to avoid a possible influence of the target conditions. Denoted MC(1) in the figure is our nominal physics input discussed above, with the possible variations in the yield due to the layer thickness uncertainties, which are shown with dotted lines. Separate Monte Carlo runs with varied thickness inputs were performed for the latter. MC(2) is using
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the original $d\mu t$ formation from Ref. [70–72,133] without the low energy modification, while for MC(3) an energy independent rate $\lambda_{d\mu t}^c = 5 \, \mu s$ (also independent of $F$) was added to the nominal $d\mu t$ formation rates.

Our results are in rough agreement with the nominal MC, but using the original model of Faifman worsens the agreements at larger layer thicknesses. Many Monte Carlo calculations were performed in order to find a better agreement, varying the input such as formation rates and scattering cross sections and the transfer rates, but no simple scaling of any of these parameters allowed perfect agreement with the data.

A significant improvement is observed, however, when the constant rate $\lambda_{d\mu t}^c$ is added. This is a phenomenological parameter motivated primarily by reproducing the experimental data, and its interpretation requires careful consideration, which we shall later come back to. The dependence of the fusion yield in thick layers on $\lambda_{d\mu t}^c$ suggests the importance of, and our sensitivity to, the low energy processes in the solid state of hydrogen, but fortunately for our measurements of the epithermal molecular formation in thin layers, the low energy effects are expected to be rather small as we shall see.

In our nominal model, $\lambda_{d\mu t}^{F=0}$ at low energy is already set to a considerably higher value than in the original Faifman’s model, hence the main effect in the difference between MC(2) and MC(3) comes from the increase in the triplet formation rate. Indeed, increasing the singlet rate alone to $\lambda_{d\mu t}^{F=0} = 4000 \, \mu s^{-1}$, while keeping the triplet rate at Faifman’s value, does not change the fusion yield.

MOD yield

In the MOD yield analysis, before comparing with the MC yield, we used a simple attenuation model to analyze our data, which gave us crude yet useful insight into the physics involved.

In the one dimensional approximation, the yield $Y$ of particles after going through a
medium of thickness $d$ can be expressed as:

$$Y = A_0 \exp \left( -\frac{d}{L_{\text{att}}} \right),$$

(8.7)

where $A_0$ is the number of the original particle, $d$ medium thickness, and the $L_{\text{att}}$, the attenuation interaction length. The attenuation interaction is defined here as a process which absorbs or deflects the particle.

Fitting our data using Eq. 8.7 gave us a rather good fit with $\chi^2/dof = 1.67$ (confidence level 19%, $dof = 2$), which is somewhat surprising, considering the degree of approximation introduced\(^{10}\). Nonetheless, with some caution in its interpretation, we can extract a phenomenological parameter, the attenuation interaction length in $\mu t + d$ collisions $L_{\text{int}}^{\text{eff}} = 37.7 \pm (1.2)_{\text{fit}} \pm (2.0)_{\text{thick}} \mu g \cdot cm^{-2}$, where the first error is given by the fit, and the second error is due to the uncertainty in the layer thickness (the latter was obtained from different fits in which the thickness was varied). The error in the experimental yield used in the fit included only the relative uncertainties, since normalization errors such as muon stopping fraction and Si solid angle cancel in the thickness dependence. The value of $L_{\text{int}}^{\text{eff}}$ can be converted to the effective attenuation interaction cross section $\sigma_{\text{att}}^{\text{eff}} = 4.4 \times 10^{-20} \text{ cm}^{-2}$, using $\sigma = 1/(nL)$ with $n$ the number density. This can be compared to the $\mu t + d$ total elastic scattering cross section \([17]\) ~ $20-50 \times 10^{-20} \text{ cm}^{-2}$ at $\mu t$ lab energies of 1-20 eV, near which $\mu t$ emission is distributed. The difference in the two cross sections can be in part understood in that it takes more than one collision to attenuate the $\mu t$, especially given the scattering angular distribution, which we will shortly come to.

We now turn to the comparisons with the SMC simulations. Encouraged by its success in fitting the experimental data, we apply the attenuation model also to characterize the

---

\(^{10}\)The detection of $\mu t$ using the fusion signal is assumed to be constant in this model. This is not precisely correct due to the energy dependence of the molecular formation rates. In addition, the effect of muon decay is obviously not included in the model. The fraction of the $\mu t$ reaching DS before muon decay changes if the energy of the transmitted $\mu t$ changes.
Figure 8.18: Experimental data (EXP) and Monte Carlo results for the MOD yield measurements plotted on a log scale. MC(a) used the nominal input which had the $\mu t + d$ scattering angular distribution from Ref. [17] which is forward peaked, while MC(b) used an isotropic distribution $d\sigma_{\mu t+d}(\theta)/d(cos \theta) = \text{const}$. The solid line is a fit of the MC(a) to an expression $A_0 \exp(-x/L_{eff}^{MC(a)})$ with the dashed lines indicating variations in the slope due to the fit uncertainty and to thickness uncertainty, while the dash-dotted line is a fit to MC(b), indicating the importance of the $p$-wave scattering in the $\mu t + d$ interaction. Both fits are normalized to the experimental point at thickness 0.

MC results. Figure 8.18 illustrates our Monte Carlo analysis. Plotted with error bars are experimental data, while crosses indicate the yield from the MC. The lines are fits to the MC yield from which we extract the effective attenuation lengths as above. Shown in the solid line (MC(a)) is a fit to the MC results with the nominal input, with dotted lines indicating the variations in the slope due to the fit uncertainty and the thickness errors, where we obtained $L_{eff}^{MC(a)} = 34.9 \pm (2.4)_{fit} \pm (2.1)_{thick} \mu g \cdot cm^{-2}$. This is consistent at the 10% level with the experimental value extracted above. Indeed, the same conclusion can
Chapter 8. Analysis III – Molecular formation

<table>
<thead>
<tr>
<th></th>
<th>$L_{\text{int}}^{\text{eff}}$ ($\mu g\cdot cm^{-2}$)</th>
<th>$d\sigma^\mu t + d / d(\cos \theta)$</th>
<th>comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>This experiment</td>
<td>$37.7 \pm 1.2 \pm 2.0$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>MC(a) nominal</td>
<td>$34.9 \pm 2.4 \pm 2.1$</td>
<td>from Ref. [17]</td>
<td>$p$-wave dominant</td>
</tr>
<tr>
<td>MC(b) isotropic</td>
<td>$16.9 \pm 2.3 \pm 2.1$</td>
<td>Constant</td>
<td>$s$-wave assumed</td>
</tr>
</tbody>
</table>

Table 8.15: Summary of the attenuation analysis using the MOD yield data

be made from a direct comparison of the Monte Carlo and the data without the use of the intermediate approximation of the attenuation model.

On the other hand, given in the dot-dashed line is a fit to the MC assuming an isotropic angular distribution in $\mu t + d$ elastic scattering with the total cross section kept the same (MC(b)), where we obtained $L_{\text{MC(b)}}^{\text{eff}} = 16.9 \pm (2.3)_{\text{fit}} \pm (2.1)_{\text{thick}} \mu g\cdot cm^{-2}$. The Monte Carlo with an isotropic angular distribution is in disagreement with our data. In Table 8.15 we present the summary of our attenuation analysis. We shall defer the discussion of the implications of these measurements to Chapter 9, but for now it suffices to state that a reasonable agreement of our $L_{\text{int}}^{\text{eff}}$ with the MC gives us some confidence in our model of $\mu t$ moderation in a deuterium layer.

8.3.2 Time-of-flight analysis

In this section, we shall extract two important physics parameters, the rate for the resonant molecular formation and its resonant energies. We show in Fig. 8.19 the results of our Monte Carlo calculations for a 3 T-l DS layer using the nominal input to illustrate the features of our TOF measurements. Plotted are simulated two dimensional scatter-plots of the fusion time and the energy at which $d\mu t$ formation takes place. The direct events (top) refer to a process in which $\mu t$ forms $d\mu t$ directly, before being scattered by $D_2$ in the DS layer, while the total events (bottom) include also indirect processes where the $\mu t$ first loses energy in collision with another $D_2$ molecule in the DS layer. The
Figure 8.19: The simulated correlations between the fusion time and the energy at which dpt molecular formation takes place for the TOF measurements using 3 T-l layer.
direct process shows a strong correlation between the fusion time and formation energy exhibiting individual resonance structure as shown on the projection on the time axis, whereas for the total events the correlation is obscured by the indirect processes. The role of the indirect processes is more prominent, hence the correlations further weakened, in thick DS layer measurements as shown Fig. 8.20, in which only the total events are plotted (the direct process is nearly independent of layer thickness in these examples). In addition, the thick layer measurements are sensitive to the low energy process as also seen in the US measurement, thus there are relatively large theoretical uncertainties due to solid state effects (notice that a considerable amount of fusion is taking place at very low energies in Fig. 8.20). This is why we focused our measurements and analysis on thin layers. It should be stressed, however, that despite the presence of the indirect process, a significant correlation is preserved between the fusion time and formation energy in the
thin layer measurements.

In extracting physical quantities, formation rate and resonance energy, our approach is to perform iterative fits to the data using Monte Carlo calculations with varied input parameters. We varied the formation rate $\lambda_{d\mu t}$ and the resonant energies $E(d\mu t)$ by scaling

$$\lambda_{d\mu t} \rightarrow S_{\lambda} \lambda_{d\mu t} \quad (8.8)$$

$$E(d\mu t) \rightarrow S_{E} E(d\mu t), \quad (8.9)$$

where $S_{\lambda}$ is taken as energy-independent, and $S_{E}$, rate-independent. The fusion probability $W$ was kept at Faifman's value during $\lambda_{d\mu t}$ scaling.

We made full use of our accurate absolute fusion yield determination. In each fit (i.e., fit to each Monte Carlo result for a particular physics scaling input), the $\chi^2$ was minimized by varying the overall normalization by the Monte Carlo spectrum by a factor $\alpha_f$. But since our MC yields are already normalized, taking into account factors such as the number of incident muons, the muon stopping fraction, and the Si solid angle, together with experimental corrections due to nitrogen contamination (if present), $d\mu d$ proton contribution, the energy cut efficiency, and systematic effect in the background subtraction, all of which have been discussed in great detail in this thesis, we expect $\alpha_f = 1$ in an ideal case. Thus constraining $\alpha_f$ to its uncertainty $\delta\alpha_f$ (which is the relative error in our overall normalization), we define our $\chi^2$ as:

$$\chi^2 = \sum_{i=1}^{N} \frac{\left( \frac{h_i^{exp} - \alpha_f h_i^{MC}}{(\delta h_i^{exp})^2 + (\alpha_f \delta h_i^{MC})^2} \right)^2 + \left( \frac{1 - \alpha_f}{\delta\alpha_f} \right)^2}{}, \quad (8.10)$$

where $h_i^{exp}$, $h_i^{MC}$ are experimental and simulated counts in bin $i$, with $\delta h_i^{exp}$, $\delta h_i^{MC}$ being the respective uncertainties. Thus the likelihood function is maximized, hence $\chi^2$ minimized, when the shape and the yield match between experiment and Monte Carlo.
Figure 8.21: Fit of the calculated MC spectrum (upper histogram) to the experimental data (error bars) for 3 T-l in Series A, giving $\chi^2/dof = 0.964$ (confidence level 54.6%). The contribution from the direct process is also plotted (lower histogram). Both the data and MC results are normalized to GMU.

**Fit results**

We show in Fig. 8.21 a fit to the 3 T-l data from Series A ($c_t = 0.1\%$) with the nominal physics input, where we obtained $\chi^2/dof = 0.964$ and confidence level 54.6%, with the normalization factor $\alpha_f = 1.001(31)$. The fit is very good, perhaps somewhat accidentally. But the situation is less perfect in the case for the 3 T-l in series B ($c_t = 0.2\%$) shown in Fig. 8.22, in which we obtained $\chi^2/dof = 1.47$ and CL 1.67% with $\alpha_f = 0.78(4)$.

The result of the formation rate scaling is presented in Fig. 8.23, where the total $\chi^2$...
Figure 8.22: Fit of the calculated MC spectrum to the experimental data for 3 T·l in Series B, giving $\chi^2/dof = 1.47$ (confidence level 1.67%).

is plotted against the log of the formation scaling parameter $S_\lambda$. The log scaling of the horizontal axis was chosen to give relatively symmetrical shape of the resulting curve, compared to linear scaling. More physically motivated scalings (such as $(1 - \exp(-S_\lambda))$) were tried, but none of them symmetrized the curves for both Series A and Series B simultaneously. The points near the minimum (indicated by filled squares) were fitted with a quadratic function to obtain the best fit value and its error was estimated from finding the $S_\lambda$ in which $\chi^2$ is increased by 1 from the minimum. The dotted line in the figure indicates reduced $\chi^2$ (i.e., $\chi^2/dof$) of one. The results for two separate measurements (Series A and B) are given in Table 8.16. They are in an apparent disagreement by two standard deviations, indicating the existence of an unanticipated systematic uncertainty,
which we shall discuss below.

The results for the resonant energy scaling are shown in Fig. 8.24. The horizontal axis is a linear scale here due to a reasonably symmetric distribution of the data points. A quadratic fit similar to above was performed to obtain the best fit, $S_E$, and its error. Table 8.17 summarizes the energy scaling measurement. The results for two series of runs are in agreement within one standard deviation for the resonant energy measurements. Note that Series A had a higher sensitivity for both the rate and the energy measurements due to better statistics.

We note that, for both formation rate scaling and resonance energy scaling measurements, there are some uncertainties in the determination of the best fit value and its error due to the non quadratic shapes of the $\chi^2$ curves. But these effects are relatively small compared to other uncertainties involved in the measurements.

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Formation rate scaling $S_\lambda$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Series A</td>
<td>0.88$^{+0.11}_{-0.10}$</td>
</tr>
<tr>
<td>Series B</td>
<td>0.55$^{+0.12}_{-0.10}$</td>
</tr>
</tbody>
</table>

Table 8.16: The results of $\chi^2$ fit for $d\mu t$ formation rate.

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Resonance energy scaling $S_E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Series A</td>
<td>0.928 $\pm$ 0.040</td>
</tr>
<tr>
<td>Series B</td>
<td>0.994 $\pm$ 0.087</td>
</tr>
</tbody>
</table>

Table 8.17: The results of $\chi^2$ fit for $d\mu t$ formation resonance energy.
Figure 8.23: Total $\chi^2$ versus $d\mu t$ formation rate scaling factor $S_\lambda$, for Series A and Series B.
Figure 8.24: Total $\chi^2$ versus the scaling factor of the resonant energy for $d\mu t$ formation factor $S_E$, for Series A and Series B.
8.3.3 Monte Carlo uncertainties

Many of the systematic effects in our Monte Carlo modelling were tested by running the MC calculations with the parameters in question varied in the input. In some cases we estimate the uncertainties without the Monte Carlo, relying on, for example, an analytical approximation. We treat these Monte Carlo errors separately from our measurement errors given in the previous section. In most of these tests, we ignore the difference in spectrum shape and consider only the changes in the fusion yield due to the input variations, which is usually a good approximation.

Table 8.18: Estimated effects of various systematic uncertainties on the TOF fusion yield. Values denoted * were either ignored or included in other errors.

<table>
<thead>
<tr>
<th>MC error source</th>
<th>Estimated $\Delta Y/Y$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Layer thickness</td>
<td>10.3</td>
</tr>
<tr>
<td>Muon beam size</td>
<td>2.2</td>
</tr>
<tr>
<td>Nonuniform $\mu$ stopping in layer</td>
<td>3.3</td>
</tr>
<tr>
<td>US–DS spacing</td>
<td>4.8</td>
</tr>
<tr>
<td>RT minimum energy</td>
<td>2.0</td>
</tr>
<tr>
<td>Low energy processes</td>
<td>9.0</td>
</tr>
<tr>
<td>(formation, thermalization, backdecay energy)</td>
<td></td>
</tr>
<tr>
<td>MC error total</td>
<td>15.2</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Exp fusion yield error source</th>
<th>Estimated $\Delta Y/Y$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Muons stopping fraction</td>
<td>5.0</td>
</tr>
<tr>
<td>Si solid angle</td>
<td>4.2</td>
</tr>
<tr>
<td>Time zero shift</td>
<td>$&lt; 0.5^*$</td>
</tr>
<tr>
<td>Si double peaking</td>
<td>$&lt; 1.0^*$</td>
</tr>
<tr>
<td>$N_2$ contamination</td>
<td>2.5 (0)</td>
</tr>
<tr>
<td>Proton from $d\mu d$</td>
<td>0.9*</td>
</tr>
<tr>
<td>Energy cuts</td>
<td>$&lt; 0.5^*$</td>
</tr>
<tr>
<td>Background subtraction</td>
<td>6.4 (4.0)</td>
</tr>
<tr>
<td>Si1 (Si2) Exp error total</td>
<td>9.5 (7.7)</td>
</tr>
<tr>
<td>Si1 (Si2) statistical error</td>
<td>3.4 (6.6)</td>
</tr>
</tbody>
</table>
Our estimate of the Monte Carlo uncertainties in the time-of-flight fusion yield are given in Table 8.18. By performing fits with intentionally varied Monte Carlo amplitudes, we found that 15.2% error in the fusion yield translates into a relative 8.6% uncertainty in the formation rate strength parameter $S_\lambda$. Also given in Table 8.18 are the statistical and the experimental uncertainties in the fusion yield which have been extensively discussed and already included in the fit.

Regarding the resonance energy measurements, our major uncertainty is expected to come from the US-DS target foil spacing, and (US) layer thickness, whose effects in energy we estimate to be about 6% each\(^\text{11}\), resulting in about an 8.5% error. Note that the energy measurement is not very sensitive to small variations in the fusion yield.

### 8.3.4 Formation rate and resonance energy results

We combine the values of scaling parameters and uncertainties to give the final results on the formation rate and the resonance energy for $d\mu t$ muonic molecules. For the formation rate measurements, the best $S_\lambda$ from Series A and B disagree by more than the error bars as given in Table 8.16.

This discrepancy is curious. The difference in the run conditions, between the two series, in addition to the tritium concentration difference (i.e., $c_t = 0.1\%$ vs. $0.2\%$), was the presence of a 500 T·l $H_2$ substrate under the thin $D_2$ layer for Series A. Comparisons of two runs for other observables such as the US fusion yields and the $pt$ transfer time suggest that there is no problem in the $\mu t$ production in the emission layer. There is also no evidence from our run record that there was a problem in target preparation for either run. One possible effect is that due to the presence of the thick $H_2$ substrate in Series

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\(^{11}\)The former was estimated by doing similar $\chi^2$ minimization with fits from varied MC geometry, but with much reduced number of fit points, while the latter was estimated by observing the shift in mean time-of-flight with one fixed $S_E$.\]
A, \( \mu t \) which passed through a thin \( \text{D}_2 \) layer may thermalize in \( \text{H}_2 \), and may in turn be re-emitted back into the \( \text{D}_2 \) layer to form \( d\mu t \) and fuse. In fact this effect was already taken into account in our analysis; using our Monte Carlo we estimated that the re-emission would increase the fusion yield by a factor of \( 1.05 \pm 0.05 \), where the uncertainty is estimated taking into account the lack of knowledge of formation rates as well as scattering cross section at very low energies. However, estimating these effects is difficult without proper solid state cross sections, and in light of our recent observation of \( \mu p \) emission from a pure \( \text{H}_2 \) layer [216], it may still be underestimated. Note, however, that due to a high \( p\mu t \) formation rate, low energy \( \mu t \) emission from \( \text{H}_2 \) should be somewhat suppressed compared to \( \mu p \).

Another possibility is the presence of some unaccounted errors in our values of \( \lambda_{pt} \) and \( \lambda_{ppp} \) (the \( pt \) transfer rate and \( ppp \) formation rate, respectively), which were measured with our solid targets [83]. There is no strong reason to doubt those values except perhaps that \( \mu p \) emission just mentioned was not considered at the time. Using the theoretical values from Refs. [17] and [51] would reduce the discrepancy significantly but not completely.

Accepting the discrepancy as a measure of an unaccounted systematic uncertainty in our measurement, we shall increase our errors in accordance with Particle Data Group's procedure [229, 230]. Thus our final result for the resonant molecular formation rate is:

\[
S_\lambda = 0.747 \pm (0.161)_{\text{meas}} \pm (0.086)_{\text{model}},
\]

where the first error is the combined average error increased by a factor of 2.1, and the second error is due to the MC model uncertainty discussed in the previous section.

As for the resonance energy measurement, since the values from Series A and B agree within the error, we take the standard weighted average to obtain:

\[
S_E = 0.940 \pm (0.036)_{\text{meas}} \pm (0.085)_{\text{model}},
\]

(8.12)
where similarly the first error is a combined measurement error and the second error is due to uncertainties in the MC modelling.
9.1 Muonic tritium scattering

9.1.1 The Ramsauer-Townsend effect

We have observed emission of $\mu t$ from a hydrogen layer into vacuum via imaging of muon decay electrons. From the position and time of the muon decay, the information on the energy distribution of the emitted $\mu t$ is obtained. Comparing the electron time spectrum in the vacuum region with detailed Monte Carlo calculations, we determined the Ramsauer-Townsend energy minimum to be $E_R = 13.6 \pm 1.0$ eV, in agreement with the theoretical prediction of 12.4 eV by Chiccoli et al. using the multi-level calculations in the Adiabatic Representation of the three body Coulomb problem (the Nuclear Atlas [17]). Molecular and condensed matter effects are not included in our analysis, but their influence on $\mu t$ transport properties is expected to be negligible at these energies. Figure 9.1 illustrates the preferred variations (shaded band) of cross section from this measurement together with theoretical values (dashed line) [17]. Our results are consistent with the emission probability of about 15% per muon stopped in the production layer.

In addition to the above MWPC measurements from Run Series I, the $d\mu t$ fusion measurements from Run Series II which were performed in a quite different setup (see Table 4.1) can add some information. A preliminary result of the time spectrum of fusion at the DS thick layer with no US moderation layer is consistent with the RT scaling of
Chapter 9. Discussion and Conclusion

Figure 9.1: The preferred variations in $\mu t + p$ elastic scattering cross sections from this measurement (shown in a shaded band), together with the original theoretical cross sections from Ref. [17] (dashed line). The dot-dashed line is the constant cross sections used in the comparison given in Fig. 9.2. The box is an expansion near the Ramsauer-Townsend minimum plotted on a linear scale.

1.05 ± 0.05 when all other nominal physics input, and the nominal US–DS target spacing, are assumed. Given the considerable difference in the setup and detection method, this gives us further confidence in our measurement of the RT minimum reported in this thesis.

We note that preliminary results of recent measurements [234] performed by our collaboration, but using a different X-ray technique and independent analysis with a
Figure 9.2: The time spectrum of $\mu t$ decay in vacuum region (error bars) compared with a Monte Carlo calculation assuming no RT minimum in the $\mu t + p$ cross section (histogram). An energy independent cross section of $5 \times 10^{-21}\text{cm}^2$ was assumed in the simulation, which gives a similar $\mu t$ yield in the vacuum region. The comparison clearly rules out the possibility of a constant cross section, establishing the existence of a minimum in $\mu t + p$ cross section.

separate MC, indicate a shift of the RT minimum ($0.4 \pm 0.05$ eV, or relative 3% shift) to lower energy, opposite to the indication given here. It should be noted that not all uncertainties were included in the quoted value; for example, the error in the target spacing has been neglected so far. The difference between the X ray measurement and the MWPC measurement reported here probably gives a measure of unexpected systematic uncertainties. We stress, however, that for our goal of molecular formation rate measurements, the confirmation of the theoretical RT minimum energy at the 10% level is sufficient, in comparison with uncertainties in other processes$^1$.

Finally, we present in Fig. 9.2 a comparison of our MWPC emission data with a Monte

$^1$The energy scaling method in Ref. [234] is slightly different from ours described here, but this difference alone would not resolve the apparent discrepancy between the two results.
Carlo calculation assuming no RT minimum. The simulation using an energy independent cross section of $5 \times 10^{-21}$ cm$^2$ gives a similar yield of $\mu t$ emitted in vacuum, but its time-of-flight distribution is very different. Thus our measurements provide direct evidence for the existence of a deep minimum in the $\mu t + p$ cross section. Note that in diffusion type measurements, which are mainly sensitive to the integrated diffusion length, it would be more difficult to rule out the possibility of an energy-independent cross section.

9.1.2 $\mu t + d$ scattering

From the measurements of the DS fusion yield with varying US overlayer thickness (MOD measurements), we have extracted the effective attenuation interaction length $L_{\text{eff att}}$, which agrees at the 10% level with the value given by the Monte Carlo using the $\mu t + d$ elastic cross section from Ref. [17]:

$$L_{\text{eff att}}(\text{exp}) = 37.7 \pm (1.2)_{\text{fit}} \pm (2.0)_{\text{thick}} \mu g \cdot cm^{-2} \quad (9.1)$$

$$L_{\text{eff att}}(\text{mc}) = 34.9 \pm (2.4)_{\text{fit}} \pm (2.1)_{\text{thick}} \mu g \cdot cm^{-2} \quad (9.2)$$

where the first error is due to the fit and the second due to the uncertainty in the US layer thickness. The attenuation length is determined mainly by the $\mu t + d$ elastic scattering process, thus the agreement between Eqs. 9.1 and 9.2 suggest confirmation of the $\mu t + d$ scattering cross section of Chiccoli [17] at the 10% level. This is the first quantitative measurement of the $\mu t + d$ cross section to our knowledge, as conventional cycling measurements in D/T mixtures are not directly sensitive to this process. We note that dependence of our results on the value of $d\mu t$ formation rate is rather weak; a change of $\lambda_{d\mu t}$ by a factor of 2 produces variations in $L_{\text{eff att}}$ similar to or smaller than those due to the thickness uncertainty.

Our data could alternatively be used to extract information on the scattering angular distribution. A MC calculation assuming isotropic scattering with the same total cross
section as Ref. [17] resulted in the effective attenuation length:

\[
L_{\text{att}}^{\text{eff}}(mc, \text{iso}) = 16.9 \pm (2.3)_{\text{fit}} \pm (2.1)_{\text{thick}} \mu g \cdot cm^{-2},
\]  

(9.3)

which is in clear disagreement with our experiment. The \( \mu t + d \) elastic cross sections are in fact predicted to have large \( p \)-wave contributions, whose angular distribution is strongly peaked forward [17]. This is due to the existence of the loosely bound \( d\mu t \) molecular state with an angular momentum of \( J = 1 \), the very state responsible for the resonant molecular formation. The energy loss of a particle in the lab frame in an elastic collision is uniquely determined by the scattering angle \( \theta \). A scattering to large angles will reduce the energy substantially (depending on the masses involved), while the particles do not suffer much energy loss for forward angle scattering. Hence, the forward peaked angular distribution results in a smaller attenuation, compared to the isotropic one for the same total cross section.

In order to reproduce our experimental attenuation length \( L_{\text{att}}^{\text{eff}} \) using the isotropic angular distribution, the total cross section needs to be half of the predicted value, which seems rather unlikely (the experimental absolute fusion yield at US thickness zero is reasonably well reproduced by either model). Thus, our results given in Table 8.15 confirm the importance of the \( p \)-wave contribution in \( \mu t + d \) scattering.

Furthermore, the suggestion of the large \( p \)-wave contribution in turn provides indirect yet intriguing experimental evidence for the existence of the \( J = 1 \) state near the \( \mu t + d \) threshold. Note that while for the \( d\mu d \) molecule there is some evidence for its \( J \) state from the fusion branching ratio of the two channels (it is predicted to be sensitive to \( J \) according to R-matrix calculations [235]), there has been little direct experimental information of the \( J \) state of the loosely bound \( d\mu t \).

Finally, our analysis presented here suggests that the measurement by Strasser et al. [195], who assumed an isotropic angular distribution in the simulation of the \( \mu d \)
deceleration in deuterium, may require re-interpretation. Since there is a significant $p$-wave contribution also in the $\mu d + d$ interaction, neglecting its angular distribution is not justified, as demonstrated in our analysis. Therefore, their conclusion, which stressed the importance of molecular effects, should be taken with caution.

9.2 Condensed matter and subthreshold effects

Despite the considerable success of our analysis in epithermal energy scattering above a few eV, low energy processes are complicated by the solid state effects on both $\mu t$ slowing and $d\mu t$ formation.

Our fusion yield dependence on layer thickness is inconsistent with that predicted by the standard Faifman model, or even with our nominal model where the low energy formation rate for $\mu t(F = 0) + D^2\text{tho}$ is set to 130 $\mu$s$^{-1}$. An improved yet still not perfect agreement has been achieved only after inclusion of a small constant term $\lambda_{d\mu t}^s \sim 5$ $\mu$s$^{-1}$ for $d\mu t$ formation. The value of $\lambda_{d\mu t}^s$ is sensitive to the detail of the $\mu t$ deceleration process, hence it should be taken as a model-dependent phenomenological parameter at this stage.

Regardless of its rate, however, there is an indication that a nonzero value for $F = 1$ formation rates plays an important role in our measurements. The nonresonant $d\mu t$ formation rate is predicted to be very small at low temperature ($\lesssim 0.5$ $\mu$s$^{-1}$), and does not appear to explain our observation regardless of the thermalization model.

Recalling that there is some evidence for subthreshold resonances for $F = 0$, the same might be possible for $F = 1$, although for the latter the resonance energies $E_r$ are expected to be more negative ($E_r \sim -50$ meV). However, if one assumed the Breit-Wigner resonance profile as a zeroth approximation (despite the fact that this form is criticized for high densities [160]), the subthreshold formation rate $\lambda_{\text{sub}}$ falls off as
\[ |E_r|^{-5/2} \] [141], and if one takes into account the experimental evidence for \( F = 0 \) that \( \lambda_{\text{sub}} \sim 300 \, \mu s^{-1} \) with \( E_r \sim -10 \, \text{meV} \), it is not completely implausible to have a few \( \mu s^{-1} \) for \( F = 1 \) at low temperature.

We note that the possibility of a nonzero \( F = 1 \) rate has not previously been ruled out experimentally either, since in the previous D/T mixture experiments at low temperatures, \( F = 1 \) rates were usually assumed to be zero in the fit (e.g. Ref. [36]). At any rate, small rates for \( F = 1 \) would be difficult to measure in the cycling experiments due to fast \( \mu t \) spin flip. Thus, our measurements in multilayers may offer a unique sensitivity to the resonance profile at large detuning energy, if theoretical uncertainties due to solid effects can be removed. To our knowledge, there are no realistic calculations of \( \lambda_{\text{sub}} \) for \( F = 1 \), and we urge theorists to extend their calculations to \( F = 1 \).

Calculations for \( \mu t \) scattering processes and molecular formation in solid hydrogen are in progress [170]. We eagerly await these new results. Fortunately, the solid state effects did not overwhelm our measurements using very thin layers.

### 9.3 Resonant molecular formation

Using the emitted beam of \( \mu t \) from a hydrogen layer, we have measured the formation rates and resonance energy of \( d\mu t \) muonic molecules. The combined results of the rate and energy scaling parameters \( S_\lambda, S_E \) from two separate sets of runs are:

\[
S_\lambda = 0.747 \pm (0.161)_{\text{meas}} \pm (0.086)_{\text{model}} \quad (9.4)
\]

\[
S_E = 0.940 \pm (0.036)_{\text{meas}} \pm (0.085)_{\text{model}}, \quad (9.5)
\]

where the first errors are experimental uncertainties and the second ones are MC modeling uncertainties (including target geometry). When the errors are added in quadrature, we have achieved accuracies of about 25% and 10% respectively for the formation rates and
energy. Our measurement of resonant molecular formation corresponds to a peak rate\(^2\) of \((8.7 \pm 2.1) \times 10^9\) s\(^{-1}\) for the reaction

\[
\mu t(F = 1) + D_2^{ortho} \rightarrow (d\mu t)dee,
\]

quantitatively confirming for the first time the existence of the strong epithermal resonance.

Our measurement of the resonance energy scaling corresponds to the position for the strongest the resonance peak in the reaction (9.6) of 423 ± 41 meV in the \(\mu t\) lab frame. If we assume the molecular spectrum of the complex \([(d\mu t)dee]\) is predicted reliably, our results can be considered a first direct measurement of the loosely bound state energy level. Our accuracy of \(\sim 25\) meV in the center of mass frame, compared to the muonic atomic energy scale [2] of \(E_\mu = (m_\mu/m_\mu)2R_y = \sim 5630\) eV, is better than 10 ppm. Indeed it is comparable to the vacuum polarization correction in the loosely bound state energy level.

Until a few years ago, the problem of muonic molecule binding energies appeared to have been completely settled at least for the \(dpd\) case. Extraction of the binding energy from the temperature dependence of \(dpd\) formation rates, as analyzed by Scrinzi \textit{et al.} [12], showed remarkable agreement with the theoretical prediction. However, new studies, both theoretical and experimental, seem to indicate the real situation is not as clear. For example, recent calculations by Harston \textit{et al.} on the \(d\mu d\) finite size effects indicates that previous values used in Ref. [12] were significantly underestimated. Similar effects are suggested for the \(d\mu t\) case by Bakalov \textit{et al.} [137]. Experimentally, new and precise measurements of \(d\mu d\) fusion at PSI [236] are not in complete agreement with those used in Ref. [12]. In addition, the assumption of complete \(\mu d\) thermalization may not be valid at low temperatures. It should also be stressed that in the \(d\mu t\) case, because

\(^2\)Assuming the Doppler width given by Faifman.
of the lack of experimental observation of the temperature dependence predicted by the standard Vesman theory, there was virtually no experimental information on the $d\mu t$ energy level. Thus our measurements of the resonance energy may provide an interesting new opportunity to test the calculations of $d\mu t$ binding energies.

Let us make a few remarks on the theoretical assumptions in our analysis.

- Inclusion of explicit back decay in the simulation is essential. The use of the renormalized effective rates without an explicit resonant scattering channel would lead to a significant overestimate in the calculated fusion yield. See appendix B for a detailed discussion.

- In our Monte Carlo simulations, the dependence of the formation rates $\lambda_{d\mu t}$ and fusion probability $W$ on the quantum states $S, K_f$ are separately averaged, i.e., the averages for $\lambda_{d\mu t}$ and $W$ over the quantum states are factorized. This is not rigorously accurate if their dependence on $S$ or $K_f$ is large. Although this is probably not a large effect, the precise estimate of the degree of accuracy requires comparisons with more complete calculations.

- Since our $D_2$ target is not in thermal equilibrium rotationally (i.e., $\omega_B(K_f) \neq \omega_K(K_f)$) the use of Eq. 2.40–2.42 assuming the Boltzmann distribution for $K_f$ may introduce some error, though judging from the difference in the results between using Eq. 2.40–2.42 and Eq. 2.37–2.39 at 300 K, shown in Figs. 10 and 11 of Ref. [133], the effects appear rather small. The $K_f$ distribution could also be affected if the rotational relaxation is substantially smaller than predicted by Ostrovskii and Ustimov [153], as suggested in Ref. [154].

\footnote{See discussion of the factorization approximation in the appendix B.}
The use of a (state independent) effective fusion rate $\tilde{\lambda}_f$ appears valid at least for its Auger decay contribution, since it is presumably independent of $\nu_f, K_f$, or $S$. However the value used, $\tilde{\lambda}_f = 1.27 \times 10^{11} \text{ s}^{-1}$, could be overestimated by $\sim 40\%$, if the lowest value amongst Armour's several predictions [152] turns out to be the correct one. This would not affect low energy formation where the fusion probability is very high, but could have a significant effect in epithermal formation, where fusion and back decay branches are comparable. On the other hand, possible collisional vibrational quenching would increase $\tilde{\lambda}_f$.

Our method of scaling the formation rate, given in Eq. 8.8, while keeping the fusion probability $W$ fixed, is phenomenological in nature. More physically motivated scaling would be to scale the matrix elements, which change both $\lambda_{d\mu t}$ and $W$. By the same token, scaling of the $d\mu t$ binding energy as input into the formation rate calculations, and comparisons with the resulting resonance structures (assuming scaled binding energies) may be more justified than our simple scaling in Eq. 8.9 for the resonance energy measurements, although these comparisons would require considerable input from theorists. Thus, our scaling results should be taken as a first step toward a more complete analysis.

In our analysis, the resonance width is given by Faifman et al., which is determined by the Doppler broadening due to the D$_2$ motion assuming a 3 K gas. In a solid, however, the Doppler broadening is expected to be larger due in part to the larger zero-point motion of a D$_2$ bound in the lattice, hence the resonance profile may be broader. The precise evaluation of this effect would require more theoretical work which is in progress [170].

Finally, let us present in Fig. 9.3 a comparison of our data with the MC calculations assuming a constant rate for $d\mu t$ formation, a value of which was chosen to reproduce
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Figure 9.3: Comparison of the experimental data (error bars) from Series A with the MC (histogram) assuming a constant formation rate. The comparison rules out the possibility of an energy independent formation rate, establishing the existence of a resonant structure in epithermal \( d\mu t \) formation.

the fusion yield in the US moderator layer. The indication is quite convincing; we have confirmed the existence of epithermal resonant \( d\mu t \) formation in \( \mu t \) collisions with \( D_2 \).

9.4 Concluding remarks

9.4.1 Improvements and Future directions

One of the difficulties of our time-of-flight measurements was its low event rate. Although our accuracy for the molecular formation rate is not dominated by the statistical uncertainty, the small size of the fusion data sample made it difficult for us to investigate
thoroughly the systematic effects such as background subtraction. More data would help us to understand these effects better, and reduce the uncertainties.

At the present moment, theoretical uncertainties due to solid state effects give a large contribution to our total uncertainty. It is hoped that the situation will be improved in the near future, as theoretical efforts are underway. As discussed in Appendix B, the evaluation of the back-decayed $\mu t$ energy would be necessary to significantly improve the accuracy of our measurements.

For better determination of the RT energy and $d\mu t$ resonance energy, more precise knowledge of the target spacing distance would be required. Measurement *in-situ* of the distance is not trivial in our setup, and appropriate methods should be investigated. Some minor modification of our gold target support (e.g., using a flat plate rather than a very thin foil for the DS) could help to better define this distance.

As a future project for our collaboration, I have spent a considerable amount of effort in the past few years, together with Peter Kammel and Glen Marshall, in developing a new method for a direct measurement of the sticking probability using the multilayer solid targets [171,179,237]. Sticking, as discussed in Chapter 1, places the most stringent limit on the fusion yield per muon, hence, has attracted much attention in this field, but the discrepancy between experiment and theory persists to date. Most previous experiments are sensitive to the final sticking, which is a combination of initial sticking and stripping, hence cannot be readily compared with theory. Taking advantage of our multilayer target, we propose to (a) experimentally separate initial sticking and stripping, and (b) unambiguously determine sticking at high density where the $\mu$CF efficiency is highest, but the discrepancy is largest. The experiment is already approved by the TRIUMF Experimental Evaluation Committee, but our situation is a little unclear due to funding difficulties. The further discussion of this experiment is beyond scope of this thesis and interested readers are referred to Refs. [171,179,237].
9.4.2 Summary

In this thesis, we have reported a new approach in \( \mu \)CF studies, \textit{viz} the time-of-flight method using an atomic beam of muonic tritium. With this new technique we have made measurements on \( \mu t \) scattering as well as epithermal \( d\mu t \) resonant formation, which have been quantitatively studied for the first time.

Various experimental challenges have been overcome in order to complete the experiments. Technical contributions of this thesis to the field of \( \mu \)CF studies include:

1. Characterization of target layer thickness and uniformity to an accuracy of up to a few tens of nanometers, and the evaluation of effective average thickness using the muon beam profile obtained from the MWPC imaging.

2. New methods for determining the stopping fraction, such as the absolute amplitude method via delayed electron coincidence, and the relative amplitude with electron energy cuts.

3. Considerations of resonant scattering in the \( \mu \)CF processes with detailed expressions for scattered \( \mu t \) energy.

The physics results of this thesis can be summarized as follows.

1. We have observed an emission of muonic tritium in vacuum via imaging of muon decay electrons. From the position and the time of muon decay, information of the \( \mu t \) energy was obtained, enabling us to spectroscopically establish the existence of the Ramsauer-Townsend effect in \( \mu t + p \) interactions. The energy of the RT minimum was measured to be \( 13.6 \pm 1.0 \) eV, in fair agreement with quantum three body calculations by Chiccoli \textit{et al.} [17].

2. Using the \( \mu t \) beam, we have confirmed theoretical \( \mu t + d \) scattering cross sections [17] to the 10\% level by measuring the attenuation of \( \mu t \) through deuterium.
Comparisons with Monte Carlo simulations, assuming different scattering angular distributions, also confirmed the importance of $p$-wave scattering in the $\mu t + d$ interaction, giving angular momentum information on the loosely bound state of the $d\mu t$ molecule.

3. The existence of the predicted large resonance in $\mu t + D_2$ collisions was directly confirmed for the first time. Our results of the resonance strength correspond to a peak rate of $8.7 \pm 2.1 \times 10^9$ s$^{-1}$ when the resonance width given by Faifman is assumed. This is more than an order of magnitude larger than room temperature rates. Our measurement of the resonance position indicates a resonance energy of $0.42 \pm 0.04$ eV for the $F = 1$ peak in ortho deuterium.

4. Assuming the theoretical $[(d\mu t)\text{dee}]$ energy spectrum, our results for the resonant energy imply sensitivity to the binding energy of the loosely bound $J = 1, v = 1$ state of the $d\mu t$ molecule, with an accuracy approaching the magnitude of the relativistic and QED corrections, providing potential future opportunities to directly test quantum few body calculations.

5. Indications of solid state effects have been observed in the layer thickness dependence of the fusion yield, but more theoretical input is needed for better understanding. Efforts have begun by theorists to calculate $\mu t$ interactions in solid hydrogen. The data obtained here will confront any future calculations.
Appendix A

Abbreviations and notation

A.1 Abbreviations

List of important abbreviations and relevant pages

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<td>ADC</td>
<td>Analog-to-digital converter</td>
<td>91</td>
</tr>
<tr>
<td>AR</td>
<td>Adiabatic Representation in three-body Coulomb problems</td>
<td>29</td>
</tr>
<tr>
<td>cl</td>
<td>Confidence level in χ² fit</td>
<td>various</td>
</tr>
<tr>
<td>Del</td>
<td>Delayed electron coincidence</td>
<td>102</td>
</tr>
<tr>
<td>Del-e</td>
<td>Delayed electron coincidence with electron scintillator</td>
<td>102, 151</td>
</tr>
<tr>
<td>Del-t</td>
<td>Delayed electron coincidence with electron telescope</td>
<td>102, 151</td>
</tr>
<tr>
<td>dof</td>
<td>Degrees of freedom in χ² fit</td>
<td>various</td>
</tr>
<tr>
<td>DS</td>
<td>Downstream target</td>
<td>60</td>
</tr>
<tr>
<td>DSY</td>
<td>(Measurement of) fusion yield in the downstream target</td>
<td>190</td>
</tr>
<tr>
<td>EVS</td>
<td>Experimental vacuum space</td>
<td>60</td>
</tr>
<tr>
<td>FWHMₜ</td>
<td>Full width at a half maximum of the Gaussian part of flat-top Gaussian distribution</td>
<td>120</td>
</tr>
<tr>
<td>Ge</td>
<td>Germanium X-ray detector</td>
<td>90</td>
</tr>
<tr>
<td>GMU</td>
<td>Scaler for the number of incident muons identified as &quot;good&quot; muons taking into account computer dead time and pile up</td>
<td>94</td>
</tr>
<tr>
<td>IA</td>
<td>Improved Adiabatic approach</td>
<td>32</td>
</tr>
<tr>
<td>LHD</td>
<td>Liquid hydrogen density</td>
<td>20</td>
</tr>
<tr>
<td>MC</td>
<td>Monte Carlo simulation or code</td>
<td>various</td>
</tr>
<tr>
<td>MMC</td>
<td>Muonic Molecular Complex [(dμt)xee] where x = p, d, t</td>
<td>260</td>
</tr>
<tr>
<td>MOD</td>
<td>(Measurement of) the downstream fusion with variable upstream moderation layer thickness</td>
<td>190</td>
</tr>
<tr>
<td>MODY</td>
<td>Yield for MOD</td>
<td>190</td>
</tr>
<tr>
<td>MWPC</td>
<td>Multi-wire proportional chamber</td>
<td>89</td>
</tr>
<tr>
<td>N1</td>
<td>NE213 neutron detector</td>
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## Appendix A. Abbreviations and notation

List of important abbreviations and relevant pages

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<td>NE213 neutron detector</td>
<td>2, 90</td>
</tr>
<tr>
<td>NE213</td>
<td>Liquid scintillating material</td>
<td>99</td>
</tr>
<tr>
<td>Q\textsubscript{Au}</td>
<td>Huff factor for muon capture on gold</td>
<td>131</td>
</tr>
<tr>
<td>RT</td>
<td>Ramsauer-Townsend (effect)</td>
<td>7</td>
</tr>
<tr>
<td>R\textsubscript{flat}</td>
<td>Radius of the flat part of flat-top Gaussian distribution</td>
<td>120</td>
</tr>
<tr>
<td>S\textsubscript{H}</td>
<td>Fraction of the muon stopping in the upstream hydrogen layer</td>
<td>130</td>
</tr>
<tr>
<td>S\textsuperscript{ABS}_H</td>
<td>Muon stopping fraction obtained from absolute amplitude method</td>
<td>150</td>
</tr>
<tr>
<td>S\textsuperscript{AR}_H</td>
<td>Muon stopping fraction obtained from absolute ratio method</td>
<td>131</td>
</tr>
<tr>
<td>S\textsuperscript{AR}_H</td>
<td>Reduced stopping fraction in the amplitude ratio method</td>
<td>132</td>
</tr>
<tr>
<td>S_E</td>
<td>Scaling parameter for the $d\mu t$ formation resonance energy</td>
<td>232</td>
</tr>
<tr>
<td>S_X</td>
<td>Scaling parameter for the $d\mu t$ formation rate</td>
<td>232</td>
</tr>
<tr>
<td>Series A</td>
<td>TOF run series with a tritium concentration $c_t = 0.1%$</td>
<td>221</td>
</tr>
<tr>
<td>Series B</td>
<td>TOF run series with a tritium concentration $c_t = 0.2%$</td>
<td>221</td>
</tr>
<tr>
<td>Si1</td>
<td>Silicon charged particle detector located on the beam-left (looking along the beam direction)</td>
<td>90</td>
</tr>
<tr>
<td>Si2</td>
<td>Silicon charged particle detector located on the beam right</td>
<td>90</td>
</tr>
<tr>
<td>SET</td>
<td>Standard emission target</td>
<td>105</td>
</tr>
<tr>
<td>$t_0$</td>
<td>Time of the muon entrance defined by $T1$</td>
<td>94</td>
</tr>
<tr>
<td>T1</td>
<td>Beam defining plastic scintillator muon counter</td>
<td>88</td>
</tr>
<tr>
<td>TBS</td>
<td>Tritium barrier space</td>
<td>65</td>
</tr>
<tr>
<td>TDC</td>
<td>Time-to-digital converter</td>
<td>91</td>
</tr>
<tr>
<td>Tel1</td>
<td>Electron telescope 1</td>
<td>102, 151</td>
</tr>
<tr>
<td>Tel2</td>
<td>Electron telescope 2</td>
<td>102, 151</td>
</tr>
<tr>
<td>T·l</td>
<td>Target thickness measured in Torr x litre</td>
<td>67</td>
</tr>
<tr>
<td>TOF</td>
<td>Time of flight</td>
<td>various</td>
</tr>
<tr>
<td>TOFY</td>
<td>(Measurement of) yield in the time-of-flight measurement</td>
<td>190</td>
</tr>
<tr>
<td>TOFS</td>
<td>(Measurement of) the time-of-flight spectrum</td>
<td>190</td>
</tr>
<tr>
<td>W\textsuperscript{F}</td>
<td>Effective fusion probability</td>
<td>47</td>
</tr>
<tr>
<td>US</td>
<td>Upstream target. The upstream target almost always had 1000 T· layer with $c_t = 0$, 0.1, or 0.2%, so sometimes US is just referred to the overlayer</td>
<td>60</td>
</tr>
<tr>
<td>USY</td>
<td>(Measurement of) yield in the upstream overlayer target</td>
<td>190</td>
</tr>
<tr>
<td>$\lambda\textsuperscript{F}_{d\mu t}$</td>
<td>$d\mu t$ molecular formation rate for $\mu t$ hyperfine states $F$</td>
<td>47</td>
</tr>
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List of important abbreviations and relevant pages

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<tbody>
<tr>
<td>$\lambda_{d\mu\tau}^F$</td>
<td>Effective $d\mu\tau$ formation rate, renormalized for the fusion probability</td>
<td>47</td>
</tr>
<tr>
<td>$\lambda_{d\mu\tau}^c$</td>
<td>Phenomenological parameter for an energy and $F$ independent $d\mu\tau$ formation rate</td>
<td>247</td>
</tr>
<tr>
<td>$\Omega_{Si}$</td>
<td>Si detector acceptance</td>
<td>127</td>
</tr>
<tr>
<td>$\phi$</td>
<td>Density in units of liquid hydrogen atomic number density $N_0 = 4.25 \times 10^{22}$ cm$^{-3}$</td>
<td>17</td>
</tr>
<tr>
<td>$\sigma_x^{WC}$</td>
<td>MWPC resolution function parameter in the $x$ axis</td>
<td>170</td>
</tr>
<tr>
<td>$\sigma_y^{WC}$</td>
<td>MWPC resolution function parameter in the $y$ axis</td>
<td>170</td>
</tr>
<tr>
<td>$\oplus$</td>
<td>Represents deposition of an additional layer (overlayer) on top of a emission layer</td>
<td>105</td>
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</table>
A.2 Error notation

Throughout this thesis, unless otherwise noted, the uncertainty for the given quantity is presented in parentheses immediately following the mean value of the quantity, with the order of the last digit of the uncertainty equal to the order of the last digit in the mean. That is, regardless of the position of the decimal point,

\[ mmmm(ee) \equiv mmmm \pm ee, \]

where \( mmmm \) is the mean value and \( ee \) is the uncertainty for that quantity. We show a few numerical examples below:

\[
\begin{align*}
23.4(7) &= 23.4 \pm 0.7 \\
23.5(12) &= 23.5 \pm 1.2 \\
0.510(8) &= 0.510 \pm 0.008.
\end{align*}
\]
Appendix B

Resonant scattering of \( \mu t \)

In this appendix, we discuss the influence of resonant scattering in the modelling of the muon catalyzed fusion processes. As we have seen elsewhere in this thesis, resonant formation of \( d\mu t \) molecular ions in a loosely bound state of \( J = v = 1 \) occurs via formation of a metastable muonic molecular complex \([((d\mu t)_{11} xee]\) (denoted MMC hereafter) in a collision of \( \mu t \) on a molecule \( DX \),

\[
\mu t + DX \rightarrow [((d\mu t)_{11} xee]^*, \tag{B.1}
\]

where \( X = H, D, T \) and \( x = p, d, t \). Here we shall denote the total rate for the formation of the complex as \( \lambda^{tot} \). After the formation of the complex, two competing processes take place: stabilization of the complex leading to fusion (mainly via Auger de-excitation of the \( d\mu t \) molecule) with the effective fusion rate \( \lambda_f \), and back decay [151] to \( \mu t + DX \) with the rate \( \Gamma_{back} \). The above rates generally depend on the quantum numbers, such as ro-vibrational, spin, and hyperfine states, of initial and final states, but in the first part of this appendix we consider the averaged rates for simplicity and drop indices for quantum numbers. This simplification does not affect our conclusion.

In conventional analyses of \( \mu CF \), the effective formation rate \( \lambda^{eff} \), a renormalized rate taking into account the back decay probability, has been widely used, and is defined (when ignoring the indices for the quantum numbers) as:

\[
\lambda^{eff} = \lambda^{tot} \cdot W, \tag{B.2}
\]
where $W$ is the branching ratio for the fusion channel

$$ W = \frac{\tilde{\lambda}_f}{\tilde{\lambda}_f + \Gamma_{\text{back}}}. $$

With the recent recognition of the importance of transport properties of muonic atoms, theoretical cross sections for muonic atom scattering have been calculated to high accuracy with sophisticated methods. Despite the vast theoretical efforts in pushing the accuracy of various scattering cross sections, including electronic screening, atomic and molecular structure, and most recently solid state effects, little detailed attention has been paid to the back decay process as a scattering mechanism of muonic atoms, except in the context of spin flip in the $d\mu d$ system \cite{34}.

In early work, resonant scattering processes were neglected or treated incorrectly. For example, in the pioneering studies of Markushin \cite{9,27,75,82}, who first performed the full three-dimensional Monte Carlo calculations of muonic processes, renormalized effective formation rates were used to account for the back decay processes. On the other hand, in the theoretical analysis of the $\mu$CF kinetics in D/T mixture \cite{238}, Somov claimed that $\mu t$ emitted after back decay of the muonic molecular complex has a thermal distribution with a temperature of the surrounding medium. Jeitler et al. followed this in their Monte Carlo analysis of D/T and H/D/T mixtures \cite{22,74}. They claim that this is justified when one assumes complete thermalization (of the translational motion) and the rotational relaxation of the MMC \cite{74}.

The purpose of this appendix is to point out the importance of the resonant scattering process resulting via backdecay of the molecular complex, particularly in the atomic beam type of experiments, as performed in this thesis. We will show first that the use of renormalized effective rates as was done in Refs. \cite{9,27,75,82} significantly overestimates the calculated fusion yield, and second that thermalized $\mu t$ emission after back decay is not justified even if one assumes the complete MMC thermalization.
Appendix B. Resonant scattering of $\mu t$

B.1 Effective formation model

B.1.1 Validity

Obviously, in transport calculations, neglecting one of the scattering channels leads to inaccurate results, if the contribution of that channel is significant. Therefore the back decay process has to be looked at carefully. On the other hand, in the description of fusion yield, the effective formation approximation has been used in many analyses (see for reviews Refs. [1–4]). Let us investigate the validity of this approximation in the fusion yield description.

We point out that in order for the effective formation approximation to be justified in terms of describing the fusion yield, at least one of the following criteria must be met: (a) trivial conditions that the back decay probability $(1 - W) \sim 0$, (b) a negligible change in $\mu t$ energy before and after back decay in the laboratory frame\(^1\), (c) fast (compared to the formation rate) "re-thermalization" of back-decayed $\mu t$ ($\mu d$) in a thermal equilibrium condition.

For example, at low temperatures the condition (a) is satisfied for $d\mu t$ formation, while the condition (c) applies for $d\mu d$ formation at least at high densities (one may need to be careful about this at very low density). In case of $d\mu t$ formation at epithermal energies, however, none of these conditions apply, hence the back decay process cannot be neglected. In the following section, we shall consider the implications of the resonant scattering.

\(^1\)This may be possible, for example, in the case of molecular formation in a solid at energies low compared to its Debye temperature, where recoilless processes analogous to the Mossbauer effect may dominate.
Appendix B. Resonant scattering of $\mu t$

B.1.2 Epithermal Collisions

Let us consider an epithermal collision between $\mu t$ and a DX molecule, with a $\mu t$ lab energy $E_{\mu t}$ (velocity $V_{\mu t}$). Throughout this appendix, solid state effects such as lattice binding and phonon exchange are neglected unless otherwise specified.

Resonant scattering may occur via the following sequence of processes: (1) the molecular complex (MMC) is formed in a collision $\mu t + DX$, (2) MMC receives a recoil from the impact of $\mu t$, (3) MMC may be (partly) thermalized via collisions with the rest of the target molecules, (4) ro-vibrational states of MMC may change as a result of collisions with other molecules, and (5) back decay occurs leaving molecule $DX$ in either the ground state (elastic channel) or in an excited state (inelastic).

The energy of the $\mu t$ after back decay is important. It can be shown that if the $\mu t$ energy is the same before and after the back decay, the effective formation approximation gives the correct fusion yield (condition (b) above). However, if the $\mu t$ energy changes such that it is removed from the resonant region, the effective formation approximation fails as we shall see. It should be noted that resonant structure has a narrow width for low temperature targets, therefore a small change in the $\mu t$ energy is sufficient to remove it from the resonance.

Even in the completely elastic case (no MMC thermalization, no MMC relaxation, and back decay via the elastic channel), the mean energy of $\mu t$ in the lab frame is significantly reduced after back decay, because of recoil of the MMC in process (2) and that of DX in (5). In reality, MMC thermalization, MMC ro-vibrational relaxation, and back decay via inelastic channels, all give contributions to reducing the $\mu t$ energy, hence the $\mu t$ is likely to be removed from the resonance region.
B.1.3 Comparison with the explicit back decay model

We illustrate the effect of back decay in the following simplified comparison. We consider a case in which a \( \mu t \) beam of resonant energy collides with a D\(_2\) layer of varying thickness, and we wish to describe the fusion yield in the D\(_2\) layer as a function of its thickness. We compare the two models (a) using effective formation rates renormalized for the back decay fraction as was used in Ref. [9,27,75,82], and model (b) using the total formation rate and explicit back decay. We consider only two competing processes of elastic scattering and molecular formation, the latter leading to either fusion or back decay, and neglect other possibilities such as scattering followed by molecular formation (muon decay is also neglected).

With these assumptions, the fusion yield as a function of layer thickness in the effective formation model (a) can be expressed as:

\[
Y_a(x) = \frac{\sigma_{eff}}{\sigma_{eff} + \sigma_{scat}}[1 - \exp\{- (\sigma_{eff} + \sigma_{scat})x\}],
\]

where \( x \) (cm\(^{-2}\)) is the thickness multiplied by number density \( n \), and cross sections \( \sigma_i \) and rates \( \sigma_i \) are related with \( \lambda = vn\sigma \), \( v \) being velocity of the \( \mu t \) for a target molecule at rest. With back decay explicitly included (model b), the fusion yield as a function of layer thickness is now written:

\[
Y_b(x) = \frac{\sigma_{tot}W}{\sigma_{tot} + \sigma_{scat}}[1 - \exp\{- (\sigma_{tot} + \sigma_{scat})x\}]
\]

These expressions can be expanded in series in \( x \):

\[
Y_a \sim \sigma_{eff}x - \frac{1}{2}(\sigma_{eff} + \sigma_{scat})x^2 + \cdots \quad \text{(B.6)}
\]

\[
Y_b \sim \sigma_{tot}Wx - \frac{1}{2}(\sigma_{tot} + \sigma_{scat})x^2 + \cdots \quad \text{(B.7)}
\]

Recalling \( \sigma_{eff} = \sigma_{tot}W \), both models give the same results to leading order. Therefore, in the limit of a thin layer, model (a) can be justified for describing the fusion yield.
Appendix B. Resonant scattering of $\mu t$

However, the next-to-leading order corrections become important at $x$ comparable to the mean interaction length, $x_{\text{int}} = (\sigma_{\text{tot}} + \sigma_{\text{scat}})^{-1}$. In the limit of a thick layer ($x \gg x_{\text{int}}$), the fusion yield is simply the coefficient in Eqs. B.4 and B.5. Comparing Eq. B.6 and B.7, it is clear that

$$Y_a > Y_b, \quad \text{unless } W = 1$$

resulting in an overestimate of fusion yield with model (a).

In our earlier analysis of the time-of-flight fusion experiments, the effective rate (model a) was used, and we observed that the calculated fusion yield was significantly larger than the experimental data. Since our thickness was comparable to the interaction length $x_{\text{int}}$, the discussion given here explains the discrepancy. Detailed Monte Carlo calculations indeed show that the fusion yield is overestimated by nearly 50% in our time-of-flight fusion measurement arrangement, compared to model (b).

It is interesting to note that Eq. B.5 suggests that, within model (b) and given the knowledge of $\sigma_{\text{scat}}$, one can determine both $\lambda_{\text{tot}}$ and $W$ by the absolute measurement of the thickness dependence of fusion yield $Y(x)$, i.e., $\lambda_{\text{tot}}$ from the curvature of $Y(x)$, and $W$ from $Y(x)$ at the limit of large $x$ (via the coefficient $\sigma_{\text{tot}}W/(\sigma_{\text{tot}} + \sigma_{\text{scat}})$). Note that it is difficult to disentangle $\lambda_{\text{tot}}$ and $W$ in conventional measurements using a homogeneous D/T mixture.

B.2 Back-decayed $\mu t$ energy distribution

B.2.1 MMC recoil and thermalization

In this section, we shall take a more detailed look at each step in the back decay in order to estimate the energy of $\mu t$ after back decay.

When the muonic molecular complex is formed in the collision, it will receive recoil
Appendix B. Resonant scattering of $\mu$

velocity $V_C = \frac{M_\mu}{M_\mu + M_{DX}} \cdot v_\mu$. For the case of $\mu + D_2$ collisions at the main resonance of $E_{\mu}^{lab} = 0.5$ eV, $V_C \sim 0.24$ cm·$\mu$s$^{-1}$. The final velocity of MMC is important since it affects the lab energy of back-decayed $\mu$. Two kinematic extremes are the complete thermalization of MMC before it decays, and no thermalization at all. For either case, the maximum possible energy of back-decayed $\mu$ (for a low temperature target) is obtained when “elastic scattering” (i.e. no excitation of the target $D_2$ molecule) takes place. However, even in elastic scattering, $\mu$ is decelerated in the lab frame due to the recoil of MMC and $D_2$ as discussed above.

Cross sections for the interaction of $[(d\mu)\bar{e}e] + D_2$ has been calculated by Padial et al. [154,239]. Extrapolating a figure given in Ref. [239] to epithermal energies, the elastic scattering cross section $\sigma_C^{el}$ appears to have a value of order $3 \times 10^{-15}$ cm$^{-2}$. The average time between elastic collision $\Delta t \sim (v_C n \sigma_C^{el})^{-1}$, where $n$ is the $D_2$ number density, can be compared to the MMC life time, $\tau_C$.

In the case of $E_{\mu}^{lab} \sim 0.5$ eV, which corresponds to the largest resonance for molecular formation in $\mu + D_2$, the MMC recoil velocity is $V_C \sim 0.24$ cm·$\mu$s$^{-1}$, hence at a molecular density of $1.4 \ N_0/2$ ($N_0 = 4.25 \times 10^{22}$ cm$^{-3}$, the atomic density of liquid hydrogen) corresponding to a solid at 3 K, $\Delta t \sim 5 \times 10^{-14}$ s. Comparing this to $\tau_C \sim 10^{-12}$ [152,156], we have

$$\Delta t \gg \tau_C, \tag{B.9}$$

suggesting a fair number of collisions occur before the MMC decays.

The average number of collisions needed to slow the MMC of $E_{MMC}^{init}$ to $E_{MMC}^{fin}$, can be roughly estimated, by generalizing the formula for neutron thermalization [240]:

$$n = \frac{1}{\xi} \ln \frac{E_{MMC}^{fin}}{E_{MMC}^{init}}, \tag{B.10}$$

where

$$\xi = 1 + \frac{(M_{MMC} - M_{DX})^2}{2M_{MMC}M_{DX}} \ln \frac{M_{MMC} - M_{DX}}{M_{MMC} + M_{DX}}. \tag{B.11}$$
For $E_{\mu t}^{\text{lab}} \sim 0.5 \text{eV}$ in $\mu t + D_2$, the MMC recoil energy is $E_{\text{MMC}}^{\text{init}} \sim 0.2 \text{eV}$, and we have $n \sim 8$ for $E_{\text{MMC}}^{\text{fin}} = 0.4 \text{meV}$. Obviously, the formula B.10 gives simply a crude estimate, but together with Eq. B.9, one can assume that the MMC slows down significantly before back decay occurs.

### B.2.2 MMC ro-vibrational relaxation

Relaxation in ro-vibrational states of MMC would change the $Q$-value available for the back decay reaction, affecting the $\mu t$ energy. According to the estimate of Lane [151], vibrational de-excitation rates appear to be a few orders of magnitude smaller than the MMC decay rate (although the former increases with the target temperature), hence we ignore its contribution here. Future calculations of the MMC vibrational relaxation would be very helpful.

Rotational transitions in the MMC have been calculated by Ostrovskii and Ustimov [153], and by Padial et al. [154] for the case of targets in thermal equilibrium. Ostrovskii and Ustimov estimate relaxation rates of order of $10^{13} \text{s}^{-1}$, while Padial et al., who claim higher accuracy, give $\sim 0.3 \times 10^{13} \text{s}^{-1}$ at 300 K. Given the MMC decay rate of $\sim 10^{12} \text{s}^{-1}$, if rotational thermalization is achieved only partially, then detailed rate equations\(^2\) should be solved to calculate the energy distribution of back-decayed $\mu t$. In our calculation below, we give the two extreme cases of complete rotational thermalization and no thermalization at all.

\(^2\)Scrinzi et al. give an example of such rate equations in Ref. [12], though for different purposes.
B.2.3 Resonant excitation of DX molecule

We should stress here the importance of resonant excitation of the DX molecule, which has not been well considered in the literature to our knowledge:

\[ \mu t + [D_2]_{\nu_i K_i} \rightarrow [(d\mu t)_{11}^{\text{dec}}]^* \rightarrow \mu t + [D_2]_{\nu_i' K_i'} \]  

(B.12)

The process when \( \nu_i', K_i' > \nu_i, K_i \) leaves the target D_2 molecule in an excited state, hence the back-decayed \( \mu t \) energy is correspondingly reduced\(^3\).

B.2.4 Back-decayed \( \mu t \) energy

As we have seen, the energy distribution of back-decayed \( \mu t \) depends on the details of back decay reactions as well as MMC interactions with the surrounding environment, which include: (a) the back decay matrix elements\(^4\) \(< \nu_f K_f | H | \nu_i' K_i' >\), (b) collisional ro-vibrational relaxation of the MMC, (c) the MMC elastic scattering cross section for \([(d\mu t)_{\text{dec}}]^* + D_2\), and perhaps (d) solid state effects such as emission of phonons upon back decay and recoilless MMC decay. Apart from the plausible yet still speculative effect (d), the processes (a) and (b) affect the \( Q \)-value of the back decay reaction, while (c) changes the motion of the MMC centre of mass frame, hence affecting the \( \mu t \) energy in the lab frame.

The complete analysis of this complex chain of processes is beyond the scope of this appendix, and we deal with two limiting cases of (b), i.e., no rotational relaxation at all, and complete rotational relaxation (no vibrational relaxation assumed in either case). We develop in the following expressions for the double differential rates for resonant scattering which depend on both outgoing and incoming \( \mu t \) energies in the lab frame

\(^3\)Note that resonant deexcitation (e.g., \( \nu_i', K_i' < \nu_f, K_f \)) is possible for high temperature targets, but is negligible at low temperature.

\(^4\)I am using the language of the perturbation theory here with some reservation, recognizing the controversy in the formulation of the MMC formation and back decay processes.
\( \lambda^F_{\text{ResScat}}(E'_{\mu}; E_{\mu}) \). Our formulation is analogous to the resonant formation rate calculations by Faifman et al. [133]. We explicitly treat the back-decayed \( \mu\)t energy \( E'_{\mu} \), paying attention to the energy balance. Thermal motion of the MMC is included to give the Doppler broadening [157] of the \( E'_{\mu} \) distribution.

In the first limit of no relaxation at all in the MMC states, the rate for the resonant scattering rate \( \lambda^F_{\text{ResScat}}(E'_{\mu}; E_{\mu}) \) can be written:

\[
\lambda^F_{\text{ResScat}}(E'_{\mu}; E_{\mu}) = \sum_{\nu_i, K_i} \omega(\nu_i, K_i) \sum_{\nu_f, K_f, S} \lambda^S_{\nu_i K_i, \nu_f K_f}(E_{\mu}) \sum_{\nu_i', K_i'} \frac{\Gamma^S_{\nu_f K_f, \nu_i' K_i'}}{\tilde{\Lambda}_f + \sum_{\nu_i', K_i'} \Gamma^S_{\nu_f K_f, \nu_i' K_i'}} \times \int dE_u D(E_u) I \left[ E'_{\mu} - E'_{\nu_i K_i}(\nu_f K_f S ; \nu_i' K_i') - E_u \right] \quad (B.13)
\]

where

\[
E'_{\nu_i K_i}(\nu_f K_f S ; \nu_i' K_i') = \frac{M_{DX}}{M_{DX} + M_{\mu}} \left[ \epsilon_{\text{res}}(\nu_f K_f) - \Delta E_{\nu_i K_i, \nu_i' K_i'} \right] \quad (B.14)
\]

is the \( \mu\)t energy from the decay channel \((\nu_f, K_f, S) \rightarrow (\nu_i', K_i', S)\) with

\[
\Delta E_{\nu_i K_i, \nu_i' K_i'} = -[E(\nu_i' K_i') - E(\nu_i K_i)] \quad (B.15)
\]

being the binding energy difference between the initial and final state of \( DX \) (i.e., excitation or de-excitation energy of \( DX \) due to the resonant scattering). Other notations used in Eq. B.13 include:

- (As usual) \( S, F \) denote the spin of \( d\mu\)t and hyperfine state of \( \mu\)t, respectively.

- \( I[\Delta] \) is the resonance intensity profile for detuning \( \Delta \) (e.g., \( I[\Delta] = \delta(\Delta) \) in Faifman’s model).

- \( D(E_u) \) is the Doppler broadening distribution due to the motion of MMC at the time of back decay (e.g., Gaussian distribution for thermalized MMC if \( E'_{\mu} \gg kT \)).

- \( \Gamma^S_{\nu_f K_f, \nu_i' K_i'} \) is the back decay width for the channel \((\nu_f, K_f) \rightarrow (\nu_i', K_i')\) with given \( S, F \).
Appendix B. Resonant scattering of $\mu$

- $\lambda_{\nu_i,K_i,v_f,K_f}(E_{\mu})$ is the total formation rate for the incident $\mu$ lab energy $E_{\mu}$.

- $\omega(\nu_i, K_i)$ is the initial $\nu_i, K_i$ distribution of target molecules $DX$.

In the other limit that MMC rotational relaxation is complete, the following substitution should occur:

$$
\frac{\Gamma_{\nu_f,K_f,v_i',K_i'}^{SF}}{\hat{\lambda}_f + \sum_{v_i',K_i'} \Gamma_{\nu_f,K_f,v_i',K_i'}^{SF}} \rightarrow \frac{\lambda^{SF}_{\nu_f,K_f,v_i',K_i'}}{\lambda_f + \sum_{v_i',K_i'} \lambda^{SF}_{\nu_f,K_f,v_i',K_i'}} \tag{B.16}
$$

$$
E_{\nu_i,K_i}(v_f K_f S; v_i' K_i') \rightarrow E'_{\nu_i,K_i}(v_f K_f S; v_i' K_i') \tag{B.17}
$$

with $\omega_B(K_f')$ being the Boltzmann distribution for the rotational states of MMC, hence simplifying the summation in Eq. B.13.

Because the expression given in Eq. B.13 is rather complicated, we can alternatively take advantage of the already calculated formation rates $\lambda_{\mu}^{SF}$ and fusion probability $W^F$ to write, in the limit of no MMC rotational relaxation, that

$$
\lambda_{ResScat}^{SF}(E_{\mu}'; E_{\mu}) \approx \lambda_{\mu}^{SF}(E_{\mu}) \cdot (1 - W^F) \cdot f^F(E_{\mu}'; E_{\mu}) \tag{B.18}
$$

where

$$
f^F(E_{\mu}'; E_{\mu}) = \sum_{\nu_f K_f} h_{\nu_i K_i S}^{SF}(\nu_f K_f; E_{\mu}) \sum_{v_i',K_i'} g_{\nu_f K_f}^{SF}(v_i', K_i') \times \int dE_u D(E_u) I [E_{\mu}'' - E_{\nu_i,K_i}(\nu_f K_f S; v_i' K_i') - E_u] \tag{B.19}
$$

is the energy distribution of back-decayed $\mu$ for a given initial energy $E_{\mu}$, and

$$
g_{\nu_f K_f}^{SF}(v_i', K_i') = \frac{\Gamma_{\nu_f K_f,v_i' K_i'}^{SF}}{\sum_{v_i',K_i'} \Gamma_{\nu_f K_f,v_i' K_i'}^{SF}} \tag{B.20}
$$

is the branching ratio for decay to the state $(v_i', K_i', S)$, given the MMC state of $(\nu_f, K_f, S)$, and

$$
h_{\nu_i K_i S}^{SF}(\nu_f K_f S; E_{\mu}) = \frac{\lambda_{\nu_i,K_i,v_f,K_f}^{SF}(E_{\mu})}{\sum_{\nu_f K_f S} \lambda_{\nu_i,K_i,v_f,K_f}^{SF}(E_{\mu})} \tag{B.21}
$$
is the conditional probability that the MMC has the state \((\nu_1, K_i, S)\), given that the MMC is formed with a \(\mu t\) of energy \(E_{\mu t}\). Again, in the other limit of complete MMC rotational relaxation, we can average \(K_f\) states over the Boltzmann distribution, and let

\[
\sum_{\nu_f K_f S} \tilde{h}^{SF}_{\nu_1 K_1}(\nu_f K_f S; E_{\mu t}) \to \sum_{\nu_f S} \tilde{h}^{SF}_{\nu_1}(\nu_f K_f S; E_{\mu t}) = \sum_{\nu_f S} \frac{\sum_{K_f} \omega_B(K_f') \lambda_{\nu_1, K_i, \nu_f, K_f'}^{SF}(E_{\mu t})}{\sum_{\nu_f S} \lambda_{\nu_1, K_i, \nu_f, K_f'}^{SF}(E_{\mu t})},
\]

together with the substitution in Eq. B.17.

Note in Eq. B.18, we made an approximation by factorizing the state dependence, and this is not rigorously accurate when the state dependence of each factor is large, hence the approach given here should be taken as a first approximation. On the other hand, the first expression given in Eq. B.13 does not rely on the factorization approximation hence it is more accurate, though more complicated.

In order to numerically evaluate these expressions, we need some several hundred matrix elements for MMC transitions. It should be stressed, however, these have been already calculated, for example by Faifman et al. for calculations of \(\lambda_{\mu t}^F\), thus with their assistance we can readily estimate the energy distribution of back-decayed \(\mu t\) (within the approximation that the MMC is translationally thermalized, and is rotationally relaxed or not relaxed at all).

We note that in case of the elastic scattering, \((\nu'_i, K'_i) = (\nu_1, K_i)\) with translationally thermalized MMC, the energy of back-decayed \(E_{\mu t}\) is given by the resonance energy \(\epsilon_{res}\) divided between \(\mu t\) and \(DX\) (convoluted with the Doppler broadening profile). Hence the claims by Somov and Jeitler [22, 74, 238] of thermalized \(\mu t\) after back decay is not physically justified. Jeitler also considered the limit of no MMC rotational relaxation at all, but the simple expression given for the \(\mu t\) energy distribution for that case \((E'_{\mu t} = E_{\mu t}(\frac{M_{MMC}}{M_{\mu t} + M_{MMC}})^2\), Eq. 4.72 in Ref. [74]) is not accurate, as the back decay into \(DX(\nu'_i, K'_i) \neq (\nu, K_i)\) (the resonant excitation of \(DX\), and the Doppler broadening due to
MMC thermal motion are neglected.

B.3 Implications for our measurements

Finally, we discuss the implication of resonant scattering in our atomic beam experiments. Regarding the validity of the effective model, we used detailed Monte Carlo simulations to compare the fusion yields in our standard time-of-flight target arrangement\(^5\) for both model (a), the effective model with the renormalized rates, and model (b), with explicit resonant scattering. Assuming that resonant scattering removes \(\mu t\) from the resonance region, which is well justified from the discussion above, we observed that model (a) overestimates the fusion yield in the DS layer by nearly 50%. The use of model (b) thus resolves the inconsistency of our data with our earlier analysis using model (a), as reported in Ref. [78].

As for our sensitivity to the scattered \(\mu t\) energy, we have performed Monte Carlo calculations with different assumptions of the back-decayed \(\mu t\) energies. As mentioned, the exact evaluation of our expressions given in this appendix for the back-decayed \(\mu t\) energy distribution cannot be performed yet, since it requires the transition matrix elements which are not currently available to us. Our Monte Carlo calculations in the time-of-flight arrangement, with \(E'_\mu t\) varied between 1 meV to 0.3 eV, suggest some 7% difference in the DS fusion yield. It should be noted that these variations depend also on the assumptions for \(\mu t\) interactions in a solid at low energies. Nonetheless, our present lack of precise knowledge of the back-decayed \(\mu t\) energy gives, a significant, yet not overwhelming contribution to the total uncertainty in our measurements of the resonant molecular formation rate, hence significant improvement in the accuracy of the latter would require, among others, the exact numerical evaluation of our \(E'_\mu t\) expression.

\(^5\)Emission target with \(c_t = 0.1\%\) with a 14 T-\(\ell\) D\(_2\) moderation layer in the US, and 3 T-\(\ell\) D\(_2\) in the DS.
In summary, we have pointed out in this appendix the importance of the resonant scattering via back decay, a process which has not previously been well-considered, and (a) showed that the use of renormalized effective formation rates leads to an overestimate of fusion yield and (b) gave detailed expressions for the energy distribution of back-decayed $\mu t$, which is considerably different from the previously assumed thermal distribution. From the full Monte Carlo calculations, the effect of (a) is estimated to be $\sim 50\%$ and (b) $\lesssim 10\%$, the latter depending on the possible solid state effects in $\mu t$ thermalization and molecular formation.
Appendix C

Muon decay electron time spectrum fit

Fits of muon decay electron time spectra to the exponential functions were performed taking into account the finite size of the time bins.

For the muon stopping in $M$ different elements, it obviously holds in the continuous approximation that

$$\frac{dN^\mu(t)}{dt} = -\sum_k \lambda_k N^\mu_k,$$

where $N^\mu_k(t)$ are the numbers of muons in the element $k$ at time $t$, in which the muon disappearance rate is $\lambda_k$, and $N^\mu(t) = \sum_k N^\mu_k(t)$. By solving Eq. C.1 for $N^\mu$, we have a number of $\mu$ decay per unit time

$$\frac{dN^\mu(t)}{dt} = -\sum_k \lambda_k N^0_k \exp(-\lambda_k t),$$

where $N^0_k$ is the number of muons at $t = 0$ in the element $k$ with $N^0 = \sum_k N^0_k$. Note that Eq. C.2 can be decomposed into a set of $M$ uncoupled independent equations.

On the other hand, the decay electron detection rate, which we measure in our experiments, is

$$\frac{dN^e(t)}{dt} = -\epsilon_k B_k \frac{dN^\mu(t)}{dt}$$

$$= \sum_k \epsilon_k B_k \lambda_k N^0_k \exp(-\lambda_k t),$$

where
where $\epsilon_k$ is the detector efficiency for decay electrons from the element $k$, and $B_k$ is the branching ratio of the muon-to-electron decay to total muon disappearance

$$B_k = \frac{Q_k \lambda_0}{Q_k \lambda_0 + \lambda_k^C + \lambda_k^X}, \quad (C.4)$$

where $\lambda_0$ is the muon decay rate in free space, and, for the element $k$, $Q_k$ is the Huff factor, $\lambda_k^C$ the nuclear capture rate, and $\lambda_k^X$ is an effective rate representing other loss mechanisms such as muon transfer to heavier elements and muonic atom emission\(^1\). The Huff factor ($Q \leq 1$) takes into account the effect of muon binding to the nucleus, resulting in reduced phase space available as well as the time dilation of the muon’s proper time with respect to the lab frame, both of which in turn lead to the reduction in muon decay rate, according to Huff [226]. Thus we have

$$\frac{dN^e(t)}{dt} = \frac{d}{dt} \sum_k M_k N^e_k(t) \quad (C.5)$$

We used $\lambda_k = Q_k \lambda_0 + \lambda_k^C + \lambda_k^X$. Note that because of muon loss channels it is $Q_k \lambda_0$, rather than $\lambda_k$, which is in the normalization factor in the final line of Eq. C.5.

The actual experimental time spectra are histogrammed in a finite size of time bin $\Delta t$, hence are discrete function of $t_i$ ($i = 1, 2, 3 \ldots$). To reflect this, Eq. C.5 is integrated over $\Delta t$,

$$\Delta N^e(t_i) = \int_{t_i-\frac{1}{2}\Delta t}^{t_i+\frac{1}{2}\Delta t} \frac{dN^e(t)}{dt} dt. \quad (C.6)$$

Evaluating Eq. C.6, we have

$$\Delta N^e(t_i) = \sum_k \frac{Q_k \lambda_0}{\lambda_k} N^0_k \times$$

\(^1\)Here we are approximating these loss processes as an exponential function with a simple “effective” rate. The real situation could be more complex.
Appendix C. Muon decay electron time spectrum fit

\[
\left\{ \exp \left[-\lambda_k \left( t_i - \frac{1}{2} \Delta t \right) \right] - \exp \left[-\lambda_k \left( t_i + \frac{1}{2} \Delta t \right) \right] \right\}.
\] (C.7)

This can be rearranged as

\[
\Delta N^e(t_i) = \sum_k^M \epsilon_k \frac{Q_k \lambda_0}{\lambda_k} N_k^0 \times 
\exp \left[-\lambda_k t_i \right] \left( \exp \left[\frac{1}{2} \lambda_k \Delta t \right] - \exp \left[-\frac{1}{2} \lambda_k \Delta t \right] \right),
\] (C.8)

which can be expanded in \( \lambda_k \Delta t \) to \( \mathcal{O}(\lambda_k \Delta t) \),

\[
\Delta N^e(t_i) \sim \sum_k^M \epsilon_k \frac{Q_k \lambda_0}{\lambda_k} N_k^0 \exp \left[-\lambda_k t_i \right] \times \lambda_k \Delta t 
= \frac{dN^e(t_i)}{dt} \Delta t.
\] (C.9)

Eq. C.9 is an explicit expression for the intuitively obvious conclusion that, in the limit of small \( \Delta t \), counts in a histogram bin are proportional to its bin size. It also shows that the use of the integrated fitting function Eq. C.10 is necessary unless \( \lambda_k \Delta t \ll 1 \), i.e., the bin size is much smaller than the lifetime\(^2\).

Thus, our fit function \( F^{fit}(t_i) \) is, with a background term \( \text{bkgd}^3 \)

\[
F^{fit}(t_i) = \Delta N^e(t_i) + \text{bkgd}
= \sum_k^M \epsilon_k \frac{Q_k \lambda_0}{\lambda_k} N_k^0 \times 
\left\{ \exp \left[-\lambda_k \left( t_i - \frac{1}{2} \Delta t \right) \right] - \exp \left[-\lambda_k \left( t_i + \frac{1}{2} \Delta t \right) \right] \right\}
+ \text{bkgd}.
\] (C.10)

\(^2\)In this thesis, histograms were collected occasionally with a 10 ns bin size, which gives \( \lambda_k \Delta t \sim 1/7 \) for \( k = \text{gold} \) (1/\( \lambda_k \sim 70 \text{ ns} \)), clearly requiring the integrated function Eq. C.10 for accurate fits.

\(^3\)In a single-hit detection mode (as opposed to a multi-hit mode), the detection efficiency is time dependent, hence the recorded histogram will have a slope even though the accidental background was constant in time.
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