CRYOGENIC HYDROGEN MASER

By

Martin Dominik Hürlimann

Dipl. Natw. ETH, Swiss Federal Institute of Technology, Zürich

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF

DOCTOR OF PHILOSOPHY

in

THE FACULTY OF GRADUATE STUDIES

PHYSICS

We accept this thesis as conforming to the required standard

THE UNIVERSITY OF BRITISH COLUMBIA

July 1989

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Department of <u>Physics</u>

The University of British Columbia Vancouver, Canada

Date July 28, 1989

Abstract

A new type of atomic hydrogen maser that operates in a dilution refrigerator has been developed. In this device, the hydrogen atoms circulate back and forth between a microwave pumped state invertor in high field and the maser cavity in zero field.

A prototype maser with a small maser cavity has been built and the results obtained so far are encouraging. Stable maser oscillations were observed for temperatures of the maser bulb between 230 mK and 660 mK and for densities up to 3×10^{12} cm⁻³. The short term frequency stability was measured with the help of two high quality quartz crystal oscillators by the three-cornered-hat method. The observed fractional frequency fluctuations for an averaging time of 1s were $6.3 \pm 3.7 \times 10^{-14}$, which is lower than the results from the best room temperature masers. In conjunction with the stability measurements, the phase noise of the maser electronics was investigated. In particular, the temperature dependence of the phase noise of the cooled preamplifier was measured and it was shown that anomalous high noise levels between 2.2 K and 4.2 K are caused by the boiling of the liquid helium.

From the temperature dependence of the maser frequency, the binding energy E_B of H on ⁴He could be determined to a high precision. The result is $E_B = 1.011 \pm 0.010$ K.

An extensive computer simulation program has been written that models the operation of the cryogenic hydrogen maser. It has been used to analyze and interpret some of the data. In addition, this simulation program is helpful for the design of an improved second generation cryogenic maser. Based on the present data and the model calculations, a new pumping scheme is proposed that is expected to increase the efficiency of the state invertor significantly.

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Acknowledgement

First of all, I would like to thank my 'Doktorvater' Walter Hardy who supervised this work over the last seven years. He conceived this project and his active participation in all aspects of the experiment were essential for its success. I also profited from the theoretical support from John Berlinsky in the early stage of the project. Rick Cline was of great help in the cryogenic design work. In addition, he built the dilution unit used in this experiment and he also assured that an efficient computer system was always operational in our laboratory. In this experiment, we relied extensively on the excellent service of the departmental electronics shop and machine shop. In particular, I would like to thank Phil Akers and George Babinger for their superb machining of the maser parts.

This project would have been impossible without the support from many people outside this university in form of advice and loan of equipment. I am especially indebted to Stuart Crampton and Fred Walls. I had many helpful discussions with Stuart Crampton about the basic maser equations and the maser computer simulation. He was also very generous in lending us two conventional hydrogen masers. We are very grateful to Fred Walls who shipped us the two best quartz crystal oscillators in possession of the National Bureau of Standards (now NIST). Without this loan, it would not have been possible to perform the reported frequency stability measurements. David Allan and Dick Davis from NBS loaned us a GPS receiver system which enabled us to calibrate our local oscillators. We also acknowledge the loan of a rubidium standard by David Wineland and the loan of magnetic shielding material by Bob Vessot.

I would like to thank Bud Bridges for his assistance in the early design work and

х

Erwin Klein for his help during a low temperature run. All the members of the atomic hydrogen group at UBC have given me countless assistance and have created a very pleasant working environment. I would like to thank in particular Ichiro Shinkoda, Meritt Reynolds, Mike Hayden, Esa Tjukanov and Chris Lusher.

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I am grateful to the University of British Columbia and the Killam Foundation for their support in form of postgraduate fellowships.

Finally, I thank my wife Elaine for her patience and for accepting the fact that low temperature experiments often take much longer than initially anticipated.

Chapter 1

Introduction

The atomic hydrogen maser is presently the most stable atomic clock for averaging times $\tau \leq 10^6$ s. In the best devices, fractional frequency fluctuations of less than 10^{-15} for $\tau \simeq 10^4$ s have been achieved [1]. This extraordinary frequency stability has led to several spectacular applications: Vessot and collaborators [2] used a space-borne hydrogen maser to measure the gravitational red shift of the earth to a precision of 7×10^{-5} . The success of the very long baseline interferometry (see e.g. [3]) also depends to a large extent on the stability of the hydrogen maser. Other applications of the hydrogen maser include the navigation of space probes in deep space.

The atomic hydrogen maser was initially conceived and developed at Harvard University in 1960 by H. M. Goldenberg, D. Kleppner and N. F. Ramsey [4]. They realized that collisions of atomic hydrogen with paraffin or Teflon coated walls do not perturb the hyperfine levels of hydrogen atoms significantly. In fact, it is possible to confine a low density sample of hydrogen atoms that radiate at the hyperfine transition for up to 1 s in a Teflon coated bulb, before collisions destroy the phase coherence. Such long interaction times between the hydrogen atoms and the microwave radiation give rise to a very narrow line width. This is an essential prerequisite for any atomic clock.

An excellent discussion of the theory and the principle of the atomic hydrogen maser by its inventors can be found in [5] and [6]. The basic principle is illustrated in fig. 1.1. Molecular hydrogen is dissociated in an rf discharge and a beam of atomic hydrogen emerges from the source. The beam is passed through a hexapole magnet. This magnet

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Figure 1.1: Basic scheme of the conventional hydrogen maser. Also shown is the hyperfine diagram and the typical behavior of the output power versus the hydrogen flux.

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acts as a state selector and focuses only atoms which are in the upper two hyperfine states into a Teflon coated bulb. This storage bulb is located inside a microwave cavity with a high quality factor which is tuned to the hyperfine transition at zero magnetic field, the famous '21 cm line'. The hydrogen maser uses the $\Delta F = 1$, $\Delta m_F = 0$ transition (| c) to | a)), because its frequency is to first order independent of magnetic field. A small bias field is applied along the cavity rf magnetic field (H_z) to remove the degeneracy with the $\Delta m_F = \pm 1$ transitions. If the flux of state selected hydrogen atoms exceeds a certain threshold value, stable maser oscillations can be observed. Efficient pumps remove all the hydrogen atoms leaving the maser bulb and keep the background pressure very low.

It is indicated in fig. 1.1 that the output power of the maser does not increase linearly with the hydrogen flux, but it turns over and the maser stops to oscillate altogether if the flux of hydrogen atoms exceeds a critical level. The maximum power output is typically limited to about 10^{-12} W. There is only a relatively small range of hydrogen fluxes or hydrogen densities at which maser action is possible. In order to understand this behavior, the effect of collisions between hydrogen atoms has to be taken into account. In particular, collisions between hydrogen atoms can lead to spin exchange which both broadens the atomic line and leads to fast relaxation between the hyperfine levels that reduces the polarization of the incoming hydrogen atoms. As the density is increased, these effects increase proportionally and they are responsible for the observed density dependence of the maser output power.

Although the frequency stability of conventional hydrogen masers is already extremely high, there is demand for clocks with even better performance. Intended applications include better deep space navigation and experiments searching for gravity waves. Another example that illustrates the need for better clocks is the fact that the long term stability of the period of the millisecond pulsar PSR 1937+21 exceeds the performance of all presently available atomic clocks [7].

The stability of state-of-the-art hydrogen masers is limited by electronic and thermal noise [1]. This will be further discussed in chapter 4 of this thesis. It is unlikely that the performance of conventional hydrogen masers, operated at room temperature, can be further improved by a significant amount, except for very long averaging times, where external factors degrade the stability. In 1978, Crampton, Phillips and Kleppner [8] pointed out that hydrogen masers operated at low temperatures might have an improved stability. Vessot et al. [1] came to the same conclusion. The main reason is the much smaller spin exchange broadening at cryogenic temperatures, which implies that the maximum available power output of the maser increases, as does the range of operating densities. In addition, the thermal noise is reduced, a cooled low noise preamplifier can be used, and the structural stability is much higher at low temperatures.

However, when a conventional hydrogen maser was cooled to low temperature, it was soon found that Teflon is not a suitable wall coating at low temperature, and no maser oscillation could be observed below about 50 K [9]. The first maser signal at 10 K was observed by Crampton et al. [10] who used frozen neon as the storage surface. Unfortunately, this cryogenic wall coating evaporates slowly and its use is therefore not very suitable in a high performance hydrogen maser.

Approximately at the same time, several groups started to develop techniques to study atomic hydrogen at temperatures below 1 K. These efforts have been directed towards the possible observation of the Bose Einstein condensation in spin polarized hydrogen and were initially completely unrelated to the maser research. In the first step, Silvera and Walraven [11] showed that atomic hydrogen can be stored successfully at low temperature in a container lined with a film of superfluid helium. In a series of experiments, Hardy et al. [12] studied the properties of atomic hydrogen at temperatures below 1 K and zero magnetic field by means of pulsed magnetic resonance at the hyperfine transition. They confirmed that the spin exchange broadening is at these temperatures about 1000 times

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smaller than at room temperature. In addition, they found that the hyperfine frequency shift due to the combined effect of the helium buffer gas and interactions with the walls goes through an extremum at around 0.5 K. This is therefore the natural temperature to operate a cryogenic hydrogen maser with ⁴He coated walls. Berlinsky and Hardy [13] analyzed in detail the theoretical frequency stability of such a device. They concluded that an improvement of up to three orders of magnitude compared to the stability of a conventional hydrogen maser operated at room temperature can be expected. This prediction has been the main motivation for the work described in this thesis.

In spring 1986, three groups reported almost simultaneously the successful operation of cryogenic hydrogen masers with helium coated walls. The first report came from the MIT group [14]. In their apparatus, a hydrogen discharge source is placed inside a high magnetic field and cooled to 0.3 K. Atoms that are predominantly in the upper two hyperfine states emerge from this source and reach a microwave cavity in zero field. After the cavity quality factor was enhanced by external feedback, self-sustained maser oscillations were observed. This apparatus was not intended to be a stable frequency source, but it was used as a rather elegant test of the efficiency of their hydrogen source. This source has been used in their experiments on the magnetic confinement of atomic hydrogen [15].

About a week after the MIT group, we observed the first maser signal in our apparatus [16]. In our maser, the hydrogen atoms circulate back and forth between a microwave pumped state selector and the maser cavity. The principle of operation and the results obtained with our prototype maser are discussed in detail in this thesis.

One month later, the joint group of Harvard University and the Smithsonian Astrophysical Observatory succeeded with their cryogenic maser [17]. The design of this maser is very similar to that of a conventional room temperature maser, except that the maser bulb is coated with a superfluid film. In order to keep the helium background pressure

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low enough and enable the beam of atomic hydrogen to reach the maser bulb, a large sorption pump was installed in their dilution refrigerator. Unfortunately, the apparatus has had problems with low temperature leaks, which has made it impossible to achieve a saturated helium wall coating, and no results of frequency stability have been reported so far. It will be interesting to compare the performance of this type of beam maser with our maser, which is based on a completely different design.

This thesis is organized as follows: In chapter 2, the hyperfine structure of hydrogen and the interaction with a classical rf field are discussed. Following a similar approach as taken in the NMR literature when effects of the so-called radiation damping are discussed, we derive the equations relevant for maser action. In chapter 3, the properties of atomic hydrogen at low temperatures that are important for the application of the maser are reviewed. Chapter 4 contains a summary of the possible sources of frequency fluctuations and we discuss the potential stability that can be obtained with a cryogenic hydrogen maser. The description of the UBC prototype maser and its principle of operation can be found in chapter 5. Chapter 6 contains all the experimental results that have been obtained so far with this apparatus. They include frequency stability measurements, the measurement of the power output, the temperature dependence of the maser frequency, from which we could determine the binding energy of H on 4 He, and extensive measurements of phase noise of the maser electronics. In order to be able to interpret some of the results, a computer simulation has been developed that models the operation of the maser. Numerical results and the model on which this simulation is based are presented in chapter 7. In the last chapter, the performance of the present prototype maser is summarized. It also includes suggestions on how the performance of future devices can be further improved.

Some of the results of this work have been published previously. After the initial reports of the successful operation of the UBC maser [16], [18], we presented a longer

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paper at the 18th International Conference on Low Temperature Physics [19]. The results of the phase noise measurements on the cooled GaAs preamplifier have been reported in [20].

Chapter 2

Theory of Maser Operation

2.1 Hyperfine states of atomic hydrogen

The Hamiltonian, describing the spin degrees of freedom of a hydrogen atom in its electronic ground state in a static magnetic field B_0 is

$$\hat{H}_{0} = a \mathbf{i} \cdot \mathbf{s} - \hbar (-\gamma_{e} \mathbf{s} + \gamma_{p} \mathbf{i}) \cdot \mathbf{B}_{0}$$
(2.1)

The most recent measurement of the hyperfine frequency $\frac{a}{h}$ is [21]

$$\frac{a}{h} = 1420.405751773(1) \,\mathrm{MHz}$$

and the accepted values for the electronic and proton gyromagnetic ratios are [22]

$$\gamma_{\rm e} = 1.7608592(12) \times 10^{11} \,{\rm s}^{-1}{\rm T}^{-1}$$

 $\gamma_{\rm p} = 2.67522128(81) \times 10^8 \,{\rm s}^{-1}{\rm T}^{-1}$

In terms of the bases $|m_s m_i\rangle$, where m_s and m_i are the electron and proton spin projections along the applied field **B**₀, respectively, the eigenstates of (2.1) are in order of increasing energy:

$$|a\rangle = \cos\theta |-\frac{1}{2},\frac{1}{2}\rangle - \sin\theta |\frac{1}{2},-\frac{1}{2}\rangle$$
(2.2)

$$|b\rangle = |-\frac{1}{2}, -\frac{1}{2}\rangle$$
 (2.3)

$$|c\rangle = \sin\theta |-\frac{1}{2}, \frac{1}{2}\rangle + \cos\theta |\frac{1}{2}, -\frac{1}{2}\rangle$$
(2.4)

$$|d\rangle = |\frac{1}{2}, \frac{1}{2}\rangle, \qquad (2.5)$$

Chapter 2. Theory of Maser Operation

where

$$\tan(2\theta) = \frac{a}{\hbar(\gamma_{\rm e} + \gamma_{\rm p})B_0} = \frac{50.607\,{\rm mT}}{B_0}$$

The corresponding energies are

$$E_{a} = -\frac{a}{4} - \frac{a}{2} \left\{ 1 + \left[\frac{\hbar(\gamma_{e} + \gamma_{p})B_{0}}{a} \right]^{2} \right\}^{\frac{1}{2}}$$
(2.6)

$$E_b = \frac{a}{4} - \frac{\hbar}{2} (\gamma_e - \gamma_p) B_0 \qquad (2.7)$$

$$E_{c} = -\frac{a}{4} + \frac{a}{2} \left\{ 1 + \left[\frac{\hbar(\gamma_{e} + \gamma_{p})B_{0}}{a} \right]^{2} \right\}^{\frac{1}{2}}$$
(2.8)

$$E_{d} = \frac{a}{4} + \frac{\hbar}{2} (\gamma_{e} - \gamma_{p}) B_{0}$$
 (2.9)

Figure 2.1 shows the field dependence of the hyperfine energies.

At low densities, the natural line width of transitions between the upper and lower hyperfine states is extremely narrow. In free space the lifetime due to spontaneous emission is 3.75×10^{14} s. Experimentally, the line width is determined by other effects such as inhomogeneities in the magnetic field, the limited confinement time of the hydrogen atoms in the bulb or interactions with the walls. By choosing the transition between the $|c\rangle$ and $|a\rangle$ state in zero magnetic field to be the maser transition, the contribution to the line width due to magnetic field inhomogeneities can be greatly reduced because this transition frequency depends only in second order on the magnetic field.

In the remainder of this chapter, we first calculate the evolution of the magnetization under the influence of an applied rf field. We then make use of a result that relates the oscillating magnetization in a microwave cavity to the induced magnetic rf field in order to take the process of stimulated emission self-consistently into account. We derive results for the population inversion in the cavity, the frequency pulling effect due to cavity mistuning and expressions for the maser output power.

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Figure 2.1: Hyperfine energies as a function of magnetic field

2.2 Interaction with rf magnetic field

The interaction of atomic hydrogen with an rf field applied along the static magnetic field is governed by the following Hamiltonian:

$$\hat{H} = \hat{H}_0 - \hbar (-\gamma_e \hat{s}_z + \gamma_p \hat{\imath}_z) \mu_0 H_z \cos \omega t \qquad (2.10)$$

Note that the electromagnetic field is treated classically and as a consequence, we will not be able to calculate effects due to spontaneous emission with this theory.

An rf field along the z-axis can only induce transitions between the $|a\rangle$ and $|c\rangle$ state (see Appendix A). We are effectively dealing with a two-level system and it is convenient to rewrite the Hamiltonian (2.10), confined to the 2 dimensional subspace of the $|a\rangle$ and $|c\rangle$ states, as the Hamiltonian of a fictitious spin $\frac{1}{2}$ particle. All the quantities within the frame of this fictitious spin $\frac{1}{2}$ particle will be labeled with a "~". The density matrix is written as an expansion of Pauli matrices σ

$$\tilde{\rho} = \frac{1}{2} \mathbf{1}^{(2)} + \langle \tilde{\mathbf{s}} \rangle \cdot \boldsymbol{\sigma}$$
(2.11)

and the Hamiltonian (2.10) is expressed as

$$\hat{H} = \frac{1}{2}\tilde{E}_0 - \hbar\tilde{\gamma}(\tilde{\mathbf{B}}\cdot\tilde{\mathbf{s}})$$
(2.12)

If we choose the effective gyromagnetic ratio $\tilde{\gamma}$ to be

$$\tilde{\gamma} = \gamma_{\rm e} + \gamma_{\rm p} \tag{2.13}$$

then it is easy to show that in zero applied field, the effective magnetic field \mathbf{B} is given by

$$\tilde{B}_x = \mu_0 H_z \cos \omega t \tag{2.14}$$

$$\tilde{B}_y = 0 \tag{2.15}$$

$$\tilde{B}_{z} = \frac{a}{\hbar(\gamma_{e} + \gamma_{p})} = 50.607 \,\mathrm{mT}.$$
 (2.16)

The expectation values for the components of the effective magnetic moment $\tilde{\mu} = \hbar \tilde{\gamma} \tilde{s}$ are related to the elements of the density matrix by

$$\langle \tilde{\mu}_x \rangle = \frac{1}{2} \hbar (\gamma_e + \gamma_p) (\rho_{ac} + \rho_{ca})$$
 (2.17)

$$\langle \tilde{\mu}_{y} \rangle = -\frac{i}{2} \hbar (\gamma_{e} + \gamma_{p}) (\rho_{ac} - \rho_{ca})$$
(2.18)

$$\langle \tilde{\mu}_z \rangle = \frac{1}{2} \hbar (\gamma_e + \gamma_p) (\rho_{aa} - \rho_{cc})$$
 (2.19)

Note that both the fictitious longitudinal magnetization and the fictitious transverse magnetization are in real space magnetizations along the z-axis (see Appendix A). With the concept of the fictitious magnetization we can map our problem onto the standard NMR problem and we arrive at the same simple equation of motion for $\langle \tilde{\mu} \rangle$:

$$\frac{d}{dt}\langle \tilde{\boldsymbol{\mu}} \rangle = \tilde{\gamma} \langle \tilde{\boldsymbol{\mu}} \rangle \times \tilde{\mathbf{B}}$$
(2.20)

2.3 Relaxation, Bloch equation

In the case of the hydrogen maser, the hydrogen atoms are confined in a bulb for an average duration of T_b , the bulb holding time. In the bulb the atoms collide with each other and with the walls. All these processes affect the evolution of the magnetic moment. In general, it is difficult to calculate the effects for all these mechanisms from first principles. In many cases it has been found that all the relaxation processes can be characterized by only two phenomenological relaxation times, T_1 and T_2 , and that the magnetization $\tilde{M} = n_H \langle \tilde{\mu} \rangle$ obeys the so-called Bloch equations (n_H is the number density of hydrogen atoms):

$$\frac{d}{dt}\tilde{\mathbf{M}}_{\perp} = \tilde{\gamma}(\tilde{\mathbf{M}} \times \tilde{\mathbf{B}})_{\perp} - \frac{\tilde{\mathbf{M}}_{\perp}}{T_2}$$
(2.21)

$$\frac{d}{dt}\tilde{M}_{z} = \tilde{\gamma}(\tilde{\mathbf{M}} \times \tilde{\mathbf{B}})_{z} - \frac{\tilde{M}_{z} - \tilde{M}_{0}}{T_{1}} + \tilde{J}_{s} \qquad (2.22)$$

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We have included a source term \tilde{J}_s , describing the rate and polarization of hydrogen atoms entering the bulb from the state selector. \tilde{M}_0 is the thermal equilibrium value of \tilde{M}_z and is given by

$$\tilde{M}_0 = \frac{1}{2}\hbar(\gamma_{\rm e} + \gamma_{\rm p})n_H \frac{1 - e^{-\frac{\hbar\omega_0}{kT}}}{1 + 3e^{-\frac{\hbar\omega_0}{kT}}} \approx \frac{1}{2}\hbar(\gamma_{\rm e} + \gamma_{\rm p})\frac{\hbar\omega_0}{4kT}n_H$$
(2.23)

It has to be emphasized that the Bloch equations are not always applicable. In the simple case where in addition to relaxation due to the finite bulb holding time there is cross relaxation present between the upper three states, more than one T_1 is required to describe the correct evolution of \tilde{M} . In chapter 7, a different approach is discussed where different processes including the nonlinear spin exchange relaxation can be treated exactly. For the rest of this chapter, we will assume that the Bloch equations (2.21) and (2.22) are valid.

We now proceed in the usual way and transform the Bloch equations to rotating coordinates [23]. This new coordinate frame is rotating around the direction of quantization (z-axis) with frequency ω . The linearly polarized rf magnetic field \tilde{B}_x is decomposed into two counter-rotating circularly polarized components. When ω is close to ω_0 , only one of these components is on resonance and will dominate the dynamics of the magnetization. The presence of the other component, off-resonant by 2ω , causes a slight shift in resonance frequency. To lowest order, this so-called Bloch-Siegert frequency shift is given by [23]:

$$\frac{\Delta\omega_{B.S.}}{\omega_0} = \left[\frac{\mu_0(\gamma_e + \gamma_p)H_z}{4\omega_0}\right]^2$$
(2.24)

In practical hydrogen masers, this shift is negligibly small and we can ignore the offresonant component of the rf field.

In the rotating frame, the magnetization and the effective magnetic field are then given by (see Fig. 2.2):

$$\tilde{\mathbf{M}} = \tilde{M}_z \hat{\mathbf{z}} + \tilde{M}_r (\sin \Delta \hat{\mathbf{j}} - \cos \Delta \hat{\mathbf{k}})$$
(2.25)

Chapter 2. Theory of Maser Operation

$$\tilde{\mathbf{H}}_{eff} = \frac{\omega_0 - \omega}{\mu_0 \tilde{\gamma}} \hat{\mathbf{z}} + \frac{1}{2} \tilde{H}_x \hat{\mathbf{j}}$$
(2.26)

where we have used the notation

$$\omega_0 = \tilde{\gamma} \tilde{B}_z = \frac{a}{\hbar}. \tag{2.27}$$

2.4 Stimulated emission

We have to take into account that in our case of interest, \tilde{H}_x is not an applied field that is controlled by the experimenter. The rf field is generated by the oscillating magnetization in the microwave cavity and it then acts back on the magnetization. This whole process is called stimulated emission of radiation. The Bloch equations (2.21) and (2.22) describe how the rf field acts back on the magnetization. In appendix B the rf field generated by an oscillating magnetization is calculated. This purely electrodynamic relationship can be easily applied to the quantities \tilde{H}_x and \tilde{M}_r in the fictitious spin system. The angle Δ (see Fig. 2.2) is determined by the detuning of the cavity and is given by (B.10):

$$\Delta = Q_l(\frac{\omega_c}{\omega} - \frac{\omega}{\omega_c}) \approx \frac{2Q_l}{\omega}(\omega_c - \omega), \qquad (2.28)$$

where Q_l is the quality factor of the loaded cavity and ω_c is its resonance frequency. The ratio of the magnitudes of the rf field to the magnetization is to second order in Δ :

$$\frac{\tilde{H}_x}{\tilde{M}_r} = \eta Q_l \left(1 - \frac{\Delta^2}{2} \right). \tag{2.29}$$

In practical devices, the rf magnetic field is not uniform over the whole bulb of the maser. The relations derived in appendix B can still be applied if we interpret \tilde{H}_x as the average z-component of the rf field (in real space) over the maser bulb. The





rf magnetization is essentially uniform inside the maser bulb due to the fast diffusion present. In this case, the filling factor η is [6, 24]

$$\eta = \frac{\left[\int_{bulb} H_z(\mathbf{r}) \, d^3 r\right]^2}{V_b \int_{cavity} H^2(\mathbf{r}) \, d^3 r}$$
(2.30)

where V_b is the volume of the bulb.

We now solve the transverse Bloch equation (2.21) in the rotating frame using the self consistent field (2.29). With (2.25), (2.26) and (2.29) we obtain to second order in Δ :

$$-\frac{1}{\tilde{M}_{r}}\frac{d\tilde{M}_{r}}{dt} = \frac{1}{2}\tilde{\gamma}\mu_{0}\eta Q_{l}\left(1-\Delta^{2}\right)\tilde{M}_{z} + \frac{1}{T_{2}}$$
(2.31)

$$\omega - \omega_0 = -\frac{1}{2} \tilde{\gamma} \mu_0 \eta Q_l \Delta \tilde{M}_z \qquad (2.32)$$

These are just the equations derived by S. Bloom [25] for radiation damping. In the case of stable maser oscillation, \tilde{M}_r has to be constant in time and from (2.31) and (2.19) we obtain the population inversion inside the maser bulb

$$n_H(\rho_{aa} - \rho_{cc}) = -\frac{4(1 + \Delta^2)}{\hbar(\gamma_e + \gamma_p)^2 \mu_0 \eta Q_l} \frac{1}{T_2}$$
(2.33)

From (2.32) and (2.33) then follows immediately

$$\omega - \omega_0 = \frac{\Delta}{T_2} \tag{2.34}$$

Equation (2.34) describes the pulling of the maser frequency by the cavity. As we will discuss in the next chapter, ω_0 is not exactly given by (2.27); the resonance frequency is slightly affected by the wall shift, buffer gas shift and spin exchange shifts. If we define an atomic quality factor by

$$Q_{atom} = \frac{1}{2}\omega_0 T_2, \qquad (2.35)$$

then equation (2.34) can be rewritten approximately as

$$\omega - \omega_0 \approx \frac{Q_l}{Q_{atom}} (\omega_c - \omega_0).$$
 (2.36)

Note that in the derivation of the population inversion and the frequency shift due to cavity pulling, we have used only the Bloch equation for the transverse magnetization and we have not relied on the existence of a well defined longitudinal relaxation time T_1 .

2.5 Power emitted by atoms

In order to calculate the power emitted by the atoms, we first determine the rf magnetic field in the cavity. For stable oscillations \tilde{M}_z has to be constant and with (2.22) and (2.29) we obtain

$$\tilde{H}_x^2 = \frac{2\eta Q_l}{\mu_0 \tilde{\gamma}} \left[-\tilde{J}_s + \frac{\tilde{M}_z - \tilde{M}_0}{T_1} \right]$$
(2.37)

The thermal equilibrium magnetization \tilde{M}_0 is given by expression (2.23) and the steady state value of \tilde{M}_z is determined by (2.19) and (2.33). In steady state, the number of atoms entering the maser bulb equals the number of atoms leaving it. We can thus write the source term \tilde{J}_s as

$$\tilde{J}_s = \frac{1}{2}\hbar\tilde{\gamma}\frac{n_H}{T_b}(\rho_{aa} - \rho_{cc})_{s.s}$$
(2.38)

where T_b is the bulb holding time and $(\rho_{aa} - \rho_{cc})_{s.s}$ is the population polarization of atoms entering the bulb from the state selector. The electromagnetic energy stored in the cavity can be expressed in terms of rf magnetic field and the filling factor by

$$W = \frac{\mu_0 V_b}{2\eta} \tilde{H}_x^2$$
 (2.39)

The power P radiated by the atoms equals the sum of the power dissipated in the cavity and coupled to the output electronics and it is related to the stored energy W by

$$P = \frac{\omega_0 W}{Q_l} \tag{2.40}$$

With equations (2.37) to (2.40) we finally arrive at an expression for the power in terms of hydrogen density and the state selector efficiency:

$$P = \frac{1}{2} \hbar \omega_0 V_b \left[\frac{(\rho_{cc} - \rho_{aa})_{s.s}}{T_b} n_H - \frac{\hbar \omega_0}{4kT} \frac{1}{T_1} n_H - \frac{4(1 + \Delta^2)}{\hbar (\gamma_e + \gamma_p)^2 \mu_0 \eta Q_l T_1 T_2} \right]$$
(2.41)

In order to study the dependence of the power on density, we have to make some assumptions on the relaxation processes. We make here the simple assumption that the only relevant relaxation processes are due to atoms leaving the bulb and spin exchange relaxation. In addition, we assume that the spin exchange relaxation rates are proportional to the total density $n_{\rm H}$ and that the ratio of the transverse to the longitudinal spin exchange relaxation times equals 2 [26]. This is a high temperature approximation and not quite accurate at the operating temperature of a cryogenic hydrogen maser, but qualitatively it is still correct and it is instructive to study this case. Numerical results with the exact treatment of spin exchange relaxation at low temperature are presented in chapter 7.

It is convenient to define the following two densities:

$$n_0 = \frac{4(1+\Delta^2)}{\mu_0 \hbar (\gamma_e + \gamma_p)^2 \eta Q_l T_b}$$
(2.42)

$$n_{se} = \frac{1}{\sigma_{se} \langle v \rangle T_b} \tag{2.43}$$

where $\langle v \rangle = \left(\frac{16kT}{\pi m_H}\right)^{\frac{1}{2}}$ is the thermally averaged collision velocity for hydrogen and σ_{se} is the thermally averaged spin exchange cross section. [In the notation used by Verhaar [27] $\sigma_{se} = \bar{\sigma}_0(\rho_{cc} - \rho_{aa}) + \bar{\sigma}_1(\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2$]. The relaxation rates can then be written as

$$\frac{1}{T_1} = \frac{1}{T_b} \left(1 + 2\frac{n_H}{n_{se}} \right)$$
(2.44)

$$\frac{1}{T_2} = \frac{1}{T_b} \left(1 + \frac{n_H}{n_{se}} \right) \tag{2.45}$$

From (2.45) the physical interpretation of n_{se} becomes obvious. At the density n_{se} , the contribution to the line width due to spin exchange equals the contribution due to

atoms leaving the bulb. The physical interpretation of n_0 is related to the steady state population inversion (2.33), which can now be written as

$$n_H(
ho_{cc}-
ho_{aa}) = n_0\left(1+rac{n_H}{n_{se}}
ight)$$

The power emitted by the atoms becomes

$$P = \frac{1}{2}\hbar\omega_0 \frac{V_b}{T_b} \left\{ -n_0 + \left[(\rho_{cc} - \rho_{aa})_{s.s} - \frac{\hbar\omega_0}{4kT} - 3\frac{n_0}{n_{se}} \right] n_H - \frac{2}{n_{se}} \left[\frac{\hbar\omega_0}{4kT} + \frac{n_0}{n_{se}} \right] n_H^2 \right\}$$
(2.46)

If we ignore any possible density dependence of $(\rho_{cc} - \rho_{aa})_{ss}$, the power is then a quadratic function of density. There is a lower threshold density n_l , below which no stable maser oscillations are possible. This threshold density is approximately given by

$$n_l \approx \frac{n_0}{(\rho_{cc} - \rho_{aa})_{s.s} - \frac{\hbar\omega_0}{4kT} - 3\frac{n_0}{n_{se}}}$$
(2.47)

Spin exchange relaxation also causes the existence of an upper critical density n_u . Above n_u the spin exchange relaxation is too fast and the atomic line width is too broad to sustain maser action.

$$n_u \approx \frac{(\rho_{cc} - \rho_{aa})_{s.s} - \frac{\hbar\omega_0}{4kT} - 3\frac{n_0}{n_{se}}}{2\left(\frac{\hbar\omega_0}{4kT} + \frac{n_0}{n_{se}}\right)} n_{se} \qquad (2.48)$$

The expressions derived here are similar to those given by Kleppner et al. [6] and by Berlinsky and Hardy [13], except that in the references mentioned, \tilde{M}_0 was assumed to be zero. At low temperatures $\frac{\hbar\omega_0}{4kT}$ is usually larger than $\frac{n_0}{n_{se}}$. It is thus important to include this term, otherwise it leads to an overestimate of the upper threshold density n_u . The additional factors of $\frac{V_b}{V_{cavity}}$ in [6, 13] stem from a different definition of the filling factor η .

Chapter 3

Properties of atomic hydrogen at low temperatures

3.1 Interactions with the walls

When a conventional hydrogen maser with a Teflon coated bulb is cooled to low temperatures, the device stops oscillating below about 50K [28]. The processes responsible for this failure are interactions between the hydrogen atoms and the walls. At room temperature, the precessing hydrogen atoms are almost unaffected by collisions with the Teflon coated walls. Historically, the discovery of such a wall coating was a crucial step in the development of the first hydrogen maser [4].

On closer inspection, the interactions between hydrogen atoms and the walls shift the maser frequency by a small amount. This wall shift is typically -20 mHz for a conventional full size maser at room temperature. The imperfect reproducibility of the Teflon coating and the associated uncertainty of the wall shift limits the accuracy of conventional hydrogen masers to about 1×10^{-12} .

We use a simple model to analyze the shift and broadening of the atomic resonance due to interactions between hydrogen atoms and walls. Attractive van der Waals forces between the substrate and the hydrogen atoms lead to a bound state with binding energy E_B . The hyperfine frequency of the weakly polarized adsorbed atoms is shifted from the free space value by ω_s . This frequency shift ω_s is expected to be slightly anisotropic with respect to the angle between the surface and the direction of quantization [29], but here we ignore this complication. We further assume that the adsorbed atoms move freely on the surface and form a two-dimensional gas which is in thermodynamic equilibrium with the three-dimensional gas. We can equate the chemical potentials to obtain the relationship between the number density of adsorbed atoms n_{σ} and the bulk density n_{H} . For low densities, where the interatomic interactions are small, this relationship is given by

$$n_{\sigma} = n_{H} \Lambda \exp\left\{\frac{E_{B}}{kT}\right\}$$
(3.1)

where $\Lambda = \left(\frac{2\pi\hbar^2}{m_H kT}\right)^{\frac{1}{2}} = 1.74 \text{ nm}\left(\frac{1\text{ K}}{\text{T}}\right)^{\frac{1}{2}}$ is the thermal de Broglie wavelength.

The ratio of the average duration of sticking $\langle \tau_s \rangle$ to the average time between sticking events $\langle \tau_B \rangle$ is given by the ratio of the total number of atoms in the bound state to the number of atoms in the 3 dimensional gas. In a bulb with surface area A_b and volume V_b we get

$$\frac{\langle \tau_s \rangle}{\langle \tau_B \rangle} = \frac{A_b n_\sigma}{V_b n_H} = \frac{A_b}{V_b} \Lambda \exp\left\{\frac{E_B}{kT}\right\}$$
(3.2)

In practical cases, this is a small number. Since $\langle \tau_B \rangle$ is proportional to the inverse of the average velocity of a hydrogen atom and therefore proportional to $T^{-\frac{1}{2}}$, the average phase shift per sticking event $\phi_0 = \omega_s \langle \tau_s \rangle$ depends on temperature as $\phi_0 \propto T^{-1} \exp\left\{\frac{E_B}{kT}\right\}$. With the simple assumption that both τ_s and τ_B are distributed according to a Poisson statistics, it is possible to derive the frequency shift and line width of the atomic resonance in the absence of other mechanisms. The results are [23]:

$$\delta\omega_w = \frac{1}{\langle\tau_B\rangle} \frac{\phi_0}{1+\phi_0} \tag{3.3}$$

$$\Gamma_w = \frac{1}{\langle \tau_B \rangle} \frac{\phi_0^2}{1 + \phi_0^2}$$
(3.4)

At high enough temperatures where $\phi_0 \ll 1$, the line broadening (3.4) is negligible and there is only a small frequency shift linear in ϕ_0 :

$$\delta\omega_w \approx \frac{\phi_0}{\langle \tau_B \rangle} = \omega_s \frac{A_b}{V_b} \Lambda \exp\left\{\frac{E_B}{kT}\right\}$$
 (3.5)

At lower temperatures when $\phi_0 \sim 1$, the process of motional narrowing is less effective and the resulting large line broadening can prevent a maser from oscillating. More detailed models for the line shape were discussed by Crampton et al. [30] and by Morrow and Hardy [31] who include the case of a sticking coefficient smaller than 1 and non-Poisson statistics for τ_B .

In order to design a successful low temperature H maser, it becomes obvious from the above simple model that a wall coating with a very small binding energy has to be used. The best candidates are weakly polarizable, low density materials, and table 3.1 lists measured binding energies in units of temperature for a few substrates chosen with these qualities in mind.

Substrate	Binding Energy	Reference
Teflon	140 K	[28, 32]
solid H ₂	.39.8 K	[30]
solid Ne	34 K	[33]
liquid He:		
⁴ He	$1.0\mathrm{K}$	[19]
³ He	0.4 K	[34]

Table 3.1: Binding energies E_B/k of atomic hydrogen to different substrates in units of temperature.

The value for E_B of Teflon was obtained by analyzing the published maser data for ϕ_0 in [28] and [32] according to our simple model. The binding energies of H on H₂, Ne and on liquid He were first determined by pulsed magnetic resonance on the hyperfine transition. Crampton et al. [10] successfully operated a hydrogen maser with neon coated walls at temperatures between 9 K and 10.5 K.

The liquid surfaces of the helium isotopes ³He and ⁴He have by far the lowest binding energies and are the only possible choices for a cryogenic hydrogen maser operated at temperatures below 1 K. They are also very attractive because they are effectively self cleaning: all neutral atoms or molecules with the exception of atomic hydrogen penetrate the helium film [35] and freeze out on the underlying substrate. In addition, at the operating temperature of our cryogenic maser, ⁴He films are superfluid. A consequence of the high mobility is that thinning of the Van der Walls coating is greatly suppressed and very reproducible wall coatings can be achieved. This is probably one of the main reasons why experimental efforts so far have concentrated on ⁴He coated walls, although ³He offers a significantly lower binding energy.

For the design of a low temperature maser with He coated walls, a detailed knowledge of the properties of H and the interactions between H and He at these temperatures is required. In the past decade, a wealth of information has been obtained by several groups who have studied spin-polarized hydrogen at low temperatures in their attempts to observe the predicted Bose-Einstein condensation. Extensive reviews of these experiments were written by Silvera and Walraven [36] and by Greytak and Kleppner [37]. In this chapter we will concentrate on the properties relevant to the operation of a cryogenic H maser. The most important results in this regard were obtained by magnetic resonance experiments at the hyperfine transition in zero field by the UBC group in the years between 1979 and 1982. Reviews of these experiments can be found in [12] and in the Ph.D. thesis of M. Morrow [38].

3.2 Recombination

Hydrogen atoms can only recombine if they interact via the electronic singlet potential. At low temperatures the radiative recombination is completely negligible [36] and therefore, two colliding hydrogen atoms can only recombine in the presence of a third body, otherwise momentum and energy cannot be conserved. At the low hydrogen densities used in a maser, the role of the third body is played predominately by helium atoms in the vapor phase at the higher temperatures and by the walls at the lower temperatures.

The number density of helium atoms in the vapor phase over a saturated film is strongly temperature dependent and in the case of ⁴He can be approximated by [39]

$$n_{4_{\rm He}}(T) = 1.505 \times 10^{21} \,{\rm cm}^{-3} \left(\frac{{\rm T}}{1\,{\rm K}}\right)^{\frac{3}{2}} \exp\left\{-\frac{7.16888\,{\rm K}}{{\rm T}}\right\}$$
 (3.6)

At T = 0.7 K, the ⁴He density is 3×10^{16} cm⁻³, which is orders of magnitude higher than typical hydrogen densities. At temperatures above 0.7 K, the recombination of hydrogen is thus mainly assisted by helium atoms and the decay of hydrogen follows a second order rate equation, where the rate constant is proportional to n_{He} .

As the temperature is lowered, the surface density n_{σ} increases and the recombination of adsorbed atoms begins to dominate. In this regime, the recombination process is described by

$$\left. \frac{d}{dt} n_{\sigma} \right|_{rec} = -\lambda v_s {n_{\sigma}}^2 \tag{3.7}$$

where λ is a recombination cross length and $v_s = \left(\frac{\pi kT}{m_H}\right)^{\frac{1}{2}} = 1.609 \times 10^4 \frac{\text{cm}}{\text{s}} \left(\frac{T}{1\text{K}}\right)^{\frac{1}{2}}$ is the average relative velocity of the adsorbed hydrogen atoms. Since the fraction of atoms on the surface is small and the surface density n_{σ} is proportional to the bulk density n_H , we can rewrite equation (3.7) as an equation for n_H with an effective recombination rate constant. Including the helium assisted recombination process, we get:

$$\left. \frac{d}{dt} n_H \right|_{rec} = -K_{eff} n_H^2 \tag{3.8}$$

with

$$K_{eff} = kn_{He} + \lambda v_s \frac{A_b}{V_b} \Lambda^2 \exp\left\{\frac{2E_B}{kT}\right\}$$
(3.9)

The recombination rate for a sample in thermal equilibrium and in zero field was measured by Hardy and collaborators and they found that the results could be described accurately by equations (3.8) and (3.9). For walls coated with ⁴He and ³He, respectively, they
obtained:

 $k(^{4}\text{He}) = 2.8 \pm 0.3 \times 10^{-33} \text{ cm}^{6}\text{s}^{-1}$ [40] $\lambda(^{4}\text{He}) = 0.21 \pm 0.03 \text{ Å}$ [41]

 $k(^{3}\text{He}) = 1.2 \times 10^{-33} \text{ cm}^{6}\text{s}^{-1}$ [42] $\lambda(^{3}\text{He}) = 0.19 \pm 0.03 \text{ Å}$ [34] (Note that in [41] and [34] a different expression for v_{s} was used). The recombination rate goes through a minimum which, for their value of $\frac{A_{b}}{V_{b}} = 4 \text{ cm}^{-1}$, occurs at $T \approx 0.65 \text{ K}$ with ⁴He surfaces and at $T \approx 0.25 \text{ K}$ with ³He surfaces. At these temperatures, the recombination process is remarkably slow. As an illustration, a sample of atomic hydrogen with a typical density for maser operation $n_{H} = 10^{11} \text{ cm}^{-3}$ confined by ⁴He coated walls has a lifetime $\tau = (K_{eff}n_{H})^{-1} = 4 \times 10^{4} \text{ s}$ at T = 0.65 K and $\frac{A_{b}}{V_{b}} = 4 \text{ cm}^{-1}$. In a high performance maser, the area-to-volume ratio $\frac{A_{b}}{V_{b}}$ is likely to be smaller by up to a factor of ten, thus further reducing the recombination rate at the lower temperatures. In the absence of other mechanisms which remove hydrogen atoms, only a very small flux of new atoms is required to keep the density in a maser experiment constant.

3.3 Hyperfine frequency shift

The temperature dependence of the hyperfine frequency shift below 1 K was first measured by Morrow et al. [41] for ⁴He surfaces and by Jochemsen et al. [34] for ³He surfaces. Just as for recombination, they found that the high temperature behavior can be explained by interactions between hydrogen and helium atoms in the vapor phase and the low temperature behavior is determined by the surface bound state.

The hyperfine frequency is proportional to the density of electron spin at the site of the proton. Therefore, interatomic collisions between hydrogen and helium atoms cause a small frequency shift. This 'buffer gas' shift is proportional to the helium density in the vapor phase and was analyzed theoretically by Jochemsen and Berlinsky [43]. They calculated the frequency shifts for several proposed H-He potentials and their results are in fair agreement with experiment. At lower temperatures, the fraction of time that atoms spend in the bound state on the surface increases and this leads to the dominant frequency shift. It was found that the simple model discussed in the first section of this chapter describes the situation accurately as long as $\phi_0 \ll 1$.

The total temperature dependent frequency shift then follows the expression:

$$\omega - \omega_0 = 2\pi \delta n_{He} + \omega_s \frac{A_b}{V_b} \Lambda \exp\left\{\frac{E_B}{kT}\right\}$$
(3.10)

The buffer gas shift coefficient δ was measured to be $\delta(^{4}\text{He}) = -1.183 \times 10^{-17} \text{ Hz cm}^{3}$ [40] and $\delta(^{3}\text{He}) = -9 \pm 1 \times 10^{-18} \text{ Hz cm}^{3}$ [12], and the surface frequency shift $f_{s} = \frac{\omega_{s}}{2\pi}$ was found to be $f_{s}(^{4}\text{He}) = -49 \pm 2 \text{ kHz}$ [41] and $f_{s}(^{3}\text{He}) = -23 \pm 2 \text{ kHz}$ [34]. It is important to note that δ and f_{s} have the same sign (negative). This was a key result for the application of the cryogenic H maser, because it implies that the frequency shift has an extremum where the temperature sensitivity of the maser frequency is minimized. A cryogenic hydrogen maser used as a frequency stable source has to be operated close to this frequency maximum, otherwise the requirements on temperature stabilization are unrealistic. The frequency extremum occurs at approximately 0.55 K with ⁴He walls and at 0.2 K with ³He walls and depends weakly on the area to volume ratio of the bulb. For a bulb with the same size as in a conventional maser (15 cm diameter), the frequency shifts at the optimal temperature are 50 mHz for ⁴He and 36 mHz for ³He walls, respectively. This is comparable to the wall shift at room temperature in Teflon coated bulbs.

We should also mention that at sufficiently low temperatures the sticking coefficient is expected to become exceedingly small [44]. At these low temperatures there is effectively no exchange between surface atoms and bulk atoms any more and the infrequent sticking events do not broaden the hyperfine line significantly. Thus it appears possible to design a maser that is not affected by wall shifts. In such a device, the increasing Kapitza resistance at low temperatures forces the density to be kept low, otherwise the recombination heating would be too high. At present, it is not known at what temperature the sticking

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coefficient becomes small enough for this experiment, but it is safe to predict that such a project will be experimentally very challenging.

3.4 Diffusion

At the operating temperature of the cryogenic H maser, the helium vapor density is high enough that it affects the transport properties of hydrogen. The diffusion constant Dof hydrogen in a ⁴He coated cell was measured by Hardy et al. [40] and they obtained an effective hard core diffusion cross section $Q_D = \frac{3\pi\bar{v}}{32n_{He}D} = 20 \pm 1$ Å² at $T \approx 1$ K (\bar{v} is the thermally averaged relative speed of a H-He pair). Jochemsen et al. [45] analyzed this problem theoretically and predicted a fairly strong temperature dependence of Q_D at temperatures below 1 K. However, the increase of the mean free path at low temperature, defined by $\lambda_D = \frac{1}{\sqrt{2}n_{He}Q_D}$, is mainly due to the temperature dependence of n_{He} . For the case of ⁴He surfaces at a temperature T = 0.5 K, Q_D is expected to be ≈ 12 Å² [45], resulting in $\lambda_D \approx 2$ cm, which is comparable to typical dimensions of our apparatus. By decreasing the temperature to 0.4 K, the mean free path increases to about 1 m and collisions with helium atoms do not appreciably impede the motion of the hydrogen atoms.

3.5 Spin exchange

So far we have only considered collisions between hydrogen and helium atoms, which cause a frequency shift without an appreciable line broadening. In comparison, collisions between two hydrogen atoms occur much less frequently at the operating temperature of a cryogenic hydrogen maser, but cause both a frequency shift and a line broadening. This line broadening limits the power output of a maser. The dominant process is electron spin exchange, effects due to dipolar interactions being much smaller. Chapter 3. Properties of atomic hydrogen at low temperatures

Т	0.5 K	300 K
$egin{aligned} &ar{\lambda}_0 \langle v angle \ &ar{\lambda}_1 \langle v angle \ &ar{\lambda}_2 \langle v angle \ &ar{\sigma}_0 \langle v angle \ &ar{\sigma}_1 \langle v angle \ &ar{\sigma}_2 \langle v angle \end{aligned}$	$\begin{array}{c} -1.72 \times 10^{-11} \\ -2.57 \times 10^{-14} \\ -1.67 \times 10^{-14} \\ 5.93 \times 10^{-14} \\ 7.59 \times 10^{-13} \\ 1.08 \times 10^{-15} \end{array}$	$\begin{array}{c} -3.2\times10^{-11}\\ 1.3\times10^{-13}\\ -2.4\times10^{-13}\\ -9.2\times10^{-14}\\ -2.1\times10^{-14}\\ 4.3\times10^{-10} \end{array}$

Table 3.2: Calculated values [46] of the spin exchange frequency shift parameters $\bar{\lambda}_i \langle v \rangle$ and broadening parameters $\bar{\sigma}_i \langle v \rangle$ in units of cm³/s.

The most extensive theoretical analysis of this problem has been recently completed by Prof. B. J. Verhaar and his collaborators [27, 46]. In their fully quantum mechanical calculation, they showed that the frequency shift $\delta \omega_c$ and the line broadening Γ_c due to hydrogen-hydrogen collisions are of the form:

$$\delta\omega_c = \left[\bar{\lambda}_0(\rho_{cc} - \rho_{aa}) + \bar{\lambda}_1(\rho_{cc} + \rho_{aa}) + \bar{\lambda}_2\right] \langle v \rangle n_H \tag{3.11}$$

$$\Gamma_c = \left[\bar{\sigma}_0(\rho_{cc} - \rho_{aa}) + \bar{\sigma}_1(\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2\right] \langle v \rangle n_H \tag{3.12}$$

where $\langle v \rangle = \left(\frac{16kT}{\pi m_H}\right)^{\frac{1}{2}} = 2.05 \times 10^4 \frac{\text{cm}}{\text{s}} \left(\frac{T}{1\text{K}}\right)^{\frac{1}{2}}$ is the thermally averaged relative velocity of the hydrogen atoms. The product of the thermal velocity and the calculated values of the spin exchange cross sections for the frequency shift $\bar{\lambda}_i \langle v \rangle$ and for the line broadening $\bar{\sigma}_i \langle v \rangle$ are listed in table 3.2 for T = 0.5 K and for room temperature [46].

For a given density, the magnitude of both the frequency shift and the line broadening are smaller at 0.5 K than at room temperature. This decrease is especially dramatic for the line width, where for equal densities, $\rho_{cc} + \rho_{aa} = 0.5$ and $\rho_{cc} - \rho_{aa} \approx 0$

$$\frac{\Gamma_c(T=0.5\,{\rm K})}{\Gamma_c(T=300\,{\rm K})} = 0.9 \times 10^{-3}$$

This implies that for the same line width Γ_c , a cryogenic maser, compared to a conventional maser, can be operated at a more than thousand times higher density. At the optimal line width for frequency stability and all other parameters being equal, it is shown in the next chapter that the power output of the cryogenic maser is also roughly 1000 times higher, which results in an increased signal-to-noise ratio and a much higher potential frequency stability for the cryogenic maser. This predicted improvement of frequency stability, which is a consequence of the anomalously small spin exchange cross section at low temperature, has been one of the main motivations for the development of cryogenic hydrogen masers [8].

The spin exchange frequency shift parameters $\bar{\lambda}_i \langle v \rangle$ do not decrease at low temperatures by the same amount as the parameters $\bar{\sigma}_i \langle v \rangle$. As the densities are increased for the low temperature operation, the magnitude of the spin exchange frequency shift becomes in fact larger than at room temperature. The first term in (3.11) does not pose any difficulty. The dynamics of the maser keeps the population inversion $n_H(\rho_{cc} - \rho_{aa})$ close to zero (see equation 2.33) and proportional to T_2^{-1} . This term can then be cancelled by proper mistuning of the cavity, which also results in a frequency shift proportional to T_2^{-1} (see equation 2.34). This is known as the 'spin exchange tuning', introduced by S. Crampton [47] and now routinely used in the operation of conventional masers.

Earlier calculations of the frequency shifts induced by spin exchange [26, 48] treated the problem in the degenerate-internal-states approximation which ignores hyperfine interactions relative to electron-exchange interactions during collisions. With these calculations only the first term in equation (3.11) was obtained. However, Verhaar et al. [27] showed that the inclusion of the hyperfine interaction during collisions gives rise to the troublesome second and third term in (3.11). These terms cannot be completely cancelled by cavity tuning, so that fluctuations in the density or in the relative populations cause directly a frequency fluctuation. These frequency fluctuations might limit the potential performance of a cryogenic maser significantly. This is discussed more quantitatively in the next chapter.

Prof. Verhaar and his collaborators also calculated the effect of spin exchange on the diagonal elements of the density matrix [49]. They found that in zero field, the following three relaxation processes are most efficient:

 $b + d \stackrel{G_{bd \rightarrow aa}}{\rightleftharpoons} a + a$ $c + c \stackrel{G_{cc \rightarrow bd}}{\rightleftharpoons} b + d$ $c + c \stackrel{G_{cc \rightarrow aa}}{\rightleftharpoons} a + a$

The backward rate constants are related to the forward rate constants by a Boltzmann factor:

$$G_{ij \to kl} = G_{kl \to ij} \exp\left\{\frac{E_i + E_j - E_k - E_l}{kT}\right\}$$
(3.13)

At higher temperatures, a longitudinal relaxation time constant $T_{1,se}$ for spin-exchange can be obtained from these rate constants. The rate equation for $n_H(\rho_{aa} - \rho_{cc}) \equiv n_a - n_c$ due to the three considered spin exchange processes is

$$\frac{d}{dt}(n_{a} - n_{c}) = -2 \left(2 G_{aa \to cc} + G_{aa \to bd} \right) n_{a}^{2} + 2 \left(2 G_{cc \to aa} + G_{cc \to bd} \right) n_{c}^{2} + 2 \left(G_{bd \to aa} - G_{bd \to cc} \right) n_{b} n_{d}$$
(3.14)

At higher temperatures, Stoof et al. [49] show that $G_{bd\to aa} \simeq G_{bd\to cc}$, in addition, the Boltzmann factors (3.13) can be replaced by unity, and close to equilibrium $n_a + n_c \simeq \frac{1}{2}n_H$. With these simplifications, the rate equation (3.14) becomes:

$$\frac{d}{dt}(n_a - n_c) \simeq -\left[\left(2G_{cc \to aa} + G_{bd \to aa}\right)n_H\right](n_a - n_c) \tag{3.15}$$

from which we identify $T_{1,se}^{-1}$

$$T_{1,se}^{-1} = (2G_{cc \to aa} + G_{bd \to aa}) n_H$$
(3.16)

Within the degenerate-internal-states approximation and at high temperatures where the Boltzmann factor is unimportant, the ratio of the longitudinal relaxation time constant $T_{1,se}$ to the transverse relaxation time constant $T_{2,se} \equiv \Gamma_c^{-1}$ for H-H collisions is simply [26]

$$\frac{T_{1,se}}{T_{2,se}} = \frac{1}{2} \tag{3.17}$$

Desaintfuscien et al. [32] verified this relationship with a H-maser operated between 77 K and 363 K. At low temperatures however, it is not possible to define a unique $T_{1,se}$, as can be seen from expression (3.14).

Morrow and Berlinsky [50] also considered spin exchange relaxation between adsorbed atoms on the helium surface. At the densities and temperatures relevant to the maser, the surface densities are so small that this mechanism is not important.

Chapter 4

Considerations on frequency stability

4.1 Characterization of frequency stability

The quality of frequency and time standards can be specified by two quantities: accuracy and stability. A clock of a certain type is accurate if its frequency is known exactly and reproducible from one device to the next. Atomic clocks are much more accurate than cavity oscillators and crystal oscillators, where the output frequency depends on some macroscopic length. At present, the most accurate established frequency standards are atomic cesium beam devices and have in fact been chosen to define the unit of second. The accuracy of the best Cs clocks, i.e. the degree to which their frequencies agree, is currently about 10^{-13} or better; this is about ten times better than the accuracy of stateof-the-art hydrogen masers. Cryogenic hydrogen masers are not expected to be much more accurate. The limiting wall shift at low temperature is comparable in magnitude to that for room temperature masers, and additional problems due to the spin exchange shifts may also play a role in determining the accuracy.

Stability is a measure of how constant the frequency of a single device is over a given averaging time. For averaging times τ between 1s and 10⁶s, conventional hydrogen masers are presently the most stable atomic clocks. Frequency fluctuations of less than 10^{-15} for $\tau = 1$ hour are measured with the best devices [1]. In this chapter we discuss the physical origin of these fluctuations and show that a low temperature operation of a hydrogen maser promises to result in substantially smaller frequency fluctuations and therefore improved stability.

The specification of frequency stability requires some care. Many stable frequency standards, such as high quality quartz crystal oscillators, exhibit a component of 1/fflicker frequency noise, in which case the classical variance of the fractional frequency averaged over a time τ diverges. For this reason, the so-called 'Allan variance' $\sigma_y^2(\tau)$ is used instead, which is easy to measure and it converges for all the major noise types observed in precision oscillators. In the frequency domain, the spectral density of phase fluctuations $S_{\varphi}(f)$ is most often used to characterize the frequency stability. The definitions of both quantities and their relationship is discussed in appendix C. In this thesis, all frequency stabilities mentioned should be interpreted as $\sigma_y(\tau)$ in the time domain or as $S_{\varphi}(f)$ in the frequency domain.

4.2 Intrinsic frequency instabilities

The dominant source of intrinsic frequency instabilities for a hydrogen maser operated at $T \approx 0.6$ K is the background of thermal photons that are present in the maser microwave cavity. The associated electromagnetic field is at random phase relative to the radiation produced by the maser process. In the high temperature limit $(T \gg \frac{\hbar\omega_0}{k} = 68.17 \text{ mK})$, the resulting frequency instability (square root of Allan variance) for $\tau \gg \frac{Q_{atom}}{\omega_0}$ is given by [51]:

$$\left. \frac{\delta f}{f} \right|_{int} = \left(\frac{kT}{2Q_{atom}^2 P \tau} \right)^{\frac{1}{2}} \tag{4.1}$$

The atomic quality factor Q_{atom} is related to the transverse relaxation time T_2 by $Q_{atom} = \frac{1}{2}\omega_0 T_2$. *P* is the power emitted by the atoms. In this expression, *T* is the temperature of the background photons which depends on the temperature of the maser cavity (and of the isolator termination at the output of the cavity, if present). In our apparatus, the hydrogen atoms are not in direct contact with the walls of the microwave

cavity. This permits, at least in principle, to reduce the intrinsic frequency instabilities by lowering the temperature of the cavity and isolator below the temperature of the hydrogen atoms and the helium film, which has to be kept at around 0.6 K in order to minimize the temperature dependence of the frequency shift. However, the intrinsic frequency instabilities cannot be decreased to arbitrary small values by this method. At lower temperatures ($T \approx \frac{\hbar\omega_0}{k}$), spontaneous emission starts to limit the intrinsic stability, and in this case, the thermal energy kT in equation (4.1) has to be replaced by the photon energy $\hbar\omega_0$ [52].

In order to gain some more physical insight, we can rewrite (4.1) approximately as

$$\left. \frac{\delta f}{f} \right|_{int} \approx \left. \frac{1}{Q_{atom}} \left(\frac{S}{N} \right)^{-1} \right. \tag{4.2}$$

where $\frac{S}{N}$ is the signal to noise ratio of the output amplitude (as opposed to the output power). At low temperatures, the noise term is obviously smaller, but at the same time the signal is larger due to the reduced spin exchange cross section. These two factors result in a large reduction of the intrinsic frequency instabilities. In order to discuss this more quantitatively, we will make use of the expressions for the power output P and the line width derived in chapter 2.

The power output P as well as the atomic quality factor Q_{atom} depend on the number density of hydrogen n_H . The intrinsic frequency stability can be optimized by operating the maser at the density n_{opt} which maximizes the product $Q_{atom}^2 P$. By using expressions (2.45) and (2.46), we obtain for n_{opt} :

$$n_{opt} = \frac{(\rho_{cc} - \rho_{aa})_{s.s} - \frac{\hbar\omega_0}{4kT} - \frac{n_0}{n_{se}}}{(\rho_{cc} - \rho_{aa})_{s.s} + 3\frac{\hbar\omega_0}{4kT} + \frac{n_0}{n_{se}}} n_{se} \approx n_{se}$$
(4.3)

In order to simply the expressions, we made in the last step the assumptions that $\frac{n_0}{n_{se}} \ll (\rho_{cc} - \rho_{aa})_{s.s}$ and that $\frac{\hbar\omega_0}{4kT} \ll (\rho_{cc} - \rho_{aa})_{s.s}$. At the operating temperature of a cryogenic hydrogen maser $(T \approx 0.6 \text{ K})$, this is usually fulfilled since $\frac{n_0}{n_{se}} \simeq \frac{0.37}{\eta Q_l}$ and

Chapter 4. Considerations on frequency stability

 $\frac{\hbar\omega_0}{4kT} = 2.84 \times 10^{-2}$. With the same simplifications, the power at n_{opt} becomes

$$P_{opt} \approx \frac{1}{2} \hbar \omega_0 \frac{V_b}{T_b} (\rho_{cc} - \rho_{aa})_{s.s} n_{se}$$

$$\tag{4.4}$$

and we arrive at

$$\frac{\delta f}{f}\Big|_{int} \simeq \left(\frac{16kT\sigma_{se}\langle v\rangle}{\hbar\omega_0^3 V_b(\rho_{cc}-\rho_{aa})_{s.s}}\right)^{\frac{1}{2}} \tau^{-\frac{1}{2}}$$
(4.5)

4.3 Electronic Noise

In all hydrogen masers it is found that the frequency fluctuations for short averaging times τ are much larger than those predicted by (4.1). This is a result of the fact that the hydrogen maser is intrinsically a low power device, and the microwave signal has to be first amplified before it can be further processed. This amplification process adds noise to the maser signal, which in fact dominates the short term stability.

For most state-of-the-art low noise microwave amplifiers, the dominant noise at the frequency and power levels of interest is white phase noise. Its magnitude can be characterized by the noise temperature T_N of the amplifier, so that the spectral density of phase fluctuations is given by

$$S_{\varphi}(f) = \frac{kT_N}{P_{amp}} \tag{4.6}$$

where P_{amp} is the power level at the input of the amplifier. In terms of the coupling coefficient β introduced in appendix B (see B.5), P_{amp} for the first amplifier is related to the power emitted by the atoms P by

$$P_{amp} = \frac{\beta}{1+\beta}P \tag{4.7}$$

The power input at subsequent amplification stages is increased by the gain of the preceding stages, so that in practice, only the white phase noise of the first stage contributes significantly to the total phase noise. Following Vessot et al. [1], the electronic noise contribution to the fractional frequency fluctuations (square root of Allan variance) is given by

$$\left. \frac{\delta f}{f} \right|_{elec} = \left(\frac{BkT_N}{P_{amp}} \right)^{\frac{1}{2}} \frac{1}{\omega_0 \tau}$$
(4.8)

where B is the noise bandwidth of the receiver system. In the derivation of (4.8), two assumptions were made: It was assumed that the Allan variance is measured with some finite dead time and therefore equation (C.12) instead of the expression in table C.1 was used for the translation from $S_{\varphi}(f)$ to $\sigma_y^2(\tau)$. In the absence of dead time, equation (4.8) has to be multiplied by $\sqrt{\frac{3}{2}}$. In addition, it was assumed that a heterodyne receiver system is used with a simple double balanced mixer. The resulting signal at the intermediate frequency f_{if} ($\ll f_0$) thus contains amplifier noise that originates not only from a bandwidth B around f_0 , but also from a bandwidth B around $f_0 \pm 2f_{if}$ (+sign if reference frequency is above f_0 , and vice versa). The effective noise bandwidth f_n in (C.12) is therefore 2B. In the case that an image rejection mixer is used instead of a simple double balanced mixer, equation (4.8) has to be reduced by $\sqrt{2}$.

The expected frequency instabilities due to electronic noise of a maser, operated at the optimal density n_{opt} for lowest intrinsic frequency fluctuations, can be calculated with the same approximations as were made for equation (4.5). The result is:

$$\frac{\delta f}{f}\Big|_{elec} = \left(\frac{2\left(1+\beta\right)BkT_N\sigma_{se}\langle v\rangle T_b^2}{\beta\,\hbar\omega_0^3\,V_b\,(\rho_{cc}-\rho_{aa})_{s.s}}\right)^{\frac{1}{2}}\,\tau^{-1} \tag{4.9}$$

With a cryogenic hydrogen maser, it is natural to take advantage of the very low noise temperatures that GaAs FET amplifiers exhibit when they are cooled to low ambient temperatures. Commercial units are available with $T_N \leq 15$ K at 1.4 GHz and at an operating temperature of 77 K [53], with even further improvements at lower temperatures. Recently, noise temperatures as low as $T_N = 1.4$ K at 1.4 GHz have been reported for experimental HEMT devices at cryogenic temperatures [54]. Care has to be taken that no other noise starts to dominate the white phase noise of the amplifier. It is well known that GaAs FET's exhibit a higher level of 1/f noise than Si bipolar transistors. This can give rise to significant flicker phase noise. The spectral density of phase fluctuations for this noise contribution is of the form

$$S_{\varphi}(f) = h_{flicker} f^{-\gamma} \tag{4.10}$$

where typically $\gamma \simeq 1$ and $h_{flicker}$ is a property of the amplifier. It is shown in appendix C that the corresponding Allan variance $\sigma_y^2(\tau)$ has also approximately a τ^{-2} dependence as in the case of white phase noise. It is thus critical to choose amplifiers with small enough coefficients $h_{flicker}$ in order to be able to take advantage of the low noise temperatures. It is important to realize that this requirement applies to all stages of amplification, not just the first stage, because $h_{flicker}$ does not in general depend on the power level. The situation for the designer is complicated by the fact that specifications for flicker phase noise are usually not available from the manufacturer and have to be determined for all prospective amplifiers.

Vessot et al. [1] showed that the frequency stability of their state-of-the-art hydrogen masers, operated at room temperature, is limited by the intrinsic thermal noise (4.5) and by the electronic white phase noise (4.9) for averaging times between $1 \le \tau \le 1$ hour. If a cryogenic hydrogen maser is limited by the same noise processes, we can estimate the expected frequency fluctuations from expressions (4.5) and (4.9). As an illustration, we choose the same parameters as Berlinsky and Hardy [13] in their first detailed analysis of the expected stability of a full size, low temperature hydrogen maser: $V_b = 1.8 \times 10^3$ cm³, $(\rho_{cc} - \rho_{aa})_{s.s} = 0.5$, $\beta = 1$ (critical coupling), B = 6 Hz, $T_N = 10$ K and $T_b = 8$ s. The predicted stability is then

$$\frac{\delta f}{f}\Big|_{T=0.6\,\mathrm{K}} = \left[\left(2.7 \times 10^{-17} \tau^{-\frac{1}{2}} \right)^2 + \left(1.1 \times 10^{-15} \tau^{-1} \right)^2 \right]^{\frac{1}{2}} \tag{4.11}$$

Based on this analysis, it appears possible that a frequency stability as good as 1.4×10^{-18} can be obtained for an averaging time $\tau = 10^3$ s. This has to be compared to $\frac{\delta f}{f} \simeq 1 \times 10^{-15}$ ($\tau = 10^3$ s) for the best room temperature masers [1]. This potential of a thousandfold improvement in frequency stability at low temperatures was first pointed out by Berlinsky and Hardy in 1981 [13].

4.4 Frequency shifts

A frequency stability of 1.4×10^{-18} which is predicted for a cryogenic maser on the basis of intrinsic thermal noise and electronic noise at an averaging time $\tau = 10^3$ s, can only be achieved if all the frequency shifts are stable to better than 2×10^{-9} Hz. Here we study the requirements of stability that this poses on parameters controlling the frequency shifts in order to decide whether this predicted performance of a cryogenic hydrogen maser is realistic.

4.4.1 Magnetic field shifts

The main dependence of the maser frequency on the magnetic field is determined by the magnetic field dependence of the energy levels. To second order in B_0 , this is given by

$$f - f_0 = \frac{1}{2\pi} \frac{\hbar (\gamma_e + \gamma_p)^2}{2a} B_0^2 = 2.7731 \times 10^{11} \,\text{Hz} \left(\frac{B_0}{1 \,\text{T}}\right)^2$$
(4.12)

In order to remove the threefold degeneracy of the upper three hyperfine energy levels at zero field and to suppress cross relaxation among them, a small bias field B_0 has to be applied at the maser cavity. In a room temperature maser, this field is typically of the order of $1 \text{ mG} = 10^{-7} \text{ T}$. With this bias field, a stability of

$$\delta B_0 < 3.6 \times 10^{-14} \,\mathrm{T}$$

is required for $\frac{\delta f}{f} = 1.4 \times 10^{-18}$. At low temperatures, this level of magnetic field stability can be attained with the help of superconducting shields.

On the other hand, if one would like to use a cryogenic hydrogen maser as a sensitive detector for small changes in magnetic fields, it is best to operate the maser in a larger field $B_0 \gg 50.6$ mT or alternatively, to use the transition between the $|d\rangle$ and $|a\rangle$ levels in zero field as the maser transition. For the latter approach, the sensitivity is given by

$$\delta f = \frac{\gamma_{\rm e} - \gamma_{\rm p}}{4\pi} \, \delta B_0 = 1.399 \times 10^{10} \, \frac{\rm Hz}{\rm T} \, \delta B_0$$

This way, sensitivities much higher than those obtained with SQUIDS can be achieved at least in principle, provided that a high enough intrinsic frequency stability can be retained. The main experimental difficulty lies in the severe requirement on the homogeneity of the applied magnetic field. Without a high homogeneity, the atomic line width becomes too broad for maser action.

4.4.2 Buffer gas and wall shifts

The buffer gas and wall shifts were already discussed in chapter 3. They are given by

$$f - f_0 = \delta n_{He} + f_s \frac{A_b}{V_b} \Lambda \exp\left\{\frac{E_B}{kT}\right\}$$
(4.13)

This combined frequency shift has an extremum as a function of temperature which occurs for ⁴He coated walls at around $T_{ext} \simeq 0.55$ K. We are forced to operate the maser very close to this temperature T_{ext} in order to minimize the sensitivity to temperature fluctuations. When the maser is kept at T_{ext} , the temperature has to be kept stable to at least 10 μ K if a frequency stability of $\frac{\delta f}{f} = 1.4 \times 10^{-18}$ should be reached. This level of temperature stability is well within the capability of modern low temperature technology.

There are other mechanisms related to the helium buffer gas that set similar requirements on the temperature stability. The strong temperature dependence of the helium vapor density implies that temperature fluctuations cause fluctuations in the dielectric constant ε of the buffer gas and therefore fluctuations in the resonance frequency of the maser cavity. According to (2.36), the resulting fluctuations of the maser frequency are attenuated by $\frac{Q_l}{Q_{atom}}$. If we make the pessimistic but simplifying assumption that the helium buffer gas permeates the whole cavity, we can easily estimate the importance of this effect:

$$\frac{\delta f}{f}\Big|_{maser} = \left. \frac{Q_l}{Q_{atom}} \frac{\delta f}{f} \right|_{cavity} \simeq -\frac{1}{2} \frac{Q_l}{Q_{atom}} \delta \varepsilon \tag{4.14}$$

For ⁴He, the dependence of the dielectric constant on the number density n_{He} is accurately described by

$$\varepsilon - 1 = 2.60 \times 10^{-24} \,\mathrm{cm}^3 \,n_{He}(T)$$
 (4.15)

Together with (3.6) this allows us to relate a temperature fluctuation to a change of the maser frequency. For $\frac{Q_l}{Q_{atom}} = 10^{-5}$ and $T_{ext} = 0.55$ K, a temperature fluctuation of $\delta T = 3 \,\mu\text{K}$ gives rise to $\frac{\delta f}{f} = 1.4 \times 10^{-18}$. In actual devices, the effect will be considerably smaller, because the helium buffer gas is confined to the bulb in the center of the cavity where the electric fields are weak. The size of the effect can be determined with a perturbation calculation.

4.4.3 Cavity pulling and spin exchange shifts

As we have seen, frequency detuning of the maser cavity causes frequency shifts in the maser which are attenuated by $\frac{Q_i}{Q_{atom}}$:

$$f - f_0 = \frac{1}{2\pi} \frac{\Delta}{T_2} \simeq \frac{Q_l}{Q_{atom}} (f_c - f_0)$$

For a potential stability $\frac{\delta f}{f} = 1.4 \times 10^{-18}$ and a ratio $\frac{Q_l}{Q_{atom}} = 10^{-5}$, this implies that the resonance frequency of the microwave cavity has to be stable to at least 0.2 mHz. Thermal effects on the size of the cavity are not serious at low temperatures, because

the coefficients of thermal expansion become exceedingly small at temperatures below 1 K. However, great care is still required to assure that the tuning assembly, the output coupling and the possibly varying input impedance of the preamplifier do not cause excessive fluctuations in the resonance frequency f_c or the quality factor Q_l .

In the operation of conventional masers, the maser cavity is deliberately detuned in order to compensate for the spin exchange shifts to a high degree. According to (2.34) and (3.11), the combined shift is given by

$$f - f_0 = \frac{\Delta}{2\pi} \frac{1}{T_2} + \frac{1}{2\pi} \left[\bar{\lambda}_0 (\rho_{cc} - \rho_{aa}) + \bar{\lambda}_1 (\rho_{cc} + \rho_{aa}) + \bar{\lambda}_2 \right] \langle v \rangle n_H$$
(4.16)

It was shown in (2.33) that for stable maser oscillation, the population inversion $(\rho_{cc} - \rho_{aa})$ is proportional to T_2^{-1} :

$$(\rho_{cc} - \rho_{aa}) \langle v \rangle n_H = \alpha (1 + \Delta^2) \frac{1}{T_2}$$
(4.17)

where we have used the same notation as in [27] by defining α as

$$\alpha = \frac{4 \langle v \rangle}{\hbar (\gamma_{\rm e} + \gamma_{\rm p})^2 \mu_0 \eta Q_l}$$

In conventional masers with atomic hydrogen beams, it has proved difficult to keep the beam intensity and therefore the density and relaxation time T_2 in the maser bulb constant to a high degree. Prior to the calculations of Verhaar et al. [27, 46], it was thought that $\bar{\lambda}_1 = \bar{\lambda}_2 = 0$, ¹ in which case the so-called 'spin exchange tuning' [47] of the maser cavity

$$\Delta_{se} = -\alpha \bar{\lambda}_0 \tag{4.18}$$

eliminates the possible deterioration of the frequency stability due to beam fluctuations. However, Verhaar et al. [27] showed that the shifts proportional to $\bar{\lambda}_1$ and $\bar{\lambda}_2$ are not

¹A semiclassical treatment of Crampton et al. [55] predicted $\bar{\lambda}_1 = 0$, $\bar{\lambda}_2 \neq 0$.

negligible at low temperatures and they studied the implications on the frequency stability. For this analysis, they assumed that the only density dependent part of the line width is the spin exchange contribution:

$$\frac{1}{T_2} = \frac{1}{T_0} + \left[\bar{\sigma}_0(\rho_{cc} - \rho_{aa}) + \bar{\sigma}_1(\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2\right] \langle v \rangle n_H$$
(4.19)

where T_0 does not depend on density. The term proportional to $\bar{\sigma}_0$ is very small and can be neglected. With (4.19) we get for the combined spin exchange - cavity pulling shift

$$f - f_{0} = \frac{1}{2\pi} \left[\Delta + \alpha \bar{\lambda}_{0} (1 + \Delta^{2}) \right] \frac{1}{T_{0}} + (4.20)$$
$$\frac{1}{2\pi} \left\{ \left[\Delta + \alpha \bar{\lambda}_{0} (1 + \Delta^{2}) \right] \times \left[\bar{\sigma}_{1} (\rho_{cc} + \rho_{aa}) + \bar{\sigma}_{2} \right] + \left[\bar{\lambda}_{1} (\rho_{cc} + \rho_{aa}) + \bar{\lambda}_{2} \right] \right\} \langle v \rangle n_{H}$$

The density dependence of the second term can be quite complicated, since in general $(\rho_{cc} + \rho_{aa})$ depends on n_H . In the case that this dependence can be neglected, a cavity detuning of

$$\Delta = -\alpha \bar{\lambda}_0 - \frac{\bar{\lambda}_1(\rho_{cc} + \rho_{aa}) + \bar{\lambda}_2}{\bar{\sigma}_1(\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2}$$

$$(4.21)$$

leaves the maser frequency insensitive to density fluctuations, but offset from the unperturbed value f_0 by

$$f - f_0 = -\frac{1}{2\pi} \frac{\bar{\lambda}_1(\rho_{cc} + \rho_{aa}) + \bar{\lambda}_2}{\bar{\sigma}_1(\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2} \frac{1}{T_0}$$
(4.22)

With this tuning and at a temperature T = 0.5 K, $(\rho_{cc} + \rho_{aa}) = 0.5$ and $n_H = 10^{11}$ cm⁻³, the required stability of T_0^{-1} and of $(\rho_{cc} + \rho_{aa})$ in order to be able to reach $\frac{\delta f}{f} = 1.4 \times 10^{-18}$ is

$$\delta T_0^{-1} < 1.6 \times 10^{-7} \,\mathrm{s}^{-1}$$

 $\delta(\rho_{cc} + \rho_{aa}) < 3.8 \times 10^{-6}$

These are very demanding limits and it is not obvious that they can be met experimentally. There are different tunings that eliminate the dependence of the maser frequency on the fluctuations of T_0^{-1} or $(\rho_{cc} + \rho_{aa})$ to first order, but it is impossible to make the frequency insensitive to simultaneous uncorrelated fluctuations in n_H , T_0^{-1} and $(\rho_{cc} + \rho_{aa})$. It thus appears likely that at least for long averaging times τ , these spin exchange induced frequency shifts will prevent us from reaching the limits of frequency stability set by the intrinsic thermal noise (4.1) and the electronic noise (4.9). It remains to be seen at which level they become significant. This will depend to a high degree on the ingenuity of the actual design of the cryogenic maser. Finally, it is important to verify experimentally the calculations of the spin exchange cross sections, which are rather sensitive to the interatomic potentials used.

We note here that the Bloch-Siegert shift mentioned in chapter 2 is also a density dependent shift. For a maser operated at 0.5 K and at the optimal density n_{opt} , this shift is however only (2.24)

$$\frac{\Delta\omega_{B.S}}{\omega_0} \simeq \frac{\mu_0 \hbar (\gamma_e + \gamma_p)^2 \eta Q_l (\rho_{cc} - \rho_{aa})_{s.s}}{16 \,\omega_0^2 \, T_b} n_{se} \simeq \frac{8.5 \times 10^{-21} \, \mathrm{s}^2}{T_b^2} \eta Q_l (\rho_{cc} - \rho_{aa})_{s.s}$$

which is too small to be of any practical importance.

Chapter 5

The UBC cryogenic hydrogen maser

The incorporation of liquid helium walls in a low temperature hydrogen maser poses some serious design problems. In the traditional beam geometry, the background pressure in the state selector and outside the maser cavity has to be kept very low to avoid impeding the atomic beam. In a low temperature maser with liquid helium coated walls below 1 K, the He wall coating flows rapidly towards the warm regions around the discharge, evaporates and recondenses on the cold surfaces. This process not only spoils the high vacuum, but also represents a large heat load on the refrigerator. The Harvard-Smithsonian group [17] attacked these problems by installing a giant sorption pump outside the maser cavity, which pumps away all the helium emerging from the cavity. During the maser operation, helium is continuously fed into the maser cavity to coat the walls. However, they have so far not been able to achieve a saturated liquid ⁴He film which is needed for stable maser oscillations. Ultimately of course, this scheme limits the period of continuous operation before the sorption pump needs to be regenerated, but careful design may render the restriction unimportant. Finally, the flowing helium film and the associated ⁴He vapor density will have to be extremely carefully controlled to reach the expected improvement in frequency stability.

In order to circumvent the problem of refluxing helium, we chose a radically different design, based on an idea of Walter Hardy. This closed cycle scheme is explained in the following section.

5.1 Principle of closed cycle cryogenic hydrogen maser

At temperatures below 1 K, only moderately high magnetic fields are required to reach the condition where the electronic Zeeman energy of hydrogen atoms exceeds their thermal energy. Hydrogen atoms in different states can then be efficiently separated by means of magnetic field gradients. This technique is used in our closed cycle cryogenic maser whose principle will be now explained with the help of fig. 5.1.

The maser, which is shown schematically at the bottom of fig. 5.1 consists of two main parts: a microwave cavity tuned to the 1420 MHz hyperfine transition in zero field, the so-called 'maser cavity', and a 'state invertor' which for historical reasons is usually referred to as 'state selector'. It consists of a 40 GHz microwave cavity inside a 1.4 T solenoid. The energy splittings as a function of position along the axis of the cryogenic maser are sketched in fig. 5.1.

All the inside walls of the maser are coated with a superfluid film of ⁴He. Initially, a density of hydrogen atoms is built up in the apparatus with a low temperature discharge source (not shown in fig. 5.1). In order to understand the operation of the maser, consider first a hydrogen atom in the $|c\rangle$ state, situated in the high field region. The field gradients eject it into the low field region and the maser cavity. If the population inversion is sufficient, stable maser oscillations will be observed. Atoms in the $|a\rangle$ state emerging from the maser cavity are sucked back into the high field region. In order to close the cycle, the $|a\rangle$ atoms have to be pumped up into the $|c\rangle$ state. Due to the small transition probability for direct transitions between the $|a\rangle$ and $|c\rangle$ state under microwave irradiation (see appendix A), we are forced to execute this in two steps. First, a magnetic foil, whose properties are discussed below, equilibrates the $|a\rangle$ atoms are then pumped up into the $|c\rangle$ state with the applied microwave radiation which is tuned to the

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Figure 5.1: Principle of UBC cryogenic hydrogen maser. A very schematic diagram of the UBC maser is shown at the bottom, together with the Breit-Rabi diagram of H at the top. In the center of the figure, the energy splittings are indicated as a function of position along the maser.



Figure 5.2: Schematic drawing of relaxing foil. Details are discussed in text.

strong ESR resonance. This closes the cycle. Due to the longevity of the hydrogen atoms, the atoms complete this cycle many times before they recombine to hydrogen molecules. Only a very small flux of 'new' hydrogen atoms is required to keep the operating density constant. In the present apparatus, the necessary flux is estimated to be of the order of a few times 10⁷ atoms/s.

The magnetic foil is prepared following a method developed for the magnetic recording industry [56]: an aluminum foil is anodized in a dilute solution of sulfuric acid. Under certain conditions, this forms a hexagonal array of cylindrical micropores, whose spacing and size can be varied by changing the anodizing voltage [57]. Cobalt is then electrodeposited into these micropores. Figure 5.2 shows a schematic cross section through the foil.

In the maser, we use foils where the diameter of the micropores is chosen to be about 120 Å and their spacing about 300 Å. The idea is that hydrogen atoms moving with thermal velocity over the magnetic foil (separated by the thickness of the liquid helium film) experience magnetic field fluctuations which are centered around the transition frequency between the $|a\rangle$ and $|b\rangle$ state in the appropriate field (783 MHz at 1.4 T).

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This increases the efficiency of the foil.

5.2 General characteristics of the design of the prototype apparatus

In order to simplify the cryogenic engineering, we decided to use a small split-ring resonator [58] as the maser cavity. Typical quality factors of such a resonator are an order of magnitude smaller than those achieved with a full size cavity ($Q \simeq 40000$). In contrast to the situation at room temperature, a very high Q cavity is not essential at low temperature for successful maser operation. The reason is again the strong temperature dependence of the spin exchange cross section. From expression (2.46) it is evident that for maser oscillations, the ratio

$$\frac{n_0}{n_{se}} \equiv \frac{4(1+\Delta^2)\sigma_{se}\langle v\rangle}{\mu_0\hbar(\gamma_e+\gamma_p)^2\eta Q_l}$$

has to be much smaller than 1. At room temperature, this can only be achieved with a high quality factor Q_l . At low temperatures however, the spin exchange broadening $\sigma_{se} \langle v \rangle$ is about a thousand times smaller and as a consequence, the requirement on Q_l is reduced by the same amount. A low quality factor Q_l increases the lower threshold density n_l (see 2.47), but at the same time it decreases the sensitivity of the maser frequency to fluctuations in the cavity resonance frequency (see 2.36).

The small volume of our maser bulb $(V_b = 5.15 \text{ cm}^3)$ limits the total power emitted by the atoms to much smaller values than would be possible with a conventional full size cavity. This implies that the potential stability of our prototype device is more than a factor of 10 worse than the stability anticipated for a full size cryogenic maser as given in (4.11).

A simplified diagram of our prototype apparatus is shown in fig. 5.3. The maser is mounted at the bottom of a ${}^{3}\text{He}$ - ${}^{4}\text{He}$ dilution refrigerator. The refrigerator used is an Oxford Instrument Model 400 refrigerator system. Due to problems with the original



Figure 5.3: Simplified diagram of the cryogenic hydrogen maser drawn approximately to scale. A: waveguide for 40 GHz pump microwaves; B: 1.4 T superconducting magnet with persistent switch; C: 40 GHz microwave cavity; D: relaxing foil region; E: atom transport tube; F: coaxial cable output from the 1420 MHz maser cavity; G: orifice that sets bulb holding time; H: silver sinter; I: variable coupling capacitor; J: coupling loop; K: outer wall of resonator chamber; L: split-ring resonator; M: Pyrex maser bulb; N: outer Pyrex bulb to hold liquid ⁴He cooling bath; O: lead plated brass cylinder; the μ -metal shield, the bias coil and the copper heat shield are not shown; P: thermal impedance; Q: bottom plate of vacuum can.

dilution unit, the dilution unit was replaced by a unit designed by R. Cline and built by members of the UBC Physics machine shop.

For the area to volume ratio of our maser bulb, $\frac{A_b}{V_b} = 4.9 \text{ cm}^{-1}$, the frequency extremum occurs at $T_{ext} = 0.60 \text{ K}$. At this temperature, the relatively high ⁴He vapor density impedes the hydrogen atoms sufficiently to degrade the performance of the closed cycle maser. For this reason, the temperature of the state selector and the atom transport tube (E in fig. 5.3) is reduced to typically 0.4 K.

The state selector and the atom transport tube are thermally anchored to the mixing chamber by four copper braids which are each 25 cm long and 4.5 mm in diameter. A heater at the upper end of the transport tube together with a carbon resistance thermometer on the lower end allow one to regulate the temperature of the state selector to the desired value.

The brass cone, with wall thickness of about 0.75 mm, between the atom transport tube and the maser cavity provides the weak thermal link between the maser cavity and the state selector. A nonmagnetic heater and three different thermometers as described below are in contact with the top plate of the cavity. In order to increase the flexibility of this prototype apparatus and in particular to enable one to cool the maser cavity to temperatures below the state selector temperature, we have also installed a direct thermal link between the maser cavity and the mixing chamber. It consists of a thin piece of copper braid, 40 cm long and with a diameter of 2 mm.

In previous experiments on atomic hydrogen, excessive spin relaxation has been observed which was attributed to microscopically small ferromagnetic particles impeded in the metal walls that confine the atomic hydrogen gas [59]. These particles could be magnetic impurities which are normally present in the construction materials used, or they could have been deposited during the machining process. In order to avoid this spin relaxation, all metal pieces in contact with the hydrogen gas in the maser are briefly

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etched and then coated with a thin layer of polystyrene. This is achieved by first dissolving the polystyrene in toluene, coating the metal piece with this solution and letting the toluene evaporate. Some pieces were later coated with a thin layer of clear epoxy.¹

5.3 Maser cavity

The design of the maser cavity shown in fig. 5.4 is inspired by Mike Morrow's design [38] of the zero field apparatus used to study atomic hydrogen at low temperatures. In fact, we use the same split-ring resonator [58] which is tuned to 1420 MHz.

5.3.1 Tuning and coupling of maser cavity

The copper resonator with a Teflon spacer in the gap is suspended from the top plate of the resonator housing by three Teflon supports. The tuning and coupling can be mechanically changed from the top of the cryostat. The resonance frequency of the maser cavity can be varied ($\Delta f \approx 35$ MHz) by sliding a copper plate vertically along the Teflon spacer. The position of the copper plate determines the total capacitance across the gap. The physical transmission between the rotation of a knob at the top of the cryostat to the movement of the copper plate is through a system of fiberglass rods, Nylon gears and bakelite rods. This system is less microphonic than the pulley system used by M. Morrow in his apparatus, however friction in the gear assembly can generate a substantial amount of heat so that the tuning can only be changed slowly. A similar rod/gear system is used to change the coupling capacitor. The variable capacitor assembly is mounted on the inside of the resonator top plate and is schematically shown in fig. 5.5.

The center conductor of the nonmagnetic semi-rigid coaxial cable is connected to one ¹Stycast 1266, Emerson & Cumming Inc.



Figure 5.4: Design of maser cavity. A: atom transport tube; B: helium fill capillary; C: bursting seal consisting of thin brass foil and tungsten needle; D: helium reservoir; E: brass cone with orifice; F: cooling bath; G: coupling loop; H: split-ring resonator; I: maser storage bulb; J: movable tuning plate riding on a Teflon guide, that also fills the gap of the split-ring resonator; K: Nylon gears and screw for tuning; L: Ag sinter; M: metal to metal seals with annealed Cu washers. Not shown are the thermometry tower, the variable coupling assembly, the coaxial output cable and the bakelite/copper supports. Chapter 5. The UBC cryogenic hydrogen maser



Figure 5.5: Schematic drawing of variable coupling assembly at the maser cavity. A: output coaxial cable; B: top plate of resonator housing; C: copper foil, hardsoldered to the inner conductor of the coaxial cable. On the other end, it is screwed to the Teflon body that contains copper screw and where it forms one of the electrodes; D: copper screw that determines the coupling between the two electrodes that are screwed to the outside the Teflon housing. A slab of fiberglass is glued into the slot on top of the screw; E: copper foil that forms the second electrode on one end and the coupling loop to the split-ring resonator on the other end; F: split-ring resonator; G: Teflon support for split-ring resonator; H: slotted gear, fiberglass slab sits loosely in the slot and transmits a rotation to the copper screw. of the electrodes of the coupling capacitor. The other side of the capacitor consists of a 0.25 mm thick annealed Cu strip which ends in a 3/4 turn coupling loop. The tapered copper screw in the coupling capacitor is connected to the gears sitting in the top plate of the resonator. By screwing this Cu piece further in or out of the Teflon holder, the coupling can be varied from overcoupling ($\beta > 1$) to almost zero coupling ($\beta \simeq 0$) without affecting the tuning of the cavity significantly. At low temperatures, the unloaded quality factor Q_0 of the electropolished resonator was initially 2850. In a later run, an electronic tuning assembly was added that lowered Q_0 to about 2200.

5.3.2 Maser bulb

A Pyrex bulb confines the hydrogen atoms to a region in the center of the resonator. A commercial glass to metal seal joins the Pyrex bulb to a short section of copper tube which is epoxied (Stycast 1266) into the copper plate covering the resonator. The tip of the bulb extends into a copper tube that acts as a waveguide beyond cutoff. This tip contains a small puddle of liquid helium which ensures that a saturated ⁴He film covers all the surfaces in contact with hydrogen. The placement of the pool of ⁴He outside the rf field minimizes possible cavity tuning instabilities.

The bulb holding time T_b is determined by the orifice located at the bottom of the brass cone (see fig. 5.4). The diameter of the orifice is 0.65 mm. This results in $T_b = \frac{4V_b}{A\overline{v}} = 0.6$ s at T = 0.6 K, where A is the cross section of the orifice, $V_b = 5.15$ cm³ is the volume of the bulb and \overline{v} is the average velocity of a hydrogen atom. We cannot use indium or lead O-ring seals between the brass cone and the copper pieces below and above it, because these superconducting rings would trap magnetic flux and lead to field inhomogeneities. We have successfully used 0.25 mm thick annealed and etched copper washers instead. The flanges of the brass cone and of the atom tube have machined knife edges. The forces acting on these joints are reduced by three additional supports

between the maser cavity and a platform bolted to the state selector. They consist of a lower section of bakelite and an upper section of copper, where their relative lengths are chosen in such a way that their total thermal contraction matches that of the combined atom tube - brass cone. The bakelite has a low thermal conductivity which ensures that the thermal anchoring of the maser cavity is not affected by these additional supports. These seals with copper washers have withstood more than a dozen cooldowns without developing a leak.

5.3.3 Cooling bath

The volume between the inner and outer Pyrex bulb is filled with liquid ⁴He. The high thermal conductivity of the superfluid ensures that the temperature of the inner Pyrex bulb is uniform. Silver sinter $(0.1 \text{ cm}^3, 700 \text{ Å} \text{ particle size})$ reduces the thermal resistance between the cooling bath and the resonator top plate. This way, the temperature of the hydrogen atoms in the maser cavity can be controlled by regulating the temperature of this top plate.

The outer bulb has to be completely filled with helium, otherwise the sinter is not immersed in the liquid helium. To ensure this, an additional buffer volume is mounted on top of the resonator and connected to the cooling bath by a short capillary. It can contain up to 4.4 cm^3 of liquid helium, which is much larger than the uncertainty in the volume of the cooling bath ($V_{c.b} = 9.7 \text{ cm}^3$). In order to protect the apparatus in case of a blocked filling capillary, the buffer volume is equipped with a bursting seal. The top of the volume consists of a 0.025 mm thin brass foil which is punctured by a tungsten needle in case of a large overpressure in the helium bath. The position of the needle is adjusted at room temperature to break the seal at a pressure difference of about 1.5 atm. Chapter 5. The UBC cryogenic hydrogen maser

5.3.4 Thermometry

The temperature of the maser cavity is regulated by a heater and three different thermometers. We use the resistive element of a Beckman 89PR500 Trimpot ² for the construction of a nonmagnetic heater ($\simeq 500 \Omega$). Brass leads are connected with silver paint and then the heater is coated with a thin layer of black epoxy (Stycast 2850FT) to electrically insulate it from the thick copper foil that is wrapped around it. The heater is screwed into the top plate of the resonator.

Three different types of thermometers are simultaneously used to monitor the temperature of the maser cavity: a ³He melting curve thermometer, a germanium resistance thermometer and a carbon resistance thermometer. The ³He melting curve thermometer is used to calibrate the resistance thermometers. It was built by R. Cline following the design by Greywall and Busch [60]. The germanium resistance thermometer is a Lake Shore model GR 200A-50. It is mounted into a gold plated copper holder and the electrical leads are well heatsunk to the body. A 51 Ω 1/8 W Matsushita resistor is used for the carbon resistor thermometer. The resistor is ground down to a thickness of about 0.15 mm and then installed in a slotted gold plated copper body.

All these thermometers contain small amounts of ferromagnetic or superconducting materials and therefore must not be mounted directly onto the maser cavity. For this reason, a gold plated copper post with a cross section of 90.7 mm^2 and a length of 9 cm is screwed into the gold plated copper piece containing the silver sinter. The three thermometers are then screwed into the top end of this post. This results in a low thermal resistance between the hydrogen gas in the maser cavity and the thermometers, but keeps the thermometers far enough away to avoid a degradation of the magnetic field

²This trimpot does not contain any ferromagnetic or superconducting parts according to information obtained from the manufacturer. The resistor consists of a film of 'Cervit' deposited on a slab of alumina. The conducting pads are formed from a Ag-Pd alloy.

homogeneity at the maser bulb.

5.4 State selector

The details of the state selector or state invertor are shown in fig. 5.6. The majority of the high field seeking atoms emerging from the low field region enter first the space with the layers of magnetic foil. From there they can reach the microwave cavity through four small holes of 0.35 mm diameter. Atoms that are pumped up into the $|c\rangle$ state leave the cavity predominantly through the 2.5 mm wide extension at the bottom of the cavity. This prevents the $|c\rangle$ atoms from coming in contact with the magnetic foil which would cause transitions to the $|d\rangle$ state and lower the population inversion $n_H(\rho_{cc} - \rho_{aa})_{s.s}$ outside the maser bulb.

5.4.1 Microwave cavity

The microwave cavity is placed in the center of the superconducting magnet. We use the TM₀₁₀ mode, which is for this copper cavity resonant at 39.46 GHz and has a quality factor $Q_l \simeq 2600$ at low temperatures. The lid of the cavity consists of a 0.25 mm thick copper disk which is glued to the rest of the cavity with conducting silver epoxy (epo tek 410-E). A copper adapter piece is hard soldered to the rectangular coin silver wave guide. The orientation of the waveguide with respect to the coupling iris is adjusted at room temperature so that the coupling coefficient is $\beta \approx 0.35$. It is then joined to the cavity with silver epoxy and some additional layers of clear epoxy (Stycast 1266). At low temperature, the resulting coupling to the state selector cavity is then close to critical coupling.

Hydrogen atoms are dissociated in a low temperature source which is connected to the waveguide above the state selector. The source is described later in this chapter. The



Figure 5.6: Details of the state selector region. A: waveguide for 40 GHz pump microwaves, B: 1.4 T superconducting magnet with end corrections, C: gradient coil, D: coupling iris between waveguide and microwave cavity, covered with mylar window with a pinhole which allows H atoms to enter cavity from discharge source (not shown), E: 40 GHz microwave cavity, F: region with 10 layers of relaxing foil, G: indium O-ring seal, H: atom transport tube.

atoms enter the state selector and the rest of the maser through a pinhole in the 0.04 mm thick mylar window that covers the coupling iris. The characteristic time for atoms to leave the state selector through this pinhole was measured to be of the order of 40 s in the present apparatus.

5.4.2 Relaxing foil

The magnetic foil is produced from $13 \,\mu m$ thin aluminum foil ³ according to the recipe of Abe et al. [56]. Ten layers of this foil are inserted into the state selector. An indium O-ring joint between the state selector and the atom transport tube makes it possible to disassemble this part of the apparatus nondestructively and replace the foil later, if necessary.

5.4.3 State selector magnet

The superconducting magnet is wound on an aluminum former that fits snuggly around the state selector. The main solenoid consists of 19 layers of 0.19 mm diameter NbTi wire with Cu cladding with a total of 4497 turns. A 0.025 mm thick mylar sheet is inserted after every second layer and the whole magnet is potted with GE 7031 varnish. The persistence switch and the superconducting joint are heatsunk to the 1 K pot of the dilution refrigerator. The superconducting joint has a critical current in excess of 18 A and is constructed by sandwiching bare NbTi wire between Nb foils, following the procedure described by Blair et al. [61].

The magnet was originally designed to be placed inside a 15.4 cm long hollow NbTi cylinder with 3.35 cm inner diameter. The magnet was to be charged up only after this cylinder had become superconducting, the intended purpose of the shield being to

³This thin aluminum foil was obtained from the UBC Pit burger bar where it was intended to be used as hamburger wrapper.

attenuate the magnetic field outside the magnet exponentially and eliminate possible field gradients at the maser cavity due to the state selector magnet. With this scheme, the homogeneity inside the magnet is also predicted to improve. In appendix D, the magnetic field produced by a solenoid inside an infinitely long superconducting shield is calculated. It is also outlined how to determine the optimal end corrections in this case in order to achieve the highest homogeneity close to the center of the solenoid.

However, after an unsuccessful trial we discovered that the superconducting cylinder is not able to shield the 1.43 T strong magnetic field, which is required to bring the $|b\rangle$ to $|c\rangle$ transition into resonance with the 39.46 GHz microwave radiation. The critical current of the NbTi cylinder is much smaller than that of work hardened NbTi wire because of a lack of enough strong pinning sites in the annealed material. In fact, the shielding properties of a similar NbTi shield had previously been studied by Muething et al. [62] and they came to the same conclusion, that only small fields can be confined this way. For higher fields, magnetic flux starts to leak out of the superconducting cylinder. This has forced us to remove the NbTi shield around the state selector magnet and modify the end correction coils.

When the magnet is charged up for the first time, the current has to be ramped up very slowly. It appears that flux suddenly jumps to an energetically more favorable position. In these events, large amounts of heat are released. At the operating temperature of the magnet, the specific heat is low and the thermal anchoring is also weak. Therefore, already with a small disturbance the condition for a flux jump can be reached. These processes are difficult to analyze exactly because they not only depend on the properties of the magnet, but also on the power supply used. After the first charging, this behavior is not observed again. The magnet can then be completely de-energized and re-energized without any anomalous heating.

Flux trapping in the windings might also be responsible for the homogeneity of the
magnetic field over the volume of the microwave cavity being only 9×10^{-4} . The theoretical prediction is 1.1×10^{-5} . A first order gradient along the z-direction $\frac{\partial B_z}{\partial z}$ can be applied with the gradient coil, however it improves the homogeneity only by a small amount. Although the homogeneity achieved is rather disappointing, it is nevertheless sufficient for this experiment.

5.5 Magnetic shielding of the maser cavity

At the maser cavity, a small uniform bias field has to be applied in order to remove the threefold degeneracy of the upper hyperfine states. In our experiments, a typical value of the bias field is $8 \mu T$ (= 80 mG). This is rather large compared to the fields used in other hydrogen masers, but it is necessary to suppress cross relaxation between the upper three states. The relaxation is presumably caused by some magnetic contamination in or close to the maser bulb. It will be essential to reduce such relaxation in future versions that seek to achieve very high stabilities.

The bias field has to be kept very stable and in addition, the maser cavity has to be shielded from external magnetic fields. Both requirements can be met by using a superconducting shield around the maser cavity. The whole assembly, consisting of four concentric cylinders, is bolted to the bottom plate of the vacuum can.

The outermost layer is a shield made from high permeability foil (co-netic). A ten turn demagnetization coil allows to demagnetize this shield *in situ* and to reduce the ambient field to a small value. The high- μ metal shield is wrapped around the solenoid that provides the bias field. This one-layer solenoid has 151 turns over a length of 22.8 cm and is wound on an aluminum former of 12.7 cm diameter.

The superconducting shield is situated inside the solenoid. It consists of a 30.7 cm long brass cylinder with inner diameter of 9.8 cm and wall thickness of 1.8 mm, and has

copper flanges soldered on at both ends. A $15 \,\mu$ m thick layer of lead is electrodeposited on the inside of the cylinder. The lead is coated with a thin layer of polystyrene that prevents it from oxidizing. Allen Bradley carbon resistor thermometers are located on both flanges. In addition, a nonmagnetic heater is wrapped around the top flange. The superconducting shield is separated by a 1.1 mm thick and 8.5 mm wide greased bakelite ring from a copper support which is well heatsunk to 4.2 K. The support positions the shield such that the maser cavity is exactly at its center.

For adjusting the bias field, the entire superconducting shield is first driven normal by supplying enough power to the heater on the top flange. Then the desired bias field is applied with the solenoid. Following this, the power to the heater is slowly reduced. As the shield is cooled from the bottom side with a temperature gradient sustained, the interface between the superconducting and normal phase of Pb moves slowly upwards. This not only leads to trapping of the applied bias field but in principle should also make the magnetic field inside the shield more homogeneous.

For a shield heated to above the critical temperature of lead (7.19 K), the direct heat load onto the maser cavity due to the blackbody radiation would be of the order of $10 \,\mu$ W. This is too large for the weak thermal link connecting the cavity to the refrigerator and for this reason a copper heat shield, heatsunk to 4.2 K, was inserted between the lead shield and the maser cavity.

We have observed that the bias field trapped in by the lead shield is indeed very stable. In addition, frequency shifts induced by a magnetic field from a solenoid wrapped around the dewar have also been measured. The deduced attenuation by the shield is $\frac{\Delta B_{maser}}{\Delta B_{ext}} = 1.25 \times 10^{-4}$ On the other hand, our process of trapping in the bias field does not appear to improve the homogeneity of the externally applied bias field noticeably. In particular, the measured relaxation time constants T_2 of the free induction decay with the shield in the normal or superconducting state are equal. This indicates that the lead

shield freezes in all flux lines penetrating the superconducting film and the lead film ends up in a mixed state.

5.6 Hydrogen discharge source

Atomic hydrogen is produced by rf dissociation at low temperature. The source, designed and constructed by W. Hardy, is identical to the source used by B. Statt et al. in the ESR experiment and is described in detail in [63]. The top and bottom of the source, which is shown in fig. 5.7, is heatsunk to a special heat exchanger in the refrigerator that is typically operated at 0.4 K.

Hydrogen molecules, condensed onto the inner surfaces are dissociated by rf pulses at the resonance frequency of the resonator, which is 45 MHz. Baffles at the bottom of the source thermalize the produced hydrogen atoms. The bottom tube of the source is connected to the waveguide above the state selector via thin nickel bellows and stainless steel tubes. The hydrogen atoms enter the waveguide below a mylar window vacuum seal that is epoxied across the waveguide. From there, the atoms can reach the state selector cavity and the rest of the cryogenic maser as described before.

The hydrogen molecules are originally deposited in the source through the fill tube capillary which can be heated above the boiling temperature of H₂ (\sim 21 K) while the source is kept cold. After the heat on the capillary is removed, the same fill tube is used to admit enough helium until all the surfaces are covered with a saturated ⁴He film and a small puddle forms at the tip of the inner maser bulb.

A small radioactive source of 60 Co ($\sim 20 \,\mu$ C) provides free charges and helps to start the discharge. Only a few pulses of typically 10 μ s duration and 0.4 W peak power are required to produce a high enough atomic hydrogen density in the maser for stable maser oscillations. Depending on the circumstances, the maser could continue to oscillate for





longer than an hour before the discharge has to be pulsed again.

5.7 Electronics for state selector microwave pump

The 40 GHz electronics for the selector pump sits on top of the cryostat and is shown schematically in fig. 5.8. The microwave radiation to drive the ESR transition in the state selector cavity is generated by a backward wave oscillator, ⁴ whose frequency can be swept in the interval between 26.5 GHz and 40 GHz. The microwave power is attenuated and transmitted to the state selector cavity via Ku-band waveguide. Typically, about $50 \,\mu W$ of power is dissipated in the state selector cavity. A fraction of the reflected signal is coupled to a crystal detector with a 10 dB directional coupler. This is initially used to identify the cavity resonance, and its resonance frequency can be determined approximately with the wavemeter preceding the detector. In order to monitor the frequency of the microwave radiation accurately, a 3 dB directional coupler diverts power into a harmonic mixer, where it is mixed with the 16th harmonic of the output from a HP 8663A frequency synthesizer, set at 2468.77 MHz. The resulting if signal at around 40 MHz is amplified and fed into a counter. After the initial warm up period of the backward wave oscillator, we have found that the frequency of the 39.46 GHz signal is stable to better than 0.4 MHz over the period of about an hour. This fractional frequency instability of 10^{-5} is considerably smaller than both the fractional inhomogeneity of the state selector magnet (9×10^{-4}) and the inverse quality factor of the state selector cavity (4×10^{-4}) . Therefore, it is not necessary to further stabilize the frequency of the microwave generator. It would be straight-forward to phase lock a 40 GHz source to a 10 MHz quartz crystal oscillator, or even better to the maser signal.

The waveguide used to pipe the microwave signal from the generator down to the

⁴Weinschel model 221 microwave sweep oscillator with a model H 2640 rf oscillator plugin.

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cavity has to be constructed from several sections with different thermal properties. Starting from the bottom, a 9 cm long section of coin silver waveguide is joined to the state selector cavity. Via a hardsoldered phosphor bronze 90° bend, the waveguide is continued in a piece of copper waveguide. At the upper end of this piece, a 0.05 mm thick Mylar window is epoxied (Stycast 1266) across the waveguide and forms a vacuum seal. It confines the superfluid helium film and the hydrogen atoms, which enter the waveguide below the window from the discharge source. The temperature gradient between the state selector (0.4 K) and the top of the vacuum can (4.2 K) is sustained with a 34 cm long section of thin wall stainless steel waveguide. In order to decrease the microwave attenuation without increasing the thermal conductivity appreciably, the inside of the waveguide is tinned with a thin layer of Pb-Sn solder which becomes superconducting at these temperatures. The 90 cm long section of stainless steel waveguide between the vacuum can and the top of the cryostat has a $2\,\mu m$ thick layer of electrodeposited copper on the inside. A second Mylar window is glued across the waveguide on top of the cryostat. The waveguide section between the two Mylar windows can be evacuated through a small hole to the vacuum can above the lower seal. At room temperature, the microwave loss of this section is 3.4 dB at 40 GHz.

5.8 1420 MHz detection system

We use a two stage heterodyne system to detect the maser signal. Also incorporated in this spectrometer is a pulse generator that allows us to apply a simple pulse sequence and study the response of the hydrogen atoms at densities below the maser threshold. The spectrometer was built by W. N. Hardy for the previous experiments [12] on hydrogen at the hyperfine transition and it is only slightly modified for our application. A schematic diagram of the spectrometer is shown in fig. 5.9.



Figure 5.9: Schematic diagram of the spectrometer. F: Filter, SSB Generator: Single-Side-Band Generator. Briefly, the maser signal is amplified with two low-noise amplifiers and then mixed with a local oscillator signal at 1420 MHz in an image rejection mixer. The local oscillator signal is generated by direct frequency multiplication (\times 142) of the 10 MHz reference signal, which might come from a rubidium standard or a high quality quartz crystal oscillator, depending on the experiment. By using an image rejection mixer, the resulting signal at the intermediate frequency of 405 kHz does not contain any contributions from noise components of the maser signal at 1419.595 MHz. The signal at 405 kHz is then filtered and amplified, before it is mixed with the second local oscillator signal, derived from a frequency synthesizer. This low frequency synthesizer is a HP 3330A and we use the same 10 MHz reference source as before. The exact output frequency of the synthesizer is adjusted to give the desired beat frequency. When we observe stable maser oscillation, we choose the beat frequency to be about 10 Hz; for measurements of the free induction decay, the beat frequency is set higher, typically several times the line width of the hyperfine transition.

We can apply a simple pulse sequence at the maser cavity, such as a single $\pi/2$ pulse or a $\pi - \pi/2$ pulse sequence. The pulses are generated from the two local oscillator signals at 1420 MHz and 405 kHz with a single-sideband generator. The phase shifter, used to produce the two phases of the 405 kHz signal for the SSB generator, is gated. This avoids any interference problems at the hyperfine frequency, because the generated signal at the resonance frequency of the hydrogen atoms is only present for the duration of the pulses. A 20 dB directional coupler ⁵ mounted on top of the vacuum can at 4.2 K is used to couple the pulses into the coaxial cable leading to the maser cavity. In order to set the tuning and coupling of the maser cavity, we can inject a swept frequency signal through the same directional coupler. The reflected signal is monitored with a crystal detector after the second amplifier.

⁵Technical Research and Manufacturing Inc., model DBS-205

For absolute power measurements, we have to calibrate the sensitivity of the heterodyne detection system. For this purpose, a second directional coupler ⁶ with opposite directivity is installed between the maser cavity and the preamplifier. We use a HP 8663A frequency synthesizer with well calibrated output power levels to generate the calibration signal at 1420.405 MHz.

Special care is required for all the components between the maser cavity and the preamplifier. Any loss will increase the effective noise temperature and degrade the short term stability of the maser. The first 35 cm section of semi-rigid coaxial cable in the immediate vicinity of the maser cavity has copper inner and outer conductor with an outer diameter of 0.085 inch. In order to keep the heatload low, we use stainless steel coaxial cable with outer diameter of 0.141 inch for the next section up to the vacuum flange. The inner conductor is replaced by a stainless steel wire that is tinned with soft solder, which becomes superconducting and reduces the microwave losses. At the vacuum can, a commercial ceramic seal provides a superleak tight microwave connection.

The two directional couplers, the preamplifier and the two isolators are located directly on top of the vacuum can and are kept immersed in liquid helium at 4.2 K. The GaAs MESFET preamplifier was built by W. N. Hardy for a previous experiment. It is based on a design by Williams et al. [64] and uses a Mitsubishi MGF 1412 and a Mitsubishi MGF 1402 FET in the first and second stage, respectively. At 4.2 K, the gain is 21.0 dB with a 3 dB bandwidth of 395 MHz and the noise temperature is $T_N = 20$ K. The results of flicker phase noise measurements on this device are presented in the next chapter. The isolators ⁷ at the input and output of the preamplifier are surrounded by superconducting lead jackets that shield the isolators from changing magnetic and electric fields, especially at the line frequency. At the operating temperature of 4.2 K,

⁶Midwest Microwave, model 5010-20R with 20 dB coupling

⁷Pamtech, model LTE 1102

the isolators have a bandwidth of about 300 MHz, and at 1.42 GHz an insertion loss of 0.25 dB and an isolation of 29.4 dB and 21.1 dB, respectively.

All three coaxial cables leading from the vacuum flange at 4.2 K to the top of the cryostat at room temperature have 0.141 inch diameter stainless steel outer conductor and silver-plated copper-clad steel inner conductor. The amplifier ⁸ preceding the image rejection mixer has a noise temperature of 440 K. Since the gain of the preamplifier is only 21 dB, this results in a 3.5 K contribution to the overall noise temperature, which is referred to the input of the preamplifier. In later measurements, we have reduced the contribution of the second and further stages to less than 1 K by adding a lower noise amplifier in front of the Watkins Johnson amplifier. This additional amplifier is identical to the GaAs preamplifier, however it is operated at room temperature, where its noise temperature is about 90 K.

The final filter at the output determines the overall noise bandwidth B of the receiver system and therefore affects directly the signal to noise ratio. It is this noise bandwidth Bthat enters in the expression (4.8) for the maser frequency instabilities due to electronic white phase noise.

When we observe free induction decays, the output of the final mixer is usually directly connected to the input of a Tektronix oscilloscope ⁹. In this case, the variable input filter of the oscilloscope is the final filter indicated in fig. 5.9. However, when the maser is oscillating and we want to make a precise measurement of the frequency fluctuations, we use a separate narrow-band-pass filter at the output of the final mixer. This active filter is centered at 10 Hz and has a 3 dB bandwidth of 3 Hz. Its noise bandwidth B is 5.1 Hz. In addition to a direct buffered output, the filter has a second output that delivers an approximate square wave. This output signal is derived from the filtered

⁸Watkins Johnson, model WJ 737-303S

⁹Tektronix 5440 oscilloscope with a 5A22N differential amplifier.

signal by a simple circuit that amplifies the signal and subsequently clips the amplitude. The timings of the zero crossings of both filter outputs are identical, however the slopes at the zero crossings are much larger for the 'square wave' output. As will be discussed in greater detail later, the relative frequency stability of the maser is measured by recording the time intervals between a set number of zero crossings of the beat signal. These measurements are affected by fluctuations in the dc offset of the signal and in the trigger level of the counter. These effects are minimized by using the 'square wave' output'. It is important to note, that the effective noise bandwidth B is the same for both outputs.

Chapter 6

Experimental Results

The first results on the cryogenic hydrogen maser were obtained on April 14, 1986, when we observed the first maser oscillations in our apparatus. In this chapter, we first describe the experimental procedures used to start up the maser, before we concentrate on the results, which include the frequency stability and the measurement of the binding energy.

6.1 Start up procedure

6.1.1 Cool down

Several frustrating months of leak checking preceded the breakthrough of the observation of the first maser signal. Only a few months after delivery, the Oxford Instruments refrigerator started to develop low temperature leaks on the top flange of the vacuum space, presumably in one or many of the numerous stainless steel welds. For several months, we tried to localize these leaks, however without any noticeable success. In our desperation, we turned to alcohol, and to our surprise, it solved some of the problems. We used the following procedure:¹ We first cool the refrigerator with liquid nitrogen to 77 K, then we quickly lower the dewar and spray anhydrous ethanol onto the top flange of the vacuum flange. The alcohol freezes immediately on the cold surface and forms a several millimeter thick smooth layer that becomes our new low temperature seal. With this method, we can usually reduce the leak rate sufficiently to operate the dilution

¹This method, to our knowledge first used at Cornell, was brought to our attention by Prof. T. J. Greytak.

refrigerator. The residual helium leak however limits the base temperature of the mixing chamber to 100 mK or higher. In later runs, we have installed three charcoal sorption pumps in the vacuum can, two of them mounted on the underside of the top flange and the third on the bottom plate of the vacuum can.

After the initial cool down, we load the discharge source with molecular hydrogen. The source is kept at a temperature of around 0.5 K, while 5×10^{-3} moles of H₂ are deposited onto the inner walls of the discharge source through a heated capillary, as already described. Then the capillary heater is turned off and about 3×10^{-3} moles of ⁴He are added to the maser through the same capillary. We are now ready to fire the discharge a few times and detect the atoms in the maser cavity by pulsed magnetic resonance. The maser cavity is tuned to the hyperfine transition frequency and the coupling is adjusted to reach critical coupling. Typically, we apply $\pi/2$ pulses and observe the free induction decay with the spectrometer.

6.1.2 Adjustment of bias field at the maser cavity

Before the state selector magnet is charged up, we adjust the bias field at the maser cavity to the desired value. The high μ metal shield has been demagnetized at room temperature and also at 77 K, so that the initially trapped field in the Pb shield is already quite uniform and usually of the order of 1 μ T in amplitude. The bias field can be changed after driving the superconducting shield normal, as was described in the previous chapter. The bias coil is calibrated by measuring the frequency of the free induction decay as a function of the solenoid current and fitting it to the parabola described by equation (4.12). At low density, radiation damping and spin exchange relaxation are unimportant and it is adequate to fit the signal to an exponentially damped sine wave.

When the z-component of the bias field is exactly zero, the direction of quantization is orthogonal to the rf field and we can observe the transverse transitions between the

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 $|d\rangle$ and $|a\rangle$ states and between the $|b\rangle$ and $|a\rangle$ states (see appendix A). From the beat frequency of these two signals, the transverse component H_{\perp} of the residual static field can be determined. A typical value for H_{\perp} is $0.2 \,\mu$ T. If we assume that the bias coil is perfectly aligned with the resonator and does not produce a net transverse field at the maser cavity, we can determine the total bias field that the hydrogen atoms are exposed to in the resonator by combining this measurement with the calibration of the bias coil. In most conventional hydrogen masers, the magnetic field is measured by determination of the rf frequency that induces transitions between the $|c\rangle$ and $|b\rangle$ state and between the $|c\rangle$ and $|d\rangle$ state. The copper walls of our resonator are much thicker than the skin depth at the appropriate frequency (usually a few kHz), which rules out this method in our case.

6.1.3 Cross relaxation in the maser bulb

The observed relaxation times T_2 are shorter than the bulb holding time T_b ($\simeq 0.6$ s) and are field dependent as shown in fig. 6.1. This indicates that a fairly strong relaxation mechanism between the upper three states is present. When the applied magnetic field is increased, it splits the degeneracy of the three states and thus suppresses the cross relaxation process for energetical reasons. This interpretation can be confirmed by measuring the relaxation of the longitudinal magnetization with a $\pi - \pi/2$ pulse sequence.² In our analysis, we assume that there are three important processes that determine the recovery of the longitudinal magnetization:

(i) Flow in and out of the maser bulb. The rate for the atoms to leave the bulb is $\Gamma_b = T_b^{-1}$.

²We are using here the language of the fictitious spin $\frac{1}{2}$, which was introduced in chapter 2.



Figure 6.1: Transverse relaxation time T_2 versus bias field. The temperature of the hydrogen gas is 500 mK and the densities are low enough to avoid large radiation damping and spin exchange effects.

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(ii) Cross relaxation between $|c\rangle$ and $|b\rangle$ and between $|c\rangle$ and $|d\rangle$. We assume that the relaxation rates are equal and we denote them with Γ_c .

$$b \stackrel{\Gamma_{c}}{\rightleftharpoons} c \stackrel{\Gamma_{c}}{\rightleftharpoons} d$$

(iii) We also include longitudinal relaxation between the upper three states and the $|a\rangle$ state with relaxation rate Γ_l :

$$a \stackrel{\Gamma_l}{\leftarrow} b, a \stackrel{\Gamma_l}{\leftarrow} c, a \stackrel{\Gamma_l}{\leftarrow} d$$

At low densities, spin exchange relaxation can be neglected and it is then easy to find the solution of the set of linear equations describing the longitudinal relaxation with the above assumptions. In thermal equilibrium, the population difference $(n_a - n_c)_{eq}$ is given by

$$(n_a - n_c)_{eq} \equiv n_H (\rho_{aa} - \rho_{cc})_{eq.} \simeq \frac{1}{4} n_H \frac{\hbar \omega_0}{kT}$$

After a π pulse at the maser transition, the population difference $n_a - n_c$ gets inverted and it relaxes back to the equilibrium value according to

$$\delta(n_a - n_c)(t) = -\frac{1}{2} n_H \frac{\hbar\omega_0}{kT} \left\{ \frac{2}{3} e^{-(\Gamma_b + 4\Gamma_l)t} + \frac{1}{3} e^{-(\Gamma_b + \Gamma_l + 3\Gamma_c)t} \right\}$$
(6.1)

If we apply a $\pi/2$ pulse at a time t after the π pulse, the amplitude of the free induction decay is proportional to $(n_a - n_c)(t)$ and we can verify expression (6.1) experimentally. A typical example from an early run is shown in fig. 6.2, where the normalized deviation of the longitudinal magnetization, proportional to $\delta(n_a - n_c)(t)$, is plotted versus the time t between the π and $\pi/2$ pulse.

The data is well described by the two exponentials of (6.1) and it is obvious that a single relaxation time T_1 is not sufficient to characterize the longitudinal relaxation. The dotted line in fig. 6.2 indicates the long time behavior and it extrapolates back to a normalized magnitude of $\frac{2}{3}$ at t = 0. This confirms our prediction that $\Gamma_c \gg \Gamma_l$.



Figure 6.2: Relaxation of the longitudinal magnetization in the maser bulb versus time. The solid line is a fit to (6.1) and the dotted line indicates the asymptotic behavior. The temperature of the maser bulb is 460 mK and the bias field is $13 \,\mu$ T.

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This cross relaxation is most likely caused by some magnetic contamination on or near some part of the surface of the maser storage region. It is not caused by magnetic field gradients across the resonator. The evidence is that at higher temperatures where the vapor pressure of ⁴He is so high as to effectively immobilize the hydrogen atoms, the relaxation time T_2^* becomes much longer. If there were large field gradients across the bulb present, we would expect T_2^* to become shorter, because there is no motional averaging possible any more. On the other hand, if magnetic particles on the surface were responsible for the relaxation, at higher temperatures only a few spins could come in contact with them and be relaxed. This results in a longer effective relaxation time, which is in qualitative agreement with the observations.

The choice of bias field is determined by a compromise between a low field for minimal sensitivity of the frequency to field fluctuations and a high field for maximal relaxation time T_2 . In the last run, we have trapped in a field of $8 \,\mu$ T. When the Pb shield becomes superconducting, we do not observe any effect on the frequency or T_2^* of the free induction decay.

6.1.4 State selector magnetic field

The state selector magnet has to be charged up very slowly for the first time; it takes typically about an hour to reach the operating field of 1.433 T. The frequency of the Kuband microwave radiation is adjusted to the center of the state selector cavity resonance. At the same time $\pi/2$ pulses are applied to the maser cavity. As soon as the resonant magnetic field is reached, the free induction decay signal inverts and, if the density is high enough, maser oscillations are observed. After fine adjustments of the microwave frequency and the magnetic field for maximal population inversion, the magnet is put into persistent mode.

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6.2 Power versus density measurements

6.2.1 Density calibration

An important parameter for the operation of the maser is the total density of hydrogen atoms in the maser bulb, n_H . In the present apparatus, it is not possible to measure n_H directly, as would be possible, for instance, with a calibrated capacitive pressure gauge. However, the technique of magnetic resonance can be used to infer n_H . In thermal equilibrium, the static magnetization \tilde{M}_z is proportional to n_H and is given by (2.23):

$$\tilde{M}_z = \frac{1}{2}\hbar(\gamma_e + \gamma_p)\frac{\hbar\omega_0}{4kT}n_H$$
(6.2)

With a $\pi/2$ pulse this static magnetization can be converted into a precessing transverse magnetization \tilde{M}_r with the same magnitude as \tilde{M}_z . In this case, the power emitted by the atoms immediately after the pulse is

$$P = \frac{\mu_0 \omega_0 V_b \eta Q_l}{2} \left| \tilde{M}_r \right|^2 = \frac{\mu_0 \omega_0 V_b \eta Q_l}{2} \left[\frac{\hbar^2 (\gamma_e + \gamma_p) \omega_0}{8kT} \right]^2 n_H^2$$
(6.3)

For critical coupling, half of the power P reaches the preamplifier. By measuring the power P absolutely, it is thus possible to determine n_H . We have used this method to measure n_H in the maser bulb. We first turn off the state selector by either slightly mistuning the microwave radiation applied to the state selector cavity or by changing the magnetic field at the state selector by a small amount. Both methods bring the microwave radiation off resonance with the ESR transition in the state selector cavity, but the second method is preferred for the following reason. Mistuning the microwave frequency changes the power dissipation in the cavity and can lead to small changes in the state selector temperature, which should be avoided. After the transients have decayed, we apply a $\pi/2$ pulse at the maser cavity and measure the initial power of the free induction decay.

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However, it is difficult to obtain accurate results for n_H with this method for several reasons. First of all, it is difficult to determine the filling factor η for the split ring resonator used in the maser. The exact rf field distribution is not known and the filling factor cannot be calculated from the known geometry, but it has to be determined experimentally. M. Morrow [38] measured the filling factor for the same split ring resonator, but with a larger outer resonator can, and obtained $\eta = 0.42$. An additional difference to the situation of M. Morrow is that the maser glass bulb extends further out to weaker rf fields compared with the bulb used in the earlier experiment. Both differences tend to decrease the filling factor.

There are more fundamental problems with the density determination. Ideally, a low temperature value at the orifice of the maser bulb, that could be closed in a time short compared with T_b , would confine the atoms in the maser bulb. After the atoms reach their thermal equilibrium, the density n_H could be determined from the measured power P with equation (6.3). In the derivation of (6.3), it is assumed that the spin system is in thermal equilibrium described by the temperature T. However, the hydrogen atoms in the maser bulb are also in contact with the state selector, whose temperature is in general lower. During the bulb holding time T_b , hydrogen atoms encounter many collisions with the walls. Only a few of these collisions are necessary to thermalize the translational degrees of freedom, which therefore reach their equilibrium in a time much shorter than T_b . The spin degrees of freedom however are only thermalized by hydrogenhydrogen collisions or by interactions with magnetic field gradients, produced for example by magnetic impurities on the walls. At low densities, the collision rate between hydrogen atoms is small and the thermal spin equilibrium in not completely reached within the bulb holding time. Therefore, the static magnetization M_z is larger (for $T_{s,s} < T_{maser}$) than what would be expected based on (6.2). Equation (6.3) leads then to an overestimation of the density in the maser bulb.

So far we have implicitly assumed that the total density in the maser bulb with the state invertor on and off are equal. This is not exactly fulfilled. In the absence of magnetic field gradients between different parts of the apparatus, the ratio of the total densities in different regions of the maser is completely determined by the temperatures of the regions; it does not depend on the population distribution. This is no longer true when different parts of the maser are in different magnetic fields, as is the case for the cryogenic maser. When the microwave radiation in the state invertor is on resonance, it converts high field seeking atoms into low field seeking atoms. As a consequence, the total density n_H in the maser bulb is higher when the state invertor is on compared to when it is off.

This effect and the effect of incomplete thermalization discussed before compensate each other to a certain degree. The size of the combined effects has been estimated with a numerical simulation of the maser and is discussed in detail in chapter 7. The results of the simulation indicate that for the present apparatus, operated at $T_{maser} = 509 \text{ mK}$ and $T_{s.s} = 465 \text{ mK}$, the real densities are approximately 40% higher than the densities deduced from the initial power of the free induction decay with equation (6.3). The size of the correction depends on the relaxation rates and temperatures of the different parts of the maser and is predicted to be much larger at lower temperatures.

This makes it desirable to find an additional method to determine the density in the maser bulb. To a first approximation, the only density dependent contribution to the line width $\Gamma (=T_2^{-1})$ of the maser while it is oscillating is due to spin exchange.

$$\Gamma = \Gamma_0 + \left[\bar{\sigma}_0(\rho_{cc} - \rho_{aa}) + \bar{\sigma}_1(\rho_{cc} + \rho_{aa}) + \bar{\sigma}_2\right] \langle v \rangle n_H$$
(6.4)

where Γ_0 does not depend on density and includes contributions from the flow out of the maser bulb and from the cross relaxation mechanism. At the operating conditions of the maser and based on the calculations of Koelman et al. [46], the spin exchange contribution is dominated by the term proportional to $\bar{\sigma}_1(\rho_{cc} + \rho_{aa})$. For the present situation, $\rho_{cc} + \rho_{aa}$ can be estimated based on the following observations. The strong cross relaxation in the present maser tends to equalize the upper three populations in the maser bulb, $\rho_{bb} \simeq \rho_{cc} \simeq \rho_{dd}$. In addition, according to equation (2.33) the population difference $\rho_{cc} - \rho_{aa}$ is strongly reduced by the maser action. We therefore expect $\rho_{cc} + \rho_{aa} \simeq$ 0.5, a result which is confirmed by the numerical simulations presented in chapter 7. Making use of the calculations for $\bar{\sigma}_1$ by Koelman et al. [46], the line width is then expected to depend on the density n_H as

$$\Gamma \simeq \Gamma_0 + \bar{\sigma}_1 (\rho_{cc} + \rho_{aa}) \langle v \rangle n_H = \Gamma_0 + 3.8 \times 10^{-13} \, \frac{\text{cm}^3}{\text{s}} \, n_H \tag{6.5}$$

for $T_{maser} = 0.5$ K. The line width Γ can be measured experimentally by studying the dependence of the maser frequency on the detuning of the maser cavity. Equation (2.34) describes this dependence and we rewrite it as

$$\omega - \omega_0 = \Delta \Gamma \simeq \frac{2Q_l}{\omega_0} (\omega_c - \omega_0) \Gamma$$
(6.6)

The measured line widths are shown in fig. 6.3. We use these measurements to calibrate the density determination as follows. At each density, we have measured the line width Γ of the oscillating maser and then the initial power of the free induction decay with the state selector turned off. This power is then converted into density with equation (6.3). The filling factor η , which has to be interpreted now as an effective filling factor, is adjusted in such a way that the data in fig. 6.3 has the slope required by eq. (6.5). The effective filling factor obtained by this procedure is $\eta_{eff} = 0.11$. The real electromagnetic filling factor. Based on the simulation results presented in chapter 7, we estimate that this correction factor is about 1.4, which implies that $\eta \simeq 0.22$. This is lower than the value obtained by M. Morrow, but note that the density calibration does not depend



Figure 6.3: Maser resonance line width Γ versus total density n_H in the maser cavity with $T_{maser} = 509 \,\mathrm{mK}$ and $T_{s.s} = 465 \,\mathrm{mK}$. The solid line is a least-square-fit to the data. As explained in the text, the slope is related to the spin exchange broadening parameters and is used to calibrate the density determination. From the fit, the density independent line width Γ_0 is found to be $\Gamma_0 = 4.23 \pm 0.07 \,\mathrm{s}^{-1}$.

on the exact determination of η . The calibration is correct provided that the density correction factor is independent of density and that the theoretical calculations of the spin exchange broadening parameters [46] are correct.

6.2.2 Recombination

At the beginning of an experiment, the discharge source is fired until the density of atomic hydrogen has built up to the desired value. In most cases, measurements are then performed while the density is slowly decaying. We have also tried to fire the discharge periodically (e.g. every 1 s) in order to obtain a constant density over a long period of time. We have found that even if we operate the discharge just above its threshold, the source in general produces too many atoms and the resulting quasi-equilibrium densities in the maser are too high for many experiments of interest. So far, we have not further addressed the problem of keeping the density constant to a high degree. However, this will eventually be required if one is to avoid the spin exchange shifts and if a very high frequency stability is to be achieved over longer averaging times, as discussed in chapter 4.

Except at the low temperatures, the hydrogen atoms recombine slowly, as discussed in chapter 3. The maser can oscillate for several hours without the need to replenish the atoms. The recombination follows a second order rate equation:

$$\frac{d}{dt}n_H = -Kn_H^2 \tag{6.7}$$

which has the solution

$$\frac{1}{n_H(t)} = \frac{1}{n_H(0)} + Kt \tag{6.8}$$

A typical example of a decay is shown in fig. 6.4. The data is well described by (6.8) and for $T_{maser} = 509 \text{ mK}$ and $T_{s.s} = 465 \text{ mK}$ we obtain $K = 8.3 \times 10^{-16} \frac{\text{cm}^3}{\text{s}}$.



Figure 6.4: Decay of density n_H in maser bulb while maser is oscillating. Straight line is a least-square-fit to (6.8) with $K = 8.3 \times 10^{-16} \frac{\text{cm}^3}{\text{s}}$. $T_{maser} = 509 \text{ mK}$ and $T_{s.s} = 465 \text{ mK}$.

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It is difficult to compare the measured recombination rates with theoretical predictions. For an exact prediction, it is not sufficient to know all the populations and areato-volume ratios in the different parts of the maser, because the recombination rate is also affected by the density and the decay rate of atoms in the space between the source and the waveguide above the state selector cavity. We anticipate that the effective rate constant K can be reduced by installing a large atom storage container (with a small area-to-volume ratio) below the source.

6.2.3 Power output of maser

The output power of the maser, measured at the input of the preamplifier, is shown in fig. 6.5 as a function of the total density n_H in the maser bulb for two different runs. For the first run (triangles, May 1986), the loaded cavity quality factor Q_l was 1700, whereas in the second run (circles, May 1987) Q_l was reduced to 1100 due to an added electronic tuning circuit. The state selector power was 50 μ W in both runs, the temperatures of the maser bulb were 500 mK and 509 mK for the first and second run, respectively. For the first set of data, the temperature of the state selector was 415 mK, in the second run this temperature was held at 465 mK.

The measured power levels are much smaller than expected. Based on the simple theory presented in chapter 2 and with $(\rho_{cc} - \rho_{aa})_{s.s} = 0.5$, the power at the input of the preamplifier is expected to be about 9×10^{-13} W for a density $n_H = 10^{12}$ cm⁻³ and increasing linearly for the range of densities of interest here. The data in fig. 6.5 display a definite curvature, and in the second run the maser turned off completely above a density of only 3×10^{12} cm⁻³. Our simple theory predicts for $(\rho_{cc} - \rho_{aa})_{s.s} = 0.5$ the upper threshold density should be more than ten times higher than this.

The behavior of the power is not caused by faster than anticipated spin exchange processes in the maser bulb, but is caused by a much lower than expected efficiency





Figure 6.5: Output power of the maser versus total density of hydrogen atoms in the maser bulb n_H . For the first run (triangles) $Q_l = 1700$, and for the second run (circles) $Q_l = 1100$. The temperatures of the maser bulb are 500 mK and 509 mK for the first and second run, respectively. The state selector temperatures are 415 mK and 465 mK, respectively.

of the state selector. The population inversion $(\rho_{cc} - \rho_{aa})_{s.s}$ achieved in the atom tube outside the maser bulb must be considerably smaller than 0.5 and $(\rho_{cc} - \rho_{aa})_{s.s}$ must also be decreasing with density in order to explain the data.

We found that the maser output power is not sensitive to changes in the power level of the microwave pump signal in the state selector. All the evidence therefore indicates that the low efficiency is caused by insufficient relaxation by the magnetic foil in the state selector: the magnetic foil is the bottleneck in the state inversion from $|a\rangle$ to $|b\rangle$ in high field. At higher densities, spin exchange relaxation effects in the atom transport tube lower the efficiency even further. This interpretation of the data is supported by the results of the numerical simulation to be presented in the next chapter.

The difference in maser output power for the two different runs can be at least partially explained by the slightly different temperatures of the state selector $(\delta T_{s.s} = 50 \text{ mK})$. Physically, the compression factor for $|a\rangle$ atoms, $\exp\left\{\frac{E_a(B)-E_a(0)}{kT}\right\}$, which relates the $|a\rangle$ densities in high and low field, was larger in the first run because of the lower temperature of the state selector. As a consequence, for a fixed density n_H at the maser bulb, the total number of $|a\rangle$ atoms in the region of the magnetic foil was higher, which led to a larger production of $|b\rangle$ atoms and consequently $|c\rangle$ atoms in high field. This then resulted in a higher population inversion outside the maser bulb and higher output power. This simple picture is confirmed by numerical simulations, where competing spin exchange relaxations are also taken into account. However, based on the numerical results, it is not clear whether this temperature effect can explain the whole difference between the two sets of data. It is also conceivable that the magnetic foil deteriorated in the time between the two runs, for instance by oxidation or by deposition of other nonmagnetic impurities on top of the magnetic particles.

6.3 Temperature dependence of maser frequency

We were able to observe stable maser oscillations for temperatures of the maser bulb between 230 mK and 660 mK. At higher temperatures, the ⁴He vapor density is in excess of 1.5×10^{16} cm⁻³. As a result, the diffusion time constant between the state selector and the maser cavity becomes rapidly longer and the flux of $|c\rangle$ atoms entering the maser bulb drops below the threshold flux necessary for maser action.

At the lower end of the range of operating temperatures, the fraction of hydrogen atoms adsorbed on the ⁴He coated walls increases rapidly and this results in fast recombination. It is therefore necessary to fire the discharge source with a higher and higher repetition rate. At the lowest temperatures, fast recombination prevents the hydrogen atoms from circulating back and forth between the state selector and maser cavity many times and the operation of the maser approaches a 'single shot' operation, similar to the MIT maser [14]. Below 230 mK, it is impossible with our source to create a high enough hydrogen density in the maser bulb for sustained maser action.

We have measured the frequency of the maser as a function of the temperature of the maser bulb. The results are shown in fig. 6.6. The maser signal was measured against a reference signal derived from a rubidium gas cell standard ³ which in turn was calibrated with a GPS receiver.⁴ The frequency of the beat signal was measured with a HP 5345A electronic counter that could be controlled from a PC via a GPIB link.

The temperature of the maser bulb was measured with both a germanium and a carbon resistance thermometer. At the end of the run, these thermometers were calibrated against a ³He melting curve thermometer. The state selector was always slightly colder than the maser bulb and the density of hydrogen atoms in the maser bulb was kept low

³This was an Efratom model FRK-L rubidium frequency standard, on loan from D. Wineland, NBS Boulder.

⁴This Global Positioning System receiver and monitor was developed by the US National Bureau of Standards and loaned to us by D. W. Allan and \hat{R} . Davis of NBS Boulder.





Figure 6.6: Observed frequency of the maser -1420405000 Hz versus temperature of the maser bulb

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in order to minimize the spin exchange frequency shifts.

The temperature dependence of the frequency shift is then dominated by the wall shift at low temperatures and the ⁴He buffer gas shift at the higher temperatures and is given by eq. (3.10):

$$f - f_0 = \delta n_{He} + f_s \frac{A_b}{V_b} \Lambda \exp\left\{\frac{E_B}{kT}\right\}$$
(6.9)

By fitting the data in fig. 6.6 to eq. (6.9), we can extract the surface frequency shift f_s and the binding energy E_B . Only data points between 250 mK and 520 mK were included in the fit. Below 250 mK, we expect to see deviations form the simple expression (6.9) because the phase shift per sticking event ϕ_0 is not much smaller than 1 any more. Above 520 mK, the frequency shifts $f - f_0$ become very small and spin exchange shifts and their temperature dependence start to play a role. In addition, we use the value for $\delta = -1.183 \times 10^{-17} \,\text{Hz cm}^3$ that was measured by Hardy et al. [40] at $T \simeq 1 \,\text{K}$, which is not fully justified since δ is expected to be weakly temperature dependent [43]. For these reasons, we have excluded the data points above 520 mK.

In our least-square fitting routine, we fit $\ln\{(f_0 - f)\sqrt{T}\}$ and not $f_0 - f$ versus T^{-1} . The factor of \sqrt{T} is included to cancel the temperature dependence of the de Broglie wavelength Λ . If we assume f_s to be temperature independent, the low temperature behavior of $\ln\{(f_0 - f)\sqrt{T}\}$ is a linear function in the inverse temperature T^{-1} with slope $\frac{E_B}{k}$ and intercept $\ln\{f_s \frac{A_b}{V_b} \Lambda \sqrt{T}\}$. In this way, all data points included are given about equal weight in our fitting procedure. This is desirable, since the main experimental uncertainties are associated with the temperature determination and not with the frequency measurements.

The best fit to the data is shown in fig. 6.7, and yields for the three adjustable parameters:

$$f_0 = 1420\,405\,769.89\pm0.02\,\mathrm{Hz}$$

$$\frac{E_B}{k} = 1.011 \pm 0.010 \text{ K}$$
$$f_s = -71.5 \pm 3.0 \text{ kHz}$$

We have used $\frac{A_b}{V_b} = 4.87 \text{ cm}^{-1}$, which was calculated from the known geometry of the bulb.

The frequency f_0 corresponds to the unperturbed hyperfine frequency in the applied bias field and the value obtained in the fit is consistent with the bias field trapped in by the lead shield. The value for the binding energy of hydrogen to ⁴He is the most precise determination of E_B reported so far. In fig. 6.8, we compare our value for E_B with values previously published in the literature.

There is a surprisingly large spread in the published values for E_B . The first determination of E_B (a in fig. 6.8) was obtained in a way similar to the present method: the temperature dependence of the frequency shift and recombination rate at zero field was measured by pulsed magnetic resonance on a sealed sample [41]. Both frequency and recombination data indicated $E_B = 1.15$ K. However, the frequency data was analyzed under the assumption that the sticking coefficient over the temperature range of interest is only 0.046. Subsequent measurements of the sticking coefficient s by Berkhout et al. [72] have revealed that $s = 0.33 \frac{T}{1\text{ K}}$ for temperatures between 80 mK and 0.5 K and that Morrow et al. have therefore overestimated the corrections to the simple expression (6.9), which have to be applied when the phase shift per sticking event ϕ_0 becomes comparable to 1. The point indicated by b in fig. 6.8 was obtained from a reanalysis of their frequency data. The corresponding frequency shift f_s becomes -61 kHz, instead of -49 kHz.

With the exception of measurement i in fig. 6.8, all other values of E_B were obtained by the analysis of the temperature dependence of recombination rates in high field and rely on the assumption that the intrinsic recombination cross section has no temperature



Figure 6.7: Observed and fitted temperature dependence of the maser frequency shift from the unperturbed value in the measured bias field. The frequency was measured in Hz, the temperature in K. The solid line is a fit to equation (6.9) as described in text and yields $E_B = 1.011 \pm 0.010$ K and $f_s = -71.5 \pm 3.0$ kHz.





Figure 6.8: Compilation to the measured values of the binding energy of H to liquid ⁴He plotted in temporal order. The circles represent measurements from the University of British Columbia [(a) and (b); (h) and (i) from [41], [59, 63] and [65] respectively and (k) from present work]; the diamonds from the University of Amsterdam [(c) and (e) from [66] and [67]]; the squares from MIT [(d) and (g) from [68] and [69]]; the triangle (f) from Cornell [70]; and the cross (j) from Harvard [71]. Point (b) is obtained by a reanalysis of the data of (a) as explained in the text.

dependence. We believe that the surface frequency shift is likely to be less temperature dependent than the recombination cross section. A temperature dependence as weak as $T^{\frac{1}{2}}$ leads to an error of the order of 5% or greater in the binding energy and could explain some of the scatter in the published values of E_B . Errors in thermometry might also have affected some of the reported values.

There is a discrepancy between our value for f_s and the value obtained by Pollack et al. [29], which has not yet been resolved. In a field of 6.8 T, Pollack et al. succeeded in observing the $|a\rangle$ to $|b\rangle$ NMR free induction decay signal of atoms adsorbed on the surface and measured directly the surface shift. Scaled to zero field, their value corresponds to $f_s = -43.0 \pm 0.8$ kHz. Our value for f_s depends directly on the value of $\frac{A_b}{V_b}$. The quoted uncertainty in f_s (3 kHz) does not contain a contribution from the uncertainty in $\frac{A_b}{V_b}$, but even with a pessimistic error bar of 10%, our value becomes $f_s = -71.5 \pm 7.8$ kHz and is still clearly in disagreement with [29].

It is conceivable that the frequency of adsorbed atoms is not only shifted by Van der Waals effects, but also by interactions with magnetic impurities. As long as an atom samples many impurities within a time interval $\frac{1}{2\pi f_s}$, motional averaging will prevent a large line broadening. In this case, it will be difficult to distinguish this shift from the Van der Waals shift.

6.4 Frequency stability

6.4.1 Three-cornered-hat method

In the time domain, the frequency stability of an oscillator is usually characterized by the so-called 'Allan variance' $\sigma_y^2(\tau)$, which is defined in appendix C. It can be determined by measuring the fluctuations of the output frequency, averaged over a time τ . Experimentally, the maser signal at 1420.405 MHz is translated down in two stages to a
beat signal with frequency of approximately 10 Hz. The frequency fluctuations in this low frequency signal can now easily be measured with an electronic counter. The counter registers the time intervals between zero crossings of the beat signal and averages over a given number n of periods (n = 10, 100, 1000...). These measurements of period are then converted into frequency fluctuations, averaged over $\tau = \frac{n}{10 \text{ Hz}}$, from which the Allan variances $\sigma_y^2(m\tau)$ can be calculated (m = 1, 2, 3...). The dead time between subsequent measurements is one period, i.e. approximately 0.1 s in our case.

The Allan variance measured this way describes the frequency fluctuations of the maser only if the reference oscillator has a superior performance and does not contribute significantly to the measured frequency fluctuations. In the case of the cryogenic hydrogen maser, there are no such reference oscillators available (at least for short averaging times). Nevertheless, with the so-called three-cornered-hat method it is still possible to determine the contribution of the cryogenic maser alone. This method requires two high quality reference oscillators, whose frequency fluctuations are uncorrelated to each other and to the maser. The measured Allan variance is then just the sum of the intrinsic Allan variance of the two frequency sources involved, e.g.

$$\sigma_y^2(\text{maser}, \text{ref. 1}) = \sigma_y^2(\text{maser}) + \sigma_y^2(\text{ref. 1})$$
(6.10)

From the measured values of the three relative stabilities between the three oscillators, the contribution from the maser alone can be deduced:

$$\sigma_y^2(\text{maser}) = \frac{1}{2} \left\{ \sigma_y^2(\text{maser, ref. 1}) + \sigma_y^2(\text{maser, ref. 2}) - \sigma_y^2(\text{ref. 1, ref. 2}) \right\}$$
(6.11)

It is obvious from (6.11) that the three relative stabilities have to be measured with a rather high accuracy, otherwise the uncertainty in σ_y^2 (maser) will be large and it is even possible to obtain negative results for σ_y^2 (maser) from (6.11).

6.4.2 Details of stability measurements

We have used the three-cornered-hat method to estimate the frequency stability of the maser for averaging times between 1s and 10s. The two reference oscillators were very high quality quartz crystal oscillators, on loan from F. Walls at NBS Boulder.⁵ Ideally, the three relative stability measurements should be performed simultaneously, but a lack of the necessary hardware forced us to conduct them sequentially. The set-up for each of the three measurements is shown schematically in fig. 6.9. They are all variations of the 1420 MHz spectrometer shown in fig. 5.9. The dotted boxes indicate the three sources for 1420 MHz or 1420.405...MHz. The output signal of the cryogenic hydrogen maser is first amplified by the cooled GaAs FET preamplifier, followed by a second identical amplifier at room temperature and in the third stage by a Watkins Johnson amplifier that is based on Si bipolar transistors.

The quartz crystal oscillator B5400 with serial number 144 is always used in conjunction with a homemade doubler, a homemade distribution amplifier and the multiplier in the spectrometer to produce a signal at 1420 MHz. The quartz crystal oscillator with serial number 369 is used as the reference oscillator for the HP 8663A synthesizer, a stateof-the-art instrument with exceptionally low residual phase noise close to the carrier.

The signals are mixed down to approximately 405 kHz with the image rejection mixer and finally mixed down to 10 Hz with a simple double balanced mixer. The local oscillator signal at 405 kHz is synthesized by the HP 3330A instrument from one of the quartz crystal oscillator outputs.

With the measurement scheme shown in fig. 6.9, any phase fluctuation produced in the frequency doubler and the frequency multiplier ($\times 2, \times 71$) or in the HP 8663A synthesizer

⁵We are very grateful to F. Walls for lending us the two best quartz crystal oscillators in possession of the National Bureau of Standards. Both oscillators were Oscilloquartz B5400 oven-controlled crystal oscillators with an AT cut quartz resonator and an output frequency of 5 MHz.



Figure 6.9: Simplified block diagrams of the three separate arrangements of the electronics used to measure the maser frequency stability with the three-cornered-hat method. Not shown are isolators and directional couplers at the output of maser, the amplifier following $\times 71$ multiplier and filters in the 405 kHz section for reason of clarity.

Chapter 6. Experimental Results

will increase the apparent frequency instabilities of the quartz crystal oscillators, but should not affect the deduced stability of the maser. However, excessive phase noise in the three amplifiers following the maser cannot be subtracted out. The low frequency synthesizer HP 3330A can also affect the result because any excessive phase noise can only be partially cancelled by the three-cornered-hat method. Systematic phase noise measurements of the amplifiers and of the receiver electronics are presented in section 6.5 where their contribution to the measured frequency stability are also discussed.

6.4.3 Results of the stability measurements

With each of the three set-ups shown in fig. 6.9 we measured the relative frequency fluctuations, averaged over 1 s, for more than half an hour. The input power levels at the image rejection mixer were kept the same for all three measurements. The maser bulb was operated at a temperature of 600 mK, the maser cavity was critically coupled to the preamplifier and the power level of the maser signal at the input of the preamplifier varied between 1.5×10^{-13} W and 1.0×10^{-13} W as the hydrogen atoms recombined slowly during the measurements. From the measured relative frequency fluctuations, we then calculated the Allan variances for $\tau = 1$ s, 3 s and 10 s for the three frequency sources and the results are shown in fig. 6.10.

The measured Allan variances for the quartz crystal oscillators do not depend on the averaging time τ between 1s and 10s. As shown in appendix C, this is characteristic of flicker frequency noise. This is the expected noise type for crystal oscillators. The fractional frequency fluctuations of the cryogenic hydrogen maser decrease from $6.3 \pm 3.7 \times 10^{-14}$ at $\tau = 1$ s to $2.8 \pm 13.1 \times 10^{-14}$ at $\tau = 3$ s. At $\tau = 10$ s, the value obtained for σ_y^2 (maser) by (6.11) is negative and only the upper limits of the 1 σ uncertainty of σ_y (maser, 10 s) are indicated in fig. 6.10. It is obvious that for longer averaging times τ , the ratio of the stability of the cryogenic maser to the stability of the two crystal



Figure 6.10: Fractional frequency fluctuations (square root of Allan variance) $\sigma_y(\tau)$ of the cryogenic hydrogen maser (diamonds) and of the two Oscilloquartz crystal oscillators (circles: S/N 144; cross: S/N 369), determined by the three-cornered hat method as described in text.

oscillators becomes too large for this technique to be practical, unless reference oscillators with stabilities comparable to the maser are available.

We can estimate the contributions of the intrinsic thermal noise and the electronic noise (white phase noise) to the total frequency fluctuations of the present maser with equations (4.1) and (4.8). We can use (4.8) directly since there is dead time between the measurements and there is one simple double mixer in our receiver, as was assumed in the derivation of (4.8). The presence of the image rejection mixer in the first stage of the heterodyne receiver does not increase the effective noise bandwidth and no correction factors are thus necessary in (4.8). With the relevant parameters for our cryogenic maser $(T = 0.6 \text{ K}, Q_{atom} \simeq 10^9, P_{amp} \simeq 1.2 \times 10^{-13} \text{ W}, P = 2P_{amp}, B = 5.1 \text{ Hz}, T_N \simeq 25 \text{ K})$, we obtain

$$\frac{\delta f}{f} = \left[\left(\frac{4.15 \times 10^{-15}}{\sqrt{\tau}} \right)^2 + \left(\frac{1.36 \times 10^{-14}}{\tau} \right)^2 \right]^{\frac{1}{2}}$$
(6.12)

This is compared with the measured performance in fig. 6.11. Also included in this figure are the measured stability of a state-of-the-art room temperature hydrogen maser [1], the measured performance of the superconducting cavity stabilized oscillator [73] and the prediction of a full size cryogenic hydrogen maser as given in eq. (4.11). The four open circles are measurements of the frequency fluctuations between the cryogenic maser and a single quartz crystal oscillator (S/N 144) and are therefore only upper limits of the frequency instabilities of the maser.

Fig. 6.11 shows that already for our prototype maser the short-term stability is superior to those of the best conventional masers. This is very encouraging. However, the measured fluctuations of the cryogenic maser are larger than predicted by (6.12). The most likely source of this discrepancy are noise contributions of the reference oscillators and of the receiver electronics that could not be cancelled completely in our scheme of stability measurements. This is analyzed in more detail below.



Figure 6.11: Relative frequency fluctuations σ_y versus averaging time τ for cryogenic hydrogen maser and other high quality frequency sources. Diamonds: stability of cryogenic maser measured by three-cornered-hat method. Circles: relative frequency fluctuations between cryogenic maser and the better of the quartz crystal oscillator (Oscilloquartz B5400, S/N 144). This is only an upper limit for the cryogenic maser. Curve *a*: predictions for cryogenic maser, see eq. (6.12). Curve *b*: measured performance of a state-of-the-art room temperature hydrogen maser [1]. Curve *c*: predictions for full size cryogenic maser, see eq. (4.11). Curve *d*: measured performance of superconducting cavity stabilized oscillator [73]. Great care is required to avoid interference of the maser signal with pickup signals (e.g. 60 Hz). An interference signal (with power $P_{interf.}$) weakly modulates the maser signal (P_{maser}) and gives rise to a contribution to the measured Allan variance of size

$$\left. \frac{\delta f}{f} \right|_{interf} = \left(\frac{P_{interf}}{P_{maser}} \right)^{\frac{1}{2}} \frac{1}{\omega_0 \tau} \tag{6.13}$$

All interference signals have to be therefore more than 78 dB weaker than the maser signal, otherwise they dominate the preamplifier white phase noise in our prototype maser. For this reason, it would have been advantageous to measure the frequency stability of the maser in the frequency domain, where such interference effects can be easily identified.⁶ However, it is unlikely that we were bothered by this problem since we used a final filter that suppressed interference signals outside its narrow bandwidth.

In 1986, Mann et al. [74] reported measurements on the flicker phase noise in GaAs MESFET amplifiers, operated at X-band, in the temperature range between 300 K and 2 K. On cooling of their devices, they observed a dramatic increase in phase noise. This was an alarming result in view of application to the cryogenic hydrogen maser. Excessive flicker phase noise in the preamplifier of the maser results in a frequency stability σ_y that is approximately proportional to τ^{-1} , as shown in appendix C, and could explain the difference between the measured and predicted frequency stability of the prototype maser. The contribution to σ_y due to flicker phase noise, unlike the other noise sources considered, cannot be decreased by a higher power level in the maser signal. As a consequence, the short-term frequency stability of a full size maser would not be any higher than of the prototype maser if flicker phase noise is the limiting noise source. For this reason, it was essential to measure the low temperature flicker phase noise of our preamplifier and estimate the importance of its contribution to the frequence fluctuations. The results together with measurements of the phase noise of other components of the

⁶This was first pointed out to us by F. Walls, private communication.

receiver are presented in the following section.

6.5 Flicker phase noise measurements

6.5.1 Maser preamplifier

It was mentioned already in chapter 5 that our two-stage 1.42 GHz preamplifier is based on the design by Williams et al. [64] and uses a Mitsubishi MGF 1412 and an MGF 1402 FET in the first and second stages, respectively. The 3 dB bandwidth is 395 MHz, and the gain is 18.4 dB at room temperature, increasing to 21.0 dB at 4.2 K. The 1 dB gain compression point is -0.7 dBm. As first built, the preamplifier had a minimum noise temperature of 7 K at a physical temperature of 4.2 K. However, the maser microwave cavity represents a highly reactive input load and caused the amplifier to oscillate at low temperatures. Damping was added to the input circuit, which increased the measured noise temperature to $T_N = 20$ K at 4.2 K and $T_N = 90$ K at 300 K.

A conventional phase bridge set-up [75] was used for the phase noise measurements. The amplifier, with a 30 dB attenuator at its input, was placed inside a glass Dewar flask. The signal transmitted through this arm of the bridge was mixed with the reference signal, set accurately to quadrature with a phase shifter, and then amplified and fast-Fourier-transformed on an HP 3582A spectrum analyzer covering the range of sideband frequencies between 0.15 Hz and 1 kHz. After each measurement the background of the measuring system was recorded and subtracted from the measurement.

In our experiment we varied the power in the carrier P_c between -20 dBm and -30 dBm and observed a noise power proportional to P_c . This results in a spectral density of phase fluctuations $S_{\varphi}(f)$ independent of P_c : it shows that, in this range of P_c , the measured values of $S_{\varphi}(f)$ are proportional to the spectral density of fluctuations in transit time of the signal through the device, the quantity of interest. At higher power levels effects due to nonlinearities may become visible. At lower power levels and large sideband frequencies, additive white noise starts to become important and contributes to the measured spectral density of phase fluctuations $S_{\varphi}(f)$ according to

$$S_{\varphi}(f) = \frac{kT_N}{P_c} \tag{6.14}$$

Measured spectral densities of phase fluctuations of the amplifier are shown in fig. 6.12 for different physical temperatures. At 77 K the amplifier was surrounded by nitrogen exchange gas, whereas at temperatures of 4.2 K and below the device was immersed in the helium bath. All the data shown were taken with an input power level of -20 dBm and a carrier frequency of 1.42 GHz.

When the amplifier is cooled to 77 K, the magnitude of $S_{\varphi}(f)$ does not change very much, but for frequencies larger than 10 Hz the frequency dependence changes from an f^{-1} to an $f^{-1.5}$ behavior. At 4.2 K we note a substantial increase in $S_{\varphi}(f)$ and an anomalous frequency dependence for frequencies less than 100 Hz. We attribute this to the helium cooling bath, based on the following observations. The phase fluctuations continue to increase when the temperature is lowered further. As soon as the λ -point of helium is passed and the helium stops bubbling, the phase noise drops abruptly to a level below the room temperature values. For sideband frequencies between 10 Hz and 100 Hz a drop of about 40 dB in $S_{\varphi}(f)$ can be seen in fig. 6.13 at the superfluid transition temperature of helium. Below 2 K the temperature dependence is weak and there are no signs of a low temperature divergence as in the amplifier of Mann et al. [74].

Flicker phase noise in the maser preamplifier of amplitude

$$S_{\varphi}(f) = 10^{-9} f^{-1} \left[\frac{\text{rad}^2}{\text{Hz}} \right]$$
 (6.15)

causes short-term fluctuations in the maser frequency that approximately equal the contributions due to white phase noise in our cryogenic maser (see eq. 6.12). This noise level









Figure 6.13: Temperature dependence of spectral density of phase fluctuations (relative to $1 \frac{\text{rad}^2}{\text{Hz}}$) of maser preamplifier for sideband frequencies of 10 Hz, 40 Hz and 100 Hz. Broken line indicates superfluid transition temperature of helium, and connecting lines are a guide to the eye.

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(6.15) is indicated in fig. 6.12 by the dotted line. Even with the preamplifier operated in the boiling ⁴He bath at 4.2 K, the measured flicker phase noise lies below (6.15) and therefore does not contribute significantly to the short-term frequency fluctuations in our maser. However, in a full size cryogenic maser the requirements on flicker phase noise are more than 20 dB more stringent. In order to realize the exceedingly high short-term stability predicted in (4.11), it will be necessary to avoid the excess phase noise caused by the boiling helium, either by operating the preamplifier at temperatures below the superfluid transition of ⁴He or by placing it outside the cryogenic liquid.

6.5.2 Phase noise of receiver

The phase noise of the Watkins Johnson amplifier, which was used at the input of the image rejection mixer for all stability measurements, was determined with the same method as described above. From the results for this amplifier, shown in fig. 6.14 (diamonds), it can be concluded that it does not degrade the performance of our prototype maser.

The HP 8663A synthesizer does not increase the measured frequency fluctuations of the cryogenic maser if its phase noise for carrier frequencies f_c of 1420 MHz and 1420.405 MHz are equal and constant over time. It does however affect the deduced stability of the quartz crystal oscillator (S/N 369). With the available instrumentation we were not able to measure the phase noise of this synthesizer at the two relevant carrier frequencies, but we recorded the phase noise for $f_c = 1280$ MHz (circles, fig. 6.14), which is expected to be of comparable size. For this measurement, the reference signal was obtained by frequency doubling of the 640 MHz auxiliary output of the synthesizer. This output signal is generated by direct frequency multiplication from the 10 MHz reference signal, without any phase locked loops, and the manufacturerer paid special attention to low additional phase noise in the multiplication process. The reference signal at 1280 MHz is then mixed with the synthesized signal at the same frequency from the main output



Figure 6.14: Measured spectral densities of phase fluctuations of various components of the maser receiver for sideband frequencies between 0.1 Hz and 10 kHz. Diamonds: Watkins Johnson amplifier, $f_c = 1420$ MHz; circles: HP 8663A synthesizer, $f_c = 1280$ MHz; triangles: homemade multiplier chain (×2, ×71), $f_c = 1420$ MHz; squares: HP 3330A low frequency synthesizer, $f_c = 10$ MHz. The dotted line represents eq. (6.15) and indicates the level of flicker phase noise that causes the same frequency fluctuations in the present prototype maser as the anticipated contribution of the preamplifier white phase noise (see eq. 6.12).

of the instrument and analyzed on the spectrum analyzer as before. From the results shown in fig. 6.14, it appears unlikely that the observed excess frequency fluctuations of the maser are caused by the HP 8663A synthesizer.

The phase noise of the multiplier chain (×2, ×71) which is part of the spectrometer was measured at a carrier frequency of 1420 MHz by comparing the signal from the multiplier chain with the signal form the HP 8663A synthesizer. The obtained values for $S_{\varphi}(f)$ (triangles, fig. 6.14) lie by more than 10 dB above the noise level (6.15), indicated by the dotted line. Ideally, this phase noise does not affect the stability of the maser obtained with the three-cornered-hat method and only degrades the apparent stability of the quartz crystal oscillator with serial number 144. However, if the level of phase noise in the multiplier chain changes between the two stability measurements involving this quartz crystal oscillator, the noise contribution from the multiplier chain can no longer be cancelled completely and it will affect the deduced stability of the maser. Such changes could be caused for instance by a slight change of load impedance or by temperature changes.

Although the frequency of the HP 3330A synthesizer (405 kHz) is more than 3500 times smaller than the maser frequency, the requirements on its phase noise are still approximately given by (6.15). The phase noise at the output of an ideal mixer is the sum of the phase noise of the two inputs. Therefore the phase noise of the HP 3330A contributes fully to each of the three relative stability measurements in the three-cornered-hat method. As can be seen from (6.11), only half of this contribution can be cancelled in our scheme. We have measured the phase noise of the HP 3330A synthesizer at a carrier frequency of 10 MHz (squares, fig. 6.14), which should be an upper limit for $S_{\varphi}(f)$ at $f_c = 405$ kHz. At low sideband frequencies it falls below the dotted line and should therefore contribute only insignificantly to the measured frequency fluctuations of the maser.

From the data shown in fig. 6.14, it is clear that the measurements of frequency stability by the three-cornered-hat method requires great care. It is likely that some of the frequency fluctuations attributed to the maser are in fact caused by phase fluctuations in the measuring electronics. It would be very advantageous to be able to perform the three necessary measurements simultaneously. Furthermore, the phase noise of many of the components is too high for a full size cryogenic maser, even under ideal measuring conditions, because the requirements of $S_{\varphi}(f)$ are more than 20 dB more stringent. In that case, it will be necessary to improve these components, or alternatively, construct more than one maser and compare them directly with as little additional electronics as possible.

Chapter 7

Computer simulation of the maser

We have developed a model that simulates the operation of the maser in quasi-equilibrium. This model is used, for instance, to calculate the dependence of the output power or the population inversion on such parameters as the density in the maser bulb, the temperatures and different relaxation rates in the four regions or the pumping power in the state selector. This simulation is not only useful for the analysis of data obtained with our maser and to understand some of the dependences quantitatively, but it should be also a valuable tool to design an improved second generation cryogenic maser.

7.1 Model of the maser

The maser is divided into four distinct regions which are connected as shown in fig. 7.1. The four regions represent:

- 1. Region in high field with spin-relaxing magnetic foil.
- 2. State selector microwave cavity.
- 3. Atom transport tube between state selector magnet and maser cavity.

4. Maser cavity.

Within a particular region all spatial inhomogeneities are neglected and it is assumed that each of the regions can be characterized by a set of parameters which include temperature, magnetic field and relaxation rates as discussed below. We then write down the



Figure 7.1: Model of the cryogenic maser used in the computer simulation.

rate equations for the 16 densities $n_k^{(i)}$, where k indicates the spin state of the hydrogen atom $(k = |a\rangle, |b\rangle, |c\rangle, |d\rangle)$ and i labels the region (i = 1, 2, 3, 4). In the rate equations we include the following processes:

- Flow between the different regions.
- One-body spin relaxation, which is induced by the magnetic foil in the state selector and by magnetic impurities on the walls in the rest of the system.
- Microwave pumping at the ESR transition in the state selector cavity.
- Collisions between hydrogen atoms. The dominant process among them is spin exchange relaxation.
- Maser action.

In this simulation, recombination of hydrogen atoms into molecules is neglected, as is the exchange of atoms through the pinhole in the Mylar window between the state selector cavity and the waveguide above it. This is justified because the associated time constants are much longer than the dominant time constants of the processes included in our model. As a consequence, for any given total number of hydrogen atoms in the maser, the hydrogen atoms are in a quasi-equilibrium state, in which the distribution of the hydrogen densities between the different regions and the population of the spin states are determined only by the processes considered in our model. Note that all processes included in the simulation conserve the total number of atoms and that this quasi-equilibrium state is therefore the stationary solution of our rate equations. The exchange of atoms with those in the waveguide affects the total number of atoms in the maser and the slow decay of it. However, the simulation does not attempt to model this time evolution and we treat the total number of atoms in the maser as a free parameter.

7.1.1 Flow

The mean free path of a hydrogen atom at the operating temperatures of the cryogenic maser is limited by collisions with ⁴He atoms in the gas phase. Since the mean free path is larger than the typical dimensions of the orifices connecting the different regions in our maser, the flow between the four volumes is governed by effusion. If the magnetic field differs on the two sides of the orifice, we have to include an appropriate Boltzmann factor to take the difference in spin energy into account. The contribution to the rate equations due to flow for the density of atoms in the state $|k\rangle$ (k = a, b, c, d) and in volume *i* is

$$\frac{d}{dt}n_k^{(i)}\Big|_{flow} = -\left[\sum_{j=1,j\neq i}^4 \Gamma_k^{(i\rightarrow j)}\right]n_k^{(i)} + \left[\sum_{j=1,j\neq i}^4 \Gamma_k^{(i\leftarrow j)}n_k^{(j)}\right]$$
(7.1)

where the outgoing and the ingoing rate constants are

$$\Gamma_k^{(i \to j)} = \frac{A_{ij} v_i}{4V_i} \min\left\{1, \exp\left(\frac{E_k^{(i)} - E_k^{(j)}}{kT_i}\right)\right\}$$
(7.2)

$$\Gamma_k^{(i \leftarrow j)} = \frac{A_{ij} v_j}{4V_i} \min\left\{1, \exp\left(\frac{E_k^{(j)} - E_k^{(i)}}{kT_j}\right)\right\}$$
(7.3)

In these expressions, A_{ij} is the effective cross section of the orifice between region iand j, $v_i = \left(\frac{8kT_i}{\pi m_H}\right)^{\frac{1}{2}}$ is the average thermal velocity of hydrogen atoms in region i, V_i is the volume of region i and $E_k^{(i)}$ is the spin energy of a hydrogen atom in state $|k\rangle$ and in volume i.

It is easy to see that in equilibrium, the ratio of the total densities of hydrogen atoms in the maser bulb $n_H^{(4)}$ and in the atom transport tube $n_H^{(3)}$ only depends on the respective temperatures T_4 and T_3 .

$$\frac{n_H^{(4)}}{n_H^{(3)}} = \left(\frac{T_3}{T_4}\right)^{\frac{1}{2}}$$
(7.4)

This ratio is independent of all the relaxation rates and holds both when the state selector is on or off. However, for all other ratios between the total densities in different regions we cannot find such a simple relationship. These ratios depend on the relative populations and therefore on the relaxation and pumping rates because the relevant Boltzmann factors are not all equal.

7.1.2 One-body spin relaxation

Interactions between a single hydrogen atom and inhomogeneous magnetic fields can lead to spin relaxation. In high field (volume 1 and 2), we only consider the transitions $a \rightleftharpoons b$ and $c \rightleftharpoons d$, which are essentially NMR transitions (see appendix A).

$$\frac{d}{dt}n_a^{(i)}\Big|_o = -\frac{d}{dt}n_b^{(i)}\Big|_o = -g_{1\uparrow}^{(i)}n_a^{(i)} + g_{1\downarrow}^{(i)}n_b^{(i)}$$
(7.5)

$$\frac{d}{dt}n_{c}^{(i)}\Big|_{o} = -\frac{d}{dt}n_{d}^{(i)}\Big|_{o} = -g_{2\uparrow}^{(i)}n_{c}^{(i)} + g_{2\downarrow}^{(i)}n_{d}^{(i)}$$
(7.6)

where i is either 1 or 2. The up- and down-rate constants are related by Boltzmann factors, e.g.

$$g_{1\uparrow}^{(i)} = g_{1\downarrow}^{(i)} \exp\left\{\frac{E_a^{(i)} - E_b^{(i)}}{kT_i}\right\}$$
(7.7)

In our maser, we tried to optimize the magnetic foil placed in volume 1 such as to produce a large rate constant $g_1^{(1)}$. The two rate constants $g_1^{(1)}$ and $g_2^{(1)}$ can in principle be calculated with the theory developed by Berlinsky et al. [76] if the exact size and distribution of the magnetic particles on the foil is known. Since we do not have this information, we treat the rates as adjustable parameters. Furthermore, the two NMR transition frequencies are roughly equal ($\nu_{ab} = 0.78 \text{ GHz}$, $\nu_{cd} = 0.64 \text{ GHz}$ at B = 1.4 T) and their transition probabilities are practical identical, so that we set $g_{1\downarrow}^{(1)} = g_{2\downarrow}^{(1)}$ in our simulation.

In the state selector cavity (volume 2), we tried to minimize the one-body spin relaxation. In order to be able to study the effect of magnetic impurities that might be impeded in the cavity walls, we still include the relaxation (7.5) and (7.6) for volume 2 in our simulation, but it can be turned off easily if desired.

In low field (volume 3 and 4), the considered one-body transitions are cross relaxations within the three upper hyperfine states (rate constant Γ_c) and transitions between the upper three states and the lower state. In general, the rates for these transitions with $\Delta m = 0$ and $|\Delta m| = 1$ can be different and we denote the rate constants Γ_{l0} and Γ_{l1} , respectively.

$$b \stackrel{\Gamma_{c}}{\leftarrow} c \stackrel{\Gamma_{c}}{\leftarrow} d$$
$$c \stackrel{\Gamma_{l0}}{\leftarrow} a$$
$$d \stackrel{\Gamma_{l1}}{\leftarrow} a \qquad b \stackrel{\Gamma_{l1}}{\leftarrow} a$$

The ratio of the up- and down-rates for the transitions between the upper and lower hyperfine states are given by the appropriate Boltzmann factor analogous to (7.7). In matrix form, the contribution of one-body relaxation processes to the rate equations in volume i = 3, 4 is then

$$\frac{d}{dt} \begin{pmatrix} n_{a}^{(i)} \\ n_{b}^{(i)} \\ n_{c}^{(i)} \\ n_{d}^{(i)} \end{pmatrix}_{o} = \begin{pmatrix} -\Gamma_{l0\uparrow}^{(i)} - 2\Gamma_{l1\uparrow}^{(i)} & \Gamma_{l1\downarrow}^{(i)} & \Gamma_{l0\downarrow}^{(i)} & \Gamma_{l1\downarrow}^{(i)} \\ \Gamma_{l1\uparrow}^{(i)} & -\Gamma_{c}^{(i)} - \Gamma_{l1\downarrow}^{(i)} & \Gamma_{c}^{(i)} & 0 \\ \Gamma_{l0\uparrow}^{(i)} & \Gamma_{c}^{(i)} & -2\Gamma_{c}^{(i)} - \Gamma_{l0\downarrow}^{(i)} & \Gamma_{c}^{(i)} \\ \Gamma_{l1\uparrow}^{(i)} & 0 & \Gamma_{c}^{(i)} & -\Gamma_{c}^{(i)} - \Gamma_{l1\downarrow}^{(i)} \end{pmatrix} \begin{pmatrix} n_{a}^{(i)} \\ n_{b}^{(i)} \\ n_{c}^{(i)} \\ n_{d}^{(i)} \end{pmatrix}_{o}$$
(7.8)

In our case, these relaxations are most likely caused by magnetic impurities in the walls. For a random orientation, we expect $\Gamma_{l1} = \Gamma_{l0}$.

In the maser cavity (volume 4), we can measure these rate constants by pulsed spectroscopy on the maser transition. It was already shown in chapter 6 that in the low density limit, the population difference $n_a^{(4)} - n_c^{(4)}$ relaxes to its equilibrium value after a π -pulse according to the sum of two exponentials (see fig. 6.2 and eq. (6.1)). The two inverse time constants are $\Gamma^{(4\to3)} + 4\Gamma_l^{(4)}$ and $\Gamma^{(4\to3)} + 3\Gamma_c^{(4)} + \Gamma_l^{(4)}$. In this analysis, we assumed that $\Gamma_{l1} = \Gamma_{l0}$ and we neglected the difference between the up- and down-rates. With the same approximations, it can be shown [77] that in the low density limit T_2 is related to the relaxation rates by

$$\lim_{\substack{n_H^{(4)} \to 0}} T_2^{-1} = \Gamma^{(4 \to 3)} + \Gamma_c^{(4)} + 2\Gamma_l^{(4)}$$
(7.9)

From the measurement of the longitudinal and transverse relaxation in the limit of zero density, it is therefore possible to determine $\Gamma^{(4\to3)}$, $\Gamma_c^{(4)}$ and $\Gamma_l^{(4)}$. For the one-body relaxation rates in the atom transport tube, we do not have any direct information and we are forced to treat them as adjustable parameters.

7.1.3 Microwave pumping in the state selector cavity

In the state selector cavity, the applied microwave radiation induces resonant transitions between the $|b\rangle$ and $|c\rangle$ states:

$$\frac{d}{dt}n_b^{(2)}\Big|_{mw} = -\frac{d}{dt}n_c^{(2)}\Big|_{mw} = -w_{bc}\left(n_b^{(2)} - n_c^{(2)}\right)$$
(7.10)

The transition rate w_{bc} depends on the power level P_{mw} of microwaves delivered to the cavity, the loaded quality factor Q of the cavity and the magnetic field inhomogeneity $\frac{\Delta B}{B}$ inside the cavity:

$$w_{bc} = \frac{\sqrt{2\pi}\mu_0 \gamma_e}{\omega_{bc}} \frac{Q}{V^{(2)}B} \left(\frac{\Delta B}{B}\right)^{-1} P_{mw}$$
(7.11)

If we decrease in our maser the magnetic field around the state selector from 1.433 T to 1.382 T, we bring the $|a\rangle$ to $|d\rangle$ ESR transition instead of the $|b\rangle$ to $|c\rangle$ transition into resonance with the microwave radiation at 39.46 GHz. The transition probability w_{ad} is practically equal to w_{bc} , except that we have to adjust B in (7.11) to the lower value.

We have included the possibility of driving this other ESR transition in our simulation as an option. In this case, (7.10) has to be replaced of course by:

$$\frac{d}{dt}n_a^{(2)}\Big|_{mw} = -\frac{d}{dt}n_d^{(2)}\Big|_{mw} = -w_{ad}\left(n_a^{(2)} - n_d^{(2)}\right)$$
(7.12)

7.1.4 Spin exchange relaxation

For the effects of collisions between hydrogen atoms, we make use of the theoretical results from the group of Prof. Verhaar. In high field (volume 1 and 2), the dominant spin exchange process is

$$b+d \rightleftharpoons a+c$$

with a calculated rate $G_{bd\to ac} = 4.3 \times 10^{-13} \frac{\text{cm}^3}{\text{s}}$ at a field of 1.4 T [49]. At this field, Stoof et al. [49] found that the relaxation rates of all other spin exchange and dipole relaxation processes are more than two orders of magnitude smaller. Therefore we neglect these additional processes in our simulation and we obtain the following spin exchange contribution for i = 1, 2:

$$\frac{d}{dt}n_{a}^{(i)}\Big|_{se} = \frac{d}{dt}n_{c}^{(i)}\Big|_{se} = -\frac{d}{dt}n_{b}^{(i)}\Big|_{se} = -\frac{d}{dt}n_{d}^{(i)}\Big|_{se} = -\frac{d}{dt}n_{d}^{(i)}\Big|_{se} = G_{bd\to ac}n_{b}^{(i)}n_{d}^{(i)} - G_{ac\to bd}n_{a}^{(i)}n_{c}^{(i)}$$
(7.13)

The forward and backward rates $G_{bd \rightarrow ac}$ and $G_{ac \rightarrow bd}$ are related by Boltzmann factors:

$$G_{ac \to bd} = G_{bd \to ac} \exp\left\{\frac{E_a + E_c - E_b - E_d}{kT}\right\}$$
(7.14)

For zero field (volume 3 and 4) we use the results of Koelman et al. [46]. For $0.2 \text{ K} \leq T \leq 0.6 \text{ K}$, the rate constants for the three dominant processes can be approximated by

$$b + d \rightleftharpoons a + a \qquad G_{bd \to aa} = 4.5 \times 10^{-13} \frac{\text{cm}^3}{\text{s}} \left(\frac{T}{0.6 \text{ K}}\right)^{0.08}$$
$$c + c \rightleftharpoons b + d \qquad G_{cc \to bd} = 4.1 \times 10^{-13} \frac{\text{cm}^3}{\text{s}} \left(\frac{T}{0.6 \text{ K}}\right)^{0.19}$$
$$c + c \rightleftharpoons a + a \qquad G_{cc \to aa} = 2.2 \times 10^{-13} \frac{\text{cm}^3}{\text{s}} \left(\frac{T}{0.6 \text{ K}}\right)^{0.07}$$

The inverse reaction rates are related to the rates above by the appropriate Boltzmann factor. In volume 3 and 4, we have then the following contribution to the rate equation:

$$\frac{d}{dt} \begin{pmatrix} n_{a}^{(i)} \\ n_{b}^{(i)} \\ n_{c}^{(i)} \\ n_{d}^{(i)} \end{pmatrix}_{se} = \begin{pmatrix} -2G_{aa \rightarrow bd} - 2G_{aa \rightarrow cc} & 2G_{bd \rightarrow aa} & 2G_{cc \rightarrow aa} \\ G_{aa \rightarrow bd} & -G_{bd \rightarrow aa} - G_{bd \rightarrow cc} & G_{cc \rightarrow bd} \\ 2G_{aa \rightarrow cc} & 2G_{bd \rightarrow cc} & -2G_{cc \rightarrow aa} - 2G_{cc \rightarrow bd} \\ G_{aa \rightarrow bd} & -G_{bd \rightarrow aa} - G_{bd \rightarrow cc} & G_{cc \rightarrow bd} \end{pmatrix} \begin{pmatrix} n_{a}^{(i)^{2}} \\ n_{b}^{(i)} n_{d}^{(i)} \\ n_{c}^{(i)^{2}} \end{pmatrix}$$

$$(7.15)$$

7.1.5 Maser action

The rate of change of $n_a^{(4)}$ and of $n_c^{(4)}$ due to maser action can be obtained from eq. (2.20):

$$\frac{d}{dt} \left(n_a^{(4)} - n_c^{(4)} \right)_{maser} = i(\gamma_e + \gamma_p) \mu_0 H_z \cos \omega t (\rho_{ac} - \rho_{ca}) n_H^{(4)}$$
(7.16)

It depends on the rf magnetic field H_z in the maser cavity and the off-diagonal element ρ_{ac} of the density matrix. These quantities have to be determined in a self-consistent way as was already outlined in chapter 2. Equation (7.16) can therefore not be applied directly in our simulation without including additional equations for H_z and ρ_{ac} . A more direct approach would be based on the relationship

$$\frac{d}{dt}n_a^{(4)}\Big|_{maser} = -\frac{d}{dt}n_c^{(4)}\Big|_{maser} = \frac{1}{V_4}\frac{P}{\hbar\omega}$$
(7.17)

between the rate of change of the two densities and the power emitted by the atoms. However, the expression (2.41) for the power P which was derived in chapter 2, is only valid if a well defined T_1 exists. In the previous chapter is was shown that in the present maser we need at least two time constants to describe the longitudinal relaxation at low densities, and spin exchange relaxation complicates the situation at higher densities even more.

Nevertheless, it is possible to take maser action self-consistently into account. Since we are only interested in the stationary solution of the rate equations, we can use directly

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expression (2.33).¹ In steady state, the population inversion inside the maser bulb for stable maser oscillations has to be

$$n_{c}^{(4)} - n_{a}^{(4)} = \frac{4(1 + \Delta^{2})}{\hbar(\gamma_{e} + \gamma_{p})^{2} \mu_{0} \eta Q_{l}} \frac{1}{T_{2}}$$
(7.18)

In the derivation of this expression, no assumptions on the longitudinal relaxation were made. It was only assumed that the resonance line can be approximated by a Lorentzian line shape and can therefore be described by the time constant T_2 . In terms of the relaxation rates discussed above, the line width $\Gamma = T_2^{-1}$ is given by

$$T_{2}^{-1} = \Gamma^{(4\to3)} + \Gamma_{c}^{(4)} + 2\Gamma_{l}^{(4)} +$$

$$+ \bar{\sigma}_{0} \langle v \rangle \left(n_{c}^{(4)} - n_{a}^{(4)} \right) + \bar{\sigma}_{1} \langle v \rangle \left(n_{c}^{(4)} + n_{a}^{(4)} \right) + \bar{\sigma}_{2} \langle v \rangle \left(n_{a}^{(4)} + n_{b}^{(4)} + n_{c}^{(4)} + n_{d}^{(4)} \right)$$

$$(7.19)$$

The density independent line width is identical to (7.9) and it was assumed that inhomogeneities in the bias field which only dephase the precessing magnetization without causing transitions between the hyperfine states are unimportant. The spin exchange broadening was discussed earlier (see eq. (3.12) and table 3.2).

7.1.6 Overall rate equations

For the overall rate equations that simulate the maser, it is best to perform a transformation of variables from the variables $\{n_a^{(i)}, n_b^{(i)}, n_c^{(i)}, n_d^{(i)}\}$ to the new variables $\{n_a^{(i)} + n_c^{(i)}, n_a^{(i)} + n_c^{(i)}, n_a^{(i)} + n_c^{(i)}, n_a^{(i)} + n_d^{(i)}, n_b^{(i)} - n_d^{(i)}\}$. From the previous discussion, it is straightforward to derive the set of 16 coupled nonlinear equations that describe the steady state of the

¹This approach was first suggested to us by Prof. S. Crampton.

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maser in the case of no maser action (i = 1, 2, 3, 4)

To this set of equations, we add the constraint that the total density in the maser bulb $n_H^{(4)}$ or in other cases that the total number of hydrogen atoms in the whole maser is fixed at some predetermined value. We then solve these equations numerically with the Newton-Raphson method [78]. If the population inversion in the maser bulb $n_c^{(4)} - n_a^{(4)}$ exceeds the critical value given by the right hand side of equation (7.18), the maser will start to oscillate. Note that $\frac{d}{dt}(n_a^{(4)} - n_c^{(4)})$ is the only rate equation which is affected by maser oscillation. We can therefore simply replace the equation in (7.20) for $\frac{d}{dt}(n_a^{(4)} - n_c^{(4)})$ by equation (7.18) and obtain so the steady state density distribution of the oscillating maser.

7.2 Results of the simulation

7.2.1 Choice of parameters

It was already mentioned that several parameters needed in the simulation cannot be measured directly in the present apparatus, in particular the one-body rates outside the maser bulb. We can adjust these rates so that the calculated density dependence of the output power agrees with the measured dependence. However, this method does not yield a unique set of parameters and we have not tried to minimize the discrepancies between measurement and simulation in the examples shown below.

To illustrate the dependence on the relaxation rate of the magnetic foil in the state selector, we show some numerical results where this rate is varied between 0.15 s^{-1} and

 $1.0 \,\mathrm{s^{-1}}$. For this simulation, the temperature of the maser bulb is chosen to be 509 mK and the temperature of the state selector to be 465 mK. This corresponds to the condition of the second run shown in fig. 6.5 for the power output and of the data in fig. 6.3 which shows the dependence of the maser resonance line width on the density $n_H^{(4)}$ and which has been used to calibrate the density determination.

The relevant flow rates can be calculated from the known volumes of the different regions and from the cross sections of the orifices connecting them. For the maser bulb holding time we obtain $T_b = 0.6 \,\mathrm{s} \,(= 1/\Gamma^{(4\to3)})$. This is consistent with the analysis of the relaxation rates of the transverse and longitudinal magnetization \tilde{M}_r and \tilde{M}_z as discussed before. From this analysis we also obtain $\Gamma_c^{(4)} = 1.94 \,\mathrm{s}^{-1}$. The rates $\Gamma_l^{(4)}$ are quite small and cannot be reliably extracted from the experimental data. For the results shown, we set $\Gamma_{l1}^{(4)} = \Gamma_{l0}^{(4)} = 0.33 \,\mathrm{s}^{-1}$. In the atom tube, we have to choose some more or less arbitrary values for $\Gamma_l^{(3)}$ and $\Gamma_c^{(3)}$. The rather large value of $\Gamma_c^{(4)}$ is presumably due to some magnetic dirt on the walls. The copper tube and the brass cone forming the region 3 have been coated carefully with a layer of non-magnetic Stycast 1266 and for this reason, we do not expect to have one-body relaxation rates as large as those encountered in the maser bulb. In the simulation shown, we set $\Gamma_{l0}^{(3)} = \Gamma_{l1}^{(3)} = 0.01 \, \mathrm{s}^{-1}$ and $\Gamma_c^{(3)} = 0.1 \, \text{s}^{-1}$. The power of the microwave signal injected into the state selector cavity is approximately 50 μ W, resulting in $w_{bc} \simeq 2 \times 10^2 \, \text{s}^{-1}$. This is two to three orders of magnitude larger than the relaxation rates of the magnetic foil used in the calculation and therefore the results of the simulation are not sensitive to variations in the power level P_{mw} . This is in agreement with experimental results.

7.2.2 Power output of maser and state selector efficiency

The power emitted by the atoms is given by

$$P = \frac{1}{2}\hbar\omega V_4 \frac{d}{dt} \left(n_a^{(4)} - n_c^{(4)} \right)_{maser}$$
(7.21)

In steady state, the rate of change of $n_a^{(4)} - n_c^{(4)}$ due to maser action has to be compensated by all the other contributions. The power output (7.21) is thus equal to

$$P = -\frac{1}{2}\hbar\omega V_4 \left[\frac{d}{dt} \left(n_a^{(4)} - n_c^{(4)} \right)_{flow} + \frac{d}{dt} \left(n_a^{(4)} - n_c^{(4)} \right)_o + \frac{d}{dt} \left(n_a^{(4)} - n_c^{(4)} \right)_{se} \right]$$
(7.22)

which can be easily calculated by the expressions given above if the density distribution is known.

In fig. 7.2, the calculated power output of the maser is shown as a function of the total density $n_H^{(4)}$ in the maser bulb for three different relaxation rates $g_1^{(1)} = g_2^{(1)}$ of the magnetic foil. It can be seen that the model describes the experimental data of fig. 6.5 at least qualitatively quite well. For the parameters chosen, it is clear that the magnetic foil relaxation forms a bottleneck. With a more efficient foil, the output power and the range of densities that result in oscillations can be greatly increased. The curvature in the power versus density plots is predominantly caused by spin exchange relaxation in the maser bulb. This was already shown in the simplified theory presented in chapter 2. However, some additional curvature is due to the density dependence of the population inversion $\rho_{cc}^{(3)} - \rho_{aa}^{(3)}$ outside the maser bulb. This is shown in fig. 7.3.

The population inversion in the atom tube is rather low and decreasing rapidly with density. The main mechanism for this decrease is again spin exchange relaxation, but this time in the atom tube. The increased population inversion for the more efficient magnetic foil is the main reason that the power output of the maser is in that case so much larger.



Power Emitted by Atoms (W)



Figure 7.2: Calculated power emitted by atoms versus total density in maser bulb for three different values of the relaxation rate of the magnetic foil as indicated. The values of all the other parameters are discussed in the text.





Figure 7.3: Calculated population inversion $\rho_{cc}^{(3)} - \rho_{aa}^{(3)}$ outside the maser bulb versus total density inside the maser bulb $n_H^{(4)}$ for three different values of the relaxation rate of the magnetic foil as indicated. All the other parameters used are discussed in the text.



Figure 7.4: Calculated values of $\rho_{aa}^{(4)} + \rho_{cc}^{(4)}$ versus total density in maser bulb for three different values of the relaxation rate of the magnetic foil as indicated.

7.2.3 Simulation results relevant for density calibration

Figure 7.4 shows the calculated values for $\rho_{aa}^{(4)} + \rho_{cc}^{(4)}$, the fraction of atoms in the states $|a\rangle$ and $|c\rangle$ in the maser bulb. In chapter 6, we argued that $\rho_{aa}^{(4)} + \rho_{cc}^{(4)}$ has to be close to 0.5 because of the strong cross relaxation present in the maser bulb. This was used in the theoretical prediction of the density dependence of the maser line width which then allowed us to obtain values for the density in the maser bulb. From fig. 7.4 it can be seen that our assumption $\rho_{aa}^{(4)} + \rho_{cc}^{(4)} = 0.5$ is quite well fulfilled and is certainly not the main source of any error in the density determination. The discontinuities in the slope of the curves of fig. 7.4 occur at the lower and upper threshold densities for maser oscillations, where $\rho_{aa}^{(4)} - \rho_{cc}^{(4)}$ also changes discontinuously.

With the computer simulation, we can also study how well the density in the maser



Figure 7.5: Calculated values for the density correction factors versus the total density in the maser bulb for three different values of the relaxation rate of the magnetic foil.

bulb can be measured with the pulsed magnetic resonance technique described in the previous chapter. Our method is based on the assumption that the total density in the maser bulb $n_H^{(4)}$ while it is oscillating is proportional to the population difference $\left(n_a^{(4)} - n_c^{(4)}\right)_{off}$ while the state selector is turned off. The population difference $\left(n_a^{(4)} - n_c^{(4)}\right)_{off}$ can be easily measured with a $\frac{\pi}{2}$ pulse. If the system is in thermal equilibrium, the proportionality constant is given (in the high temperature limit $kT_4 \gg \hbar\omega$) by

$$\left(\frac{n_H^{(4)}}{n_a^{(4)} - n_c^{(4)}}\right)_{therm.eq.} = \frac{4kT_4}{\hbar\omega}$$
(7.23)

In general, eq. (7.23) does not apply exactly to the oscillating maser and the right hand side has to be multiplied by a density correction factor. The calculated values for the correction factor are shown in fig. 7.5. For all three relaxation rates of the magnetic foil, the correction factors are roughly equal and independent of density. This implies that our density calibration which is based on the measured line width of the maser transition should be essentially correct, because it only relies on a constant correction factor. The upturn at low densities in fig. 7.5 however indicates that we systematically underestimate densities below about 5×10^{11} cm⁻³.

With the numerical simulation program, we have found that the density correction factor is unfortunately rather sensitive to the temperature of the state selector. Therefore, for every temperature of the state selector, a new density calibration has to be performed by measuring the line width versus density. It would be desirable to find a more direct and satisfactory method of density determination.

Chapter 8

Conclusions and outlook

8.1 The present cryogenic maser

The apparatus described in this thesis is a prototype hydrogen maser that has been primarily designed to study the potential of a new pumping scheme: hydrogen atoms are cycled many times back and forth between the maser cavity and the state selector by means of magnetic field gradients in combination with microwave pumping. In this essentially closed system, no large external pumps are required, in contrast to masers with the traditional beam geometry.

The results obtained with our prototype maser are very promising and they indicate that cryogenic hydrogen masers have indeed the potential to improve the state-of-the-art in frequency stability significantly. We have been able to observe stable maser oscillations over the range of temperature between 230 mK and 660 mK and for densities up to 3×10^{12} cm⁻³. The analysis of the temperature dependence of the maser frequency has resulted in a precision determination of the binding energy E_B of atomic hydrogen on ⁴He. Although the maser was not built for optimal frequency stability, the short term stability of our prototype device already exceeds the performance of the best conventional hydrogen masers. The frequency stability of our maser for averaging times τ larger than 10 s could not be measured, since reference oscillators with high enough stability for these averaging times were not available to us. In conjunction with the experimental work, an extensive computer simulation program was developed that helps us to understand the operation of the present maser. In addition, it is useful to guide the development of the second generation maser design.

8.2 Suggested improvements of the cryogenic maser design

8.2.1 Full size maser cavity

For the prototype maser, we chose a small size maser cavity in order to keep the design and construction as simple as possible. However, both the frequency instabilities due to thermal noise (4.1) and due to white phase noise in the electronics (4.8) decrease with increasing power. To first order, the maser output power is expected to scale linearly with the volume of the maser bulb. Therefore, a cryogenic hydrogen maser with the highest possible frequency stability has to have a full size microwave cavity.

High performance room temperature masers use the TE₀₁₁ mode of a silver plated high Q cavity. The hydrogen atoms are confined by a quartz bulb to a region in the center of the cavity where the phase of the rf magnetic field has the same sign. This design is not very well suited for a cryogenic maser because it would be difficult to control the temperature of the maser bulb to a high degree. An attractive alternative is a dielectrically loaded cavity, where the dielectric medium confines the hydrogen atoms and at the same time reduces the overall size of the maser cavity. The dielectric medium must exhibit low microwave losses, high thermal conductivity and a reasonably high dielectric constant. The best choice is single crystal sapphire with a dielectric constant $\varepsilon \approx 12$ and a loss tangent tan δ of less than 10^{-7} at low temperatures. A sapphire loaded cavity in the TE₀₁₁ mode has been used successfully by the Harvard-Smithsonian group [17] in their cryogenic maser.

The desired TE_{011} mode is not the fundamental mode in such a cavity. Since the cost of a large single crystal sapphire is high and the cutting and polishing of the surfaces
is demanding and time consuming, the design of the cavity has to be chosen carefully in order to avoid an accidental degeneracy with one of the many other modes. For this reason, the frequencies of all the modes are determined in appendix E. The calculation is complicated by the fact that sapphire is anisotropic and has two dielectric constants ε_z and ε_{\perp} .¹

8.2.2 Proposed new pumping scheme

If the split-ring resonator were to be replaced with a much larger volume sapphire loaded cavity, it is not clear whether the state selector in the present state would be efficient enough for maser oscillations and whether a larger power output would result. Based on the analysis of the experimental results and the simulation calculations, it is obvious that the present apparatus is not optimized. Two of the main problems that have been identified are the low efficiency of the magnetic foil in the high field region and the strong cross relaxation in the maser bulb.

However, even if these problems could be solved, the performance of the maser would not improve as drastically as one might naively expect. The present pumping scheme has some intrinsic limitations.

The spin exchange relaxation in low field leads to a large background of atoms in the $|d\rangle$ state in the maser bulb. Two atoms in the $|a\rangle$ state or two atoms in the $|c\rangle$ state can be relaxed to a $|b\rangle$ and a $|d\rangle$ atom in a collision.

$$a + a \rightleftharpoons b + d$$

 $c + c \rightleftharpoons b + d$

In the absence of strong cross relaxation, these spin exchange processes determine the

¹A similar calculation was performed by Folen et al. [79], but their results are not available to non-US citizens. However, we were informed (A. J. Berlinsky, private communications) that these authors did not take the anisotropy of the sapphire correctly into account and obtained erroneous results.

 $|d\rangle$ density in the maser bulb and the atom transport tube. The $|b\rangle$ atoms are sucked back into the high field region of the state selector, resulting in a depletion of $|b\rangle$ atoms in low field. The backward rates for the two spin exchange processes are therefore low and a large $|d\rangle$ density builds up in the low field regions until the backward rate balances the forward rate. This leads to a low population inversion.

In order to avoid this problem, a new pumping scheme is proposed which is a variation on the present one. The maser consists again of a region in zero field with the maser cavity, and a microwave cavity in a high magnetic field. In this new scheme, the $|a\rangle$ to $|d\rangle$ ESR transition is pumped instead of the $|b\rangle$ to $|c\rangle$ transition. In the atom transport tube, a small magnetic field is applied and an rf field, tuned to the $|c\rangle$ to $|d\rangle$ transition, equalizes the densities of $|d\rangle$ and $|c\rangle$ atoms. This way, a population inversion close to 50% is obtained outside the maser bulb. With this pumping scheme, no large buildup of $|d\rangle$ atoms in the maser bulb is possible any more. The $|b\rangle$ atoms that are attracted to the high field region do not get trapped in the ESR cavity. They relax with $|d\rangle$ atoms, which are always present in the ESR microwave cavity, into an $|a\rangle$ and $|c\rangle$ atom by spin exchange. We have modified the simulation computer program and confirmed that, at least theoretically, this pumping scheme results in much higher population inversions and power outputs than the scheme used in the present maser.

The additional rf field in the new scheme adds some complexity. However, it gives the experimenter more control and circumvents all the difficulties associated with the magnetic foil. One of the intrinsic problems with the foil that we have not mentioned so far is that its efficiency necessarily decreases with time due to the buildup of a layer of molecular hydrogen on top of it. Statt et al. [59] observed in their ESR experiment, that after atomic hydrogen recombined to H_2 and froze out on top of the magnetic impurities in the wall of their container, the one-body relaxation rate decreased significantly. The same process will affect the long term stability of the present maser with the magnetic foil.

In the new scheme, there are three regions with three different magnetic fields. It is therefore important to shield each of them carefully. This can be achieved with superconducting shields that have high critical current densities, as discussed in appendix D. Alternatively, the shield can be replaced by a counterwound solenoid. This technique was successfully used by Israelsson and Gould [80] in shielding a superconducting magnet.

The applied magnetic field in which the $|c\rangle$ and $|d\rangle$ densities are equalized by an rf field should be low enough in order to avoid the degradation of the magnetic field homogeneity at the maser cavity. On the other hand, it should be high enough to remove sufficiently the degeneracy between the $|d\rangle$ to $|c\rangle$ transition and the $|c\rangle$ to $|b\rangle$ transition. A reasonable choice seems to be a field of a few mT, which corresponds to a transition frequency of a few tens of MHz. At these fields, the $|c\rangle$ to $|d\rangle$ transition is essentially an electronic spin flip (see appendix A).

8.2.3 Optimization of maser

There are many parameters in the cryogenic maser that have to be optimized for the best performance, especially the volumes of the different regions and the sizes of the orifices connecting them. In the absence of any relaxation mechanisms, the inverse holding time for an atom in state $|k\rangle$ and in volume *i* is given by

$$\frac{1}{\tau_k^{(i)}} = \sum_{j,j \neq i} \Gamma_k^{(i \to j)}$$
(8.1)

where $\Gamma_k^{(i \to j)}$ is given by (7.2) and depends on the volume of region *i* and the cross sections of the orifices. Ideally, all the desired relaxation and pumping rates in volume *i* affecting $|k\rangle$ atoms are large compared with the rate (8.1), and the undesired relaxation rates, such as cross relaxation or possibly spin exchange relaxation rates, are much smaller than (8.1). This design rule helps to determine the optimal values for the different volumes and the orifice sizes. It is of course difficult to anticipate correctly the one-body relaxation rates in the different volumes.

A holding time of special importance is the maser bulb holding time T_b . Its proper choice is to a large extent determined by the intended application of the maser. Within the approximations used in chapter 4, the optimal intrinsic frequency stability (4.5) does not depend on the value of T_b . However, the best operating density $n_{opt} \simeq n_{se}$ is proportional to the inverse bulb holding time (see 2.43). A low operating density reduces effects due to spin exchange shifts and recombination. Therefore, for the best long-term stability of a cryogenic maser, the maser bulb holding time should be chosen as large as the cross relaxation rates permit.

If the user of the hydrogen maser is primarily interested in its excellent short-term stability, the maser bulb holding time should be adjusted to a smaller value, as can be seen from eq. (4.9). A small bulb holding time results in a larger flux of incoming state selected atoms and in a larger power output, but at the cost of a broader atomic line. For averaging times τ where the electronic noise contribution dominates, a reduction of T_b therefore improves the frequency stability.

It is not obvious what value should be chosen for the magnetic field B at the microwave cavity. An increase in B also increases the resonant frequency of the ESR transition which implies that the volume of the microwave cavity decreases. This is offset by a larger Boltzmann factor and therefore a larger density of high field seeking atoms in the cavity. It has to be checked with a computer model calculation whether the faster spin exchange and recombination processes due to larger densities cause any difficulties. We also point out that with a large field B, small temperature fluctuations in the state selector can cause significant density fluctuations in the low field regions. This can then lead to frequency fluctuations via the spin exchange shift.

8.3 Open questions

With the present work, we have not addressed several important questions, whose answers will affect the ultimate frequency stability that can be obtained with a cryogenic hydrogen maser. One of them deals with the frequency instabilities induced by the density dependent spin exchange shifts. It will be important to verify experimentally the theoretical predictions for the spin exchange shift cross sections, which were calculated by Koelman et al.[46]. In the prototype maser, no attempt has been made to stabilize the density and the populations. It is therefore not known at the present, to which degree these quantities can be held constant and at what level these frequency fluctuations will limit the performance of a cryogenic hydrogen maser.

At least for short averaging times, the cryogenic hydrogen maser promises to improve the state-of-the-art in frequency stability by at least an order of magnitude (see fig. 6.11). However, in order to take advantage of this high frequency stability, close attention has to be paid to all electronic components. Meeting all the stringent requirements on phase noise will be a major challenge.

Appendix A

Magnetic moment operators

The interaction of atomic hydrogen with a classical rf magnetic field is determined by the Hamiltonian

$$\hat{H}_{rf} = -\hat{\boldsymbol{\mu}} \cdot \mathbf{B}_{rf}(t) \tag{A.1}$$

where the magnetic moment operator $\hat{\mu}$ has a contributions from the electron spin and the proton spin:

$$\hat{\boldsymbol{\mu}} = -\hbar\gamma_{\rm e}\hat{\mathbf{s}} + \hbar\gamma_{\rm p}\hat{\boldsymbol{i}} \tag{A.2}$$

For the calculation of the transition probabilities induced by the rf magnetic field in first order perturbation theory, the matrix elements of $\hat{\mu}$ between initial and final state $\langle f | \hat{\mu} | i \rangle$ have to be evaluated. We are interested in transitions between the eigenstates $| a \rangle, | b \rangle, | c \rangle$ and $| d \rangle$ of the Hamiltonian \hat{H}_0 (2.1). Below we list the matrix elements of $\hat{\mu}_x$, $\hat{\mu}_y$ and $\hat{\mu}_z$ between all these states, or what is equivalent, we list the three operators in basis of the eigenstates of \hat{H}_0 . These expressions are valid for an arbitrary applied dc magnetic field B₀.

$$\hat{\mu}_{x} = \frac{\hbar}{2} \begin{pmatrix} 0 & \gamma_{e} \sin \theta + \gamma_{p} \cos \theta & 0 & -\gamma_{e} \cos \theta - \gamma_{p} \sin \theta \\ \gamma_{e} \sin \theta + \gamma_{p} \cos \theta & 0 & \gamma_{e} \cos \theta + \gamma_{p} \sin \theta & 0 \\ 0 & -\gamma_{e} \cos \theta + \gamma_{p} \sin \theta & 0 & -\gamma_{e} \sin \theta + \gamma_{p} \cos \theta \\ -\gamma_{e} \cos \theta - \gamma_{p} \sin \theta & 0 & -\gamma_{e} \sin \theta + \gamma_{p} \cos \theta & 0 \end{pmatrix}$$

$$\hat{\mu}_{y} = \frac{\hbar}{2i} \begin{pmatrix} 0 & \gamma_{e} \sin \theta + \gamma_{p} \cos \theta & 0 & \gamma_{e} \cos \theta + \gamma_{p} \sin \theta \\ -\gamma_{e} \sin \theta - \gamma_{p} \cos \theta & 0 & \gamma_{e} \cos \theta - \gamma_{p} \sin \theta & 0 \\ 0 & -\gamma_{e} \cos \theta + \gamma_{p} \sin \theta & 0 & \gamma_{e} \sin \theta - \gamma_{p} \cos \theta \\ -\gamma_{e} \cos \theta - \gamma_{p} \sin \theta & 0 & -\gamma_{e} \sin \theta + \gamma_{p} \cos \theta & 0 \end{pmatrix}$$

$$\hat{\mu}_{z} = \frac{\hbar}{2} \begin{pmatrix} (\gamma_{e} + \gamma_{p})\cos 2\theta & 0 & (\gamma_{e} + \gamma_{p})\sin 2\theta & 0 \\ 0 & \gamma_{e} - \gamma_{p} & 0 & 0 \\ (\gamma_{e} + \gamma_{p})\sin 2\theta & 0 & -(\gamma_{e} + \gamma_{p})\cos 2\theta & 0 \\ 0 & 0 & 0 & -\gamma_{e} + \gamma_{p} \end{pmatrix}$$

The mixing angle θ is given by

$$\tan(2\theta) = \frac{a}{\hbar(\gamma_{\rm e} + \gamma_{\rm p})B_0}$$

For zero magnetic field $\theta = \frac{\pi}{4}$, whereas for high fields $B_0 \gg \frac{a}{\hbar(\gamma_e + \gamma_p)} = 50.6 \text{ mT}$, θ can be approximated by

$$\theta \approx \frac{a}{2\hbar(\gamma_{\rm e} + \gamma_{\rm p})B_0} = \frac{25.304\,{\rm mT}}{B_0}$$

Appendix B

The Slater equation

We want to calculate the rf magnetic field that is produced by an oscillating magnetization inside a microwave cavity. This problem was first treated by Slater [81], but we follow here the simpler derivation of Wineland et al. [82].

We assume that there is only one cavity mode coupled to the magnetization and we take the magnetization to be oscillating along the z-axis. The microwave cavity, coupled to the output electronics which has impedance Z_0 , and containing the rf magnetization $M_z(t)$ can then be modeled by the electrical circuit shown in fig. B.1. The magnetic field H_z in the coil with inductance L is proportional to the current I flowing through the coil:

$$H_z = kI \tag{B.1}$$

The voltage across the inductance is then given by

$$V_L = \frac{1}{k} \frac{L}{\mu_0} \frac{d}{dt} B_z(t) = \frac{1}{k} L \frac{d}{dt} \left[H_z(t) + \eta M_z(t) \right]$$
(B.2)

where η is a filling factor. The voltage across the whole circuit has to vanish.

$$0 = V_{tot} = \frac{1}{k} \left[L \frac{d}{dt} \left[H_z(t) + \eta M_z(t) \right] + \left(R + \frac{Z_0}{n^2} \right) H_z(t) + \frac{1}{C} \int H_z(t) dt \right]$$
(B.3)

This equation can be rewritten with the following notations for the resonance frequency of the cavity ω_c , the coupling coefficient β , and the unloaded and loaded cavity quality factors Q_0 and Q_l :

$$\omega_c^2 = \frac{1}{LC} \tag{B.4}$$

Appendix B. The Slater equation

$$\beta = \frac{Z_0}{n^2 R} \tag{B.5}$$

$$Q_0 = \frac{\omega_c L}{R} \tag{B.6}$$

$$Q_l = \frac{Q_0}{1+\beta} \tag{B.7}$$

Differentiation of equation (B.3) yields

$$\eta \frac{d^2}{dt^2} M_z(t) + \frac{d^2}{dt^2} H_z(t) + \frac{\omega_c}{Q_l} \frac{d}{dt} H_z(t) + \omega_c^2 H_z(t) = 0.$$
 (B.8)

We now assume that the dominant time dependence of $M_z(t)$ and $H_z(t)$ is an oscillation with frequency ω , and we write

$$M_z(t) = M_z e^{i\omega t}$$

 $H_z(t) = H_z e^{i\omega t}$

 M_z and H_z are the amplitudes and assumed to vary only slowly. In the case of a free induction decay, the time constant for amplitude changes is T_2 ; in the case of stable maser oscillations, the magnitudes are constant. Since the time constant of the oscillation $\tau_{osc} = \omega^{-1}$ is much shorter than the cavity response time $\tau_{cav} = \frac{2Q_I}{\omega_c}$ and the relaxation time T_2 , (B.8) leads in the absence of externally applied rf fields to the relation

$$H_z(t) = \eta Q_l \frac{\omega}{\omega_c} \frac{1}{\Delta + i} M_z(t)$$
(B.9)

where Δ is given by

$$\Delta = Q_l \left(\frac{\omega_c}{\omega} - \frac{\omega}{\omega_c} \right) \tag{B.10}$$

To second order in Δ and with the approximation $\frac{\omega}{\omega_c} \approx 1 - \frac{\Delta}{Q_l} \approx 1$, equation (B.9) becomes:

$$H_z(t) = \eta Q_l \left(1 - \frac{\Delta^2}{2}\right) e^{i\left(-\frac{\pi}{2} + \Delta\right)} M_z(t)$$
(B.11)

The ratio of the amplitudes H_z and M_z is therefore given by $\eta Q_l \left(1 - \frac{\Delta^2}{2}\right)$ and Δ is the deviation from phase quadrature.

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Figure B.1: Electrical model of microwave cavity coupled to output electronics and containing rf magnetization M_z .

Appendix C

Measures of frequency stability

In this appendix we summarize different measures of frequency stability and the relationships between them. For more extensive treatment of this subject, the reader is referred to [83] and references therein.

We assume that the output signal of the frequency standard, which might be a voltage, is of the form

$$V(t) = [V_0 + \epsilon(t)] \sin[2\pi\nu_0 t + \varphi(t)]$$
(C.1)

In many cases, the amplitude noise $\epsilon(t)$ can be neglected. The instantaneous frequency $\nu(t)$ is defined by

$$\nu(t) \equiv \nu_0 + \frac{1}{2\pi} \frac{d\varphi}{dt}$$
(C.2)

It is customary to introduce y(t), the fractional frequency deviation:

$$y(t) \equiv \frac{\nu(t) - \nu_0}{\nu_0} = \frac{1}{2\pi\nu_0} \frac{d\varphi}{dt}$$
 (C.3)

In the time domain, the preferred measure of frequency stability is the so-called 'Allan variance' or 'two-sample variance':

$$\sigma_y^2(\tau) \equiv \left\langle \frac{1}{2} (\bar{y}_{k+1} - \bar{y}_k)^2 \right\rangle \tag{C.4}$$

where \bar{y}_k is the fractional frequency y(t) averaged over the time interval t_k to $t_k + \tau$.

$$\bar{y}_{k} = \frac{1}{\tau} \int_{t_{k}}^{t_{k}+\tau} \frac{\nu(t) - \nu_{0}}{\nu_{0}} dt = \frac{\varphi(t_{k}+\tau) - \varphi(t_{k})}{2\pi\nu_{0}\tau}$$
(C.5)

In the definition of the Allan variance, it is assumed that there is no dead time between subsequent intervals, i.e. $t_{k+1} = t_k + \tau$. Slow frequency fluctuations over time scales $\gg \tau$

do not affect the Allan variance $\sigma_y^2(\tau)$, and as a consequence, $\sigma_y^2(\tau)$, unlike the classical variance, is finite for all noise types typically encountered in frequency standards.

In the frequency domain, both the spectral density of phase fluctuations $S_{\varphi}(f)$ and the spectral density of the fractional frequency deviations $S_y(f)$ are used to characterize the stability of a time of frequency standard. They are defined for $f \ge 0$:

$$S_{\varphi}(f) = 2 \int_{-\infty}^{+\infty} \langle \varphi(t)\varphi(t+\tau) \rangle_{t} e^{i2\pi f\tau} d\tau \qquad (C.6)$$

and

$$S_y(f) = 2 \int_{-\infty}^{+\infty} \langle y(t)y(t+\tau) \rangle_i e^{i2\pi f\tau} d\tau \qquad (C.7)$$

From the definition of y (see C.3), it is obvious that $S_{\varphi}(f)$ and $S_{y}(f)$ are simply related by

$$S_y(f) = \left(\frac{f}{\nu_0}\right)^2 S_\varphi(f) \tag{C.8}$$

There is exactly the same information contained in $S_y(f)$ and $S_{\varphi}(f)$. The transformation between $S_y(f)$ and the Allan variance $\sigma_y^2(\tau)$ is given by:

$$\sigma_y^2(\tau) = 2 \int_0^\infty S_y(f) \frac{\sin^4(\pi f \tau)}{(\pi f \tau)^2} df$$
 (C.9)

It has been found that in many practical situations, $S_y(f)$ can be approximated by a sum of up to five power-law noise terms:

$$S_y(f) = \sum_{\alpha=-2}^{+2} h_{\alpha} f^{\alpha}$$
 (C.10)

In table C.1 we list the resulting contribution to the Allan variance from each of the five noise processes considered in (C.10).

For α in the range $-2 \leq \alpha \leq 0$, the Allan variance is proportional to τ^{μ} , where $\mu = -\alpha - 1$. However, table C.1 shows that it is not practical to distinguish between white phase noise ($\alpha = 2$) and flicker phase noise ($\alpha = 1$) from the dependence of σ_y^2 on

α	$S_y(f)$	$\sigma_y^2(\tau)$
-2	$h_{-2}f^{-2}$	$h_{-2}rac{2\pi}{3} au$
-1	$h_{-1}f^{-1}$	$h_{-1}2\ln 2$
0	h_0	$h_0 \frac{1}{2} \tau^{-1}$
+1	$\begin{cases} h_1 f^1 & \text{for } f \leq f_n \\ 0 & \text{otherwise} \end{cases}$	$h_1 \frac{3\gamma - \ln 2 + 3\ln(2\pi f_n \tau)}{(2\pi)^2} \tau^{-2}$ for $2\pi f_n \tau \gg 1$
+2	$\begin{cases} h_2 f^2 & \text{for } f \leq f_n \\ 0 & \text{otherwise} \end{cases}$	$h_2 \frac{3f_n}{4\pi^2} \tau^{-2}$ for $2\pi f_n \tau \gg 1$

Table C.1: Transformations between power-law spectral densities $S_y(f)$ and Allan Variance $\sigma_y^2(\tau)$. (Euler's constant $\gamma = 0.577215...$)

 τ . The two processes can be separated by analyzing the different dependence of $\sigma_y^2(\tau)$ on the bandwidth f_n , which can be decreased in the software. The so-called 'modified Allan variance' is based on this idea and is further discussed in [83].

In many experimental situations, dead time between subsequent measurements of \bar{y}_k cannot be completely avoided. Barnes et al. [84] discuss the effect of dead time on the obtained value for $\sigma_y^2(\tau)$. In general, it gives rise to only small corrections as long as the dead time is short compared to τ . However, in the case of white phase noise $(\alpha = 2)$, a dead time comparable to the inverse bandwidth f_n has a significant effect on the measured value of $\sigma_y^2(\tau)$. This can be understood by rewriting the Allan variance $\sigma_y^2(\tau)$ in terms of the phase:

$$\sigma_y^{\ 2}(\tau) = \frac{\langle [\varphi(t+\tau) - \varphi(t^+) - \varphi(t^-) + \varphi(t-\tau)]^2 \rangle_t}{8\pi^2 \nu_0^2 \tau^2}$$
(C.11)

For white phase noise, the correlation time for φ is determined by the bandwidth f_n . For a bandwidth large compared to the inverse dead time $t^+ - t^-$, it follows that

$$\frac{\sigma_y^2(t^- \neq t^+)}{\sigma_y^2(t^- = t^+)} = \frac{4\langle \varphi^2(t) \rangle}{6\langle \varphi^2(t) \rangle} = \frac{2}{3}$$

In the case of short but finite dead time, the expression in table C.1 for $\sigma_y^2(\tau)$ ($\alpha = 2$), obtained by assuming no dead time, has therefore to be replaced by

$$\sigma_y^2(\tau) = h_2 \frac{f_n}{2\pi^2} \tau^{-2}.$$
 (C.12)

Appendix D

Magnetic field of a solenoid inside a long superconducting shield

In this appendix, we calculate the magnetic field that is produced by a simple solenoid placed coaxially inside an infinitely long cylindrical superconducting shield. The radius of the solenoid is denoted by a, its length by $2z_0$ and the radius of the shield by b. The geometry is sketched in fig. D.1. First we restrict ourselves to a single layer solenoid with current I and n windings per unit length. We approximate the solenoid by a current sheet with uniform density of current. It is also assumed that the shield is a type I superconductor with negligible penetration depth and infinitely large critical current density.

As a first step, we solve a related problem. Consider a semi-infinite solenoid of radius a extending from $-\infty$ to $z = z_0$ with the same current density inside a superconducting shield identical to the previous one. For $z > z_0$, the magnetic field can be expanded into a series of cylindrical waveguide modes beyond cutoff, in our case at zero frequency. The coefficients of expansion can be determined with the techniques developed in section 8.11 of the textbook on electrodynamics by Jackson [85]. The result is

$$H_{z}(\rho, z > z_{0}) = In \sum_{j=1}^{\infty} c_{j} J_{0}(\chi_{j} \frac{\rho}{b}) \exp\left\{-\chi_{j} \frac{z - z_{0}}{b}\right\}$$
(D.1)

$$H_{\rho}(\rho, z > z_0) = In \sum_{j=1}^{\infty} c_j J_1(\chi_j \frac{\rho}{b}) \exp\left\{-\chi_j \frac{z - z_0}{b}\right\}$$
(D.2)

where the coefficients c_j are given by:

$$c_{j} = \frac{a J_{1}(\chi_{j} \frac{a}{b})}{\chi_{j} b J_{0}^{2}(\chi_{j})}$$
(D.3)



Figure D.1: Single layer solenoid inside superconducting shield.

and χ_j is the j-th zero of the Bessel function $J_1(x)$ $(J_1(\chi_j) = 0)$.

For $z < z_0$, the magnetic field can be calculated by considering the superposition of the following two situations. We take the magnetic field of an infinitely long solenoid with the same radius and current density as before inside the superconducting shield. This field is uniform and is given by

$$H_z = \begin{cases} In\left(1 - \frac{a^2}{b^2}\right) & \text{for } \rho < a \\ -In\frac{a^2}{b^2} & \text{for } a < \rho < b \end{cases}$$
(D.4)

From this field, we now subtract the field produced by a semi-infinite solenoid extending from z_0 to ∞ with radius a. The magnetic field in and around the semi-infinite solenoid $(-\infty \text{ to } z_0)$ is therefore

$$H_{z}(\rho < a, z < z_{0}) = In\left[\left(1 - \frac{a^{2}}{b^{2}}\right) - \sum_{j=1}^{\infty} c_{j} J_{0}(\chi_{j} \frac{\rho}{b}) \exp\left\{-\chi_{j} \frac{z_{0} - z}{b}\right\}\right]$$
(D.5)

$$H_z(a < \rho < b, z < z_0) = In \left[-\frac{a^2}{b^2} - \sum_{j=1}^{\infty} c_j \ J_0(\chi_j \frac{\rho}{b}) \exp\left\{ -\chi_j \frac{z_0 - z}{b} \right\} \right]$$
(D.6)

$$H_{\rho}(\rho, z < z_0) = In \sum_{j=1}^{\infty} c_j J_1(\chi_j \frac{\rho}{b}) \exp\left\{-\chi_j \frac{z_0 - z}{b}\right\}$$
(D.7)

With (D.1) to (D.3) and (D.5) to (D.7) we have the complete solution of the semi-infinite solenoid. From this solution, it is now easy to obtain the field of a finite solenoid of length $2z_0$ (see fig. D.1). The field is the difference between the fields produced by a semi-infinite solenoid extending from $-z_0$ to ∞ and another one extending from z_0 to ∞ . In the region $-z_0 < z < z_0$, the magnetic field is then given by

$$H_{z}(\rho < a, |z| < z_{0}) = In\left[\left(1 - \frac{a^{2}}{b^{2}}\right) - 2\sum_{j=1}^{\infty} c_{j} \exp\left\{-\chi_{j}\frac{z_{0}}{b}\right\} J_{0}(\chi_{j}\frac{\rho}{b})\cosh(\chi_{j}\frac{z}{b})\right] (D.8)$$

$$H_{z}(a < \rho < b, |z| < z_{0}) = In \left[-\frac{a^{2}}{b^{2}} - 2\sum_{j=1}^{\infty} c_{j} \exp\left\{ -\chi_{j} \frac{z_{0}}{b} \right\} J_{0}(\chi_{j} \frac{\rho}{b}) \cosh(\chi_{j} \frac{z}{b}) \right]$$
(D.9)

$$H_{\rho}(\rho, |z| < z_0) = 2In \sum_{j=1}^{\infty} c_j \exp\left\{-\chi_j \frac{z_0}{b}\right\} J_1(\chi_j \frac{\rho}{b}) \sinh(\chi_j \frac{z}{b})$$
(D.10)

and outside the solenoid $(|z| > z_0)$, we obtain

$$H_{z}(\rho, |z| > z_{0}) = 2In \sum_{j=1}^{\infty} c_{j} \sinh(\chi_{j} \frac{z_{0}}{b}) J_{0}(\chi_{j} \frac{\rho}{b}) \exp\left\{-\chi_{j} \frac{|z|}{b}\right\}$$
(D.11)

$$H_{\rho}(\rho, |z| > z_0) = 2In \frac{z}{|z|} \sum_{j=1}^{\infty} c_j \sinh(\chi_j \frac{z_0}{b}) J_1(\chi_j \frac{\rho}{b}) \exp\left\{-\chi_j \frac{|z|}{b}\right\}$$
(D.12)

where the coefficients c_j are given by (D.3). Far away from the solenoid $(|z| \gg z_0)$, only the first mode contributes significantly and the magnetic field decays like exp $\left\{-3.832\frac{|z|}{b}\right\}$.

The generalization to a multi-layer solenoid is straightforward. Each layer can be approximated by a current sheet of different radius and the contribution to the magnetic field of each layer can be determined by the expressions (D.8) to (D.12).

In order to improve the homogeneity inside the solenoid, correction coils are often added at the ends of the solenoid. We illustrate this on a simple practical example. The main solenoid is taken to consist of N layers with radii a_i (i = 1, ..., N) and extending from $-z_0$ to z_0 . On both ends of the main solenoid, N_c layers with radii d_k $(k = 1, ..., N_c)$ and width w form the correction coils (see figure D.2). For simplicity, we assume that the current density is identical in all N layers of the main solenoid and N_c layers of the corrections.

Inside the main solenoid, but outside the correction coils $(|z| < z_0 - w)$, the contribution to the magnetic field from the correction coils is:

$$H_{z}^{c}(\rho, z) = 2In \sum_{i=1}^{N_{c}} \sum_{j=1}^{\infty} c_{j}(d_{i}) \ e^{-\chi_{j} \frac{z_{0}}{b}} \left(e^{\chi_{j} \frac{w}{b}} - 1 \right) J_{0}(\chi_{j} \frac{\rho}{b}) \ \cosh(\chi_{j} \frac{z}{b})$$
(D.13)

$$H^{c}_{\rho}(\rho, z) = -2In \sum_{i=1}^{N_{c}} \sum_{j=1}^{\infty} c_{j}(d_{i}) \ e^{-\chi_{j} \frac{z_{0}}{b}} \left(e^{\chi_{j} \frac{w}{b}} - 1 \right) J_{1}(\chi_{j} \frac{\rho}{b}) \ \sinh(\chi_{j} \frac{z}{b}) \quad (D.14)$$

where $c_j(d_i)$ is given by (D.3) with a replaced by d_i . The major contribution to the inhomogeneities close to the center of the solenoid are due to the low modes. If the width w of the correction coils is chosen to be

$$w = \frac{b}{\chi_1} \ln \left[1 + \frac{\sum_{i=1}^N a_i J_1(\chi_1 \frac{a_i}{b})}{\sum_{k=1}^{N_c} d_k J_1(\chi_1 \frac{d_k}{b})} \right]$$
(D.15)



Figure D.2: Multi-layer solenoid with correction coils inside superconducting shield.

(where $\chi_1 = 3.8317...$), then the total contributions of the lowest mode (j = 1) to the magnetic field inside the solenoid vanishes exactly. With more complicated correction coils, it is also possible to cancel higher modes.

Appendix E

Eigenmodes of a sapphire loaded microwave cavity

In this appendix, the eigenmodes of a cylindrical metallic cavity, partially filled with a ring of anisotropic dielectric material are calculated. In particular, we have in mind the geometry of fig. E.1, where the dielectric ring consists of a single crystal sapphire, oriented along the z-axis. Sapphire has two dielectric constants, ε_z along the z-axis and ε_{\perp} perpendicular to it.

E.1 General Solution

We treat here the slightly more general case where both the permeability and the dielectric properties are anisotropic. Inside the cavity, the solutions have to satisfy the charge-free and current-free Maxwell equations subject to the appropriate boundary conditions:

$$\nabla \cdot \boldsymbol{D} = 0 \tag{E.1}$$

$$\nabla \times H = \frac{\partial D}{\partial t}$$
(E.2)

$$\nabla \times E = -\frac{\partial B}{\partial t}$$
(E.3)

$$\nabla \cdot \boldsymbol{B} = 0 \tag{E.4}$$

where

$$\boldsymbol{D} = \varepsilon_0 \begin{pmatrix} \varepsilon_{\perp} & 0 & 0 \\ 0 & \varepsilon_{\perp} & 0 \\ 0 & 0 & \varepsilon_z \end{pmatrix} \boldsymbol{E}$$
(E.5)



Figure E.1: Schematic drawing of the sapphire loaded microwave cavity.

$$\boldsymbol{B} = \mu_0 \begin{pmatrix} \mu_{\perp} & 0 & 0 \\ 0 & \mu_{\perp} & 0 \\ 0 & 0 & \mu_z \end{pmatrix} \boldsymbol{H}$$
(E.6)

The tensor character of ε and μ gives rise to additional terms in the wave equations. For instance, the divergence of E does in general not vanish, but it is given by

$$\nabla \cdot E = \frac{\varepsilon_{\perp} - \varepsilon_z}{\varepsilon_{\perp}} \frac{\partial}{\partial z} E_z$$
 (E.7)

and similarly the divergence of H is

$$\nabla \cdot \boldsymbol{H} = \frac{\mu_{\perp} - \mu_z}{\mu_{\perp}} \frac{\partial}{\partial z} H_z$$
(E.8)

The symmetries of the problem imply that the solutions are of the form:

$$\boldsymbol{E}(\rho,\varphi,z,t) = \boldsymbol{E}(\rho) e^{\pm ip\varphi} e^{\pm ik_z z} e^{\pm i\omega t}$$
(E.9)

$$\boldsymbol{H}(\rho,\varphi,z,t) = \boldsymbol{H}(\rho) e^{\pm ip\varphi} e^{\pm ik_z z} e^{\pm i\omega t}$$
(E.10)

where the angular quantum number p is an integer (p = 0, 1, 2, ...). An important combination of the frequency ω and the propagation constant k_z is

$$\beta_0^2 = \varepsilon_\perp \mu_\perp \left(\frac{\omega}{c}\right)^2 - k_z^2 \tag{E.11}$$

An equation for E_z can be derived by calculating the z-component of $\nabla \times (\nabla \times \mathbf{E})$ and using the equations (E.1) to (E.10). We obtain

$$\frac{1}{\rho} \frac{\partial}{\partial \rho} \left(\rho \frac{\partial E_z}{\partial \rho} \right) + \left(\beta_E^2 - \frac{p^2}{\rho^2} \right) E_z = 0$$
(E.12)

where

$$\beta_E^2 = \frac{\varepsilon_z}{\varepsilon_\perp} \beta_0^2 = \varepsilon_z \mu_\perp \left(\frac{\omega}{c}\right)^2 - \frac{\varepsilon_z}{\varepsilon_\perp} k_z^2$$
(E.13)

Similarly, an equation for H_z can be derived from $\nabla \times (\nabla \times H)$:

$$\frac{1}{\rho}\frac{\partial}{\partial\rho}\left(\rho\frac{\partial H_z}{\partial\rho}\right) + \left(\beta_H^2 - \frac{p^2}{\rho^2}\right)H_z = 0$$
 (E.14)

where

$$\beta_H^2 = \frac{\mu_z}{\mu_\perp} \beta_0^2 = \varepsilon_\perp \mu_z \left(\frac{\omega}{c}\right)^2 - \frac{\mu_z}{\mu_\perp} k_z^2 \qquad (E.15)$$

The electric field and the magnetic field are completely determined if the z-components are known. The transverse components of E and H are related to E_z and H_z by:

$$\begin{pmatrix} E_{\rho} \\ E_{\varphi} \\ H_{\rho} \\ H_{\varphi} \end{pmatrix} = \frac{1}{\beta_{0}^{2}} \begin{bmatrix} \frac{\partial^{2}}{\partial\rho\partial z} & -\mu_{0}\mu_{\perp}\frac{1}{\rho}\frac{\partial^{2}}{\partial t\partial\varphi} \\ \frac{1}{\rho}\frac{\partial^{2}}{\partial\varphi\partial z} & \mu_{0}\mu_{\perp}\frac{\partial^{2}}{\partial t\partial\rho} \\ \varepsilon_{0}\varepsilon_{\perp}\frac{1}{\rho}\frac{\partial^{2}}{\partial t\partial\varphi} & \frac{\partial^{2}}{\partial\rho\partial z} \\ -\varepsilon_{0}\varepsilon_{\perp}\frac{\partial^{2}}{\partial t\partial\rho} & \frac{1}{\rho}\frac{\partial^{2}}{\partial\varphi\partial z} \end{bmatrix} \begin{pmatrix} E_{z} \\ H_{z} \end{pmatrix}$$
(E.16)

The solutions of (E.12) and (E.14) for E_z and H_z are Bessel functions of the first and second kind. Since Bessel functions of the second kind are not bound at the origin, only Bessel functions of the first kind are physical solutions in the central region (labeled by superscript (1)) of the cavity. In the region of the dielectric ring (superscript (2)), the general solution consists of a linear combination of Bessel functions of the first and second kind:

$$E^{(1)}(\rho < \rho_1) = a_1 J_p(\beta_E^{(1)} \rho)$$
(E.17)

Appendix E. Eigenmodes of a sapphire loaded microwave cavity

$$H^{(1)}(\rho < \rho_1) = b_1 J_p(\beta_H^{(1)} \rho)$$
(E.18)

$$E^{(2)}(\rho_1 < \rho < \rho_2) = a_2 J_p(\beta_E^{(2)} \rho) + a_3 Y_p(\beta_E^{(2)} \rho)$$
(E.19)

$$H^{(2)}(\rho_1 < \rho < \rho_2) = b_2 J_p(\beta_H^{(2)} \rho) + b_3 Y_p(\beta_H^{(2)} \rho)$$
(E.20)

In the case that $\beta_0^{(1)}$ or $\beta_0^{(2)}$ is imaginary, the Bessel functions in the corresponding region have to be replaced by the modified Bessel functions, i.e. $J_p(\beta \rho)$ has to be replaced by $I_p(|\beta|\rho)$ and $Y_p(\beta \rho)$ by $K_p(|\beta|\rho)$.

E.2 Boundary conditions

The possible values of the longitudinal propagation constant k_z , the frequency ω and the radial wavenumber β are determined by the boundary conditions. We assume perfect metallic walls with infinitely high conductivity at z = 0, z = L and $\rho = \rho_2$. The components of the electric field parallel to the walls must therefore vanish exactly at the walls. The requirements $E_{\varphi}(z=0) = E_{\varphi}(z=L) = 0$ and $E_{\rho}(z=0) = E_{\rho}(z=L) = 0$ lead immediately to the quantization of the propagation constant k_z :

$$k_z = n \frac{\pi}{L}$$
 (n = 0, 1, 2, ...) (E.21)

It is more difficult to calculate the quantized values of the radial wavenumbers β , but as we can see from the expressions (E.11), (E.13) and (E.15) this is equivalent to finding the eigenfrequencies ω . The solutions of the eigenmodes have to meet the following conditions:

- $E_z(\rho = \rho_2) = 0$
- $E_{\varphi}\left(\rho=\rho_{2}\right)=0$
- E_z is continuous at $\rho = \rho_1$

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- E_{φ} is continuous at $\rho = \rho_1$
- H_z is continuous at $\rho = \rho_1$
- H_{φ} is continuous at $\rho = \rho_1$

[A] =

If the four components E_z , E_{φ} , H_z and H_{φ} are continuous at the interface between the two dielectric media, it follows that D_{ρ} and B_{ρ} are also continuous. The six conditions mentioned above lead to a set of linear equations for the six coefficients a_i and b_i (i = 1, 2, 3), defined in eq. (E.17) to (E.20):

$$A] \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ b_1 \\ b_2 \\ b_3 \end{pmatrix} = 0$$
(E.22)

In the case that $\beta_0^{(1)}$ and $\beta_0^{(2)}$ are both real, the matrix A is given by:

$$\begin{pmatrix} 0 & J_{p}(\beta_{E}^{(2)}\rho_{2}) & Y_{p}(\beta_{E}^{(2)}\rho_{2}) & 0 & 0 & 0 \\ 0 & \frac{c_{0}^{(2)}}{\rho_{2}}J_{p}(\beta_{E}^{(2)}\rho_{2}) & \frac{c_{0}^{(2)}}{\rho_{2}}Y_{p}(\beta_{E}^{(2)}\rho_{2}) & 0 & c_{H}^{(2)}J_{p}'(\beta_{H}^{(2)}\rho_{2}) & c_{H}^{(2)}Y_{p}'(\beta_{H}^{(2)}\rho_{2}) \\ -J_{p}(\beta_{E}^{(1)}\rho_{1}) & J_{p}(\beta_{E}^{(2)}\rho_{1}) & Y_{p}(\beta_{E}^{(2)}\rho_{1}) & 0 & 0 & 0 \\ -\frac{c_{0}^{(1)}}{\rho_{1}}J_{p}(\beta_{E}^{(1)}\rho_{1}) & \frac{c_{0}^{(2)}}{\rho_{1}}J_{p}(\beta_{E}^{(2)}\rho_{1}) & \frac{c_{0}^{(2)}}{\rho_{1}}Y_{p}(\beta_{E}^{(2)}\rho_{1}) & -c_{H}^{(1)}J_{p}'(\beta_{H}^{(1)}\rho_{1}) & c_{H}^{(2)}J_{p}'(\beta_{H}^{(2)}\rho_{1}) & c_{H}^{(2)}Y_{p}'(\beta_{H}^{(2)}\rho_{1}) \\ 0 & 0 & 0 & -J_{p}(\beta_{H}^{(1)}\rho_{1}) & J_{p}(\beta_{H}^{(2)}\rho_{1}) & Y_{p}(\beta_{H}^{(2)}\rho_{1}) \\ -c_{E}^{(1)}J_{p}'(\beta_{E}^{(1)}\rho_{1}) & c_{E}^{(2)}J_{p}'(\beta_{E}^{(2)}\rho_{1}) & c_{E}^{(2)}Y_{p}'(\beta_{E}^{(2)}\rho_{1}) & -\frac{c_{0}^{(1)}}{\rho_{1}}J_{p}(\beta_{H}^{(1)}\rho_{1}) & \frac{c_{0}^{(2)}}{\rho_{1}}J_{p}(\beta_{H}^{(2)}\rho_{1}) & y_{p}(\beta_{H}^{(2)}\rho_{1}) \end{pmatrix}$$

(E.23)

where the following abbreviations have been used:

$$c_0^{(i)} \equiv p n \frac{\pi}{L} \left(\frac{1}{\beta_0^{(i)}}\right)^2$$
 (E.24)

$$c_E^{(i)} \equiv \epsilon_0 \, \varepsilon_\perp^{(i)} \omega \, \frac{\beta_E^{(i)}}{\left(\beta_0^{(i)}\right)^2} \tag{E.25}$$

$$c_{H}^{(i)} \equiv \mu_{0} \, \mu_{\perp}^{(i)} \omega \, \frac{\beta_{H}^{(i)}}{\left(\beta_{0}^{(i)}\right)^{2}} \tag{E.26}$$

and the derivatives of the Bessel functions are indicated by a '.

In the case that $\beta_0^{(1)}$ or $\beta_0^{(2)}$ is imaginary, the matrix A has to be modified as follows: the imaginary values of β have to be replaced by their absolute values and the appropriate Bessel functions have to be replaced by the modified Bessel functions.

Eigenmodes are characterized by non-trivial solutions of eq. (E.22). Therefore, the determinant of the matrix A has to vanish for all eigenfrequencies:

$$\det(A) = 0 \tag{E.27}$$

This equation determines the eigenfrequencies. The left hand side of (E.27) depends in a rather complicated way on ω , namely through the parameters $c_0^{(i)}$, $c_E^{(i)}$ and $c_H^{(i)}$ and through the radial wavenumbers $\beta_0^{(i)}$, $\beta_E^{(i)}$ and $\beta_H^{(i)}$. In general, (E.27) can only be solved numerically.

In a simple cylindrical cavity with metallic walls (but without different dielectric media), all the eigenmodes can be categorized either as TM_{pmn} mode or as TE_{pmn} mode. In a TM mode, the magnetic field has only transverse components and no z-component, and in a TE mode, it is the electric field that has this property. For both types of mode, the eigenmode can be identified with three quantum numbers: the angular quantum number p, the radial quantum number m and the longitudinal quantum number n. The radial quantum number m (m = 1, 2, 3, ...) labels the modes with identical p and n in order of increasing frequency. In general, the eigenmodes of a dielectrically loaded cavity do not fall in any of the two categories: both the electric field and the magnetic field of most eigenmodes have a non-vanishing z-component and the modes are therefore neither pure TE or TM modes but hybrid modes. The only exceptions are the TE_{0mn} , the TM_{0mn} and the TM_{pm0} modes (TE_{pm0} modes do not exist). In these cases, $c_0^{(i)}$ vanishes and the matrix A can be split into two independent 3×3 matrices. For all other combinations of quantum numbers it is not possible to satisfy all the continuity conditions at $\rho = \rho_1$ simultaneously if $E_z = 0$ or $H_z = 0$.

E.3 Application to the cryogenic hydrogen maser

In this section, the cavity mode calculations are applied to find the best configuration of a sapphire loaded cavity that is to be used in a cryogenic hydrogen maser. The frequency of the operating mode has to coincide with the hyperfine frequency of hydrogen, which is 1420.405 MHz.

The choice of the operating cavity mode is determined by the requirements of the hydrogen maser. The radiating hydrogen atoms couple only to the z-component of the magnetic rf field and therefore, all TM modes are excluded. In addition, the rf field H_z should be as uniform as possible in the entire region that is filled with hydrogen atoms in order to suppress the first order Doppler broadening. This applies not only to the magnitude of H_z but also to its phase. In particular, if the spatial average of H_z is zero, the first order Doppler broadening is not suppressed at all and it results in an unacceptable broadening of the maser line. If we assume that the hydrogen atoms fill the entire region (1) and no diaphragms are used to divide up this space, the requirements on the angular and longitudinal quantum numbers are p = 0 and $n \leq 1$. Only the TE_{0m1} modes satisfy all the requirements, of which the TE₀₁₁ mode is by far the best choice.



Figure E.2: Required length of the sapphire loaded cavity that brings the TE₀₁₁ frequency in resonance with the hyperfine frequency of hydrogen. The outer diameter of the sapphire piece is fixed at 11.5 cm. The dielectric constants for sapphire are $\varepsilon_{\perp} = 9.264$ and $\varepsilon_z = 11.341$.

As an illustration, we calculate here the size of a sapphire cavity whose TE_{011} mode is on resonance with the maser transition, with the additional restriction that the outside diameter of the sapphire ring $(2\rho_2)$ is 11.5 cm. As the inner diameter of the sapphire piece $(2\rho_1)$ is varied, the length has to be adjusted to keep the frequency of the TE_{011} mode at its desired value. Equation (E.27) can be used to calculate the required length. For the TE_{011} mode, the matrix A is only a 3×3 matrix (the last three columns of row 2, 4 and 5 in eq. (E.23)) and we solve numerically eq. (E.27) for L while we fix ω at the hyperfine frequency of hydrogen. Figure E.2 shows the resulting length of the sapphire loaded cavity as a function of the inner diameter.

In this calculation we assume that in the inner region $\varepsilon = \mu = 1$. The temperature dependence of the dielectric constants of sapphire has been measured by Loewenstein et al. [86] and by Shelby et al. [87]. Extrapolated to T = 0 K, Loewenstein et al. obtained $\varepsilon_{\perp} = 9.290 \pm 0.012$ and $\varepsilon_z = 11.264 \pm 0.012$, and Shelby et al. obtained $\varepsilon_{\perp} = 9.264 \pm 0.006$

and $\varepsilon_z = 11.341 \pm 0.006$. Note that the two measurements are in slight disagreement. In the present calculations, we have used the values quoted by Shelby et al. [87] and we also set $\mu_{\perp} = \mu_z = 1$.

In the application of the hydrogen maser, it is important that the TE_{011} mode is well separated in frequency from all the other modes. We have calculated the eigenfrequencies of all modes below 2 GHz as a function of the inner diameter of the sapphire piece, where its length is varied according to the results of fig. E.2. The results are shown in fig. E.3.

It is clear from fig. E.3 that the exact dimensions of the sapphire cavity have to be chosen carefully in order to avoid an accidental degeneracy of the TE_{011} mode with any of the numerous other modes. We note that it is possible to shift the frequency of an undesired mode sufficiently by a slight modification of the cavity geometry, such as a groove or a ridge. The resulting shifts with the modified geometry can then be calculated from the solutions of the present geometry by the method of cavity perturbation theory.

The final design of a sapphire loaded cavity should not only take the results of fig. E.3 into account. For the application of the cryogenic hydrogen maser, it is also important to optimize the volume of the inner region and the filling factor η . The filling factor η (see eq. 2.30) can be easily calculated from the present solution of the TE₀₁₁ eigenmode.



Figure E.3: Frequencies of all cavity modes below 2 GHz in the sapphire loaded cavity as a function of the inner diameter of the sapphire ring. The outer diameter is fixed at 11.5 cm and the length is adjusted as shown in fig. E.2 in order to keep the frequency of the TE₀₁₁ mode at 1420.405 MHz. The hybrid modes are only labeled with their angular and longitudinal quantum numbers p and n.

Bibliography

- [1] R. F. C. Vessot, M. W. Levine, and E. M. Mattison. Comparison of theoretical and observed hydrogen maser stability limitation due to thermal noise and the prospect for improvement by low-temperature operation. In *Proceedings of the Ninth Annual Precise Time and Time Interval (PTTI) Applications and Planning Meeting*, page 549. NASA Technical Memorandum No. 78104, 1978.
- [2] R. F. C. Vessot, M. W. Levine, E. M. Mattison, E. L. Blomberg, T. E. Hoffman, G. U. Nystrom, B. F. Farrel, R. Decher, P. B. Eby, C. R. Baugher, J. W. Watts, D. L. Teuber, and F. D. Wills. Test of relativistic gravitation with a space-borne hydrogen maser. *Phys. Rev. Lett.*, 45(26):2081, 1980.
- [3] W. E. Carter, D. S. Robertson, and J. R. MacKay. Geodetic radio interferometric surveying: Applications and results. J. Geophys. Res., 90(B6):4577, 1985.
- [4] H. M. Goldenberg, D. Kleppner, and N. F. Ramsey. Atomic hydrogen maser. *Phys. Rev. Lett.*, 5(8):361, 1960.
- [5] Daniel Kleppner, H. Mark Goldenberg, and Norman F. Ramsey. Theory of the hydrogen maser. Phys. Rev., 126(2):603, 1962.
- [6] D. Kleppner, H. C. Berg, S. B. Crampton, N. F. Ramsey, R. F. C. Vessot, H. E. Peters, and J. Vanier. Hydrogen-maser principles and techniques. *Phys. Rev.*, 138(4A):A 972, 1965.
- [7] L. A. Rawley, J. H. Taylor, M. M. Davis, and D. W. Allan. Millisecond pulsar PSR 1937+21: A highly stable clock. *Science*, 238:761, 1987.

- [8] S. B. Crampton, W. D. Phillips, and D. Kleppner. Proposed low temperature hydrogen maser. Bull. Am. Phys. Soc., 23:86, 1978.
- [9] R. F. C. Vessot, E. M. Mattison, and E. L. Blomberg. Research with a cold hydrogen maser. In Proc. 33th Annual Frequency Control Symposium, page 511, 1979.
- [10] S. B. Crampton, K. M. Jones, G. Nunes, and S. P. Souza. Hydrogen maser oscillation at 10K. In Proceedings of the Sixteenth Annual Precise Time and Time Interval (PTTI) Applications and Planning Meeting, page 339. NASA Technical Memorandum No. 8756, 1985.
- [11] Isaac F. Silvera and J. T. M. Walraven. Stabilization of atomic hydrogen at low temperature. *Phys. Rev. Lett.*, 44(3):164, 1980.
- [12] W. N. Hardy, M. Morrow, R. Jochemsen, and A. J. Berlinsky. Magnetic resonance of atomic hydrogen at low temperatures. *Physica*, 109 and 110B:1964, 1982.
- [13] A. J. Berlinsky and W. N. Hardy. Cryogenic masers. In Proceedings of the Thirteenth Annual Precise Time and Time Interval (PTTI) Applications and Planning Meeting, page 547. NASA Conference Publication No. 2220, 1982.
- [14] Harald F. Hess, Gregory P. Kochanski, John M. Doyle, Thomas J. Greytak, and Daniel Kleppner. Spin-polarized hydrogen maser. Phys. Rev. A, 34(2):1602, 1986.
- [15] Harald F. Hess, Greg P. Kochanski, John M. Doyle, Naoto Masuhara, Daniel Kleppner, and Thomas J. Greytak. Magnetic trapping of spin-polarized atomic hydrogen. *Phys. Rev. Lett.*, 59(6):672, 1987.
- [16] M. D. Hürlimann, W. N. Hardy, A. J. Berlinsky, and R. W. Cline. Recirculating cryogenic hydrogen maser. *Phys. Rev. A*, 34(2):1605, 1986.

- [17] Ronald L. Walsworth Jr., Isaac F. Silvera, H. P. Godfried, C. C. Agosta, Robert F. C. Vessot, and Edward M. Mattison. Hydrogen maser at temperatures below 1 K. Phys. Rev. A, 34(3):2550, 1986.
- [18] Martin D. Hürlimann, A. John Berlinsky, Richard W. Cline, and Walter N. Hardy. A recirculating cryogenic hydrogen maser. *IEEE Trans. on Instr. and Measurement*, IM-36(2):584, 1987.
- [19] W. N. Hardy, M. D. Hürlimann, and R. W. Cline. Application of atomic hydrogen at low temperatures: The recirculating cryogenic hydrogen maser. Japanese Journal of Applied Physics, 26-3:2065, 1987. Proc. 18th Int. Conf. on Low Temperature Physics.
- [20] M. D. Hürlimann and W. N. Hardy. Measurement of flicker phase noise of 1.4 GHz MESFET amplifier at temperatures between 300K and 1.26K. *Electron. Lett.*, 23(6):283, 1987.
- [21] P. Petit, M. Desaintfuscien, and C. Audoin. Temperature dependence of the hydrogen maser wall shift in the temperature range 295-395K. *Metrologia*, 16:7, 1980.
- [22] E. Richard Cohen and Barry N. Taylor. The 1986 adjustment of the fundamental physical constants. *Reviews of Modern Physics*, 59(4):1121, 1987.
- [23] A. Abragam. The Principle of Nuclear Magnetism. Clarendon Press, Oxford, 1961.
- [24] C. Audoin, J. P. Schermann, and P. Grivet. Physics of the Hydrogen Maser, volume 7 of Advances in Atomic and Molecular Physics, page 1. Academic Press, 1971.
- [25] Stanley Bloom. Effects of radiation damping on spin dynamics. Journal of Applied Physics, 28(7):800, 1957.

- [26] L. C. Balling, R. J. Hanson, and F. M. Pipkin. Frequency shifts in spin-exchange optical pumping experiments. *Phys. Rev.*, 133(3A):A607, 1964. *Phys. Rev.*, 135(7AB):AB1, 1964.
- [27] B. J. Verhaar, J. M. V. A. Koelman, H. T. C. Stoof, O. J. Luiten, and S. B. Crampton. Hyperfine contribution to spin-exchange frequency shifts in the hydrogen maser. *Phys. Rev. A*, 35:3825, 1987.
- [28] R. F. C. Vessot, E. M. Mattison, and E. Imbier. Cold hydrogen maser research at SAO and related developments. In Proc. 37th Annual Frequency Control Symposium, page 49, 1983.
- [29] L. Pollack, S. Buchman, Y. M. Xiao, H. F. Hess, G. P. Kochanski, and T. J. Greytak. Direct observation of a two-dimensional gas of spin-polarized atomic hydrogen. *Phys. Rev. B*, 34(1):461, 1986.
- [30] S. B. Crampton, J. J. Krupczak, and S. P. Souza. Temperature dependence of hydrogen atom adsorption on molecular hydrogen surfaces. *Phys. Rev. B*, 25:4383, 1982.
- [31] M. Morrow and W. N. Hardy. The magnetic resonance lineshape for atomic hydrogen confined by liquid helium walls. Can. J. Phys., 61(6):956, 1983.
- [32] M. Desaintfuscien, J. Viennet, and C. Audoin. Discussion of temperature dependence of wall and spin exchange effects in the hydrogen maser. *Metrologia*, 13:125, 1977.
- [33] K. E. Anderson, S. B. Crampton, K. M. Jones, G. Nunes, and S. P. Souza. Hydrogen atom adsorption on solid neon. In U. Eckern, A. Schmid, W. Weber, and H. Wuhl,

editors, Proc. 17th Int. Conf. on Low Temperature Physics, page 539, Amsterdam, 1984. North-Holland.

- [34] R. Jochemsen, M. Morrow, A. J. Berlinsky, and W. N. Hardy. Interaction of hydrogen atoms with helium films: Sticking probabilities for H on ³He and ⁴He, and the binding energy of H on ³He. *Phys. Rev. Lett.*, 47(12):852, 1981.
- [35] K. E. Kürten and M. L. Ristig. Atomic and molecular hydrogen isotopes in liquid helium. Phys. Rev. B, 31(3):1346, 1985.
- [36] I. F. Silvera and J. T. M. Walraven. Spin polarized atomic hydrogen. In D. Brewer, editor, Progress in Low Temperature Physics X, page 139, Amsterdam, 1986. North-Holland.
- [37] T. J. Greytak and D. Kleppner. New Trends in Atomic Physics, page 1127. North-Holland, Amsterdam, 1984.
- [38] Michael Robert Morrow. Magnetic Resonance on Atomic Hydrogen Confined by Liquid Helium Walls. PhD thesis, University of British Columbia, 1983.
- [39] W. N. Hardy and M. Morrow. Prospects for low temperature H masers using liquid helium coated walls. *Journal de Physique*, Colloq. 42(12):C8-171, 1981.
- [40] W. N. Hardy, M. Morrow, R. Jochemsen, B. W. Statt, P. R. Kubik, R. M. Marsolais, and A. J. Berlinsky. Magnetic-resonance of gaseous atomic hydrogen confined at 1K and zero magnetic field. *Phys. Rev. Lett.*, 45(6):453, 1980.
- [41] M. Morrow, R. Jochemsen, A. J. Berlinsky, and W. N. Hardy. Zero-field hyperfine resonance of atomic hydrogen for 0.18≤ T ≤ 1K: the binding energy of H on liquid ⁴He. Phys. Rev. Lett., 46(3):195, 1981. Phys. Rev. Lett., 47(6):455, 1981.

- [42] R. Jochemsen, M. Morrow, A. J. Berlinsky, and W. N. Hardy. Magnetic resonance studies of atomic hydrogen at zero field and low temperature: Recombination and binding on liquid helium. *Physica*, 109 and 110B:2108, 1982.
- [43] R. Jochemsen, M. Morrow, and A. J. Berlinsky. The hyperfine frequency shift of atomic hydrogen in the presence of helium buffer gas. Can. J. Phys., 60:252, 1982.
- [44] Victor V. Goldman. Kapitza conductance between gaseous atomic hydrogen and liquid helium. Phys. Rev. Lett., 56(6):612, 1986.
- [45] R. Jochemsen, A. J. Berlinsky, and W. N. Hardy. The diffusion cross section for atomic hydrogen in helium gas at low temperature and the H-He potential. *Can. J. Phys.*, 62(8):751, 1984.
- [46] J. M. V. A. Koelman, S. B. Crampton, H. T. C. Stoof, O. J. Luiten, and B. J. Verhaar. Spin-exchange frequency shifts in cryogenic and room-temperature hydrogen masers. *Phys. Rev. A*, 38(7):3535, 1988.
- [47] S. B. Crampton. Spin-exchange shifts in the hydrogen maser. Phys. Rev., 158:57, 1967.
- [48] A. J. Berlinsky and B. Shizgal. Spin-exchange scattering cross sections for hydrogen atoms at low temperatures. Can. J. Phys., 58(6):881, 1980.
- [49] Henk T. C. Stoof, J. M. Vianney A. Koelman, and Boudewijn J. Verhaar. Spinexchange and dipole relaxation rates in atomic hydrogen: rigorous and simplified. *Phys. Rev. B*, 38:4688, 1988.
- [50] M. Morrow and A. J. Berlinsky. Theory of low temperature spin exchange scattering for a physisorbed two-dimensional gas of hydrogen atoms. Can. J. Phys., 61(7):1042, 1983.
- [51] Jon H. Shirley. Dynamics of a simple maser model. American Journal of Physics, 36(11):949, 1968.
- [52] K. Shimoda, T. C. Wang, and C. H. Townes. Further aspects of the theory of the maser. Phys. Rev., 102(5):1308, 1956.
- [53] Model L 1.4-30, Berkshire Technologies Inc., 5427 Telegraph Ave., Suite B2, Oakland Ca 94609.
- [54] Marian W. Pospieszalski, Sander Weinreb, Roger D. Norrod, and Ronald Harris. FET's and HEMT's at cryogenic temperatures-their properties and use in low-noise amplifiers. *IEEE Trans. Microwave Theory Tech.*, MTT-36(3):552, 1988.
- [55] S. B. Crampton and H. T. M. Wang. Duration of hydrogen-atom spin-exchange collisions. *Phys. Rev. A*, 12(4):1305, 1975.
- [56] M. Abe, M. Gomi, and F. Yokoyama. Magneto-optical effect in anodized Al₂O₃ film with micropores electrodeposited with Co. J. Appl. Phys., 57(1):3909, 1985.
- [57] F. Keller, M. S. Hunter, and D. L. Robinson. Structural features of oxide coatings on aluminum. J. Electrochem. Soc., 100:411, 1953.
- [58] W. N. Hardy and L. A. Whitehead. Split-ring resonator for use in magnetic resonance from 200-2000 MHz. Rev. Sci. Instrum., 52(2):213, 1981.
- [59] B. W. Statt, A. J. Berlinsky, and W. N. Hardy. Electron-spin resonance, nuclear polarization, and spin relaxation of spin-polarized atomic hydrogen. *Phys. Rev. B*, 31:3169, 1985.
- [60] Dennis S. Greywall and Paul A. Busch. ³He-melting-curve thermometry. J. Low Temp. Phys., 46(5/6):451, 1982.

- [61] David G. Blair, HoJung Paik, and Robert C. Taber. How to make high critical current joints in Nb-Ti wire. Rev. Sci. Instrum., 46(8):1130, 1975.
- [62] K. A. Muething, D. O. Edwards, J. D. Feder, W. J. Gully, and H. N. Scholz. Small solenoid with a superconducting shield for nuclear-magnetic-resonance near 1 mK. *Rev. Sci. Instrum.*, 53(4):485, 1982.
- [63] B. W. Statt, W. N. Hardy, A. J. Berlinsky, and E. Klein. ESR studies of spinpolarized atomic hydrogen using a 114-GHz heterodyne spectrometer. J. Low Temp. Phys., 61:471, 1985.
- [64] D. R. Williams, W. Lum, and S. Weinreb. L-band cryogenically-cooled GaAs FET amplifier. *Microwave Journal*, 23:73, 1980.
- [65] M. W. Reynolds, I. Shinkoda, W. N. Hardy, A. J. Berlinsky, F. Bridges, and B. W. Statt. Electron-spin-resonance studies of spin-polarized hydrogen on the surface of liquid ⁴He. *Phys. Rev. B*, 31:7503, 1985.
- [66] A. P. M. Matthey, J. T. M. Walraven, and Isaac F. Silvera. Measurement of pressure of gaseous H₁: Adsorption energies and surface recombination rates on helium. *Phys. Rev. Lett.*, 46(10):668, 1981.
- [67] R. Sprik, J. T. M. Walraven, G. H. van Yperen, and Isaac F. Silvera. Statedependent recombination and suppressed nuclear relaxation in atomic hydrogen. *Phys. Rev. Lett.*, 49:153, 1982.
- [68] R. W. Cline, T. J. Greytak, and D. Kleppner. Nuclear polarization of spin-polarized hydrogen. Phys. Rev. Lett., 47:1195, 1981.
- [69] Harald F. Hess, David A. Bell, Gregory P. Kochanski, Daniel Kleppner, and

Thomas J. Greytak. Temperature and magnetic field dependence of three-body recombination in spin-polarized hydrogen. *Phys. Rev. Lett.*, 52(17):1520, 1984.

- [70] B. Yurke, J. S. Denker, B. R. Johnson, N. Bigelow, L. P. Levy, D. M. Lee, and J. H. Freed. NMR-induced recombination of spin-polarized hydrogen. *Phys. Rev. Lett.*, 50:1137, 1983.
- [71] H. P. Godfried, E. R. Eliel, J. G. Brisson, J. D. Gillaspy, C. Mallardeau, J. C. Mester, and Isaac F. Silvera. Interaction of atomic hydrogen with undersaturated helium films. *Phys. Rev. Lett.*, 55(12):1311, 1985.
- [72] J. J. Berkhout, E. J. Wolters, R. van Roijen, and J. T. M. Walraven. Vanishing sticking probabilities and enhanced capillary flow of spin-polarized hydrogen. *Phys. Rev. Lett.*, 57(19):2387, 1986.
- [73] S. R. Stein. Space applications of superconductivity: Resonators for high stability oscillators and other applications. *Cryogenics*, page 363, July 1980.
- [74] L. D. Mann, D. G. Blair, and K. J. Wellington. Flicker noise in GaAs MESFET X-band amplifiers in the temperature range 300 K to 2 K. *Electron. Lett.*, 22:1037, 1986.
- [75] F. L. Walls and A. Demarchi. RF spectrum of a signal after frequency multiplication; measurement and comparison with a simple calculation. *IEEE Trans.*, IM-24:210, 1975.
- [76] A. J. Berlinsky, W. N. Hardy, and B. W. Statt. Theory of nuclear spin-lattice relaxation of spin-polarized hydrogen on liquid-helium-coated surfaces due to magnetic particles in the substrate. *Phys. Rev. B*, 35(10):4831, 1987.

- [77] M. Desaintfuscien and C. Audoin. Relaxation des niveaux de structure hyperfine d'atomes d'hydrogène dans l'état fondamental. Journal de Physique, 35:829, 1974.
- [78] William H. Press, Brian P. Flannery, Saul A. Teukolsky, and William T. Vetterling. Numerical Recipes, The Art of Scientific Computing. Cambridge University Press, Cambridge, 1986.
- [79] V. J. Folen, W. G. Maisch, J. D. White, C. A. Bartholomew, and A. H. Frank. Hybrid modes in anisotropic dielectrics. Memorandum Report 4870, U.S. Naval Research Laboratory, 1982.
- [80] U. E. Israelsson and C. M. Gould. High-field magnet for low-temperature low-field cryostats. Rev. Sci. Instrum., 55(7):1143, 1984.
- [81] J. C. Slater. Microwave electronics. Rev. Mod. Phys., 18:441, 1946.
- [82] D. J. Wineland and N. F. Ramsey. Atomic deuterium maser. Phys. Rev. A., 5:821, 1972.
- [83] Samuel R. Stein. Precision Frequency Control, volume 2: Oscillators and Standards, chapter 12: Frequency and Time-Their Measurement and Characterization, page 192. Academic Press Inc., Orlando Florida., 1985.
- [84] J. A. Barnes, A. R. Chi, L. S. Cutler, D. J. Healey, D. B. Leeson, T. E. McGunigal, J. A. Mullen Jr., W. L. Smith, R. L. Sydnor, R. F. C. Vessot, and G. M. R. Winkler. Characterization of frequency stability. *IEEE Trans. Instrum. Meas.*, IM-20:105, 1971.
- [85] J. D. Jackson. Classical Electrodynamics. John Wiley & Sons, New York, 2nd edition, 1975.

Bibliography

- [86] Ernest V. Loewenstein, Donald R. Smith, and Robert L. Morgan. Optical constants of far infrared materials. 2: Crystalline solids. Applied Optics, 12(2):398, 1973.
- [87] Robert Shelby, John Fontanella, and Carl Andeen. The low temperature electrical properties of some anisotropic crystals. J. Phys. Chem. Solids, 41:69, 1980.