Quantum Coherent Control and Compensation of Temporal Scattering

by

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Abstract

The experimental work in this thesis is divided into two distinct parts. In both parts, broadband femtosecond laser pulses are “shaped” by adjusting the relative phase and amplitude of spectral components.

In the first set of experiments, time-dependent perturbation theory is used to show that the probability of a quantum transition in atomic rubidium can be substantially enhanced or suppressed using pulse shaping, compared to the probability of transition observed when a transform-limited or “flat phase” optical pulse is used. These enhancement or suppression effects are also demonstrated experimentally. As quantum interference (the material phase having been transferred from the optical phase) is used to enhance or diminish a particular final quantum state, this can be classified as a quantum coherent control experiment.

In the second set of experiments, an optical pulse is scattered into a train of pulses by a layered structure. The layered structure is used to simulate the effect of optical pulses travelling through certain types of complex media. One consequence of the disruption of a single pulse into a train of pulses is lower per-pulse peak intensity, and thus a greatly diminished nonlinear signal. It is shown that spectral pulse shaping (in phase only) is sufficient to pre-compensate for the scattering structure, allowing a single transform-limited pulse to be obtained at the output.
Preface

This thesis is original, unpublished, independent work by the author, Timothy DeWolf.
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Dedication

I dedicate this thesis to the Only True and Living God.

I also thank my wife Chelsea and my two little sons Aaron and Enoch. I love them.
Part I

Motivation and Introduction
Chapter 1

The Goal of Controlling Matter with Light

My personal interest in studying the topics that appear in this thesis stems from a long-time interest in the idea that electromagnetic fields can both get information from and change the “state” of material systems. I am especially interested in eventually applying these ideas to complex biological systems.

Using electromagnetic fields to read information from molecular systems is one type of spectroscopy. Distinct ranges of the electromagnetic spectrum give information about particular characteristics of a target system [1]. For example, infrared (IR) radiation can elicit quantum rotational and vibrational transitions. Infrared absorption spectra allow accurate identification of characteristic functional groups based on rotational and vibrational structure; often they facilitate exact identification of simple molecular species. Radiofrequency (RF) radiation is capable of causing nuclear spin state transitions, giving rise to a field of study known as nuclear magnetic resonance (NMR) spectroscopy. Advanced NMR techniques (making use of RF pulse sequences that have been designed using quantum mechanics) can obtain enough information to allow the reconstruction of both the three-dimensional structure and the temporal dynamics of proteins [2]. Often, spectroscopic methods that employ electromagnetic fields collect information non-invasively. In contrast, some types of spectroscopy (e.g. mass spectroscopy) and many other common chemical (e.g. titration) and biochemical methods of analysis destroy the sample of interest. Often a combination of non-destructive and destructive methods is of use; for example, purification methods that isolate only the protein of interest from a cell are often used to increase signal-to-noise and reduce spectral congestion in NMR spectroscopy.

But what about using light to lastingly change or control the state of a particular target system, not just read information?
1.1 Biological Control with Incoherent Fields

Biologists have discovered a number of systems which can be “controlled” with incoherent light: the specific design of the each system allows it to respond in a certain functionally significant way to electromagnetic fields.

One example of such a system is vitamin D\(_3\) (cholecalciferol). Vitamin D\(_3\) is produced in the skin from 7-dehydrocholesterol (provitamin D\(_3\)). 7-dehydrocholesterol is converted to previtamin D\(_3\) after the absorption of a photon of ultraviolet (UV) light. Upon relaxation to the ground state, previtamin D\(_3\) is converted to (at least) six different products, among which is vitamin D\(_3\), an essential nutrient. It is an equilibrium process with spectral sensitivity to the wavelengths of incoherent radiation used \([3, 4]\).

A more elegant example is that of rhodopsin. This protein contains the retinal molecule as a cofactor; incoming light induces a conformational charge of 11-cis-retinal to all-trans-retinal. This conformational change induces a subsequent cascade of signal transduction events in the eye, enabling vision \([5]\). Many other examples of biological systems functionally responsive to incoherent electromagnetic fields by design exist.

1.2 An Early Attempt at Molecular Control with Monochromatic Fields

Is it possible to optically control an arbitrary chemical (or biological) system, namely one that has not been designed to respond to optical excitation in any especial functionally significant way? (This is in contrast to the biological examples given above.)

To answer this, consider for example the following early attempt \([6]\) to optically break a chemical bond in a molecule. Similar to the way that single-photon absorption causes a state transition in an atomic species, a narrow-bandwidth (nearly monochromatic) laser beam is tuned to excite a particular rovibrational mode in a molecule. The idea justifying this approach is that as long as the energy of excitation remains localized, the resulting product states will be influenced—breaking, for example, a particular chemical bond. There are a few special cases where this method succeeds, and the success of these cases correctly suggests that one may use optical fields to control arbitrary chemical and biological systems.

But this simple control scheme based on monochromatic excitation fails in the majority of cases. The term intramolecular vibrational redistribution (IVR) describes the fact that such excitations do not remain localized \([6]\).
1.3 Quantum Coherent Control

An excitation intended to break a given chemical bond moves away from the original site before product formation is complete. This naive approach to control has ignored the true time evolution of the target, which is defined by the molecular Hamiltonian (including, e.g., in the Born-Oppenheimer approximation, the potential energy surface). Successful control schemes employ the full molecular Hamiltonian, correctly treating the temporal evolution of the system. In this way, the process of IVR that follows monochromatic field excitation is correctly predicted.

1.3 Quantum Coherent Control

Two successful control schemes based on the full molecular Hamiltonian are introduced here. Both rely on coherent broadband optical fields. Such fields are readily obtained using pulsed lasers (Chapter 3). A coherent field has a well-defined phase relationship across the whole bandwidth, and via Fourier analysis, can give rise to pulses that are short in time.

1.3.1 Tannor-Rice Coherent Control

Tannor-Rice or “pump-dump” coherent control \[6–8\] is a scheme specific to the control of molecular species described using wavepackets. Consider the following example \[6\]. A triatomic molecule ABC may dissociate in two ways,

\[
\begin{align}
  \text{ABC} & \rightarrow \text{AB} + \text{C} \\
  \text{ABC} & \rightarrow \text{AC} + \text{B}
\end{align}
\]  

(1.1a) (1.1b)

An initial pump pulse prepares an excited state wavepacket, moving vertically from the ground to the excited state surface. This excited state then evolves in time, and the key to the method is the timing of the subsequent dump pulse. At certain time, the dump pulse will overlap better with an exit channel for the process of Eq. 1.1a whereas at another time process Eq. 1.1b is favored. The method has been confirmed experimentally \[6, 9\], and it is seen that the production of AB+C or AC+B may have an oscillatory character with respect to the pulse separation time. The mathematical description of this method \[6, 7\] is straightforward but will not be presented here.
1.3. Quantum Coherent Control

1.3.2 Brumer-Shapiro Coherent Control

The basic principle of Brumer-Shapiro control \[6, 10, 11\] is to look around for initial and final states of a system connected by two (or more) independent pathways. One chooses amplitudes and phases of the incoming field, which are transferred to the relative amplitudes and phases in the pathways between initial and final states in the material system. Control is gained as the pathways leading to a particular final state interfere constructively or destructively, thereby enhancing or diminishing the presence of any observables associated with the system having amplitude in that final state. Thus a particular Brumer-Shapiro control scheme is just the clever use of quantum interference.

Consider the simple two level system in Fig. 1.1. Each path from initial state \(|i\rangle\) to final state \(|f\rangle\) (via intermediate state \(|m_1\rangle\) or \(|m_2\rangle\)) involves two photons; this allows unique optical phases to be assigned to each path when using a broadband input field. As optical phase is transferred to quantum phase (see \[10\] or apply time-dependent perturbation theory), the final probability of finding the system in state \(|f\rangle\) is given by

\[
P_f = |c_1 + c_2|^2
\]

where \(c_j\) is the quantum transition amplitude of each two photon path \(j\). If one varies only the phase, control is gained through the interference term that will appear in the expansion of \(P_f\). Amplitude modulation may also be used.

Although Brumer-Shapiro control experiments typically require coherent
fields, it is possible to construct control methods that use optical frequencies from two different laser sources that do not have a well-defined phase relationship between them [6, 12]. The method makes use of virtual transitions, constructing a control sequence that effectively cancels out the phase dependence.

Optical coherent control schemes are not limited to controlling the dynamics of simple molecular systems in solution. The technique is very general, and has been applied to quantum dots and spin systems [13]. Quantum coherent control schemes have also been successfully applied to biological systems [14]. Some examples in protein systems are the chirped-pulse control of fluorescence in green fluorescent protein [15] and control of energy flow between intra- and intermolecular channels in a particular light-harvesting complex [16].

In this work, Brumer-Shapiro quantum coherent control is applied to two photon absorption in an atomic system (see Chapter 7).

### 1.4 Spectral Pulse Shaping

The Brumer-Shapiro method described in Section 1.3.2 and depicted in Fig. 1.1 requires the ability to adjust the phase and/or amplitude of individual frequency components in a broadband optical field. When such spectral adjustments are made to an electromagnetic field that is pulsed in time, it is called pulse shaping.

Optical pulse shapers typically operate in the frequency domain; a prism or diffraction grating maps spectral components of the optical field onto a spatial plane, where some type of 1D mask is placed. This mask may be fixed or programmable; it may allow the phase, amplitude and/or polarization of individual frequency components in the pulse to be altered. As an example, it is possible to develop a static mask which converts optical pulses from having a Gaussian temporal envelope to a square temporal envelope. This was shown to improve the switching behavior of an all-optical switch [17]. Two examples of programmable masks in common use are acousto-optic modulators (AOMs) [18] and liquid-crystal spatial light modulators (LC-SLM). In this thesis, the LC-SLM mask will be used exclusively. It contains a row of finite-width pixel elements.

As this optical pulse shaper shapes in the frequency domain, it is also called a spectral pulse shaper (or Fourier pulse shaper). The details of spectral shaping are presented in Chapter 5.
Chapter 2

The Goal of Controlling Matter with Scattered Light

2.1 The Problem of Optically Scattering Material

The introduction of coherent optical fields has introduced a new challenge in the case when the light must first pass through optically scattering material before reaching the control target. When a laser beam passes through an optically scattering material it will be distorted spatially; for example, a speckle pattern is produced. This speckle pattern results from multi-path interference between the different spatial components of the input beam as they traverse the scattering material; this process is shown schematically in the inset in Fig. 2.1. Some non-collinear optical paths at the output constructively interfere, producing bright spots in the speckle field; others destructively interfere, producing dark regions.

If the beam is pulsed (corresponding to a finite spectral bandwidth) the temporal profile of the pulses will also be altered. The ballistic pulse is defined as that part of the input pulse that travels straight through without scattering. As the effective thickness of the scattering material is increased, less and less of the ballistic pulse survives. The intensity of this ballistic pulse can be described by

$$I_b = I_0 e^{-\mu x}. \tag{2.1}$$

$I_0$ is the input beam intensity, $1/\mu$ is the scattering length (a measure of the strength of scattering), and $x$ is the total thickness of scattering material that has been traversed. The meaning of this exponential decay is shown in the main part of Fig. 2.1; the solid black represents the ballistic pulse, while gray part represents the scattered light, forming a temporally delayed and stretched pulse. Temporal disruption also means spectral disruption, as the two are connected via Fourier transformation.

Thus, attempts to do coherent control with light that has passed through a spatially scattering material are hindered in two ways:
2.1. The Problem of Optically Scattering Material

Figure 2.1: The effect that multiply scattering material has on a broadband pulse. As the unscattered or ballistic pulse passes through the material, light is scattered along various optical paths. The \( x \) coordinate is the thickness of the scattering material in the direction of the input beam. This figure is from [19].

- the originally well-collimated beam is broken up into a speckle field, so that the intensity of each speckle is a fraction of the original beam intensity, and,
- the spectral phase relationships in the scattered light have been disrupted.

The first point (loss of peak intensity) means that multiphoton optical processes, an important part of Brumer-Shapiro coherent control (Section 1.3.2), will be reduced, as these optical processes are intensity dependent (Chapter 4). The second item means that the chosen phase relationships in the input field may not match those in the output field, detrimental for Brumer-Shapiro coherent control where proper phase relationships are critical.

One way to cope with such spatial and spectral scattering is to simply treat it as noise, and do as much as possible with the transmitted ballistic pulse. For example, an experiment [19] was done where a broadband pulsed laser source was aimed through a thin layer of raw chicken breast onto a pH-sensitive probe molecule capable of two-photon absorption and fluorescence. Two phase optimizing masks were then discovered via an adaptive search algorithm (see Section 6.2), each mask maximizing the fluorescence signal in a distinct pH region. These coherent control experiments were successful simply because the ballistic pulse was sufficiently strong; the scattered field
2.2. Experimental Compensation of Spatial Scattering

Experiments have shown that the amount of coherent control available after light passes through a scattering material is not limited to the availability of a ballistic (or unscattered) light. Thus, one can go beyond the ballistic pulse regime discussed above.

Consider the following experiment by Vellekoop and Mosk [20]. A strongly scattering material, a 10.1 µm thick layer of TiO$_2$, is used. Only spatial speckle (and not spectral) compensation is investigated, as the sample is illuminated with a monochromatic HeNe laser. The incoming beam is strongly disrupted, and a spatial speckle pattern is observed (Fig 2.2(a)).

Vellekoop and Mosk then introduce a 2D spatial light modulator (SLM) into the beam. A 2D SLM is a programmable 2D array of pixel elements; each pixel modulates the phase of part of the spatial wavefront of the beam. (This is different from the spectral shaper containing a 1D SLM introduced previously in Section 1.4 as the pixel elements modulate across the spatial profile of the beam instead of modulating the spectral components.) The incoming wavefront is subdivided into $N$ segments, corresponding to the available pixels of the 2D SLM (or small blocks of pixels). $A_n$ and $\phi_n$ are respectively the amplitude and phase of the incoming optical field in segment $n$. $t_{mn}$ is a transfer matrix representing the effect of the scattering material on the impingent field. The output field is then described by

$$E_m = \sum_{n=1}^{N} t_{mn} A_n e^{i\phi_n}. \quad (2.2)$$

When the phases on the 2D SLM are set to zero, $\phi_n = 0$, the output field is again the speckle pattern, shown in Fig. 2.2(a). Compensation is achieved in the following way:

- An optical detector sensitive to the total light intensity in a localized region is placed, for example, near the center of the speckle field.

- The phase $\phi_n$ of a pixel on the 2D SLM is scanned from 0 through $2\pi$. At each value of phase, the intensity at the detector is measured. The phase of that pixel that maximizes the amount of light at the detector is recorded.
2.2. Experimental Compensation of Spatial Scattering

Figure 2.2: The transmission of light through a thin, strongly scattering layer of TiO$_2$, as demonstrated by Vellekoop and Mosk. a. The transmission micrograph of the speckle pattern that results when a flat phase incident beam is scattered by the sample. b. and c. Demonstrations of single-spot and five-spot focusing, following the iterative procedure described in the text whereby the incoming beam is pre-compensated. d. The pre-compensation phase mask used in (c), restricted to the area of the impingent beam. This is the original figure, taken from their paper [20].
2.2. Experimental Compensation of Spatial Scattering

- Subsequent pixels are scanned in the same way, repeating the previous step. Each time a pixel is scanned, all other pixels are set to zero.

- At the end of this process, the optimal phase values found for each pixel are applied to the 2D SLM. This is possible as the transformation in Eq. \(2.2\) is linear, meaning that the global optimum is the superposition of per-pixel optima.

Let this be called the “method of Vellekoop and Mosk,” as it will be referred to several times in what is to follow. The result of following this procedure with a single point detector and with five point detectors arranged in a ring is shown in Fig. 2.2(b) and 2.2(c), respectively. The intensity of the single-spot focus found is 1000 times above the intensity of speckle-pattern background level before compensation. The compensating mask found can be viewed as being the conjugate or inverse of the phase mask applied by the scattering material.

2.2.1 Experimental Compensation for Temporal Scattering

The above technique of Vellekoop and Mosk has been repeated with a broadband pulsed laser \([21, 22]\). It was found that the temporal disruption of a pulse was automatically compensated simply by bringing the beam to a spatial focus. This was possible because the temporal disruptions are caused by the same mechanism as spatial speckle—they are both caused by multiple scattering events inside the medium. The conjugate phase mask that compensates the spatial scattering then also mitigates the spectral disruption.

However, such temporal compensation depends upon the scattering in the material being weak. This means for example that the mask that recovers a focus at one frequency is effective at all other frequencies. There also exists another regime, that of strong scattering. In this case, the temporal disruption of the pulse by the scattering material is enough that the mask found via the method of Vellekoop and Mosk for one frequency does not automatically compensate at all other frequencies. In this second regime, automatic temporal (or spectral) compensation is not possible. (The transition between the two regimes is discussed and quantified in \([23]\).)

The individual speckles resulting from a spatially scattering material can also be temporally pre-compensated using a spectral shaper \([24]\). In this work the possibility of compensating temporal scattering using a spectral shaper will be further explored. Instead of a bulk 3D material, however, a 1D system is used to scatter the incoming pulse. This is the topic of
Chapter 8 and contributes the second portion of experimental work done herein.

The method of Vellekoop and Mosk is not able to compensate 1D scattering of the type used in this work for two reasons. First, there is no spatial variation of the scattering in the plane of the 2D SLM. Secondly, the scattering studied here falls into the “strong” scattering regime, as introduced above. Thus even if spatial speckle were present, the spatial compensation would not ensure automatic temporal compensation.

### 2.2.2 The Idea That Scattering Media Can Enhance Control

Up until this point, scattering has been portrayed as something that must be compensated, something that is a nuisance to coherent control. However, spatially scattering media of various types can actually enhance various aspects of signal transmission, or in other words, add possibilities beyond what was available with free-space propagation.

For example, again using the method of Vellekoop and Mosk, a focus was achieved past a scattering material [25]. The focus achieved with the scattering medium present beats the diffraction limit—by as much as ten times, in one experiment. This idea, that scattering materials can enhance control, will not be explored in this thesis.
Part II

Tools Used and Developed
Chapter 3

Femtosecond Lasers

3.1 Overview

A two-stage laser system is used. In the first stage, a femtosecond oscillator generates seed pulses. Subsequently, these seed pulses are fed into a regenerative amplifier. This two-stage laser seed production and amplification scheme is illustrated schematically in Fig. 3.1 and described in the following sections. The resulting high-power beam is split at the output; a portion of this beam is fed into a pulse shaper (Chapter 5), which is then used in the pulse shaping experiments presented here. The rest of this output beam feeds other experiments in the lab and also powers two optical parametric amplifiers (OPAs).

3.2 The Femtosecond Oscillator

The laser oscillator used here is a “Synergy,” manufactured by Femtosecond Produktions GmbH. It is a Ti-sapphire mode-locked laser. A 532 nm green pump laser pumps a titanium-sapphire lasing crystal inserted into the cavity at the Brewster angle. Ti-sapphire supports a broad fluorescence bandwidth.

![Schematic diagram of the two-stage amplification scheme used. The femtosecond oscillator creates a train of femtosecond pulses, which seed the regenerative amplifier (RGA).](image-url)
and is thus able to support the formation of a broad bandwidth laser pulse. At startup, the laser initially runs in a non-modelocked or continuous wave (CW) mode: the various cavity modes present at this stage do not have a well-defined phase relationship. By perturbing the length of the cavity after a period of thermal stabilization, the modes can be locked into a well-defined relationship with each other. This allows the cavity to emit a train of femtosecond-duration pulses instead of a continuous wave.

Several processes contribute to stable, modelocked operation [27]. When the cavity is initially perturbed to initiate modelocking, a small subset of modes can be produced which have a phase relationship between them favorable to the production of temporal spikes in intensity in the laser cavity. In order to create a single, strong pulse from any number of intensity spikes, a process known as Kerr focusing is used. (This is related to the Kerr effect [28]; see also Chapter 4 for a general introduction to nonlinear optics.) Kerr focusing means that these higher-intensity temporal spikes are brought to a closer focus in the cavity than those weak in intensity. By aligning the (focused) pump beam into the crystal such that it overlaps with the high-intensity Kerr-focused cavity modes, a type of self-amplitude modulation (SAM) is implemented: low intensity fluctuations that would disrupt the formation of a stable, equilibrium pulse are trimmed away. The Kerr effect occurring in the crystal due to the presence of high intensity optical fields is also responsible for self-phase modulation (SPM), whereby all available cavity modes can be brought into a modelocked relationship during the initial stages of pulse formation. Both SAM and SPM contribute to long term pulse stability, imparting tolerance to intra-cavity noise.

Finally, as the laser pulse must pass through the gain medium, it is necessary to compensate for dispersion. Special layered mirror structures [29] are employed which compensate dispersion, thus shortening the pulse temporally. This train of ultrashort, broadband pulses is then fed in to the regenerative amplifier. The frequency of pulses in the output train is 78 MHz.

3.3 The Regenerative Amplifier

The regenerative amplifier (RGA) is a Spitfire Pro, made by Spectra-Physics [30]. It is a laser, capable of running independently (initially seeded, like the femtosecond oscillator, from cavity fluctuations). However, it is specifically designed to use femtosecond pulse seeds from a laser oscillator. The RGA also uses a Ti-sapphire lasing crystal, and an external pump.
The intra-cavity pulse intensities that are reached in this RGA lasing cavity would be above the damage threshold (10 GW/cm²) for Ti:sapphire if short femtosecond seed pulses were used unmodified. To prevent this damage, a three stage amplification process is instead employed: the pulse is “stretched,” “amplified,” then “compressed.” This is called **chirped pulse amplification**, and will now be explained.

The pulse is stretched in time, so that the same energy is present, but the peak intensity drops. This allows the stretched pulse to be amplified many times without damaging the crystal. A grating-based pulse stretcher is used to stretch the pulse in time, creating what is termed positive **group velocity dispersion** (GVD) on the pulse; the resulting beam has been stretched in time and has the same energy but much lower peak intensity.

The next stage is amplification. At a regular interval, a pulse from the seed train is allowed to enter the cavity. This pulse then undergoes several rounds of amplification, causing broadband spontaneous emission from the Ti:sapphire gain medium. The pulse is then dumped to the compressor.

In order to select (“pick”) a pulse from the seed train and confine it in the cavity, a Pockels cell will turn on (voltage will be applied), causing it to rotate the polarization as a 1/4 waveplate. This pulse then passes twice through a (passive) 1/4 waveplate inside the cavity, and back through the active Pockels cell. The cavity is built to retain only horizontally polarized light. In this way, only the selected pulse enters and remains in the cavity. The Pockels cell then switches off before the next seed pulse; the next seed pulse still enters the cavity, but subsequently has its polarization rotated to vertical and leaves the cavity. This continues while the selected horizontal pulse passes repeatedly through the cavity and is amplified. A second Pockels cell can be used to dump (like a Q-switched laser) the cavity at the appropriate point (once the gain medium inversion has been depleted sufficiently).

The compressor is also a grating-based optical component. It adds negative GVD to the previously stretched pulse, causing the output pulse to be brought (close) to its minimal temporal length, as dictated by Fourier theory. This minimal temporal length happens when the spectral components of the pulse all have the same phase (i.e., zero phase difference). This is known as the Fourier transform limit, and such a pulse is called a **transform-limited** (TL) pulse. The spectrum of the approximately transform-limited output pulse obtained in the lab is shown in Fig. 3.2. The frequency of pulses in this final output train from the RGA is 1 kHz.
3.3. The Regenerative Amplifier

Figure 3.2: The typical power spectrum of the laser light used in this work (all parts of the thesis), measured with a 300 lines/mm grating spectrometer.
Chapter 4

Nonlinear Optics

The study of the optical phenomena occurring when intense fields interact with material systems is known as nonlinear optics [28]. Specifically, sufficiently intense optical fields (such as the pulses from the RGA) elicit a nonlinear polarization response from the medium, which in turn leads to re-radiation of new harmonic frequencies.

4.1 Nonlinear Polarization and the Introduction of New Frequencies

The polarization $P(t)$ of a material is a measure of the dipole moment per unit volume present because an electric field $E(t)$ has been applied. The spatial dependence has been omitted in this notation. If the polarization response is nonlinear, it can be expressed as a power series,

$$P(t) = \epsilon_0 \left( \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \cdots \right)$$

in powers of the incident field. $\epsilon_0$ is the permittivity of free space, and $\chi^{(n)}$ is the $n$th order susceptibility. In an anisotropic medium, the $\chi^{(n)}$ become $n+1$ rank tensors. Also, the assumption that the medium responds instantaneously is implicit in this expression [28].

4.1.1 Second Harmonic Generation

If an intense incident optical field described by

$$E(t) = \tilde{E} e^{-i\omega t} + \tilde{E}^* e^{i\omega t} \equiv \tilde{E} e^{-i\omega t} + c.c.$$  \hspace{1cm} (4.2)

is incident on an appropriate material (non-centrosymmetric), Eq. 4.1 says it will have a second-order polarization response

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} E^2(t)$$

$$= 2\epsilon_0 \chi^{(2)} \tilde{E} \tilde{E}^* + (\epsilon_0 \chi^{(2)} \tilde{E}^2 e^{-2i\omega t} + c.c.).$$ \hspace{1cm} (4.3)
c.c. stands for the complex conjugate of the preceding term(s). The zero frequency component or DC term is a static electric field, and the process is called optical rectification. The terms at ±2ω represent the second harmonic of the fundamental. Using Maxwell’s equations, this polarization leads to the generation of an optical field containing these new frequencies (see Section 4.2).

Second harmonic generation will be used for two pulse characterization techniques known as MIIPS (Section 5.5.4) and FROG (Section 5.5.3); these will be introduced later. Second harmonic generation will also be used as the nonlinear feedback signal in the work on the compensation of temporal scattering in Chapter 8.

4.1.2 Sum-Frequency Generation

If an intense incident optical field described by

\[ E(t) = \tilde{E}_1 e^{-i\omega_1 t} + \tilde{E}_2 e^{-i\omega_2 t} + c.c. \]  

(4.4)

is incident on an appropriate material (non-centrosymmetric), it will yield the following second-order polarization response [28],

\[ P^{(2)}(t) = \varepsilon_0 \chi^{(2)}(t) = \varepsilon_0 \chi^{(2)}(\tilde{E}_1^2 e^{-2i\omega_1 t} + \tilde{E}_2^2 e^{-2i\omega_2 t} + 2\tilde{E}_1 \tilde{E}_2 e^{-i(\omega_1 + \omega_2) t}) \]

\[ + 2\tilde{E}_1 \tilde{E}_2^* e^{-i(\omega_1 - \omega_2) t} + c.c.) + 2\varepsilon_0 \chi^{(2)}(\tilde{E}_1 \tilde{E}_1^* + \tilde{E}_2 \tilde{E}_2^*) \]  

(4.5)

In this expansion one sees terms corresponding to optical rectification and second harmonic generation. One also observes components appearing at both the sum of and the difference between of the incident frequencies, called sum- and difference-frequency generation, respectively.

In this work, sum-frequency generation is important for a closely related variant of the FROG pulse characterization scheme, known as XFROG (see Section 5.5.3).

4.2 The Nonlinear Wave Equation

It is now shown how Maxwell’s equations may be used with such a nonlinear polarization response. An important result that will arise is the prediction of what is known as phase-matching. This important concept lays the foundation for efficient second-harmonic generation in the lab.

Several approximations will be made to simplify this treatment; assume that the nonlinear response happens in a material that
4.2. The Nonlinear Wave Equation

- contains no free charges: $\rho = 0$,
- contains no free currents: $\vec{J} = 0$,
- is nonmagnetic, such that $\vec{B} = \mu_0 \vec{H}$, and,
- is isotropic.

The displacement field $\vec{D}$ is $\vec{D} = \epsilon_0 \vec{E} + \vec{P}$, with $\vec{P} = P^L + P^{NL}$ written to show the linear and non-linear polarization terms explicitly.

Maxwell’s equations in a medium \[28\],

\[
\begin{align*}
\vec{\nabla} \cdot \vec{D} &= \rho \\
\vec{\nabla} \cdot \vec{B} &= 0 \\
\vec{\nabla} \times \vec{E} &= -\frac{\partial \vec{B}}{\partial t} \\
\vec{\nabla} \times \vec{H} &= \frac{\partial \vec{D}}{\partial t} + \vec{J},
\end{align*}
\]

give the classical evolution of the electromagnetic fields in the system. These equations, with the help of the above approximations, yield the wave equation,

\[
\vec{\nabla} \times \vec{\nabla} \times \vec{E} + \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}.
\]

(4.7)

A substantial simplification follows if $\vec{\nabla} \cdot \vec{E} = 0$, which is not true in general but is true, for example, if $\vec{E}$ takes the form of an infinite plane wave. The vector identity

\[
\vec{\nabla} \times \vec{\nabla} \times \vec{E} = \vec{\nabla}((\vec{\nabla} \times \vec{E}) - \nabla^2 \vec{E})
\]

(4.8)

gives Eq. 4.7 to be written as

\[
- \nabla^2 \vec{E} + \epsilon^{(1)} \frac{\partial^2 \vec{E}}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}^{NL}}{\partial t^2}
\]

(4.9)

where $\epsilon^{(1)}$ is the scalar relative permittivity of the material. $\epsilon_0 \vec{E} + \vec{P}^L \equiv \epsilon_0 \epsilon^{(1)} \vec{E}$ for an isotropic material.

This equation shows how frequency components appearing in the nonlinear polarization response generate new optical fields at the same frequencies: the nonlinear polarization acts as the driving term in this wave equation for the electric field $E$. 
4.3 Sum-Frequency Generation and Phase-Matching

Eq. 4.9 can now be used to predict phase-matching in the case of sum-frequency generation. The basic ideas along with the solution are presented [28].

First, Eq. 4.9 rewritten in a form capable of describing the evolution of a single frequency component. One may simply Fourier transform it, or may follow the approach used by Boyd of substituting in a summation over frequency components $\omega_n$. Either way, the end result is the same; Boyd’s method yields the equation

$$-\nabla^2 \vec{E}_n + \frac{\epsilon^{(1)}(\omega_n)}{c^2} \frac{\partial^2 \vec{E}_n}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}_{NL}^n}{\partial t^2}$$

for frequency component $n$. $\epsilon^{(1)}(\omega_n)$ is the frequency component at frequency $n$, essentially the Fourier transform of the scalar permittivity defined previously.

For sum-frequency (or second-harmonic generation) one uses this equation to investigate how the sum-frequency component, $\omega_3$, generated from $\omega_1$ and $\omega_2$, behaves. (Broadband nonlinear generation around $\omega_3$ is an extension of this treatment.) Let

$$E_i(z, t) = A_i e^{i(k_i z - \omega_i t)} + c.c.$$  

(4.11)

describe the fields in system, all propagating collinearly in direction of the $z$ coordinate with equal optical polarization, and where

$$k_i = \frac{\sqrt{\epsilon^{(1)} \omega_i}}{c}.$$  

(4.12)

(An detailed derivation including the effects of non-collinear beam propagation is possible [31].) The nonlinear polarization term for this system ($\vec{P}_{NL}$) can be written as (see reference [28])

$$P_3 = 2\epsilon_0 \chi^{(2)} E_1 E_2 (e^{-i\omega_3 t} + c.c.).$$  

(4.13)

Putting these definitions into Eq. 4.10 one obtains

$$I_3 = \frac{2(\chi^{(2)})^2 \omega_1^2 I_1 I_2 L^2 \sin^2 \left( \frac{\Delta k L}{2} \right)}{n_1 n_2 n_3 \epsilon_0 c^2},$$  

(4.14)
4.4 Second-Harmonic Generation in a Birefringent Crystal

describing the intensity at $\omega_3$. $I_i = 2n_i\varepsilon_0c|A_i|^2$ give the intensity of the waves, with $n_i$ the refractive index at $\omega_i$, and $L$ gives the length of the medium, for example, $L$ could give the thickness of a crystal.

In order to arrive at Eq. 4.14 $A_1$ and $A_2$ have been taken as constant (initially they could have had $z$ dependence), undepleted input beams. The input beams often become significantly depleted; this approximation, however, allows for simplified mathematics which correctly predict phase-matching, even though the expression for $I_3$ as a whole is not valid [28].

Note the introduction of

$$\Delta k \equiv k_1 + k_2 - k_3.$$  

(4.15)

As an argument of the sinc function, this says that $I_3$ generation is maximized when $\Delta k = 0$. Physically, this says that the input and output waves (which are distinct frequencies) are in-phase with each other. The thickness of the medium $L$ also contributes to argument of sinc; excessively thick non-linear crystals increase the effect of any residual phase-mismatch $\Delta k$. This also helps in understanding why the thickness must be kept to a minimum when a broader range of frequencies must be used, as when doing broadband second-harmonic generation using femtosecond laser pulses.

4.4 Second-Harmonic Generation in a Birefringent Crystal

Having theoretically established the need for phase matching, one may briefly consider its physical realization in an arbitrary medium having normal dispersion. Using Eq. 4.12

$$\Delta kc = n_1\omega_1 + n_2\omega_2 - n_3\omega_3 = 0,$$  

(4.16)

one can compute the difference $n_3 - n_2$,

$$n_3 - n_2 = (n_1 - n_2)\frac{\omega_1}{\omega_3}.$$  

(4.17)

Normal dispersion says $n_i(\omega_i) > n_j(\omega_j)$ for $\omega_i > \omega_j$, and thus Eq. 4.17 does not have a solution in this case.

Two possibilities for realizing phase-matching in the lab are use of an anomalous dispersion material or a method known as quasi-phase-matching [28]. More commonly, however, one uses a birefringent crystal.

In birefringent crystal phase-matching, one can choose the input beam polarization(s) and the crystal angle such that the functions $n_i(\omega_i)$ take on
values that allow one to satisfy Eq. 4.17. Commercially prepared crystals are used in the lab, pre-cut such that phase matching can be achieved via rotation about a single axis.

In the experiments done in this work a 40-µm thick crystal of β-barium borate (BaB$_2$O$_4$ or BBO) is used. BBO is a nonlinear material with properties favorable [32] for use in a wide range of nonlinear optical experiments and devices, including optical parametric amplifiers (OPA).

4.5 Relation to Two-Photon Absorption

Two-photon absorption will be investigated later (Chapter 7). This is where an atomic transition is made by the absorption of two-photons of frequencies $\omega_1$ and $\omega_2$, whose sum is resonant with the energy of the total atomic transition. This process is actually very similar to the generation of second-harmonic light described here. When second-harmonic light is generated, e.g. in a BBO crystal, two-photons at $\omega$ are absorbed “virtually,” with the production of a photon at $2\omega$. The process is said to be parametric, as no net absorption of energy has taken place. The process of two-photon absorption in atom is similar, except that one now replaces one or both of the virtual levels with real levels (energy eigenstates) of the atom. The process is then non-parametric—a state transition has occurred, and instead of producing a field at the second-harmonic, relaxation pathways to other (atomic eigenstate) energy levels may dominate.
Chapter 5

Pulse Shaping and Characterization

The basic idea of spectral pulse shaping was previously introduced only briefly (Section 1.4). This chapter is an amalgamation of considerations pertinent to working with broadband pulses, considering both pulse shaping and pulse characterization methods. Pulse shaping means changing the spectral amplitude and/or phase content of a pulse; characterization means determining the amplitude and/or phase content of some optical field.

5.1 The $4f$ Pulse Shaper

The laser amplifier (RGA) is a source of approximately Gaussian-shaped pulses in the time domain. As expected from Fourier theory, the observed frequency spectrum is also approximately Gaussian. Attempting to shape the pulses directly in the temporal domain would require optical components with an extreme optical response times on the order of one femtosecond. This cannot be achieved with present day technology. Instead, one optically Fourier transforms the incoming pulse using a diffraction grating. The pulse’s spectral components are then manipulated directly, after which a subsequent optical Fourier transformation yields again a well-collimated beam of shaped pulses.

A schematic of this type of spectral pulse shaper is shown in Fig. 5.1. The purpose of the various components appearing in the figure will be described in the following subsections.

5.1.1 Before the Spatial Light Modulator (SLM)

The spectral components of an incoming beam must be spatially spread. A diffraction grating is used. A diffraction grating is a periodic structure which, when properly illuminated, leads to an interference pattern mapping spectral components of the incident light to a well-defined angular distribu-
5.1. The 4f Pulse Shaper

Figure 5.1: A schematic diagram of a spectral pulse shaper. The frequency components present in the input pulse $E_{\text{in}}(t)$ are presented along the spatial dimension of the dual-mask SLM. The SLM adjusts $E_{\text{in}}(\omega)$, yielding $E_{\text{out}}(\omega)$, which is then converted back into a well collimated beam of output pulses, $E_{\text{out}}(t)$.

\[ \sin \theta_i + \sin \theta_r = 10^{-6} n \lambda m. \]  
\[ (5.1) \]

Here, $\theta_i$ is the angle of incidence and $\theta_r$ the angle of diffraction, both taken with respect to the normal. $n$ is the spatial periodicity of “lines” in the diffraction grating, in units of lines mm$^{-1}$. The diffraction order $m \in \mathbb{Z}$ ($m$ is an integer) and the wavelength $\lambda$ in units of nm then give the spatial structure of the diffracted frequency components, order-by-order. Each diffractive order gives another copy of the spectrum.

The exact electric field produced in the far field after diffraction off the grating is given by Fraunhofer diffraction theory; it is the two-dimensional spatial Fourier transform of the aperture function $A(y, z)$, given by

\[ E(k_y, k_z) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dy \, dz \, A(y, z) e^{i(k_y y + k_z z)} \]  
\[ (5.2) \]

For the diffraction grating, the aperture function $A(y, z)$ is a product of a function specifying the physical bounds of the grating (e.g. “rectangle” functions in $y$ and $z$), multiplied by a function which describes the periodic structure of the grating (e.g. a periodic amplitude or phase modulation). For a grating with vertical lines (etched along the $z$ coordinate), the integration in the vertical domain is just the 1-dimensional transform of the beam’s (e.g. Gaussian) spatial profile. The interesting part of the transform is 1-dimensional transform of the product of the beam’s spatial profile, the grating’s physical boundaries in that dimension, and the periodic grating
5.1. The 4f Pulse Shaper

pattern. In the limit that the contributions to the transform from the first two of these are sufficiently broad and smooth features, one considers the effect of the grating only in the horizontal dimension, computing

\[ E(\vec{k}_y) = \int_{-\infty}^{\infty} dy A(y)e^{i(\vec{k}_y y)}. \]  \hspace{1cm} (5.3)

But \( A(y) \) is a unit periodic function on \([0,1)\) (with an appropriate choice of coordinate system), and so can be expressed by the Fourier series \([35]\),

\[ A(y) = \sum_{m=-\infty}^{\infty} B_m e^{i(2\pi my)} \]  \hspace{1cm} (5.4a)

\[ B_m = \int_{-1/2}^{1/2} dy A(y)e^{-i(2\pi my)} \]  \hspace{1cm} (5.4b)

\( B_m \) (Eq. 5.4b) gives the relative amplitude of diffraction order \( m \) \([36]\). This can then guide the design of a grating that contains the desired distribution of amplitudes among different orders. For the purpose of this pulse shaper, it is desirable that the power flowing into the first order (\( m = 1 \)) diffraction spot is maximized. This is achieved by the use of a blazed grating. Instead of, e.g., a sinusoidal pattern, a blazed grating chooses a repeating unit which is a wedge-like shape.

As shown in Fig. 5.1, a lens is used to bring the diffracted beam into a focus at the plane of the SLM. A spherical focusing mirror can also be used in place of the lens (with appropriate changes to the overall shaper layout). The Fourier plane then appears at \( 2f \), twice the focal length \( f \). As shown in the schematic diagram, this presents \( E_{in}(\omega) \) to the SLM masks. The operation of the SLM will be discussed in Section 5.2.

5.1.2 After the SLM

The final lens (or mirror) and diffraction grating bring the spectral components back to a time-domain pulse, \( E_{out}(t) \), effectively performing the inverse operation.

If the properly aligned SLM is set such that \( E_{in}(\omega) = E_{out}(\omega) \) (blank mask), then the resulting pulse emerges with no added dispersion. For this reason, the pulse shaper setup (the actual arrangement of the optical components) described here is said to create a zero dispersion line. This is also called a 4f-line, as the line relies on a total of four focal lengths from grating to grating \([18, 37]\).
5.2 The Liquid-Crystal SLM

A liquid-crystal SLM, manufactured by Cambridge Research & Instrumentation, Inc. (CRi), is used [38]. The orientation of the two masks in the spectral shaper was presented schematically in Fig. 5.1. Fig. 5.2 complements this figure, showing the individual pixels appearing in the two masks. Two 640-pixel masks appear back-to-back in this unit. Each pixel is 100 ± 0.005 µm wide; there exists a small 2.0 µm optical gap between each pixel that is not controlled. The two masks are aligned with each other to a tolerance of ±2.0 µm. The pixels are tall enough (5 mm) that the focused beam easily fits within the pixel area in this direction.

Each pixel is driven to a user-specified drive voltage or count, controlling the refractive index along the crystal’s extraordinary axis. The ordinary axis sees no change in index with the application of the field. The horizontally polarized input beam (see Fig. 5.1) is directed onto the first pixel mask, which has its extraordinary axis oriented at 45° to the input beam. The output field is drawn to emphasize the appearance of vertically polarized light, in addition to the horizontal component already present at the input.
second mask has its extraordinary axis oriented orthogonally to the first; if the two masks vary a pixel together, the resulting beam will be phase-modulated by this amount. If the two masks vary a pixel differently, the resulting beam will be modulated by this phase difference in polarization. If a linear polarizer is placed at the output, the resulting beam will be modulated in amplitude and phase. The intensity modulation due to a linear polarizer acting on a polarized beam is given by Malus’s Law [34],

\[ I(\theta) = I(0) \cos^2 \theta \]  

(5.5)

where \( \theta \) is the angle between a linear polarization component of the input beam and the output polarization, and \( I(0) \) is the incident intensity. It follows from the above considerations that modulation of the field due to the SLM is given by [18]

\[ E_{\text{out}}(\omega) = H(\omega) E_{\text{in}}(\omega), \]  

(5.6)

with complex transfer function

\[ H(\omega) = \exp \left( i \frac{\phi_1(\omega) + \phi_2(\omega)}{2} \right) \cos \left( \frac{\phi_1(\omega) - \phi_2(\omega)}{2} \right). \]  

(5.7)

\( \phi_n \) gives the phase appearing at an SLM pixel on the first or second mask, corresponding to \( n = 1,2 \). Note that the SLM must be calibrated in order to determine what electrical voltage, applied to a given pixel, produces an optical phase change \( \phi_n \).

### 5.3 Calibration and Operation of the SLM

In this section I discuss the process of calibration, followed by the procedure by which the SLM is used to display an arbitrary phase and amplitude mask.

#### 5.3.1 Pixel-Wavelength Calibration

The SLM is operated with a polarizer in place at the output, so that it can shape in amplitude. Amplitude shaping is used to block all but four pixels, appearing in a sufficiently intense part of the pulse spectrum, and the spectrum of the pulse past the SLM is acquired from the spectrometer (Section 5.5.1). A linear fit matches (to a good approximation) the SLM pixels to the wavelength locations of the peaks or dips in the spectrum; the wavelength matching each SLM pixel is thus determined. This is illustrated in Fig [5.3].
5.3. Calibration and Operation of the SLM

(a) Observed transmission spectrum after all but 4 pixels are blocked on the SLM. Here, for example, pixel numbers 200, 250, 345 and 400 are used.

(b) The open pixels are plotted against the peak wavelengths. The fit line indicates that pixel 0 (the y-intercept) modulates light at 752 nm and that each subsequent pixel is about 0.15 nm wide.

Figure 5.3: The pixel-wavelength calibration of the SLM.

5.3.2 Phase-Voltage Calibration

The phase-voltage calibration is simple: one needs to measure the phase modulation that the SLM produces in response to a particular voltage, for all pixels. The SLM is operated so that it can shape in amplitude and phase, with a polarizer at the output. Then the following procedure is performed:

1. All pixels on the first mask are set to the same drive voltage, starting at 500 and continuing to 1400 drive counts (in steps of 1), while the second mask is set to the midpoint voltage, namely 950 drive counts. This range of 500 to 1400 drive counts corresponds to a region of high modulation for this particular SLM, meaning that the SLM can produce relative phase changes between the masks in excess of $2\pi$.

2. Each time a flat voltage mask voltage is sent to the SLM, one must wait for the data to transfer to the SLM. One also must wait for the mask to become optically active, meaning that the liquid crystal molecules in each pixel have finished their motion in the newly applied electric field. A total wait of 500 ms is sufficient, most of which is data transfer time.

3. After the mask is active, a spectrum is acquired from the spectrometer. This spectrum shows the optical attenuation that the SLM has produced. As the voltage is swept the relative phase between masks
5.3. Calibration and Operation of the SLM

passes between multiples of $\pi$ and $2\pi$, and thus between regions of bright and dark. The pattern of bright (transmission) and dark (no transmission) is due to Eq. 5.7. Bright bands result when the phase difference $\phi_1 - \phi_2$ is 0. Dark bands result when the phase difference is $\pi$; at these spots, the light coming out from the SLM has been rotated such that it is blocked by the output polarizer.

4. The process is repeated with the first mask fixed and the second scanned. Two very similar sets of data are collected in this way, one for each mask. In Fig. 5.4 the data collected for one of the two masks is shown.

The analysis of this set of acquired spectra yields the desired phase-voltage calibration curves for each mask. The procedure is best illustrated visually, and thus each step here is accompanied by one of the plots appearing in Fig. 5.5 in order. The analysis is:

1. The entire data set is normalized to a global maximum of 1.
5.3. Calibration and Operation of the SLM

Figure 5.5: Steps of the analysis of the SLM calibration data in Fig. 5.4.
The analysis gives SLM phase-voltage curves for both masks, and at all pixels with sufficient input beam intensity. The analysis involves Eq. 5.9 as described in the text.
5.3. Calibration and Operation of the SLM

2. The response curve for a particular wavelength is isolated; this corresponds to taking one of the horizontal lines in Fig. 5.4. The peak spacing widens from left to right because the amount of SLM modulation per drive decreases. The curve is then fit to a smooth cubic spline function. This process can contribute to the minima and maxima not going to 0 and 1, beyond what is present in the original data. By finding the offsets between the minima and 0 and maxima and 1, linear functions constructed between the peaks allow the curve to be scaled to fit exactly between at 0 and 1. It is important for the following steps that this is the case.

3. The relationship between the phase difference across the two masks and the transmitted intensity is given by Eq. 5.7 in terms of intensity,

\[ I(\omega, V) = \sin^2 \left( \frac{\phi(\omega)}{2} \right) ; \] (5.8)

inverted, this is

\[ \phi(\omega) = 2 \arcsin \left( \sqrt{I(\omega, V)} \right) . \] (5.9)

\[ V \] is the drive voltage applied; it does not matter what the other mask voltage is as long as it is fixed. Thus, the first step in retrieving the phase is taking square root.

4. The sign lost in taking the square is recovered by multiplying the region between every other pair of zeros of the function by negative one.

5. The arcsin is now taken, as per Eq. 5.9.

6. This step involves the first step in unwrapping the phase, done by multiplying the curve by negative one wherever the curve sloped downward (which corresponds to the regions between every other pair of maxima in Step 3).

7. The last step in unwrapping the phase requires adding a \( \pi \) staircase function to the curve. The curve is then multiplied by two.

Some of the phase curves (for some of the SLM pixels) may come out of this analysis shifted by \( 2\pi \)—this can be corrected. The phase-voltage curves for a few of the SLM pixels (sampled at even intervals across the spectrum) is shown in Fig. 5.6. The phase values in the curves are adjusted so that \( \phi = 0 \) appears roughly in the center.
5.3. Calibration and Operation of the SLM

Figure 5.6: A sample of a few of the phase-voltage curves, sampled at evenly spaced intervals across the spectrum, used to calibrate the SLM. This is from the analysis of the data in Fig. 5.4.

The phase curves are assembled into a matrix, giving the phase at a given spectrometer wavelength and drive voltage. This calibration matrix is converted, via bilinear interpolation, into a matrix giving the phase at a given SLM wavelength and drive voltage.

5.3.3 Shaping in Phase and Intensity

In order to realize shaping in intensity and phase, one must calculate the two phase masks which will be applied to the SLM. Eqs. 5.7 and 5.9 suggest that the phases two phase masks, $\phi_1$ and $\phi_2$ respectively, may be computed using

$$\phi_1(\omega) = \phi(\omega) + \sqrt{\arccos(I(\omega))}$$  \hspace{1cm} (5.10a)

$$\phi_2(\omega) = \phi(\omega) - \sqrt{\arccos(I(\omega))}$$  \hspace{1cm} (5.10c)
5.4 Frequency-Domain Pulse Shaping

where \( I \) and \( \phi \) are the desired intensity and phase.

5.3.4 Converting Phases To Drive Counts

These phase values for each SLM mask must then be converted to drive count levels via the calibration matrix. In the process, the LabVIEW class method that handles this request does several things. First, the requested phase masks are combined (added) to the phase compensation mask (see Section 5.5.4). Then, this final requested phase is wrapped at \( 2\pi \), ensuring that no phase mask exceeds the available modulation range (e.g., outside the 500-1400 drive counts calibration range). Finally, the appropriate drive counts are determined and sent to the SLM.

5.4 Frequency-Domain Pulse Shaping

In electrical engineering, the concept of a linear, time-invariant filter can be used to describe the relationship between the temporal input and output signal in a system. This relationship is given by

\[
E_{\text{out}}(t) = E_{\text{in}} \star H(t) \equiv \int dt' E_{\text{in}}(t')H(t-t')
\]  

(5.11)

where \( E_{\text{in, out}} \) are respectively the input and output signals in the time domain, and \( H(t) \) is called the impulse response function \[39\]. Fourier transforming both sides and invoking the convolution theorem gives

\[
E_{\text{out}}(\omega) = E_{\text{in}}(\omega)H(\omega),
\]  

(5.12)

where \( H \) is now called the frequency response or complex transfer function.

As discussed in Section 5.1, the SLM sits in the Fourier plane of the lens (or spherical mirror), where it directly modulates the phase of the spectral components \( E_{\text{in}}(\omega) \). Thus \( H(\omega) \) represents the effect of intensity and phase modulation applied to the shaper and output is given by simple multiplication. This concept has of course already been introduced in Section 5.2, but here the relationship between the time and frequency domain is emphasized. Also, unlike Section 5.2, the details of the dual-mask structure are now not important; rather \( H(\omega) \) is simply the total phase and amplitude mask that will realized by the SLM upon request. The SLM allows masks in the form

\[
H(\omega) = A(\omega)e^{i\phi(\omega)}
\]  

(5.13)

where \( A \in [0,1] \) represents amplitude modulation and \( \phi \) represents the desired phase.
5.4. Frequency-Domain Pulse Shaping

At present, consider phase-only shaping,

\[ H(\omega) = e^{i\phi(\omega)}. \]  

(5.14)

Expanding \( H(\omega) \) as a Taylor series to order \( n \) gives \[ \phi(\omega) = \phi(\omega_0) + \phi^{(1)}_0 (\omega - \omega_0) + \cdots + \frac{1}{n!} \phi^{(n)}_0 (\omega - \omega_0)^n \]  

(5.15)

with \( \phi^{(n)}_0 = (d^n \phi / d\omega^n)_{\omega = \omega_0} \). The various terms each have distinct, simple effects on the resulting pulse in time domain, \( E_{\text{out}}(t) \):

- The carrier envelope phase, or the global phase of a particular pulse, is controlled by \( \phi^{(0)} \), and has units of radians.

- The pulse is delayed in time according to \( \phi^{(1)} \), having units of time. This is known as group delay.

- The pulse is temporally spread according to \( \phi^{(2)} \), having units of time squared. This is known as chirp. This is equivalent to the pulse stretching process described for the RGA (Section 3.3).

5.4.1 Numerical Fourier Transformation and Pulse Shaping

All shaping on the SLM has to be done in the frequency domain; it is sometimes desirable to predict the time-domain result of that shaping. One may use the fast Fourier transform (FFT) for this. However, some subtleties arise in using the FFT to convert an arbitrary spectrum into its time domain signal and vice versa. As such, this process is described here.

1. If the desired input spectrum is in units of wavelength it must be converted to units of frequency. This can be done simply via linear interpolation.

2. It is then advantageous to increase the resolution of, and zero pad, the input spectrum. Increasing the resolution can be done via linear interpolation. The final length of the upsampled, zero padded array may be chosen to be a power of 2, as the FFT typically operates optimally with such lengths.

3. The spectrum is mirrored onto negative frequencies, by taking the complex conjugate, as the time domain field is real.
4. Finally, the FFT is computed using standard methods. The resulting time domain points contain the real field. The two halves of the output array might have to be exchanged, an operation known as an “FFT shift” in some FFT libraries.

5. The time coordinates corresponding to these new time-domain field amplitudes can be computed using the relation

\[ \delta t = \frac{2\pi}{\Delta \omega}, \]  

(5.16)

where \( \delta t \) is the spacing in time between points in the transformed data, and \( \Delta \omega \) gives the total angular frequency extent of the input frequency array fed into the FFT.

There exist limitations to the types of output fields \( E_{\text{out}}(t) \) which can be realized with a particular SLM mask given by \( H(\omega) \). Via the Fourier transform, the temporal features which can be realized are limited because of the overall bandwidth. Also, and again via Fourier transformation, the pixel resolution limits the maximum length of the shaped pulse.

5.4.2 A Gallery of Shaped Pulses

In this section, a number of frequency-domain masks \( H(\omega) \) will be presented, along with their resulting effect on the pulse in time. These masks produce time domain behavior more complicated than the simple constant, linear and quadratic phase terms discussed, and have been computed using the numerical strategy outlined in Section 5.4.1. These masks are important because they are similar to the SLM masks used in the experiments appearing in Part III.

A Gaussian input pulse is used for the numerical pulse shaping computations in this work, as shown in Fig. [5.7]. The bandwidth of this numerical input pulse matches (to a good approximation) that of the laser amplifier used in the lab. The time domain electric field amplitude is also shown, computed via Fourier transformation. Second harmonic spectra are also shown in this section as they are important to the work in Chapter 8.

The \( \pi \) Step

Consider the phase mask shown in Fig. [5.8]

\[ \phi(\omega) = \pi \theta(\omega - \omega_0). \]  

(5.17)
5.4. Frequency-Domain Pulse Shaping

Figure 5.7: The Gaussian input pulse used for the numerical calculations performed in this work, along with its appearance in time and associated second-harmonic spectrum. This is provided for reference, as later results will provide only the SLM mask applied to this input.

\( \omega_0 \) is the pulse center frequency (\( \omega_0 = 2.355 \text{ fs}^{-1} \) corresponds to 800 nm), \( \theta \) is the Heaviside step function, and \( \phi(\omega) \) is used in Eq. 5.14. This delays half of the frequency components in the pulse such that the two “halves” of the pulse interfere, producing the shown time domain field.

Although the \( \pi \) step mask shown here is not used in this work, a variety of masks with sharp step-like features appear in Chapter 7.

### Pulse Trains

One may create a closely spaced sequence of pulses (known as a pulse train) from a single input pulse by using a certain type of SLM mask. As pulse trains will be very important to the work appearing in Chapter 8, they will be discussed here in some detail.

A structure occurring with periodicity \( \Delta \omega \) in the frequency domain (e.g. on the SLM phase mask) will lead to a repeating structure in the time
5.4. Frequency-Domain Pulse Shaping

Figure 5.8: The effect of a $\pi$-step phase mask; the phase mask steps from 0 to $\pi$ at the location of the center of the input pulse spectrum.
domain (e.g. in the pulse train coming out from the shaper) with periodicity \( \Delta t \), related by [40]

\[
\Delta \omega = \frac{2\pi}{\Delta t}.
\]  
(5.18)

The above equation does not address the question of how the intensity is distributed among the pulses appearing in the train. Recall that a blazed diffraction grating tailors the shape of each groove in order to change the spatial intensity of each diffraction order (see Section [5.1] and Eq. [5.4b]). Similarly, varying the structure of the periodic unit appearing on the SLM will change the relative intensity of pulses appearing in the pulse train. In the following, two different periodic units will be introduced.

A commonly used periodic structure that can be applied to the SLM mask is the sinusoidal phase function,

\[
\phi(\omega) = \alpha \sin(\tau(\omega - \omega_0)).
\]  
(5.19)

The result of applying Eq. [5.19] with \( \alpha = 1 \) and \( \tau = 300 \) ps is shown in Fig. [5.9]. Note that the resultant pulse train contains replicas of the input pulse with varying amplitude, and are spaced in time by the period \( \tau \).

Instead of a sinusoidal mask, consider an alternating binary mask; now each periodic phase unit is set to half zero, and half \( \pi \). Applying such a mask to the input pulse yields the pulse train shown in Fig. [5.10]. Note that Eq. [5.18] correctly relates the 20.9 ps\(^{-1} \) periodicity in the frequency domain (on the SLM mask) to the observed 300 fs pulse spacing. Compare the time domain distribution of pulses observed in here in Fig. [5.10] with Fig. [5.9]. The binary unit leads to a much longer train of pulses; the shape of the periodic unit is also seen to change the “sharpness” of the fringes appearing in the second harmonic spectrum.

5.5 Pulse Characterization Techniques

Pulse characterization refers to a method which calculates the amplitude (intensity), phase, or amplitude and phase of an unknown optical field.

5.5.1 Spectrometer

A spectrometer is a device which takes an unknown field, \( E(t) \), and returns the intensity spectrum, \( I(\omega) = |E(\omega)|^2 \). In the spectrometer used here, light first enters a narrow slit. The slit is used for choosing the resolution of the spectrometer. Making the slit narrow increases the spectral resolution, but limits the amount of light entering the spectrometer. Basic optics
Figure 5.9: The effect of a sinusoidal phase mask, \( \phi(\omega) = \alpha \sin(\tau(\omega - \omega_0)) \), \( \alpha = 1 \) and \( \tau = 300 \) ps. A train of pulses separated by \( \tau \) arises.
5.5. Pulse Characterization Techniques

Figure 5.10: The effect of a binary repeating unit used as an SLM mask. The periodicity of the mask is $20.9 \text{ ps}^{-1}$ in the frequency domain, chosen to again yield a 300 fs pulse separation. Note that the second harmonic spectrum reflects the change in repeating unit; all fringe peaks are now “sharp.”
5.5. Pulse Characterization Techniques

combined with a diffraction grating map the spectral components onto a charge-coupled device (CCD)-based camera that can detect the intensity of the spectral components.

The grating is rotated via a computer-controlled motor such that the current spectral window can be centered on an arbitrary wavelength (within a certain optical range). The grating line density determines the width of the spectral window observed at a given wavelength.

In order to calibrate the spectrometer, an atomic lamp (for example, krypton) with a well-known spectrum is used to illuminate the spectrometer, with a narrow slit width. Four spectral lines at known wavelengths are chosen, and a linear fit is used to determine the relationship between a pixel on the CCD and the wavelength of the light hitting it.

5.5.2 Autocorrelation

Autocorrelation can give a measure of the pulse length [41]. The PulseScout Autocorrelator made by Newport is used here.

The incoming pulsed laser beam is split with a beamsplitter, and one of the beams is delayed temporally by $\tau$, by allowing it to travel a longer distance. The distance is controlled via an internal motor in the PulseScout. Then the two beams are overlapped at a focal point in a nonlinear crystal (Section 4.4). They are focused to maximize the intensity of the nonlinear signal.

The two beams are thus $E(t)$ and $E(t - \tau)$. The generated second-harmonic signals for such a field can be found in

$$
(E(t) + E(t - \tau))^2 = E^2(t) + E^2(t - \tau) + 2E(t)E(t - \tau).
$$

Three beams emerge from the interaction. Second-harmonic associated with $E^2(t)$ continues to travel in the original direction of $E(t)$, and likewise for the second-harmonic generated from $E(t - \tau)$. The cross term $E(t)E(t - \tau)$ travels in a direction given by the vector sum of the wave vectors of the two incoming pulses, as required by photon momentum conversation [18].

Intensity Autocorrelation

If the beams overlap non-collinearly, the cross term travels in a spatially distinct direction, and can be isolated onto a detector. This is shown in Fig. 5.11. If the detector is a photodiode, it will integrate the ultrashort
Figure 5.11: An experimental realization of optical intensity autocorrelation. The input pulse is split, and one copy is delayed in time. The two pulses $E(t)$ and $E(t - \tau)$ are then brought to a focus non-collinearly in the second harmonic generation (SHG) crystal. This non-collinear arrangement allows the detector to be placed to detect only the sum-frequency generation (SFG) signal, or the cross term $E(t)E(t - \tau)$.

The pulse over time; the detector thus measures a signal proportional to

\[
M(\tau) = \int_{-\infty}^{\infty} dt \mid E(t)E(t - \tau)\mid^2 = \int_{-\infty}^{\infty} dt I(t)I(t - \tau)
\]

which is exactly the definition \cite{42} of autocorrelation for a signal $I$.

This method is thus known as intensity autocorrelation. The full-width at half maximum (FWHM) of the autocorrelation envelope, $\tau_{\text{autocor}}$, computed from a Gaussian input pulse that has a FWHM $\tau_{\text{input}}$ is found according to:

\[
\tau_{\text{input}} = \frac{\tau_{\text{autocor}}}{\sqrt{2}}.
\]

This numerical factor is different for non-Gaussian pulse envelopes; it is known as the deconvolution factor (see the measurement in \cite{43} for example). Thus the PulseScout, operating in non-collinear overlap mode, provides a simple way to check the temporal duration of a Gaussian-like input pulse.
5.5. Pulse Characterization Techniques

Interferometric Autocorrelation

If the two beams are collinear, the photodiode integrates the whole signal appearing in Eq. 5.20; the measurement is thus proportional to

$$M(\tau) = \int_{-\infty}^{\infty} dt |(E(t) + E(t - \tau))^2|^2.$$  \hspace{1cm} (5.23)

This method is called interferometric autocorrelation.

An optical setup which achieves interferometric autocorrelation is shown in Fig. 5.12. Interferometric autocorrelation has been introduced here because a variant of interferometric autocorrelation is used in Chapter 8 (where it is implemented using only the spectral shaper).

5.5.3 FROG

Frequency-resolved optical grating (FROG) is a pulse characterization method capable of retrieving the full complex field—amplitude and phase—of an unknown pulse. In a FROG experiment, one uses any method which can optically measure the spectrogram, defined as

$$S(\omega, \tau) = \left| \int_{-\infty}^{\infty} E(t)g(t - \tau)e^{-i\omega t} dt \right|^2.$$  \hspace{1cm} (5.24)

The gate field $g(t - \tau)$ can be the pulse $E(t)$ itself. If the gate field is not a replica of $E(t)$, the measurement is known as an XFROG (see below). Setting $g(t) = E(t)$, the thing being Fourier transformed in Eq. 5.24 is exactly the cross term of Eq. 5.20.
Thus, to collect what is termed a \textit{FROG trace} one likewise splits the input beam and sends one copy to a delay line which allows for the variation of $\tau$. The two beams are overlapped space \textit{and} in time. Again, as done in the autocorrelation measurement, the beam represented by $E(t)E(t-\tau)$ is isolated spatially. The Fourier transform is done by the spectrometer. By stepping through offsets of $\tau$ one assembles the desired spectrogram.

\textbf{XFROG}

A variant of FROG called XFROG (for cross-correlation FROG) \cite{45} overlaps two \textit{different} pulses. The spectrogram is

$$S(\omega, \tau) = \left| \int_{-\infty}^{\infty} E_{\text{ref}}(t)E_{\text{test}}(t-\tau)e^{-i\omega t} dt \right|^2,$$ \hfill (5.25)

with $E_{\text{ref}}(t)$ typically being a Gaussian-like reference pulse, typically discovered via FROG. $E_{\text{test}}$ can now be an unknown field. The nonlinear signal is now a sum-frequency signal.

Note that this use of second-harmonic or sum-frequency light is one of several methods available for generating a FROG or XFROG spectrogram; methods not involving nonlinear light generation exist.

\textbf{Field amplitude and phase retrieval}

Once a FROG (or XFROG) trace has been collected, it remains a numeric problem to figure out what real field $E(t)$ would give such a trace. The FROG trace contains enough information that simple pulse shapes can be characterized. However, theoretically the information present in the trace is insufficient, such that complex pulse shapes cannot be reliably retrieved \cite{44}.

In this work, a FROG analysis software package from Femtosoft Technologies, LLC is used, which retrieves the real field (both phase and amplitude) from a FROG trace. The actual algorithms \cite{46, 47} used by this software are complex.

\textbf{5.5.4 MIIPS}

The method known as the \textbf{multiphoton intrapulse interference phase scan} (MIIPS) \cite{48} retrieves field phase information, using information present in the spectrum of second harmonic light. This well-known method is used here to flatten the phase of laser pulses. The spectral shaper is used to make the initial field phase measurements, and to subsequently apply the
5.5. Pulse Characterization Techniques

compensating (inverse) phase mask. This compensating phase mask stays active and is added to all other phase shaping performed using the SLM.
Chapter 6

Software and Algorithms; The Genetic Search

6.1 Introduction

An extensive set of LabVIEW routines was written to communicate with external devices. These LabVIEW routines also do some basic data collection, such as that required for SLM calibration (Section 5.3), FROG (Section 5.5.3), and MIIPS (Section 5.5.4).

A program named QuantumBlackbox was then developed. Originally, this development was motivated by the desire to implement a genetic search algorithm in the lab. Due to the inherent difficulty in writing complicated algorithms in LabVIEW, and the availability of a comprehensive C++ class-based library that implements a variety of genetic search types and routines, C++ was picked as the language of choice. QuantumBlackbox was subsequently extended to make it a fully functional data collection and visualization platform; it communicates directly with the LabVIEW suite of software. Finally, a small C++ module was developed to allow direct communication between a set of Mathematica routines and LabVIEW.

A few details related to the design of this software appear in Appendix C. The remainder of this chapter is devoted to the implementation of the genetic search.

6.2 Genetic Search

In quantum control, one often doesn’t know the optimal field for directing the system to a particular target state. There exist a variety of approaches to discovering such an optimal field. In certain cases, the theoretical model of the system is well known and can be used directly to predict the optical field needed to move the system into a target state. In other cases, the theoretical model may be complex, such that predicting the optimal control field is a difficult problem. A mathematical tool known as quantum optimal
control theory has been successfully applied to a variety of such systems where the model is known but complex. Finally, in still other cases it may be that a theoretical model is poorly known or unknown, in which case one can simply employ a method which will learn the correct control field via an iterative process (see the early work by Rabitz [49]). Test fields are applied to the system, and some feedback signal allows the search algorithm being used to converge on the best control [50].

Such a search algorithm may search any parameterization of the optimal control field. For example, the search algorithm may directly search the individual pixels of the SLM. Or perhaps it is known that an optimal field for controlling the system can be obtained using a sinusoidal phase mask, but the exact frequency, amplitude and phase of the sinusoid function are unknown. One could use the search algorithm to search numeric parameters $\alpha$, $\tau$, $\delta$ and so forth, in the parameterization

$$\phi(\omega) = \alpha \sin \left( \delta + \tau (\omega - \omega_0) + \tau^2 (\omega - \omega_0)^2 + \cdots \right),$$

(6.1)

where $\phi$ is the phase and $\omega$ is frequency, afterwards attempt to decipher the meaning of the discovered optimal field [51]. In all cases, the user of the search algorithm provides a function mapping the parameterization chosen to the pixels on the SLM mask. I call this the \textit{mapping function}. The chosen algorithm operates on the returned feedback signal, called the \textit{objective score}, attempting to minimize or maximize it. The details of \textit{how} a new parameters are picked each time an objective score is returned is left up to the chosen algorithm.

One type of search algorithm is the genetic search. Genetic algorithms [52] maintain a \textit{population} of individuals; each individual has its own numeric values for the set of parameters that a particular search scheme uses. The set of parameter values that each individual has make up its \textit{genome}. At the start, initial (often random) values for the parameters are chosen. Then, \textit{each generation} involves these steps:

1. The mapping function provided by the user is called for each member of the population. The mapping function is provided a copy of the \textit{genome} for that individual, from which it calculates the objective score.

2. The objective scores are used to assign a fitness score to each member of the population.

3. One or more of the best individuals may (if desired) propagate into the next generation unchanged, called \textit{elitism}. 

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4. The genetic algorithm then uses genetic operations to generate individuals with new genomes for the next generation. The most common genetic operations used here include crossover between two genomes, and point mutation. There is also a possibility for complete replacement of certain individuals (e.g. created with random valued parameters).

Objective scores are a key to these genetic operations. For example, a particular type of selection strategy chooses some of the best individuals in a population for crossover. The rationale behind this is that a “re-combination” of these good candidates is likely to produce a better one.

The user allows the algorithm to repeat for many generations—ideally until the desired or maximal objective score has been reached, time constraints make further searching impractical, or the individuals in the population have become identical.

One item above may be clarified. A scaling scheme [52] is the method by which fitness scores are calculated from objective scores. These scale an objective score in such a way that an individual in the population with a genome that is very unique receives a higher fitness score than an individual with a comparable objective score but a less-unique genome. It is actually the fitness score that is used in selection, not the raw objective score.

There are several considerations to be made in using a genetic algorithm, which will govern how good a solution it finds:

1. The chosen parameterization. A good parameterization can substantially decrease the number of generations (and the population size) needed for the genetic algorithm to find a good solution.

2. The frequency of the crossover and mutation, and values optimal for a particular problem, will influence how fast the genetic search finds a solution. The percentage of the population replaced, and the presence of elitism, will also contribute to the rapidity with which a good solution is discovered. Good values may be discovered by trial and error, as well as intuition based on past experience with similar types of searches.

3. The noise in the system. In an experiment, the objective score is prone to experimental errors in measurement, as well as drift over time and fluctuations. A certain level of noise may necessitate refinements in the parameterization used; for example, a parameterization that
searches SLM pixels directly might need to group pixels together so that changes are measurable and appear above the noise level. Real enhancements must be accompanied by a change in the objective score that is above the level of fluctuations in the score due to the noise, in a given generation.

One of the strengths of genetic search is that it is well suited to searching a space (an $N$-dimensional hyperspace, defined by the number of parameters present) with local extrema without getting stuck at them. It combines stochastic elements with the concept of fitness, meaning that it can look for fit individuals anywhere on the search landscape, without bias. On the other hand, random sampling anywhere on the landscape may not be the best strategy in a system with a very simple, regular landscape. Another strength of the genetic search is that it routinely compares substantially different individuals, whose objective scores vary by amounts well above the noise level. Thus, it still makes progress in the search, whereas a hill-climbing method might get lost quickly in noise when comparing closely spaced points in a landscape.
Part III

The Experiments
Chapter 7

Quantum Coherent Control of a Two-Photon Transition in Rubidium

7.1 Introduction

One way to determine how to coherently control a quantum system is to start with a system that is simple, for which the theory is known. Rubidium is such a system; it is an atomic system, and electromagnetic field excitations (from a laser pulse) can be treated with time-dependent perturbation theory.

Here the transitions of interest will be two-photon transitions, as they are within the bandwidth of the laser. These transitions, as they appear in rubidium, along with a version of time-dependent perturbation theory well suited to frequency domain pulse shaping, are presented in Section 7.2. After the two-photon excitation, the rubidium atom will relax back to its ground state via several channels, including a visible fluorescence. This will be discussed in Section 7.3.

Pulse shaping of a broadband pulse is used to substantially enhance or diminish the two photon absorption in rubidium; this is measured via the fluorescence intensity subsequently observed. The experimental setup used to do this is given in Section 7.4. Numerical and experimental measurements will be compared side-by-side in Section 7.5. Finally, Sections 7.5.4 and 7.5.5 go beyond the regime of “simple” control for a known system, focusing less on known theory and more on adaptive search methods, namely genetic search (Section 6.2) and single-pixel scan methods.

7.2 Two-Photon Transitions in Rubidium

The neutral rubidium atom, studied here, is known in spectroscopic tables [53] as Rb I, and has electronic configuration 1s\(^2\)2s\(^2\)2p\(^6\)3s\(^2\)3p\(^6\)3d\(^{10}\)4s\(^2\)4p\(^6\)5s_{1/2}. It is the electronic excitations of this single valence electron which will be
7.2. Two-Photon Transitions in Rubidium

studied in this work.

A Grotrian diagram for the rubidium atom is shown in Fig. 7.1. It gives the excitation and decay pathways present in rubidium when illuminated with a laser beam with the spectrum shown in Fig. 3.2; the numerical computations done for rubidium also use this experimentally measured spectral intensity, with an assumed flat phase. It happens that all of the rubidium spectral lines within (or near) the bandwidth of this incident optical field appear in this Grotrian diagram.

The second-order transition amplitude (given by perturbation theory) of a two-photon transition from initial state $|i\rangle$ to final state $|n\rangle$ is given by

\[ c_{n}^{(2)} \sim \sum_{m} \mu_{nm}\mu_{mi} \left( \mathcal{P} \int_{-\infty}^{\infty} d\omega \frac{E(\omega_{ni} - \omega)E(\omega)}{\omega_{mi} - \omega} - i\pi E(\omega_{ni} - \omega_{mi})E(\omega_{mi}) \right). \]

(7.1)

This expression is rigorously derived in Appendix B; it uses the electric dipole Hamiltonian, derived in Appendix A. It is equivalent to the perhaps more familiar time-domain formulation. The allowed transitions are governed by the standard electric dipole selection rules; transitions forbidden by these rules will have $c_{n}^{(2)} = 0$. The states $|m\rangle$ are resonant intermediate states, meaning that the frequencies of the two photons involved in the transition are close to the frequencies of the individual transitions $|i\rangle \rightarrow |m\rangle$ and $|m\rangle \rightarrow |f\rangle$. The summation over $m$ arises when more than one intermediate state appears between $|i\rangle$ and $|f\rangle$, such that there is a multiple paths from $|i\rangle$ to $|f\rangle$. The electric dipole matrix elements $\mu_{ij}$ are given in Table 7.1.

The two terms in the brackets give the part of the amplitude associated with the spectral distribution of the electric field, $E(\omega)$. (The spectral shaper does not shape in polarization, and so the scalar field $E$ may be used, rather than $\vec{E}$. ) The first term contains the operator $\mathcal{P}$, which denotes the principal value of the integral. It causes the point where $\omega = \omega_{mi}$ to be excluded, and so this first term gives the off-resonance contribution of $E(\omega)$ to the transition. The second term gives the contribution of the field components exactly resonant with the transition, meaning that both photons involved in the transition match the frequencies of the individual transitions. This splitting, as given, into off-resonant and resonant terms removes a divergence which would otherwise appear in this expression for $c_{n}^{(2)}$.

There are three possible two-photon transitions from the rubidium ground state, shown in red in Fig. 7.1. One of these two-photon pathways connects $|5s_{1/2}\rangle$ and $|5d_{5/2}\rangle$, and thus leads to an amplitude $c_{d_{5/2}}^{(2)}$ in the excited state
7.2. Two-Photon Transitions in Rubidium

Figure 7.1: A Grotrian diagram for rubidium, showing the most relevant transitions for this experiment. All wavelength-labelled upward transitions are either within or reasonably close to the spectral bandwidth of the laser. The highlighted transition pathways are the dominant pathways; other pathways may be minor contributors. The dotted lines represent spontaneous transitions. The downward pointing blue arrows give the ultimate fate of the majority of the energy pumped into the two-photon transition: two closely spaced fluorescence lines. The side channel decay paths leading from the 6p to the 6s and 4d states are drawn as single arrows to simplify the diagram. The energy levels are not drawn exactly to scale. Data taken from various sources [53–57].
7.2. Two-Photon Transitions in Rubidium

<table>
<thead>
<tr>
<th>Transition, $i \rightarrow j$</th>
<th>Dipole Element, $\mu_{ij}$ (a.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Excitation Pathways</strong></td>
<td></td>
</tr>
<tr>
<td>$5s_{1/2} \rightarrow 5p_{1/2}$</td>
<td>4.221</td>
</tr>
<tr>
<td>$5s_{1/2} \rightarrow 5p_{3/2}$</td>
<td>5.956</td>
</tr>
<tr>
<td>$5p_{1/2} \rightarrow 5d_{3/2}$</td>
<td>1.616</td>
</tr>
<tr>
<td>$5p_{3/2} \rightarrow 5d_{3/2}$</td>
<td>0.787</td>
</tr>
<tr>
<td>$5p_{3/2} \rightarrow 5d_{5/2}$</td>
<td>2.334</td>
</tr>
<tr>
<td><strong>Relaxation Pathways</strong></td>
<td></td>
</tr>
<tr>
<td>$5d_{3/2} \rightarrow 6p_{1/2}$</td>
<td>18.106</td>
</tr>
<tr>
<td>$5d_{3/2} \rightarrow 6p_{3/2}$</td>
<td>8.160</td>
</tr>
<tr>
<td>$5d_{5/2} \rightarrow 6p_{3/2}$</td>
<td>24.491</td>
</tr>
<tr>
<td>$6p_{1/2} \rightarrow 6s_{1/2}$</td>
<td>9.684</td>
</tr>
<tr>
<td>$6p_{3/2} \rightarrow 6s_{1/2}$</td>
<td>13.592</td>
</tr>
<tr>
<td>$6p_{1/2} \rightarrow 4d_{3/2}$</td>
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</tr>
<tr>
<td>$6p_{3/2} \rightarrow 4d_{3/2}$</td>
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</tr>
<tr>
<td>$6p_{3/2} \rightarrow 4d_{5/2}$</td>
<td>6.184</td>
</tr>
<tr>
<td>$6p_{1/2} \rightarrow 5s_{1/2}$</td>
<td>0.333</td>
</tr>
<tr>
<td>$6p_{3/2} \rightarrow 5s_{1/2}$</td>
<td>0.541</td>
</tr>
</tbody>
</table>

Table 7.1: Select transition dipole matrix elements relevant for the calculation of 420 nm fluorescence in rubidium after broadband 2-photon excitation with a pulse centered near 800 nm. These values come from published tables, computed using a relativistic all-order method [58]. A dipole element of 1 a.u. (atomic units) is equivalent to $8.478 \times 10^{-30}$ C m in SI units.

given by Eq. (7.1) with no summation (a single term in the sum). The other two two-photon pathways connect $|5s_{1/2}\rangle$ and $|5d_{3/2}\rangle$, via two different resonant intermediate states, as shown; the transition amplitude $c^{(2)}_{d_{3/2}}$ is given by Eq. (7.1) with the summation being over two pathways.

The final probability of transition to a level is given by the squared modulus of the transition amplitude,

$$ P_{d_{3/2}} = \left| c^{(2)}_{d_{3/2}} \right|^2 \quad \text{and} \quad P_{d_{5/2}} = \left| c^{(2)}_{d_{5/2}} \right|^2. \quad (7.2) $$

No single-photon transitions (or contributions from higher-order perturbation theory) are necessary.
7.3 Modeling the Fluorescence Intensity

To understand how the transition probabilities of Eq. 7.2 lead to observable fluorescence signals, one considers all the possible decay channels (via spontaneous emission). These levels $|5d_{5/2}\rangle$ and $|5d_{3/2}\rangle$ decay to $|6p_{3/2}\rangle$ and $|6p_{1/2}\rangle$ as allowed by dipole selection rules, shown in Fig. 7.1. The population in these $6p$ levels then decays via three possible routes. Decay to the $|6s_{1/2}\rangle$ level is possible, as is decay to the 4d levels. It is assumed that any $6p$ population decaying to either of these side channels is lost and does not contribute directly to the observable fluorescence (as this side channel-related fluorescence light is very far from 420 nm). The possibility that these side channels feed the 5p intermediate levels, thus facilitating subsequent single-photon transitions back to 5d is a higher-order process that has not been considered. The third possible decay channel from 6p leads the atom back to the ground state and is the channel of most interest here, as it produces a pair of fluorescence lines near 420 nm.

The fact that the fluorescence signal arises from such a network of decay channels means that there is no \textit{a priori} guarantee that population appearing in a certain ratio $P_{d_{5/2}}$ to $P_{d_{3/2}}$ after two-photon excitation will appear in that same ratio in the observable fluorescence level $M$, calculated as

$$M = w_{d_{3/2}}P_{d_{3/2}} + w_{d_{5/2}}P_{d_{5/2}}.$$  \hspace{1cm} (7.3)

As a fictitious example, consider the two 5d levels equally populated after a pulse. It is conceivable that although the two levels are initially equally populated at 50% each, 25% of one level and 15% of the other actually reach the observable fluorescence. This would be because one of the 6p levels decayed more rapidly to the 6s or 4d side channel levels than the other. The weights $w_i$ must be calculated.

In order to determine these weights, one first writes down the coupled
7.3. Modeling the Fluorescence Intensity

system of rate equations describing this system. This system is

\[
\dot{N}_{5d_{3/2}} = -A_{5d_{3/2} \rightarrow 6p_{3/2}} N_{5d_{3/2}} \\
\dot{N}_{5d_{1/2}} = -A_{5d_{1/2} \rightarrow 6p_{3/2}} N_{5d_{1/2}} - A_{5d_{1/2} \rightarrow 6p_{1/2}} N_{5d_{1/2}} \\
\dot{N}_{6p_{3/2}} = A_{5d_{3/2} \rightarrow 6p_{3/2}} N_{5d_{3/2}} + A_{5d_{1/2} \rightarrow 6p_{3/2}} N_{5d_{1/2}} - A_{6p_{3/2} \rightarrow 6s_{1/2}} N_{6p_{3/2}} \\
&\quad - A_{6p_{3/2} \rightarrow 4d_{5/2}} N_{6p_{3/2}} - A_{6p_{3/2} \rightarrow 4d_{3/2}} N_{6p_{3/2}} - A_{6p_{3/2} \rightarrow 5s_{1/2}} N_{6p_{3/2}} \\
\dot{N}_{6p_{1/2}} = A_{5d_{3/2} \rightarrow 6p_{1/2}} N_{5d_{3/2}} - A_{6p_{1/2} \rightarrow 6s_{1/2}} N_{6p_{1/2}} - A_{6p_{1/2} \rightarrow 4d_{3/2}} N_{6p_{1/2}} \\
&\quad - A_{6p_{1/2} \rightarrow 5s_{1/2}} N_{6p_{1/2}} \\
\dot{N}_{5s_{1/2}} = A_{6p_{3/2} \rightarrow 5s_{1/2}} N_{6p_{3/2}} + A_{6p_{1/2} \rightarrow 5s_{1/2}} N_{6p_{1/2}}. 
\]

(7.4a) (7.4b) (7.4c) (7.4d) (7.4e)

\( N \) gives the population of a level, and \( \dot{N} \) gives the time derivative or time rate of change of a level. The Einstein coefficients \( A_{ij} \) give the rate of spontaneous transitions between levels \( |i\rangle \) and \( |j\rangle \), calculated according to

\[
A_{ij} = \frac{2\omega_{ij}^3}{3\epsilon_0 \hbar c^3} |\mu_{ij}|^2, 
\]

(7.5)

with the dipole matrix elements given in Table 7.1. The frequencies \( \omega_{ij} \) correspond to the frequency of each atomic transition from state \( |i\rangle \) to \( |j\rangle \).

The system is then solved numerically; a solution obtained using initial values \( P_{5d_{3/2}} = P_{5d_{1/2}} = 1 \) is given in Fig. 7.2. Only a fraction of excited initial population makes it back to the ground state through these 420 nm fluorescence channels. The approximate lifetimes observed in this calculation are consistent with known values [58].

If one then sets \( P_{5d_{3/2}} = 1 \) and \( P_{5d_{1/2}} = 0 \), one obtains \( w_{d_{3/2}} = P_{5s_{1/2}} = 0.1995 \). If one instead sets \( 5d_{3/2} = 0 \) and \( 5d_{1/2} = 1 \), \( w_{d_{5/2}} = P_{5s_{1/2}} = 0.2359 \). This then answers the question posed above as to whether or not the two 5d levels’ populations are equally visible in the final observable fluorescence—the initial populations of 5d appear in the observed fluorescence in roughly the same ratio. The same ratio is observed for any value \( C \) comparing \( 5d_{3/2} = C \) and \( 5d_{5/2} = 0 \) with \( 5d_{3/2} = 0 \) and \( 5d_{5/2} = C \).

The above considerations motivate calculating the fluorescence intensity \( M \) as simply

\[
M \sim P_{d_{3/2}} + P_{d_{5/2}},
\]

(7.6)
as \( w_{d_{3/2}} \approx w_{d_{5/2}} \). (In Fig. 7.4 one sees that both fluorescence lines are transmitted equally, and thus no additional weighting to account for filter transmission is needed.) The values \( P \) are given by Eq. 7.2
7.4 Experimental Apparatus

There are two naturally abundant isotopes of rubidium \( ^{60,61}\)Rb and \( ^{85,87}\)Rb (72.17\% and 27.83\% naturally abundant, respectively). Both isotopes respond identically to the type of excitations used in this work. Both isotopes have a melting point of 39.31 °C. The solid (at room temperature) rubidium metal is inside a sealed glass cell, having flat windows at both sides. The cell is held in a metal block, equipped with a heating element and thermocouples. An external temperature controller connected to a thermocouple mounted in the block switches the heating element on and off, maintaining the cell at 200 °C. The rubidium gas in the cell interacts with the incoming laser beam. This apparatus is shown schematically in Fig. 7.3.

Fluorescence is an omni-directional incoherent field associated with spontaneous emission. The interaction with the incoming laser field results in a pair of fluorescence lines near 420 nm. In addition to fluorescence, superfluorescence can also be observed. This is a directional, coherent field, and a type of stimulated emission: the superfluorescent beam comes out of the cell collinear with the laser source. To avoid collecting superfluorescence light \([57]\) the fluorescence signal is observed via a hole cut on the side of the metal block, perpendicular to the laser beam.

An interference filter with a transmission profile shown in Fig. 7.4 helps
7.4. Experimental Apparatus

Figure 7.3: The experimental apparatus used to study the quantum coherent control of two photon absorption in rubidium. The shaped pulse is focused into a rubidium vapor cell. The two photon absorption subsequently decays via several routes, including a visible fluorescence at 420 nm. This signal is imaged onto a photomultiplier tube (PMT), after being filtered to contain only the light near 420 nm with an interference filter (IF). This interference filter blocks background light. Unused input beam mixed with superfluorescence light exits the tube collinear with the input, and is blocked.

Figure 7.4: The transmission curve for the Thorlabs FB420-10 bandpass filter [62], using data provided by Thorlabs. The filter has a transmission window centered at $420 \pm 2$ nm with a FWHM of $10 \pm 2$ nm.
to ensure that only the desired fluorescence lines are observed. This filtered light is imaged onto a photomultiplier tube (PMT). The signal from the PMT is in turn fed into a Boxcar Integrator from Standford Research Systems. This instrument allows a successive averaging of a gated temporal region of the PMT signal; the PMT response of 30 to 300 laser pulses is averaged. This averaged signal is collected via a serial interface, and transmitted to QuantumBlackbox via a LabVIEW component. Although the boiling point of rubidium is 688 °C, at 200 °C there exists enough rubidium vapor (the solid phase vapor pressure [61] is 67.1 mTorr) in the cell such that the fluorescence signal collected here is readily observed, and comes in well above the noise level.

7.5 Coherent Control of Two-Photon Absorption in Rubidium

The following subsections focus on understanding and demonstrating the coherent control of two-photon absorption in rubidium. The fluorescence intensity predicted by Eq. 7.6 can be enhanced because a transform limited (flat phase) Gaussian input pulse is not optimal for two-photon excitation [55]. In particular, the non-resonant contribution from a particular two-photon pathway can be enhanced [55], the relative phase between the resonant and non-resonant component in a pathway can be tuned [63], and finally, the relative phase between different interfering pathways can be tuned when a sufficiently broadband laser pulse is used.

These three avenues to coherent control in rubidium are illustrated in the next three subsections. To simplify this discussion, in addition to the definition of $c_n^{(2)}$ given by Eq. 7.1 let

$$c_{n,PP}^{(2)} \equiv \mu_{nm} \mu_{mi} P \int_{-\infty}^{\infty} d\omega \frac{E(\omega_{ni} - \omega)E(\omega)}{\omega_{mi} - \omega}$$

(7.7)

and let

$$c_{n,res}^{(2)} \equiv \mu_{nm} \mu_{mi} i\pi E(\omega_{ni} - \omega_{mi})E(\omega_{mi}).$$

(7.8)

7.5.1 Nonresonant Contributions

$c_{n,PP}^{(2)}$ gives the contribution of the non-resonant part of the field to the two-photon transition, i.e. when $\omega \neq \omega_{mi}$. The structure of the denominator in this principal value integrand, $(\omega_{mi} - \omega)^{-1}$, is visualized in Fig. 7.5(a).

Two important facts follow from the structure of this denominator. First,
7.5. Coherent Control of Two-Photon Absorption in Rubidium

Figure 7.5: A visual comparison of the behavior of the integrand in Eq. 7.1 describing the off-resonant part of the two-photon transition in rubidium according to second-order time dependent perturbation theory. Note that as the spectral functions $E(\omega)$ are added, they have the effect of modulating the integrand such that the frequencies close to the resonance become relatively more important. Here, the 780 nm resonance is used for $\omega_{m1}$.

the structure of the field $E(\omega)$ around a resonance is weighted very strongly in its importance to the transition amplitude, relative to far off-resonant components. Second, the denominator introduces a sign change in the function around the intermediate resonance. Fig. 7.5 shows how the different components of the integrand come together to realize this behavior.

As it stands, this roughly anti-symmetric structure of the integrand leads to a low contribution from the integral to the total two-photon transition amplitude. One may enhance this off-resonant contribution to the transition in two ways.

Phase Shaping

Phase shaping can be used to impose a negative sign in an appropriate region of the integrand \[55\]. The integrand shown in Fig. 7.5(c) then becomes entirely positive (nearly symmetric, rather than antisymmetric); the presence of the broadband field is now a substantial benefit to the transition.

The resonances near the two pathways at 775.8 nm and 780 nm and 775.9 nm and 780 nm are closely spaced (approximately 4 nm apart). A convenient way to maximize the integrand in Fig. 7.5(c) is to place a $\pi/2$
7.5. **Coherent Control of Two-Photon Absorption in Rubidium**

![Graph showing π/2 window phase scans. A 4 nm wide π/2 phase window, shown in the upper plot, is scanned across the spectrum (the spectral region of the scans is shown on the x-axis of the lower plots). Several π/2 windows centered at different positions are shown on the upper plot; shades of gray suggest the motion of the phase window during the scan. The bottom plots show the ratio of enhancement vs. the center wavelength of the π/2 window. This ratio, shown in the plots, is the ratio of the fluorescence intensity calculated with a flat phase pulse to that calculated with a phase window applied. Theoretically, the maximal enhancement appears when this window is exactly centered between the two resonances, as expected. Experimentally, uncompensated phase distortions in the input pulse and interference with the pathway at 795 nm mean a departure from the ideal result (see Section 7.5.5). In both cases, however, a 5 fold enhancement in two-photon absorption is measured.](image-url)
window (which is approximately 4 nm wide) between the resonances near 776 and 780 nm (see Fig. 7.6). Then, the factors $E(\omega_{ni} - \omega)$ and $E(\omega)$ both pick up an extra $\pi/2$ phase in the 776 nm to 780 nm region; as a result, the integrand in this region is multiplied by -1, and the whole integrand becomes positive. Using the full bandwidth of the input pulse, scanning this $\pi/2$ window through this region yields the enhancement ratios shown in Fig. 7.6. The numerical result computed here matches closely with a published result [55]; the optimal enhancement is less than the value of 7 predicted in that work due to interference with the transition pathway appearing at 795 nm, due to the present use of a broader bandwidth pulse. By removing the spectral components near the 795 resonance, the numerical model used here reproduces this 7 fold enhancement.

The off-resonant contribution associated with the two-photon pathway involving resonances at 762 and 795 nm may be enhanced with a $\pi$ window applied to, e.g. the right side of, the resonance at 795 nm. This works well here because the two resonances are now far apart. The numerically computed result of scanning this window appears in Fig. 7.8, along with the SLM mask used.

**Intensity Shaping**

A second method for enhancing the off-resonant part of the two-photon transition is to simply remove the portion of the spectral content (see the SLM mask in Fig. 7.7) which picks up a negative sign relative to the other side of the resonance. In other words, enhancement is obtained by removing one half of the anti-symmetric shape shown in Fig. 7.5. The maximum enhancement available is less than that possible using phase shaping, but is still substantial. The scans shown in Fig. 7.7 demonstrate this enhancement by using a intensity window centered between the two resonant wavelengths of the transition. The maximum enhancement ratio obtained occurs when this window exactly removes the spectral components in the input which cause the integrand in $c_{np}^{(2)}$ to pick up a negative sign across the position of the intermediate resonance.

One sees that the spectral components near 762 nm and 795 nm have also been removed from the broadband pulse in this intensity scan, as they are set to zero transmission using the SLM. This ensures no interference with the other pathway at 795 nm. Instead of removing spectral components around the pathways at 776 and 780 nm, one may alternatively scan this intensity edge near 761 and 795 nm, as shown in Fig. 7.8. One sees that all three resonant pathways in rubidium behave similarly, and may be coherently
7.5. Coherent Control of Two-Photon Absorption in Rubidium

Figure 7.7: Intensity edge scans. The SLM mask uses shades of blue to show how a transmission window, centered between the two resonant wavelengths, is varied in width during the course of the scan. Two-photon absorption is maximized when all cancellation in the integrand of $c_n^{(2)}$ is eliminated. It is seen that two-photon absorption can be enhanced to just over twice the level observed when transform-limited pulses are used.

controlled in the same manner.

7.5.2 Nonresonant and Resonant Term Interference

The resonant contribution to the two-photon transition amplitude is given by $c_n^{(2)}$. This equation predicts that these resonant field components are in quadrature to those in the non-resonant contribution given by $c_n^{(2)}_{\text{PP}}$—they include this additional phase shift via the factor of $i$. The existence of this resonant term means that another possible avenue for enhancement of the two-photon transition amplitude is to manipulate the phases of resonant spectral components such that they become in-phase with the off-resonant terms.
7.5. Coherent Control of Two-Photon Absorption in Rubidium

Figure 7.8: $\pi$ and intensity edge scans at 795 nm. The top two plots show the SLM masks used for the corresponding computed scans in the bottom plots. Shades of blue and gray are again used to convey changes to the masks during the course of each scan. Compare these results of applying $\pi$ shaping and intensity edges to the pathway involving the 762 and 795 nm resonances to the scans involving the pathways at 776 and 780 nm. Both measurements have been made after removing the spectral components around 776 and 780 nm, so those pathways do not contribute to the measured transition amplitude.
7.5. Coherent Control of Two-Photon Absorption in Rubidium

One complication to this approach is that SLM pixel elements are relatively wide and so any attempt at controlling the residue term phase will also invariably change the off-resonant field contributions. These off-resonant components nearest the residue are strongly weighted (see again Fig. 7.5 and the caption), so that attempting to change a pixel at the spot of the resonance produces substantial modulation. There also exists a chance that the resonant spectral component simply overlaps with the small gaps between SLM pixels.

7.5.3 Two-Pathway Interference

One of the two-photon pathways associated with the resonances near 776 and 780 nm is summed with the pathway associated with resonances at 762 and 795 nm, according to the prescription for \( c_{5d_{3/2}}^{(2)} \) given by Eq. 7.1. By careful manipulation, one thus seeks to not only maximize the magnitude of the two pathways leading to \( |5d_{3/2}\rangle \), but also the relative phase.

7.5.4 Putting it All Together—Theory

It is possible to construct genetic search parameterization (Section 6.2) of an SLM mask which combines all three of the above methods of achieving coherent control in rubidium. The mask contains six distinct features. Three of these are pixel windows of variable widths (not all varying in the same ranges) which search near the location of the resonances, each contributing to a residue term \( c_{n,\text{res}}^{(2)} \). The widths of these windows (in pixels) are searched by the genetic scan. One of the two resonances near 776 nm is included, and both of the resonances at 762 and 795 nm are included. This inclusion of both resonances at 762 and 795 nm is redundant as far as the contribution to the residue term for the pathway is concerned, but it was found to be useful, providing an extra degree of freedom—the resonant windows shape a small portion of the off resonant components, too. The other three pixel windows are designed to provide enhancement for the non-resonant contribution to the three pathways, again having variable widths which are genetic-searched. All six windows shape in phase; the height of each phase window is searched by the genetic scan. All search parameters are restricted to take values only within a certain set of specified ranges; for the phases, this is simply between zero and \( 2\pi \). For the widths of the windows, reasonable limits are chosen such that the windows cannot overlap or come to close to neighboring transition pathways. Thus resulting SLM mask will look similar to that in Fig. 7.9(b).
7.5. Coherent Control of Two-Photon Absorption in Rubidium

(a) The variation in the population slowly disappears as the search runs. The phase mask which produces the maximum two-photon absorption in rubidium is found at the 738\textsuperscript{th} measurement and gives an enhancement ratio of 9.86.

Figure 7.9: Evolution of the genetic search, seeking to maximize broadband excitation of a two-photon transition in rubidium. Each measurement made by the search algorithm is plotted, in order. Also shown is the maximally enhancing SLM mask obtained by genetic search. The transition probability ratio shown here is computed as it was before; compare both the enhancement ratios and the optimal SLM mask obtained here with those shown in Figs. 7.6, 7.7 and 7.8.

The evolution of the genetic search, applied to the numerical model, is depicted in Fig. 7.9(a). The initial population is quite diverse; this can be seen in, e.g., the first 100 iterations. Although the individual generations are not marked, one sees a steady trend of increasing maximum ratio and diminishing diversity as individuals are tested, genetic operations are performed and generations pass. At the end, the search has essentially converged, having found an SLM mask giving an almost ten times enhancement of the two-photon transition probability calculated for rubidium. This optimal SLM phase mask is shown in Fig. 7.9(b). This solution is not unique, but it is typical. Other executions of this genetic search obtain masks that provide comparable enhancements. The features observed in this mask are typical, as a center window at a height near $\pi/2$ is present, and two side windows are present whose heights sum to $\pi$ (approximately).

A more detailed understanding of the enhancement process can be gained
7.5. Coherent Control of Two-Photon Absorption in Rubidium

Figure 7.10: The evolution of the components of the transition amplitude throughout the genetic search appearing in Fig. 7.9(a). The three lines shown in each plot represent the contribution of a particular pathway to the quantity shown below each figure. The blue line shows the contribution from the $m \rightarrow n = 5p_{3/2} \rightarrow 5d_{3/2}$ pathway, purple gives $5p_{1/2} \rightarrow 5d_{3/2}$, and olive gives $5p_{3/2} \rightarrow 5d_{5/2}$. 

(a) $|c_{m\rightarrow n,pp}^{(2)}|$. 
(b) $\text{Arg } c_{m\rightarrow n,pp}^{(2)}$. 
(c) $|c_{m\rightarrow n,\text{res}}^{(2)}|$. 
(d) $\text{Arg } c_{m\rightarrow n,\text{res}}^{(2)}$. 
(e) $|c_{m\rightarrow n}^{(2)}|$. 
(f) $\text{Arg } c_{m\rightarrow n}^{(2)}$. 

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7.5. Coherent Control of Two-Photon Absorption in Rubidium

by inspection of Fig. [7.10]. These six plots show how the three pathways which contribute to the total fluorescence intensity $M$ (Eq. 7.6) evolve as the genetic search runs. Extending the notation $c_m^{(2)}$ to $c_{m-n}^{(2)}$, where $m$ indicates the intermediate state in the two-photon transition, $M$ is now written as

$$M \sim \left| c_{5p_{3/2} \rightarrow 5d_{3/2}}^{(2)} + c_{5p_{1/2} \rightarrow 5d_{3/2}}^{(2)} \right|^2 + \left| c_{5p_{3/2} \rightarrow 5d_{5/2}}^{(2)} \right|^2. \quad (7.9)$$

(The sum over $m$ originally present in the expression for $c_n^{(2)}$ has been written out explicitly here; this is what gives rise to the summation of the two terms $c_{5p_{3/2} \rightarrow 5d_{3/2}}^{(2)}$ and $c_{5p_{1/2} \rightarrow 5d_{3/2}}^{(2)}$. This expression is still equivalent to the original expression for $c_n^{(2)}$, being only a change of notation.)

Fig. [7.10(a)] shows that the contribution to $|c_{m-5d_{3/2},PP}|$ from the two interfering pathways leading to $|5d_{3/2}\rangle$ is nearly equal. Thus, they can interfere strongly in this model. The other pathway to $|5d_{5/2}\rangle$ is somewhat higher in magnitude, due to the particular electric dipole transition matrix elements involved. Fig. [7.10(c)] shows, as it must, that the magnitude of each residue term is constant. The importance of interference to Brumer-Shapiro (Section 1.3.2) quantum control in rubidium is emphasized in Figs. [7.10(b)] and [7.10(d)]. As the search evolves, the phase difference between interfering terms is minimized. One sees in the plot that Arg $c_{5p_{3/2} \rightarrow 5d_{5/2},res}^{(2)}$ does not vary, as the frequencies associated with this term have not been included in any window appearing on the SLM mask. Figs. [7.10(e)] and [7.10(f)] summarize the total contribution coming from each pathway; they are then inserted into Eq. [7.9] and used to calculate Fig. [7.9(a)].

This same genetic search parameterization can also be used to minimize the two-photon transition probability ratio. The genetic search is instructed to minimize the objective score; doing this, one obtains a two-photon transition probability ratio of about 0.1. Plots similar to those given in Fig. [7.10] reveal that this minimization is achieved by minimizing the values $|c_{m-n,PP}^{(2)}|$ and by finding phases that lead to destructive interference.

7.5.5 Putting it All Together—Experiment

The agreement shown in Figs. [7.6] and [7.7] of theory with experiment is not great, even though the enhancement, looking at it only as a numerical value, is good.

The broadband laser pulse used for rubidium excitation is compensated using MIIPS and has also been checked using both FROG and temporal
7.5. Coherent Control of Two-Photon Absorption in Rubidium

autocorrelation. These measures all suggest that the pulse is close to being transform-limited. Despite this, Figs. 7.6 and 7.7 suggest that transform-limited pulse is not really transform limited. It may be that long-term drift of the laser phase (e.g. on the order of an hour) prevents proper pulse compensation or introduces distortions after-the-fact. The spectral intensity near the resonance at 762 nm is relatively weak, and this causes difficulty for phase compensation methods such as MIIPs. The shaper is temperature sensitive, and pulse chirp has been observed to drift appreciably during the course of operation.

Short-time pulse phase instability is also a known problem not completely solved during the time allotted for this thesis work. Particularly, it can be observed that the second-harmonic intensity spectrum, which is phase sensitive, is unstable on a very short timescale (seconds). Air currents are appreciable in the lab room where these experiments are done; these created variations in the optical path length, critical in the region of the shaper where spectral components are dispersed in space. Attempts made to enclose both the entire optical table, as well as additionally enclose the shaper and SLM, improved stability, but did not provide a total remedy to the problem.

Despite this, the presence of uncompensated phase features (those not varying on a short timescale on the order of seconds) provides a unique opportunity. This is the ability to explore a method for obtaining quantum coherent control in an atomic system where unknown phase irregularities are present (see also Chapter 8). This method makes use of the fact that spectral components close to rubidium resonances are strongly weighted in their importance to the two-photon transition probability relative to other spectral components. Implemented using the JavaScript functionality in QuantumBlackbox, this method involves the following steps, depicted in Fig. 7.11:

1. A resonance to scan around is selected, e.g. that at 775.94 nm. A starting pixel group size \( n_g \) is selected, typically \( n_g = 2 \).

2. \( n_g \) SLM pixels at the position of the resonance are scanned in phase on the interval \([0, 2\pi]\), measuring the ratio every \( \pi/4 \) radians and thus producing a set of points like those in Fig. 7.12(c). The phase yielding the highest ratio is selected and two points \( \pi/8 \) to the left and right of the maximum are also measured in just in case the additional phase granularity contributes a significantly higher enhancement. (Numerical tests using a transform-limited pulse suggest that \( \pi/4 \) phase
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Figure 7.11: Illustration of a method by which narrow groups of pixels are alternately scanned to the left and right of the rubidium resonance of interest, in order to find substantial enhancements to the transition amplitude without the complexity or slow convergence observed in a genetic search. The light green vertical lines mark single pixels on the SLM. Starting at the top, a group of pixels to the left of the resonance at 776 nm are scanned in phase, as shown by the shades of gray (not all values scanned are shown). The optimal value (e.g. producing the highest measured value of fluorescence intensity) is kept and applied to the mask. In the middle plot, a group of pixels to the right of the resonance is again scanned and the optimal phase value recorded. In the bottom plot, the alternating pattern continues, but one sees that the number of pixels involved in the scanned group has increased. As this method relies on the heavy weighting of \( E(\omega) \) close to each resonance, shown in Fig. 7.5, pixel group sizes must be increased as the distance from a resonance increases in order to produce the same measurable response in fluorescence.
granularity is a sufficient basis for excellent two-photon enhancement in rubidium.)

3. The above scans are repeated, alternating scanning $n_g$ pixels to the left or to the right of pixels already scanned. If the enhancement gained is small, $n_g$ is increased. In this way, one spends the most time scanning near the resonances, where the weight of the contribution to the two-photon enhancement pathway is the strongest.

4. These ratios are plotted to produce a plot like that of Fig. 7.12(a). One sees that as the distance from the center position (of the resonance) is increased, the increase in ratio generally plateaus. Drifts up and down in the ratio are typically due to noise—transient drifts on timescales on the order of seconds arise from the pulse instability previously discussed.

5. The maximally enhancing SLM mask is thus obtained.

This method, like the genetic scan of the numerical model performed in Section 7.5.4, also effectively combines the three methods (the first three subsections of this section) by which coherent control of two-photon absorption in rubidium may be achieved.

Fig. 7.12 shows the result of applying this method to the two transition pathways involving resonances at 776 and 780 nm. Spectral components further than about 8 nm away from these two resonances are removed, so that this scan focuses on only these pathways. Likewise, Fig. 7.13 scans the transition pathway involving the 762 and 795 nm resonances, removing the intensity around 776 and 780 nm. These scans are, in principle, capable of enhancing both the nonresonant contribution to the transition probability (Section 7.5.1) and adjusting the relative phase between resonant and nonresonant field components (Section 7.5.2). Due to phase instability across the pulse, scans employing the full spectral content (Section 7.5.3) were not attempted here.

These scans have been kept simple. One may expect that many more pixels, more allowed phase values and averaging may enhance this outcome. This scan method may find utility when attempting to do a quantum coherent control through temporally scattering media.

7.5.6 Other Experimental Investigations

Several other experiments involving two-photon absorption in rubidium were performed. Each experiment used the genetic search functionality in Quan-
7.5. **Coherent Control of Two-Photon Absorption in Rubidium**

(a) The ratio obtained as the correct phase values are picked on the SLM mask, starting at the center and alternating left and right. This process has been illustrated in Fig. 7.11.

(b) The optimal SLM mask obtained at the end of the scan.

(c) A sample of the transition probability ratio vs. phase data measured as a single pixel group is scanned.

Figure 7.12: The resonance-centered pixel-scan method described in the text, applied to the two transition pathways involving resonances at 776 and 780 nm. Spectral components further than 8 nm from these two resonances are removed (the pathway involving resonances at 762 and 795 nm is removed).
7.5. **Coherent Control of Two-Photon Absorption in Rubidium**

Figure 7.13: The resonance-centered pixel-scan method described in the text, applied to the two transition pathways involving resonances at 762 and 795 nm. Spectral components around the resonances at 776 and 780 nm are removed. The scans in c. and d. build upon those in a. and b., and thus the final phase mask includes both regions. The ratios scanned in c. do not plateau in the typical manner, but no additional enhancement is found (in this case) by going beyond the wavelengths shown.
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tumBlackbox with a particular SLM mask parameterization. One such experiment parameterized the SLM mask by dividing it up into small groups of pixels, each of which could take the value 0 or $\pi/2$ (motivated by the success of the $\pi/2$ in Fig. 7.6). This yielded a several-fold enhancement in the transition probability ratio; the optimal SLM mask found by this search features $\pi/2$ windows between 776 and 780 nm, as well as around the resonances involved in the two-photon pathway beginning at 795 nm. Using a simple sinusoidal mask (see the discussion of Eq. 6.1 in Section 6.2), other experiments were done where both enhancements and reductions in the rubidium transition probability ratio were obtained.

The observed reductions in transition probability were confirmed using the numerical model. For example, a genetic search like that presented in Section 7.5.4 was used to minimize two-photon absorption (fluorescence intensity). In so doing, the numerical search finds ways to maximize the amount of destructive interference between the non-resonant and resonant terms contributing to each pathway, and also creates destructive interference between pathways. These reductions in two-photon absorption could be made very strong, such that almost no fluorescence was observed in either the numerical or experimental results; no shaping in intensity was required.

“Traditional” electromagnetically induced transparency involves destructive interference between coupled levels [64], and can be realized using monochromatic laser light. The reduction in absorption observed here is also a type of “induced transparency,” but the lack of absorption has instead been realized by shaping the phase relationship between spectral components in a broadband laser source. The reduction of two-photon absorption observed here happens in a system where the two-photon absorption occurs with the help of resonant intermediate states; the use of phase shaping to reduce photon two-photon absorption in a non-resonant two-photon absorption (in a system where a two-photon transition occurs via only virtual intermediate states) has been studied elsewhere and requires slightly different theory [65].

Along with quantitative measurements of the fluorescence intensity $M$, direct observation of the visible superfluorescence light at 420 nm coming from the cell (as shown in Fig. 7.3) provided a simple qualitative affirmation of the two-photon transition enhancement and reduction effects reported here.
7.6 Conclusion

In this chapter, particularly in Section 7.5, it is seen that the amplitude of two-photon absorption in rubidium, in the presence of a broadband field, is due to interplay between three distinct mechanisms. Two contributions to this amplitude are off-resonant part of the field and the contribution of the exactly on-resonant parts; interference between multiple two-photon absorptions leading to the same final level was also seen to contribute.

It is then found that quantum coherent control of these two-photon absorption pathways in rubidium could be understood in terms of these three contributions to the two-photon absorption amplitude. The interplay between these contributions has been used to both substantially enhance or substantially diminish the total two-photon transition probability, when compared excitation via a transform-limited pulse (Gaussian envelope with flat phase). Further, this enhancement or suppression is achieved via only phase or only amplitude shaping, although phase-only shaping yields higher enhancements. A key result appearing in this chapter (Fig. 7.6) is the comparison of a particular phase only shaping scheme (the \( \pi/2 \) window) with previously published results [55].

The numerical and experimental tools used for this investigation also provided an opportunity to explore genetic search (adaptive learning) methods of finding optimal coherent fields (Section 7.5.4). Additionally, the presence of unsolved phase instabilities in the excitation field provided motivation for exploring a scan-based method that attempts to find optimal control fields by scanning groups of pixels in the immediate vicinity of each resonance (Section 7.5.5). This method finds good solutions in less time than the genetic search (faster convergence). In both cases (the genetic search and the pixel scans) it is seen that the availability of even limited information about the structure of the system can provide constraints on the types of searches that should be used, and thus substantially decreases the required search space.

The coherent control of two-photon transitions in simple atomic systems like rubidium provides an example of the power and utility of quantum coherent control, and its general applicability. Indeed, rubidium is not an elaborate biological system specifically designed for optical functional control (see Section 1.1). Rather, control has been facilitated by the availability of a simple and accurate model of the system, combined with the technique of pulse shaping.
Chapter 8

Regaining Coherent Control after Random 1D Glass Stacks

8.1 Introduction

The method of Vellekoop and Mosk allows for compensation of spatial scattering (Section 2.2). In the weak scattering regime, this spatial compensation also corrects for temporal scattering, when a broadband laser source is used (Section 2.2.1). The theory and experiment appearing in this chapter shows how compensation may be achieved for a strongly scattering one-dimensional system. This scattering system, realized here as a stack of glass microscope slides, is described in Section 8.2.1. Partial reflections of the incident laser pulse at each air-glass or glass-air interface give rise to non-uniformly spaced trains of pulses.

Section 8.2.2 then presents a numerical model of these glass stacks. The numerical models provide a convenient way to explore scattering compensation (Section 8.3), aiding in the development of a scan-based compensation algorithm. This compensation algorithm is presented in Section 8.3.6.

The remaining sections in this chapter report (Section 8.4) and discuss (Section 8.5) numerical and experimental compensations achieved for glass stacks that have between one and ten layers.

8.2 Glass Stacks

8.2.1 Experimentally Constructed Glass Stacks and Sample Realizations

As shown in Fig. 8.1 a stack of glass microscope cover slides is a simple way to implement a one dimensional scattering system. Each glass layer in the stack is characterized by an average thickness (100 µm), but the exact
8.2. Glass Stacks

Figure 8.1: A stack of non-uniform glass slides, also called a random stack or glass stack. The reflection of the input pulse off the glass stack is captured, using the beamsplitter technique shown. The light scattered from the stack consists of a train of pulses; pulses arise from reflections at each air-glass or glass-air interface. The reflected train is focused onto a second harmonic generation (SHG) crystal, and the spectrum acquired. Fig. 8.8 explains the purpose of the SHG crystal and spectrometer.

thickness varies randomly. The exact thickness of the air gap which appears between the sandwiched glass layers also varies. For this reason, the stack is called “random.”

The random stack is a scattering material due to the presence of air-to-glass and glass-to-air interfaces in the material. For the glass used here, the index of refraction is approximately $n_{\text{glass}} = 1.52$, and the index of air is taken to be $n_{\text{air}} = 1.0$. The percent reflection from such an index mismatch is given by

$$R = \left(\frac{n_{\text{glass}} - n_{\text{air}}}{n_{\text{glass}} + n_{\text{air}}}\right)^2 = 4.3\%.$$  \hspace{1cm} (8.1)

A reflected pulse arises at each air-glass interface. The spacing between any two reflected pulses is given by

$$\Delta t = \frac{2d \times 10^3}{c} n_{\text{glass or air}},$$  \hspace{1cm} (8.2)

where $\Delta t$ is the temporal spacing in femtoseconds, $d$ is the layer (glass or air) thickness in $\mu$m, $n$ the refractive index, and $c = 299.792458$ nm $\text{fs}^{-1}$.
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Figure 8.2: Experimentally measured fundamental spectra (left) and XFROG traces (right) of the reflected pulse train arising from one, two and three layer glass stacks. The power spectrum becomes more complex as the number of layers increases. The XFROG traces show that an approximately 1 picosecond delay, due to the approximately 100 µm thick glass layers, is observed. One also observes pairs of closely spaced pulses; these pulse pairs exist because of small air gaps between the layers of glass; these air gaps appear even when the stack is pressed tightly in a mount.
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is the speed of light in vacuum. Multiple reflections can occur inside the stack, meaning that a pulse can be reflected more than once before it exits the stack. A random pulse train exits both sides of the stack.

On the right side of the stack shown in Fig. 8.1, the pulse train that exits the stack also contains the ballistic pulse—the part of the input pulse that was never scattered. This is labeled as the transmitted beam. The other pulses in this transmitted train have been reflected at least twice, and thus are much less intense (by a factor of $R^2 \approx 0.1\%$) than the ballistic pulse. This huge contrast between the ballistic pulse and other pulses in the train is problematic for the detection scheme that will be used (Section 8.3.4).

To avoid this problem, the reflected beam which comes from the left side of the stack shown in Fig. 8.1 is used. This train of pulses contains nearly equal-amplitude pulses (i.e. of the same order of magnitude). The reflected beam is captured by inserting a beamsplitter in the input beam, as shown in the figure.

In Fig. 8.2, measured spectra and XFROG traces of reflected pulse trains arising from one, two and three layer stacks are presented. The power spectra shown in the figure demonstrate fringing. This fringing arises because two or more pulses are present in the reflected train, which interfere on the detector (on the CCD inside the spectrometer). The XFROG traces also verify that the expected approximately one picosecond spacing between reflected pulses (Eq. 8.2 using $d = 100 \mu m$) is observed.

The actual mount used for these experimental realizations of glass stacks consists of two metal plates. A large hole is drilled in the center of each plate. The glass stack is held tightly between the two plates, with the laser beam passing through the center holes and also the stack. Thickness or other surface irregularities in the glass slides can cause the glass thickness seen by different spatial components of the propagating beam to vary. Likewise, uneven pressure on different parts of the stack in the mount can cause the air gap seen by different spatial components of the beam to vary. To combat these problems, the beam is brought to a focus inside the stack using two spherical mirrors so that the beam diameter inside the stack is small. Efforts are also made to keep the pressure applied on the stack by the mount as even as possible, to eliminate the appearance of spatial speckle. In some cases, a rubber O-ring is also inserted in the mount to aid in applying sufficient regular pressure to stacks with more layers (e.g. stacks with 4 or 5 layers).
Figure 8.3: Numerically computed reflection spectra; these simulate the scenario where a Gaussian beam (shown in Fig. 5.7) illuminates one to six layer thick stacks of glass slides (see also Fig. 8.4). They are calculated by the method described in Section 8.2.2. Other realizations of glass stacks having from one to ten layers are not shown here, but their compensation, along with the compensation of the stacks presented here, appears in Section 8.4.1.
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8.2.2 Numerically Modeled Glass Stacks and Sample Realizations

A method by which glass stacks are numerically modeled is now introduced. One establishes a list of desired glass layer and air gap thicknesses, along with their respective indices of refraction. From these, the coefficients of reflection and refraction are computed. At each frequency, a transfer matrix is computed \[67\]. The transfer matrices at all frequencies are then used to compute reflection amplitudes, which are then multiplied by the input spectrum. (One can also calculate transmission spectra.) This scheme has been implemented in a set of MATLAB scripts \[68\]. One advantage of this method is that it accounts for all orders of reflections inside the stack.

A sample of the computed reflection spectra obtained using this method are shown in Fig. 8.3 and 8.4. These realizations of the numerical model have between one and six layers of glass. Other realizations of these and higher (seven to ten) layer stacks are prepared in the same way. The list of glass thicknesses is generated around the value 100.0 ± 7.5 μm. The average air gap thickness is 140 ± 35 nm. In both cases, the random variations are drawn from a uniform or flat distribution around the mean (e.g. not from a Gaussian distribution). Each glass layer and air gap is chosen independently.
8.3 Four Possible Phase Pre-Compensation Methods

(i.e. each layer in the stack is different).

8.3 Four Possible Phase Pre-Compensation Methods

From a frequency-domain pulse shaping perspective (see Section 5.4), compensation of scattering from glass stacks is simply a matter of finding out whether it is possible to construct a mask $H(\omega)$, that undoes the effect (e.g. $H^{-1}(\omega)$) of the stack.

Intensity shaping cannot be used in the compensation: from a practical perspective, the goal is to bring all of the scattered pulses back together without power loss. Phase based shaping achieves this. Thus, an efficient phase mask $\phi(\omega)$, that undoes the phase (e.g. $\phi^{-1}(\omega)$) applied by the stack, is sought, and no intensity shaping is done. The compensating phase will be applied by the pulse shaper before the light reaches the stack. This makes it a pre-compensation: a pulse train prepared by the shaper before it is directed onto the stack, and it comes out of the stack as a single pulse.

8.3.1 Phase Compensation via XFROG

XFROG, in principle, should be able to recover the unknown electric field amplitude and phase, of the pulse train coming out from the stack. Then, the phase conjugate could be applied directly.

However, this method has several disadvantages. First, the position of each pulse in the XFROG trace (Section 5.5.3) can only be known as accurately as the step size of the trace, or in other words, by the amount that the delay $\tau$ between cross-correlated pulses is stepped each time a spectrum is acquired. Second, XFROG is ill-suited to retrieving the phase of complex fields, and these random pulse trains are considered complex for XFROG. This method also requires additional optics, typically taking additional time set up each time a different random stack must be tested. Lastly, even if one measures this conjugate phase, that does not solve the problem of how to apply it. For complex compensating phases, important information can be lost in the process of interpolation, e.g., when converting the compensating phase from the high resolution of a spectrometer to a lower resolution SLM. It will be seen later (see Fig. 8.16) that even if the field phase could be retrieved exactly, other compensation methods outperform it, due to this interpolation loss.
8.3. Four Possible Phase Pre-Compensation Methods

8.3.2 A Prerequisite: Creating Irregularly Spaced Pulse Trains with a Pulse Shaper

Other methods by which the unknown compensating phase may be discovered will be introduced later—all will require the material introduced in this section. These methods do not directly retrieve the unknown spectral phase function \( \phi(\omega) \). Rather, they focus on ways of retrieving parameters characterizing a pulse train, produced by a stack. These parameters are the relative amplitude \( a_i \) and position \( t_i \) of each pulse \( i \) in the train.

Once known, these parameters are used to create a phase function \( \phi(\omega) \) that (ideally) represents the inverse of the phase of the pulse train coming out from the stack. If the \( a_i \) and \( t_i \) are discovered correctly, this computed phase should approximately cancel the spectral phase originally introduced by the stack.

This phase-only random pulse train is constructed in the following way. The time domain electric field of a random pulse train can be written as

\[
E(t) = a_0 g(t) e^{-i\omega t} + a_1 g(t - t_1) e^{-i\omega(t - t_1)} + \cdots. \tag{8.3}
\]

The temporal envelope of the input pulse is \( g(t) \); for a transform-limited Gaussian input pulse,

\[
g(t) = e^{-t^2/2b^2}. \tag{8.4}
\]

The temporal full width at half maximum \( \tau_{\text{FWHM}} \) is given by \( \tau_{\text{FWHM}} = 2\sqrt{2\ln 2} b \), and \( \omega_0 \) gives the carrier frequency. \( a_0 \) is always taken equal to one from now on; the other pulse strengths are made relative to \( a_0 \). The Fourier transform of \( \text{8.3} \) is

\[
E(\omega) = G(\omega) \left( a_0 + a_1 e^{i\omega t_1} + \cdots \right) \tag{8.5}
\]

with

\[
G(\omega) = be^{-\frac{b^2(\omega - \omega_0)^2}{2}}. \tag{8.6}
\]

\( G(\omega) \) is provided by the input pulse; factoring this out, the part of interest \( \text{8.7} \) is

\[
H(\omega) \equiv 1 + a_1 e^{i\omega t_1} + \cdots. \tag{8.7}
\]

The compensating phase function \( \phi(\omega) \) is then

\[
\phi(\omega) = \text{Arg} \, H(\omega). \tag{8.8}
\]

It is this compensating phase which is applied to the spectral shaper to pre-compensate glass stack scattering.
Example: Creating a Two-Pulse Train with a Spectral Shaper

Consider a two pulse train with \( a_0 = a_1 = 1 \) and \( t_1 = 300 \) fs:

\[
H(\omega) = 1 + e^{i\omega \cdot 300 \text{ fs}}.
\]  
(8.9)

Applying amplitude and phase together, one obtains the desired pulse train, shown in Fig. 8.5. If only the complex magnitude of this function, \(|H(\omega)|\), is used, the pulse train shown in Fig. 8.6 results. Essentially this is a perfectly phase compensated two-pulse train. For comparison, the result of applying the phase \( \text{Arg} \ H(\omega) \) only is plotted in Fig. 8.7. A comparison of Fig. 8.6 and Fig. 8.7 shows that it is the phase and not the intensity modulation done by the stack that gives the stronger contribution to the production of the correct number of pulses with the correct amplitudes. Likewise, it is seen that the depth of the fringes seen in the second-harmonic spectrum due to shaping only in intensity is less than those appearing when shaping in phase. This suggests that the phase-only compensation may work quite well; this is verified in later sections.

The phase-only two pulse train shown here is equivalent to the fixed periodic unit pulse train introduced previously (Section 5.4.2). Transforming \( H(\omega) \to H(\omega)e^{-i\omega \tau/2} \equiv \tilde{H}(\omega) \) yields

\[
\tilde{H}(\omega) = 2 \cos \left( \frac{\omega \tau}{2} \right).
\]  
(8.10)

The transformed phase, \( \text{Arg} \ \tilde{H}(\omega) \), is exactly the repeating unit phase mask shown in Fig. 5.10. The phase-only train has two equal strength pulses that appear in the center; it is the addition of the amplitude shaping that removes the outlier pulses.

8.3.3 Phase Compensation using Spectral Information

The reflected intensity spectrum, coming from a glass stack (or equivalently from its second harmonic) can be Fourier transformed to the time domain to give estimates of the pulse times appearing in the random train. A flat temporal phase across each individual reflected pulse is assumed. The timing information is good, but only within the limit of spectrometer resolution. The resolution of the spectrometer used here limits this timing accuracy to about \( \pm 50 \) fs; this comparable to the temporal pulse width and is far below the level required for good compensation.
8.3. Four Possible Phase Pre-Compensation Methods

Figure 8.5: The two-pulse train is created by shaping in intensity and phase, using $H(\omega) = 1 + e^{i\omega \cdot 300 \text{ fs}}$ (Eq. 8.9). See also Figs. 8.6 and 8.7.
8.3. Four Possible Phase Pre-Compensation Methods

Figure 8.6: A pulse train is created by shaping in intensity only, using $|H(\omega)|$. $H(\omega)$ is given by Eq. [8.9] This is essentially a phase compensated two pulse train.
8.3. Four Possible Phase Pre-Compensation Methods

Figure 8.7: A pulse train is created using phase-only shaping, using \( \phi(\omega) = \text{Arg } H(\omega) \). \( H(\omega) \) is given by Eq. [8.9].
8.3. Four Possible Phase Pre-Compensation Methods

(a) An unscattered input pulse generates a smooth second harmonic spectrum. The integral of the measured spectrum, \( \int d\omega E(\omega) \), is maximum.

(b) A pulse train, e.g. coming from a random stack, generates a second harmonic spectrum with heavy fringing. The fringing in the spectrum leads to a low value of the integral.

(c) A phase compensated pulse train has most of its energy back in a single pulse, with relatively small outlier pulses. The fringing in the spectrum has been lifted to some degree, and the numeric integral takes on an intermediate value. An adaptive search algorithm can refocus a pulse train by looking for ways to maximize this integral.

Figure 8.8: An illustration of how second harmonic generation (SHG) is used to provide a measure of how well a random pulse train, as generated by a glass stack, has been compensated.

8.3.4 A Prerequisite: A Feedback Signal that Detects when Scattering has been Compensated

Both compensating phase discovery methods previously introduced (XFROG and the use of spectral information) attempt to reconstruct the unknown compensating phase in a single shot. Two additional methods will be introduced that instead employ adaptive search algorithms. These methods iteratively test and refine a collection of pulse train parameters in order to discover the best compensation.

Before these methods are introduced, this section introduces the feedback signal used to guide these adaptive search algorithms. The feedback required by a typical adaptive search algorithm is a single numerical value which gives a measure of how good one solution is compared to another.

This feedback is implemented as shown in Fig. 8.8. The concept behind
this feedback is as follows: the presence of a multiple-pulse train leads to lower second harmonic intensity, and also leads to fringing (due to multi-pulse interference) in the second harmonic spectrum. When a set of $a_i$ and $t_i$ are discovered that bring at least one pair of pulses in the train back together (after the pre-compensated pulse train is scattered from the stack), the peak intensity available for the second harmonic generation goes up and fringing in the spectrum is lessened. The area under the curve $E(\omega)$ increases. This area of integration is used as the value of the feedback signal. The region of integration is limited to the region of the spectrum in which the spectral components appear above the noise level.

Another way in which this feedback signal could be implemented would be to use a slow photodiode, which integrates the total intensity of the second-harmonic train.

### 8.3.5 Phase Compensation using a Genetic Search

In order to use the genetic search (Section 6.2) to find the compensating field phase, a suitable parameterization of the phase must be determined. Among the possibilities for parameterization of the phase mask is the use of a discrete pixel search, where each SLM pixel (or small groups of pixels) is allowed to take values from 0 to $2\pi$.

Another possibility is to use of the pulse-train parameterization introduced above, allowing the genetic search to determine the optimal values of $a_i$ and $t_i$. The mapping function (see Section C.1.6) converts these to the appropriate phase mask, via Eq. 8.8.

This method works for simple stacks. However, at higher numbers of layers the convergence of the search is poor: the genetic search fails to find good compensating values of $a_i$ and $t_i$ in a reasonable time.

### 8.3.6 Phase Compensation using an Interferometric Autocorrelation-Like Method

Original attempts at compensating few-layer stacks in the lab [69] involved iteratively “guessing” at and searching by hand the correct pulse parameters that would optimize the compensation. In an attempt to automate this, the following method is developed. It will be subsequently referred to as the “methodic” scan, as the scans are made methodically (genetic searches on the other hand are stochastic in nature).

Each of the pulse parameters can be scanned independently, refocusing one pulse in the train at a time. This method of compensation works well
and it bypasses convergence issues encountered with the genetic search for higher-layer systems. This methodic scan performs the following steps:

1. **Uncompensated stack reference.** A flat phase mask and unit (100% transmission) intensity mask are applied to the SLM. The value of the second harmonic feedback signal (Section 8.3.4) is stored as $A_{\text{ref}}$. It is the reference value for the uncompensated stack.

2. **Scan range selection.** A list of $N$ allowed scan ranges $t_{i,\text{start}}$ to $t_{i,\text{end}}$ and $a_{i,\text{start}}$ to $a_{i,\text{end}}$, $i$ an integer from 1 to $N$, is prepared. Good compensation can typically be obtained by including one range for each first order reflection (those shown in Fig. 8.1); $N$ is thus typically equal twice the number of glass layers in the stack. It is hoped that each chosen range includes the actual values of $t_i$ and $a_i$, matching the stack. The chosen ranges may be based on estimates of pulse positions obtained using the second harmonic spectrum (see Section 8.3.3), as well as upon experience with past scans.

3. **Pulse time (position) scan.** A fixed value of $a_i$, typically near 0.8, is chosen. Then $t_i$ is scanned between $t_{i,\text{start}}$ to $t_{i,\text{end}}$. At each value of $t_i$, a phase mask—computed using Eq. 8.8—is applied to the SLM. This computed mask $H(\omega)$ always includes terms for any parameters $a_j, t_j$ previously determined for pulses $j = 1, 2, ..., i-1$. At each point, the value of the second harmonic feedback signal $A$ is measured. The second harmonic compensation ratio, defined as $A/A_{\text{ref}}$, is recorded; see Fig. 8.9(a). After the interval has been scanned, the selected value of $t_i$ is the one corresponding to the maximum second harmonic compensation ratio.

4. **Pulse amplitude scan.** Using the selected value of $t_i$, the value of $a_i$ is scanned from $a_{i,\text{start}}$ to $a_{i,\text{end}}$. The value of the second harmonic compensation ratio is recorded at each point; see Fig. 8.9(b). This amounts to determining the relative peak intensity of the pulse in the train. The selected value of $a_i$ is the one that maximizes the second harmonic compensation ratio.

5. **Repeat.** Steps 3 and 4 are repeated for each pulse $i$ from 1 to $N$. The final second harmonic compensation ratio, after all pulse terms are in place, is measured.

Each reflection at an glass-air interface, moving from glass to air, is associated with an additional $\pi$ phase shift. However, attempting to explicitly
Figure 8.9: A sample of the time and pulse amplitude scans used in the interferometric autocorrelation-like or “methodic” scan method. The stored values of $t_1$ and $a_1$ are the ones that correspond to the maximum second harmonic compensation ratio on each curve. These scans correspond to realization 1 of the 1 layer numerically modelled glass stack shown in Fig. 8.3. The final compensation—using the values determined by the scans shown here—appears in Fig. 8.10.
put this phase shift into Eq. 8.3 unnecessarily complicates the algorithm; one would have to carefully track which terms should have the phase shift and which should not, and include these while scanning. It will be seen that excellent compensation can be achieved without this. The presence of the \( \pi \) shift improves the second harmonic ratio found for realization 1 of the numerical 1 layer system by only about 1/10\(^{th}\) of one percent.

In the time domain, what is happening when the \( t_i \) are being scanned is much like interferometric autocorrelation done through a second harmonic crystal (see Section 5.5.2). One pulse is being scanned past another in time, and the collection is done in a collinear geometry. The trace obtained in this step (e.g. Fig. 8.9) thus looks very similar to that obtained by interferometric autocorrelation. It is a bit different because phase-only shaping is used here (see Fig. 8.7).

Refinements to the Methodic Method

The optical period at 800 nm is 2.67 fs; this period is observed in Fig. 8.9(a). The pulse temporal envelope FWHM is much larger than this. Thus the interferometric autocorrelation-like structure contains many peaks and valleys, spaced by 2.67 fs, within the overlap of any two pulses. Following this consideration, and in order to reduce the amount of measurement, the temporal \((t_i)\) and pulse strength \((a_i)\) scans are refined a bit:

1. For the \( t_i \) scan, first scan every e.g. third or fourth optical period. Each scanned period has a minimum and maximum.

2. Of these scanned periods, find the one containing the highest second harmonic ratio. Scan a few periods around the peak value on a coarse grid (e.g. 0.4 fs spacing). This spacing must be small enough that within the optical period, at least one or two points are likely to appear very close to a maximum.

3. Find the \( t_i \) for the maximum second harmonic ratio on this coarse grid. Then scan on a finer resolution (e.g. about 0.2 fs) grid in a reduced window around the maximum, and choose the final \( t_i \) from this.

4. Scan the \( a_i \) first on a coarse grid (spaced by 0.1) and then a fine grid (spaced by 0.03) in a reduced window around a maximum on the coarse grid.

This modification reduces total search time, and is especially important in the experimental scans.
8.3. Four Possible Phase Pre-Compensation Methods

For the numerical model searches, an additional enhancement was made to the methodic method. A couple of routines were added that (one systematically and one stochastically) test small variations of the pulse parameters found in the methodic scan, and look for improvements to the second harmonic compensation ratio. The second harmonic compensation ratio improves, but total computation time increases.

This interferometric autocorrelation-like scan method was first developed using the numerically modeled stacks; this reduced development time, as pulse shaping and second harmonic spectrum computations are much faster than the lab measurements. In the process of numerically compensating successively higher-layer systems, some important details and nuances were uncovered and understood.

Using the numeric model, it was also possible to directly monitor the temporal electric field $E(t)$, or the structure of the pulse train. While initially attempting to compensate the numerical three layer stack realizations, second harmonic compensation ratios significantly lower than expected were obtained. The issue was resolved by understanding that the maximum second harmonic ratio the algorithm was finding was not associated with moving the second pulse in the train onto the first. This was because the third and second pulses in the train, once put together, yielded a higher second harmonic ratio than the second and first pulses would have. Only pulses with $t_i > 0$ were used. By starting the refocusing process away from the origin, there was no way for the algorithm to bring the first pulse in the train in the opposite direction, and refocus it.

The simple solution was to also allow $t_i < 0$. These negative time terms move the pulses in the opposite direction. (The reason why the first pulse is not the most intense will be discussed in Section 8.5.3.)

**SLM-based Interferometry**

Subsequent to the completion of this work, it was found that SLM-based interferometric autocorrelation had been studied before. The Michelson interferometer required for SPIDER (which stands for Spectral Phase Interferometry for Direct Electric-field Reconstruction), e.g., has been implemented using an SLM-generated two pulse train [70]. As both intensity and phase shaping are used (as in Fig. 8.5), this SLM-based SPIDER scan differs from the methodic scan. The final objective of the methodic scan is not field measurement, but to produce a usable (properly compensated) field containing a single pulse.
8.4 Results: The Achieved Compensations

8.4.1 Compensated Numerical Model Realizations

In this section, the numerically modeled glass stacks introduced in Section 8.2.2 are compensated. Four different values are presented for each realization of a glass stack, shown in Table 8.1. These values are the second harmonic compensation ratios (defined in Section 8.3.6), and give a measure of the degree to which a scattered pulse train has been refocused in time.

The meaning of the different columns in the table follows. “Exact compensation” refers to the case where the phase of the stack has been compensated exactly. Typically, the resolution of the computed stack spectrum is approximately 5-7 times higher than the resolution of the SLM (which has 640 pixels). But experimentally, the compensation must be applied by the SLM. The “predicted for SLM” column gives the compensation ratio obtained when the exactly known phase mask from the stack spectrum is crudely downsampled to the resolution of the (numerically simulated) SLM.

The “methodic scan” column gives the result of compensation using the interferometric autocorrelation-like scan method introduced previously. The “genetic search refinement” column, when not empty, shows the result of feeding the pulse parameters $t_i$ returned from the methodic scan into a genetic search, and letting these parameters be refined along with finding $a_i$. At 5 layers, this method of genetic search refinements was also (in addition to a less constrained genetic search that does not have the pulse time estimates fed in) found to be ineffective, and so simple pulse parameter refinement scheme (described in Section 8.3.6) was instead built into the methodic scan.

For one layer, a two pulse train arises (excluding weak higher-order reflections). A genetic scan was performed using the single-pixel parameterization method (Section 8.3.5) with the single layer numerical model stack. The expected sawtooth-like SLM phase (Fig. 8.5 or Fig. 8.10) was indeed retrieved, after many generations. This method requires no prior knowledge of the problem, but features much slower convergence because of the large search space introduced.

For the methodic and genetic compensation methods, it is observed that the second harmonic compensation ratio of the system is relatively insensitive to variations in the single layer thickness—the ratio varies slightly for the different 1 layer stack realizations. The uncompensated and compensated second harmonic spectra and time domain electric fields for the realization 1 of the one layer stack are shown in Fig. 8.10.
### Results: The Achieved Compensations

<table>
<thead>
<tr>
<th>Realization #</th>
<th>Exact Compensation</th>
<th>Predicted for SLM</th>
<th>Methodic Scan</th>
<th>Genetic Search Refinement</th>
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<tr>
<td>1 Layer</td>
<td></td>
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<tr>
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<td>1.356</td>
<td>1.358</td>
<td>1.358</td>
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<tr>
<td>2 Layers</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1(^\d)</td>
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<td>1.415</td>
<td>1.418</td>
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<tr>
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<td></td>
</tr>
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<td>2.754</td>
<td>3.095</td>
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<td></td>
</tr>
<tr>
<td>1(^\d)</td>
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<td>3.070</td>
<td>3.273</td>
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<td></td>
</tr>
<tr>
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<td>3</td>
<td>5.342</td>
<td>3.124</td>
<td>4.418</td>
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</tr>
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</table>

Table 8.1: Second harmonic compensation ratios obtained for the computationally simulated glass stacks, for one to six layer stacks. The four columns to the right of the realization number give the ratio obtained using each of the four compensation techniques indicated. \(^\d\)See Fig. 8.10, 8.11, 8.12, 8.13, 8.14 and 8.15 for samples of systems having each number of layers present here. These results are summarized in Fig. 8.16.
8.4. Results: The Achieved Compensations

![Graph: Compensating Phase](image)

**Numerical 1 Layer Stack**

<table>
<thead>
<tr>
<th>Compensating Phase</th>
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</thead>
<tbody>
<tr>
<td><strong>Wavelength (nm)</strong></td>
</tr>
<tr>
<td>760</td>
</tr>
<tr>
<td>780</td>
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<tr>
<td>800</td>
</tr>
<tr>
<td>820</td>
</tr>
<tr>
<td>840</td>
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</table>

<table>
<thead>
<tr>
<th>Compensated Second Harmonic</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Wavelength (nm)</strong></td>
</tr>
<tr>
<td>390</td>
</tr>
<tr>
<td>395</td>
</tr>
<tr>
<td>400</td>
</tr>
<tr>
<td>405</td>
</tr>
<tr>
<td>410</td>
</tr>
</tbody>
</table>

Figure 8.10: The compensating phase, along with second harmonic spectra and time domain electric fields observed before and after compensation. This is realization 1 of the one layer numerically modeled glass stack; the stack spectrum appears in Fig. 8.3
8.4. Results: The Achieved Compensations

Figure 8.11: The compensating phase, along with second harmonic spectra and time domain electric fields observed before and after compensation. This is realization 1 of the two layer numerically modeled glass stack; the stack spectrum appears in Fig. 8.3
8.4. Results: The Achieved Compensations

**Numerical 3 Layer Stack**

![Compensating Phase](image1)

![Uncompensated Second Harmonic](image2)

![Uncompensated Time Domain](image3)

![Compensated Second Harmonic](image4)

![Compensated Time Domain](image5)

Figure 8.12: The compensating phase, along with second harmonic spectra and time domain electric fields observed before and after compensation. This is realization 1 of the three layer numerically modeled glass stack; the stack spectrum appears in Fig. 8.3.
8.4. Results: The Achieved Compensations

**Numerical 4 Layer Stack**

The compensating phase, along with second harmonic spectra and time domain electric fields observed before and after compensation. This is realization 1 of the four layer numerically modeled glass stack; the stack spectrum appears in Fig. 8.3.
8.4. Results: The Achieved Compensations

Figure 8.14: The compensating phase, along with second harmonic spectra and time domain electric fields observed before and after compensation. This is realization 1 of the five layer numerically modeled glass stack; the stack spectrum appears in Fig. 8.4.
8.4. Results: The Achieved Compensations

Numerical 6 Layer Stack

Figure 8.15: The compensating phase, along with second harmonic spectra and time domain electric fields observed before and after compensation. This is realization 1 of the six layer numerically modeled glass stack; the stack spectrum appears in Fig. 8.4.
8.4. Results: The Achieved Compensations

Figure 8.16: Trends in the second harmonic compensation ratios obtained using the numerically computed stacks, from one to ten layers. Models with genetic search refinements plot the final result of the genetic search, instead of the methodic scan results. See Table 8.1.

The extension to two layers is trivial, provided that one uses enough terms. The genetic search refinement was actually able find a better solution with fewer terms \( t_i \) by varying the exact values of a subset of the \( t_i \) and \( a_i \) found via the methodic scan. See Fig. 8.11.

Three, four, five and six layer compensations are presented in Fig. 8.12, Fig. 8.13, Fig. 8.14 and Fig. 8.15 respectively.

These ratios are also collected for systems having between seven and ten layers. Although the individual realizations are not shown in Table 8.1 above, they are included in Fig. 8.16. This figure compares all of the numerical model compensation ratios.

8.4.2 Compensated Experimental Realizations

Experimentally achieved methodic scan-based compensations will now be given. One successful scan is shown for each glass stack system from one

[103]
8.5 Discussion

<table>
<thead>
<tr>
<th>Number of Layers</th>
<th>Methodic Scan Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.42 ± 0.01</td>
</tr>
<tr>
<td>2</td>
<td>1.99 ± 0.04</td>
</tr>
<tr>
<td>3</td>
<td>2.5 ± 0.2</td>
</tr>
<tr>
<td>4</td>
<td>2.22 ± 0.09</td>
</tr>
<tr>
<td>5</td>
<td>2.2 ± 0.2</td>
</tr>
</tbody>
</table>

Table 8.2: Second harmonic compensation ratios obtained for experimentally realized glass stacks, having from one to five layers. The corresponding compensated and uncompensated second harmonic spectra is shown in Fig. 8.17, and the data in this table is plotted in Fig. 8.18.

to five layers thick. Table 8.2 gives second harmonic compensation ratio (defined in Section 8.3.6) for each stack, and Fig. 8.17 shows the associated uncompensated and compensated second harmonic spectra. The ratio values presented are each from a single compensation, or one execution of the methodic scan. The error bars are estimated using ratio obtained in other successful experimental compensations; unsuccessful compensation runs (e.g. those having significantly lower compensation ratio) have not been included in this estimate. Fig. 8.18 shows the overall trend observed in the ratios.

Experimentally, it typically takes several tries with each different layer-number stack to find the right combination of input parameters to give a successful compensation using this scan method. On some of these unsuccessful attempts, experimental noise also appears to play a role in hindering the discovery of a proper compensation. In multiple scans of the same stack, slightly different pulse time parameters and small changes in \( a_i \) are observed. The four and five layer systems required a slight revision to the mount (added rubber O-ring and a greater applied pressure) in order to hold the higher number of glass slides without slipping.

8.5 Discussion

8.5.1 The Numerical Models

The second harmonic ratio for the exactly compensated stacks, shown in Fig. 8.18, rises with the number of stack layers. This makes sense; as the
8.5. Discussion

Figure 8.17: The experimentally measured second harmonic spectra for the physical glass stacks, having one to three layers. In each figure, the gray curve gives the uncompensated second harmonic spectrum, and the teal curve gives the second harmonic spectrum measured when the SLM applies the compensating phase mask discovered with the methodic scan.
8.5. Discussion

![Graph showing experimental stack compensations](image)

Figure 8.18: The integrated second harmonic compensation ratio, obtained from physical glass stacks compensated using the methodic scan. See also Table 8.2 and Fig. 8.17.

pulse trains are get increasingly complex, there is more and more “room for improvement.” But the phase compensation is made at the resolution of the model, and so the full improvement can always be made.

On the other hand, the “predicted for SLM” result is somewhat surprising. It shows that with a careful choice of the values appearing on each SLM pixel, one can do better than is possible with a naive nearest-neighbor interpolation and an “exactly” known phase. This means that even if a very high fidelity pulse phase characterization technique is employed, it is likely to yield a poorer compensation than those discovered using the SLM.

The methodic and genetic scans are capable of making increasingly good compensations up to 6 layers. After 6 layers, the limited resolution of the spectral shaper prevents additional compensation. The phase features of the stack are too complex to be represented properly (consider the phase shown in the 6 layer numerical stack in Fig. 8.4). This is why the compensation ratio drops: as disruption to the input pulse gets increasingly worse, it is not matched by a corresponding increase in shaper resolution.
To illustrate this point, consider a numerical simulation of a long pulse train, with pulses stretching from \(-7\) ps to 7, spaced by 1 ps and having equal amplitude \(a_i = 1\), as shown in Fig. 8.19. One of the physical 4f pulse shapers used in this work is configured such that each pixel covers 0.139 nm of bandwidth, which by the Fourier transform (Eq. 5.18) corresponds to a temporal length of about 15 ps. Computationally, finite pixel resolution is simulated using a nearest neighbor interpolation scheme, to project the 640 flat pixels of the SLM onto the higher resolution input pulse. Fig. 8.19(b) demonstrates that the quality of the pulse train declines as the temporal limits are approached.

If one computes the same SLM mask and pulse train for a hypothetical SLM having eight times smaller pixel width (and eight times more pixels), the noise present in Fig. 8.19(b) disappears: the pulse train in Fig. 8.19(c) is very regular. Indeed, the 0.0173 nm pixel width of this higher resolution virtual “SLM” mask corresponds to nearly 123 ps in time. Equivalent to looking at the time-domain pulse structure, one observes this limitation simply by looking at the SLM mask in Fig. 8.19(a): the phase mask is almost entirely comprised of sharp pixel-to-pixel jumps. Observe that the use of both positive and negative pulse times \(t_i\) in the train will allow the shaper to create longer pulse trains than otherwise possible with only positive or only negative time parameters, as higher time (larger \(|t_i|\)) terms produce sharper spectral features; a more sophisticated version of the methodic scan might attempt to use this fact to enable the compensation of stacks with higher numbers of layers, by selecting where the refocused \(t = 0\) pulse appears, rather than just allowing it to appear wherever the first time scan picks the highest strength pulse.

The resolution of the spectral shaper thus sets a fundamental limit to the amount of scattering compensation available. More correctly, it is the product of the shaper resolution with the duration of the scattered train that sets this limit. A scattering system with very small layer thicknesses would allow the shaper (used here) to compensate 1D scattering from stacks with many more layers than those studied here. The synthesis of such a system was in fact attempted, using thin layer deposition techniques, but difficulties encountered in the fabrication of these very thin layers favored the use of the glass stack.

### 8.5.2 Comparing Theory and Experiment

The ratios obtained in these experimental models are not as high as those found in computational models with the same number of layers. There are
8.5. Discussion

(a) An SLM mask designed to create an equal amplitude train of pulses from $-7 \text{ ps}$ to $7$, spaced by $1 \text{ ps}$.

(b) The time domain train computed when the above mask is applied on a numerically simulated 640 pixel SLM. The resolution of this simulated shaper matches the shaper used in the lab. The temporal noise continues long past the temporal extents shown and is present at lower amplitude throughout the pulse train. The pulse amplitudes are uneven.

(c) The same time domain pulse train, now created with a numerically simulated shaper having with an eight times higher resolution (an eight times smaller SLM pixel width matched by an eight times increase in the number pixels), shaping the same bandwidth. Note that both pulse trains still have pulses appearing at the same times, but the noise is substantially less and the amplitudes are even.

Figure 8.19: A numerical demonstration of the effect that limited pulse shaper resolution has on the creation of lengthy pulse trains.
Figure 8.20: The second harmonic spectrum associated with an *exact* phase compensation (at the resolution of the numerical model) of the realization 1, four layer numerically simulated glass stack. The uncompensated spectrum and shaper-resolution compensation are found in Fig. 8.13. The important result to be noted here is that a low level of fringing is still present (and in fact the spectrum looks very similar to the case where the numerically simulated shaper is used.)

a couple of important issues to appreciate in making this observation.

First, while the numerical model used strives to be complete, accurately simulating multi-layer reflection and transmission in a stack, it does not attempt to exactly match the experimentally observed air gap thickness, nor does attempt to exactly match the amount of deviation seen in the glass layer or air gap thicknesses. Systematically increasing or decreasing the magnitude of any of these parameters can place the stack in a regime where the second harmonic compensation ratio will be either higher or lower (see Fig. 8.21); this has to do with inter-pulse interference effects (Section 8.5.3).

Second, and following from this first point, it is incorrect to think of a higher second harmonic compensation ratio as indicating a more successful scan when comparing substantially different stacks. The maximally achievable second harmonic ratio increases anytime the input pulse train is more “complex,” meaning that the relative amplitudes of the pulses in the scattered train have increased. Making the pulse train more complex can be achieved by adding layers; this is why the second harmonic ratio grows with increasing numbers of layers.

It can be seen that the fringe depth appearing in the experimentally measured *uncompensated* second harmonic spectra (Fig. 8.17) is typically less than that appearing in the uncompensated computational model spectra.
8.5. Discussion

(Figs. 8.10 through 8.15). This observation suggests that there is simply less compensation available to be done in these experimental systems—the initial integral of the uncompensated spectrum is larger, making the ratio smaller. A possible reason for this lessened fringe depth in experiment is that the stack is not one-dimensional, as the idealized model predicts. Spatial variations across the surface of the stack (Section 8.2.1) are thus encountered by the beam, leading to variation of pulse timings and amplitudes in the reflected trains, and thus a blurring of fringe intensity finally measured by the spectrometer.

When comparing the success of theory and experiment via visual inspection of the measured compensated second harmonic spectra, recall that even a perfectly phase compensated reflected pulse train does not have a “fringe-free” second harmonic spectrum. This was seen earlier in the two-pulse intensity shaping demonstration (Fig. 8.6), and can also be seen in Fig. 8.20. In this latter figure, the second harmonic spectrum for one of the four layer numerical stacks is shown for the case where the phase has been compensated exactly, to the full numerical resolution of the model. It is seen that a small degree of fringing remains.

8.5.3 Pulse Interference

When the thickness of the air gap is less than the temporal pulse width, the resulting pair of pulses will interfere. This is not the only way that pulses from the stack can interfere, however. Recall the creation of two pulses, separated by $\tau = 300$ fs, using spectral shaping (Fig. 8.5). The phase compensated two-pulse train, Fig. 8.6, is still a train of pulses, separated by $\tau$. Some of these lower-intensity pulses that appear in the compensated train can then interfere with seemingly temporally distant pulses coming from other layers of the stack.

The criteria for observing this second type of interference is as follows. Let there be a pulse at 1120 fs. According to the two-pulse example above, low-intensity pulses will appear at multiples of this time, e.g. a low intensity pulse will appear at 2240 fs. Thus pulse can then interfere with a pulse coming from another stack layer, e.g. at pulse at 2250 fs, even though the two pulses originally coming from the stack shared no temporal overlap! This is simulated using a shaper-generated pulse train, as shown in Fig. 8.21(a). A four-pulse train is created using intensity and phase shaping; the SLM mask is again made using Eq. 8.7. This four-pulse train is structured to simulate this two layer stack with air gap. As the thickness of the second layer is varied, the available second harmonic compensation ratio varies rapidly. (The
8.5. Discussion

Figure 8.21: The effect of variations in air gap and layer thickness, simulated computationally using shaper-generated four-pulse trains. Each time point used in constructing these curves effectively represents a different stack.

exact phase compensation of this train is applied at each value of thickness to measure this ratio.) This confirms the predicted interference between the two initially non-overlapping pulses. The second harmonic compensation ratio shown here oscillates at the carrier period (2.67 fs). When the layer thickness increases such that multiples of the compensated pulse at 1120 (or 1130) fs no longer overlap, the oscillations seen in the second harmonic ratio in Fig. 8.21(a) fade.

The interference at the air gap is easier to understand, as the pulses already overlap due to the closeness of two reflecting layers. As done for the layer thickness, the effect of this air gap thickness on the total available second harmonic compensation ratio can be computed. In Fig. 8.21(b), a four-pulse, two-layer stack is simulated using spectral shaping. The air gap thickness is varied by scanning the time $\Delta t_{12}$ offset of one of the pulses at the gap. When the two pulses no longer overlap, the oscillations in second
8.5. Discussion

A numerical simulation for a four layer thick glass stack, prepared using the full numerical model that includes all orders of reflection, and transmission. The reflected second harmonic spectrum is computed at each point as the air gap thickness is varied; the ratio is computed by applying the exact phase conjugate.

Figure 8.22: A numerical simulation for a four layer thick glass stack, prepared using the full numerical model that includes all orders of reflection, and transmission. The reflected second harmonic spectrum is computed at each point as the air gap thickness is varied; the ratio is computed by applying the exact phase conjugate.

The effect of air gap interference in a four-layer numerically modeled glass stack (as opposed to using a simulated pulse train) is shown in Fig. 8.22. A set of random variations in the layers and in the three air gaps are chosen once at the beginning. The actual “base” thickness of each air gap is scanned, added each time to the set of fixed air gap variations chosen before the start of the scan. One sees that the result is consistent with the simpler pulse train model: oscillations and an overall trend towards increased values of the second harmonic ratio are observed as the air gap distance grows relative to the temporal width of the pulse. The trend is that as the air gaps grow, the pulses become better separated, and the maximum achievable second harmonic tends to increase.

It is thus seen that the exact compensation ratio achieved in a particular multi-layer stack can vary widely, and that range of compensation observed when small deviations in layer thickness are present can also vary widely. The relatively low variability appearing in the achievable second harmonic compensation ratio for a given layer system amongst the computational realizations studied previously (Fig. 8.3 and 8.4) may be understood by recalling that the air gaps there chosen range from 0.70 to 1.17 fs, a difference of less than 20% of the optical period. Thus the potentially huge range of modulation predicted here is not seen in these realizations.

The presence of inter-pulse interference explains why values $a_i$ found in
the scans typically take values between 0 and 2: values above 1 occur the compensation encounters a “pulse” is actually the constructive interference between two pulses.

8.5.4 A Brief Comment on the Structure of the Search Landscape

The genetic search method converges quickly, for few-layer systems, when the $a_i$ and $t_i$ parameterization is used. It does not converge within reasonable experimental and computational time limits when higher-layer systems are scanned. In contrast, studies have been done [71, 72] to explain why the results found in genetic search methods applied to quantum systems yield such excellent solutions.

In the case of a quantum control experiment where a genetic search is used to maximize the probability of single initial to final state transition, the search landscape possesses the property that all extrema in the space correspond to optimal controls for the system [71]. In other words, every extrema is a global extrema. This surprising result helps to explain why a genetic search can rapidly discover very high quality controls in such a system, even if the system Hamiltonian is complex. On the other hand, the glass stack-scattered pulse trains have many sub-optimal extrema in the search landscape. For example, the search may discover various ways to “refocus” one or more pulses onto each other in the time domain, finding local extrema corresponding to sub-optimal control, and then get stuck there. The search will thus miss finding the global extremum existing in the search space, here corresponding to the state where all pulses have been recombined into one.

8.6 Conclusion

It has been shown that spectral shaping can be used to successfully compensate temporal scattering, even in the strongly scattering regime, in which the input pulse structure has been substantially changed. A compensation algorithm was developed, based on a phase shaping-only variant of interferometric autocorrelation. This algorithm allowed easily and automatic compensation of both numerically realized and experimentally realized glass stacks. In the numerical compensations the electric field is observed directly, and it is seen that the phase compensations are indeed refocusing the train of pulses, effectively undoing the effect of the stack. In experimental compensations, one observes this compensation indirectly by observing the lifting of fringing from the second harmonic spectra presented.
In the process of achieving compensation, it is also seen that a simple stack of glass microscope slides has provided occasion to study a number of interesting effects. These include a brief study of the limitations that shaper resolution has on the ability to compensate scattering, and the observation that adaptive search schemes may outperform direct phase characterization methods, due to losses incurred when attempting to apply the compensating phase onto the finite resolution pixels of an SLM. Pulse interference effects due to the presence of narrow gaps of air between the glass layers have also been studied; it has been seen these interference effects can also arise between even seemingly distant pulses, as they will be coupled by the train of pulses produced by the compensating phase, and are responsible in part for the range of available compensation in different stack realizations.

In Chapter 1 Brumer-Shapiro coherent control was introduced as a means by which an arbitrary atomic or molecular system could be guided to a particular quantum state. The use of broadband coherent fields, however, gives rise to the problem of optical scattering (introduced in Chapter 2). The carefully chosen phase relationships in the broadband optical field are disrupted in random ways by the scattering materials. The present chapter has thus introduced an important concept for further studies. Even if the (possibly biological) system of interest contains optically scattering material through which the frequencies relevant to the control scheme must first pass, compensation strategies exist by which the correct control fields will be seen by the target system.
Bibliography


Bibliography


[68] The generation of the stack model spectra presented is done using MATLAB code from both Valery Milner and Thomas Drane.

[69] I give credit to Thomas Drane, also in the Milner group, for this method of determining phase masks for irregular pulse trains. He first applied it to this problem, using a manual, interative approach to discover good compensating masks for few-layer glass stacks.


[79] “Qt.” [http://qt-project.org/]. A cross-platform application and UI framework for developers using C++ or QML.


[88] “jQuery UI.” [http://jqueryui.com/]


Appendix A

The Electric Dipole Hamiltonian

In quantum electrodynamics (QED), the full Hamiltonian describing a system of charges and fields in the Coulomb gauge \([73]\) is

\[
H(t) = \sum_{\alpha} \frac{1}{2m_{\alpha}} \left( \vec{p}_{\alpha} - q_{\alpha} \vec{A}(\vec{r}_{\alpha}, t) - q_{\alpha} \vec{A}_e(\vec{r}_{\alpha}, t) \right)^2 \\
+ V_{\text{Coul}} + \sum_{\alpha} q_{\alpha} U_e(\vec{r}_{\alpha}, t) + \sum_i \hbar \omega_i \left( a_i^\dagger a_i + \frac{1}{2} \right) 
\]  
(A.1)

(This Hamiltonian equation comes from Legendre transformation of the QED Coulomb gauge Lagrangian.) \(\vec{p}_{\alpha}\) describes the momentum of particle \(\alpha\), and \(m_{\alpha}\) is the mass. \(q_{\alpha}\) is the charge and \(\vec{A}\) the vector potential of the fields in the system. The terms \(q_{\alpha} \vec{A}\) give the effect of the internal fields on the particles (contributing to their motion); \(q_{\alpha} \vec{A}_e\) gives the effect of an external field on the particles (also contributing to their motion). \(V_{\text{Coul}}\) gives the all Coulomb effects in the system. \(U_e\) is the external scalar potential imposed on the system, with \(q_{\alpha} U_e\) the effect of the scalar potential on particle \(\alpha\). The final term describes the state of the internal field, an ensemble of quantum harmonic oscillators describing field normal modes having frequency \(\omega_i\).

This Hamiltonian can deal with a wide variety of scenarios, including (nonrelativistic) quantum optics experiments where the quantum evolution of the electromagnetic fields is of vital import. Fortunately, substantial simplification is possible here. The evolution of the internal field is not important, and so one can set \(\vec{A} = 0\). \(U_e\) can be ignored, as it describes the effect of external Coulomb field. In this approximation only the “perturbation” of the system by some external transverse field is treated. The Hamiltonian is then

\[
H(t) = \sum_{\alpha} \frac{1}{2m_{\alpha}} \left( \vec{p}_{\alpha} - q_{\alpha} \vec{A}_e(\vec{r}_{\alpha}, t) \right)^2 + V_{\text{Coul}}. 
\]  
(A.2)
Appendix A. The Electric Dipole Hamiltonian

In order to further simplify, one assumes that the charges (for example, those in a rubidium atom) are localized at the origin. If the interacting field wavelength is long compared to the dimensions of the localized quantum system, the Hamiltonian is written instead as

\[ H(t) \approx \sum_{\alpha} \frac{1}{2m_{\alpha}} \left( \vec{p}_\alpha - q_\alpha \vec{A}_e(0, t) \right)^2 + V_{\text{Coul}}. \tag{A.3} \]

The time-dependent unitary transformation of a time-dependent Hamiltonian is given by (see Chapter 4 in [73])

\[ H'(t) = T(t)H(t)T^\dagger(t) + i\hbar \left( \frac{dT(t)}{dt} \right) T^\dagger(t). \tag{A.4} \]

If

\[ T(t) \equiv \exp \left( -\frac{i}{\hbar} \vec{d} \cdot \vec{A}_e(0, t) \right), \text{ and,} \tag{A.5} \]

\[ \vec{\mu} = \sum_{\alpha} q_\alpha \vec{r}_\alpha \tag{A.6} \]

then still in the long wavelength approximation,

\[ H'(t) = \sum_{\alpha} \frac{\vec{p}_\alpha^2}{2m_{\alpha}} + V_{\text{Coul}} - \vec{\mu} \cdot \vec{E}_e(0, t) \tag{A.7} \]

is the known as the electric dipole Hamiltonian.

This simple Hamiltonian can be split into two parts, \( H = H_0 + V \). \( H_0 \) includes the particle velocity and Coulomb terms, but the only important thing here will be to know the energy spectrum of \( H_0 \). \( V \) is the interaction term, given by

\[ V(t) = -\mu E_e(0, t) \equiv -\mu E(t). \tag{A.8} \]

\( \mu \) is the electric dipole moment of the interaction, and \( E \) is the external electric field (e.g. the optical pulse imposed on the atom). This form of the expression applies when the field \( E \) is linearly polarized.
Appendix B

Frequency Domain
Time-Dependent
Perturbation Theory

B.1 Time-Dependent Perturbation Theory

Ideally suited for Hamiltonians of the form $H = H_0 + V$, the interaction picture [73] defines a new a time-dependent quantum state ket in the interaction picture, $|\alpha(t)\rangle_I$. This is related to the ordinary state ket in the Schrödinger picture, $|\alpha(t)\rangle_S$, via

$$|\alpha(t)\rangle_I = e^{iH_0 t/\hbar} |\alpha(t)\rangle_S. \quad (B.1)$$

It follows that an observable $V_I$ in the interaction picture is related to its definition $V$ in the Schrödinger picture by

$$V_I(t) = e^{iH_0 t/\hbar} V(t) e^{-iH_0 t/\hbar} \quad (B.2)$$

The effect of doing this is that the quantum wave equation for time-evolution is now

$$i\hbar \frac{\partial}{\partial t} |\alpha(t)\rangle_I = V_I |\alpha(t)\rangle_I. \quad (B.3)$$

By defining the time-evolution operator

$$|\alpha(t)\rangle_I = U_I(t, t_0) |\alpha(t_0)\rangle, \quad (B.4)$$

with $U_I(t_0, t_0) = 1$, Eq. [B.3] leads to the integral equation

$$U_I(t, t_0) = 1 - \frac{1}{\hbar} \int_{t_0}^{t} dt' V_I(t') U_I(t', t_0). \quad (B.5)$$

($V_I$ is the quantum propagator or kernel of this integral equation.) By
B.1. Time-Dependent Perturbation Theory

iteration, one obtains the Dyson series \[74\],

\[
U_I(t, t_0) = 1 + \cdots + \left(-\frac{i}{\hbar}\right)^n \int_{t_0}^{t} dt' \int_{t_0}^{t'} dt'' \cdots \int_{t_0}^{t^{(n-1)}} dt^{(n)} \times V_I(t') V_I(t'') \cdots V_I(t^{(n)}) + \cdots. \tag{B.6}
\]

The Dyson series can be made more useful for the present application by transforming it into a form that gives the transition amplitude between two energy eigenkets. The state \(|\alpha\rangle\) is related to its stationary state energy eigenkets \(|\alpha\rangle\) by

\[
|\alpha(t)\rangle = \sum_{a} |a\rangle \langle a| \alpha(t_0) \rangle e^{-iE_a(t-t_0)/\hbar}, \tag{B.7}
\]

where \(E_a\) is here the set of energy eigenvalues of \(H_0\). At the initial time, \(|\alpha(t_0)\rangle_I = |a\rangle\) (by appropriate choice of the initial phase). The transition amplitude between initial state \(|i\rangle\), evolved in time from \(t_0\) to \(t\), and final state \(|n\rangle\), is

\[
c_n(t) = \langle n| U_I(t, t_0) |i\rangle. \tag{B.8}
\]

Then, the terms in the Dyson series (Eq. B.6) gives rise to approximations \(c_n^{(j)}\) of order \(j\) for \(c_n(t)\). As \(U_I(t_0, t_0) = 1\),

\[
c_n^{(0)}(t) = \delta_{ni}. \tag{B.9}
\]

The first order coefficient \(c_n^{(1)}\) is identically zero when the laser bandwidth does not include spectral components with sufficient energy for the transition (this can be verified using a procedure analogous to that presented in Section B.2).

The second order coefficient is

\[
c_n^{(2)}(t) = -\frac{1}{\hbar} \sum_{m} \int_{t_0}^{t} dt' \int_{t_0}^{t'} dt'' e^{i\omega_{nm} t'} V_{nm}(t') e^{i\omega_{mi} t''} V_{mi}(t''). \tag{B.10}
\]

In general, \(V_{ij} \equiv \langle i| V | j\rangle\) and \(e^{i\omega_{ij} t} \equiv e^{i(E_i - E_j) t/\hbar}\). The sum over \(m\) is a sum over all energy eigenkets, introduced by the identity

\[
\sum_{m} |m\rangle \langle m| = 1. \tag{B.11}
\]

Expression Eq. B.10 is known as the second-order transition amplitude in time-dependent perturbation theory (TDPT).
The above perturbative expression $c_n^{(2)}$ has a simple interpretation. Roughly, one associates the four “factors” in Eq. B.10 from right to left, with transition amplitude flowing from $i$ to $m$, evolving in time, transition amplitude flowing from $m$ to $n$, and then evolving again. It thus appears as an ideal candidate for understanding two-photon transition. A frequency domain treatment, now presented, complements this time-domain interpretation, allowing the effects of frequency-domain shaping applied via the SLM to be understood directly.

### B.2 TDPT in the Frequency Domain

Starting with Eq. B.10, one uses Eq. A.8 and the Fourier transform of $E(\omega)$ to $E(t)$, given by

$$E(t) = \int_{-\infty}^{\infty} d\omega E(\omega)e^{-i\omega t},$$

(B.12)

to obtain

$$c_n^{(2)}(t) \sim \sum_m \mu_{nm}\mu_{mi} \int_{t_0}^{t} dt' \int_{t_0}^{t'} dt'' \int_{-\infty}^{\infty} d\omega' E(\omega')e^{i(\omega_{nm}-\omega)t'} \times \int_{-\infty}^{\infty} d\omega'' E(\omega'')e^{i(\omega_{mi}-\omega'')t''}.$$  

(B.13)

Reversing the order of the time and frequency integration yields

$$c_n^{(2)}(t) \sim \sum_m \mu_{nm}\mu_{mi} \int_{-\infty}^{\infty} d\omega' \int_{-\infty}^{\infty} d\omega'' E(\omega')E(\omega'') \times \int_{t_0}^{t} dt' e^{i(\omega_{nm}-\omega')t'} \int_{t_0}^{t'} dt'' e^{i(\omega_{mi}-\omega'')t''}. $$

(B.14)

As $E(t)$ is the temporal representation of a single pulse, the interaction can be taken to start at $-\infty$: let $t_0 \to -\infty$. The behavior during the optical pulse (coherent transients) is not of interest here, but rather the amplitude after the process is done. Thus let $t \to \infty$. This of course assumes that assuming no competing processes are transferring amplitude to or away from the involved levels during interaction.

With these limits, performing $t''$ time integral as is would give an indefinite oscillatory term, evaluated at $-\infty$. In order to overcome this, let (for an example, albeit with a different integrand, see [74])

$$\omega'' \to \omega'' + i\epsilon.$$  

(B.15)
B.2. TDPT in the Frequency Domain

Then, after the integration, take $\epsilon \to 0$. Thus,

$$
\int_{-\infty}^{t'} dt'' e^{i(\omega_{mi} - \omega'')t''} = \frac{e^{i(\omega_{mi} - \omega'')t'}}{i(\omega_{mi} - \omega'')}.
$$

(B.16)

The subsequent integration over $t'$ yields a $\delta$ function fixing $w'$:

$$
c_n^{(2)} \sim \sum_m \mu_{nm} \mu_{mi} \int_{-\infty}^{\infty} d\omega' \int_{-\infty}^{\infty} d\omega'' \frac{E(\omega')E(\omega'')}{i(\omega_{mi} - \omega''')} \delta(\omega_{nm} + \omega_{mi} - \omega' - \omega'')
$$

(B.17)

This $i \to n$ transition has $\omega_{ni} \equiv \omega_{nm} + \omega_{mi}$, $\forall$. Thus,

$$
c_n^{(2)} \sim \sum_m \mu_{nm} \mu_{mi} \int_{-\infty}^{\infty} d\omega \frac{E(\omega_{ni} - \omega)E(\omega)}{\omega_{mi} - \omega}.
$$

(B.18)

(Compare with [55].) Physically, this says that a given two-photon transition is comprised of all pairs of frequencies $\omega_{nm}$ and $\omega_{mi}$ in the input pulse that sum to $\omega_{ni}$. The denominator of this expression is such that a simple pole appears at the frequency of the intermediate state or resonance. The available laser bandwidth limits which resonances are involved, as only the resonances appearing in regions of nonzero spectral amplitude contribute. Thus, the sum over $m$ is reduced to a sum over bandwidth-accessible intermediate states for the transition.

Removing the Divergences

In order to meaningfully evaluate Eq. (B.18), the integrand in Eq. (B.18) is extended to the complex plane, promoting $\omega \to z$, $z \in \mathbb{C}$. Then this integrand, be known as $f(z)$, is

$$
f(z) = \frac{g(z)}{\omega_{mi} - z} = \frac{E(\omega_{ni} - z)E(\omega)}{\omega_{mi} - z}.
$$

(B.19)

Then the integral along the real line is replaced by an integral along the real axis that excludes the singularity, plus a semi-circular contour of vanishing radius that goes around the simple pole.

The integral along the real axis is defined [75], in general, by

$$
\mathcal{P} \int_{a}^{b} dx f(x) = \lim_{\epsilon \to 0^+} \left( \int_{a}^{c-\epsilon} dx f(x) + \int_{c+\epsilon}^{b} dx f(x) \right)
$$

(B.20)

and called the Cauchy principal value of the integral. The singular point is the point at $c$. For numerical integration, the problem can be simplified and
numerical stability gained. One constructs out of \( f(x) \) functions odd and even about the point \( c \),

\[
\begin{align*}
  f_{\text{odd}}(x, c) &= \frac{1}{2} (f(c + x) - f(c - x)), \quad \text{and,} \\
  f_{\text{even}}(x, c) &= \frac{1}{2} (f(c + x) + f(c - x))
\end{align*}
\]

respectively. Then a symmetric principal value integration of each of these

\[
\lim_{\epsilon \to 0^+} \left( \int_{-\epsilon}^{r} dx f_{\text{odd}}(x, c) + \int_{-r}^{-\epsilon} dx f_{\text{odd}}(x, c) \right) = 0, \quad \text{and,}
\]

\[
\lim_{\epsilon \to 0^+} \left( \int_{-\epsilon}^{r} dx f_{\text{even}}(x, c) + \int_{-r}^{-\epsilon} dx f_{\text{even}}(x, c) \right) = \lim_{\epsilon \to 0^+} 2 \int_{\epsilon}^{r} dx f_{\text{even}}(x, c)
\]

where \( r \) is a finite (user chosen) integration radius around the singular point. Only the even contributions to the principal value integral about the singular point contribute. Thus Eq. B.20 becomes

\[
P \int_{a}^{b} dx f(x) = \lim_{\epsilon \to 0^+} \left( \int_{a}^{c-\epsilon} dx f(x) + \int_{c+\epsilon}^{r} dx (f(c + x) + f(c - x)) + \int_{c+r}^{b} dx f(x) \right)
\]

Note that the divergence due to the denominator in Eq. B.18 imparts an oddness to the whole integrand, about the singular point, making the numerical stability good. This is the integral that Mathematica will perform if asked to do a principal value integration [76, 77].

The contribution from the semi-circular contour \( C_{\rho} \) going around the simple pole at \( z = \omega_{mi} \) is given by [78]

\[
\lim_{\rho \to 0} \int_{C_{\rho}} f(z) dz = -B_{0} \pi i
\]

The residue \( B_{0} \) is given by

\[
B_{0} \equiv \text{Res}_{z=\omega_{mi}} f(z) = g(\omega_{mi}) = E(\omega_{ni} - \omega_{mi})E(\omega_{mi}).
\]
Demoting $z$ back to $\omega \in \mathbb{R}$, Eq. [B.18] becomes

$$
c_n^{(2)} \sim \sum_m \mu_{nm}\mu_{mi} \left( \mathcal{P} \int_{-\infty}^{\infty} d\omega \frac{E(\omega_{ni} - \omega)E(\omega)}{\omega_{mi} - \omega} - i\pi E(\omega_{ni} - \omega_{mi})E(\omega_{mi}) \right).
$$

(B.26)
Appendix C

Software and Algorithms: QuantumBlackbox and Mathematica

C.1 QuantumBlackbox

QuantumBlackbox is written using C++. Specifically, the Microsoft Visual Studio 2010 interactive development environment (IDE) and compilers are used, along with Qt [79] version 4.8.3. As a result of the use of Qt, the resulting program is readily cross-platform portable.

Both the LabVIEW software and the components of QuantumBlackbox employ use object-oriented approach. For QuantumBlackbox, each distinct component is a C++ class object. These core components are now described.

C.1.1 Connecting to LabVIEW

One of chief purposes of this software is to provide a more flexible and convenient way to communicate with and control the lab hardware. In order to still make full use of the functionality provided by the LabVIEW software, QuantumBlackbox communicates with a special LabVIEW VI. This LabVIEW VI connects with QuantumBlackbox via a TCP/IP [80, 81], a standard networking protocol used on the Internet. It then reads from and writes to the hardware device, returning the information. On the QuantumBlackbox side, the class that implements this communication is called NetworkHardwareProxy.

An advantage of using a network protocol such as TCP/IP versus other forms of inter-process communication (IPC) typically provided by an operating system is that communication can easily happen even when the server software is running on a different machine than the client. And, if running on the same machine, the communication is extremely fast, so that there is no disadvantage to using TCP/IP.
C.1. Hardware Device Proxy Classes

Each of the important hardware devices with which QuantumBlackbox must communicate are represented by their own C++ class. These C++ classes expose sets of methods that represent each hardware object; the same is true in the LabVIEW software. Certain methods simply call on the NetworkHardwareProxy. For example, the SLMService class which represents the SLM calls the NetworkHardwareProxy when a SLM phase or amplitude mask must be applied to the SLM. Two other proxy classes include the SpectrometerService and the OtherHardwareService. These communicate with the spectrometer, and provide an means of returning IEEE 754 \[82\] double-precision floating point numbers (e.g. measurement results), respectively.

C.1.3 Numeric Routines

For convenience, a small collection of basic numeric routines exist in QuantumBlackbox. Many of these are also available for use with the genetic search component (Section 6.2). They include the following:

- A set of basic methods for adding arrays, multiplying them by constants, averaging an array of numbers and computing its standard deviation.

- Methods to shift or rotate the elements of an array (for use in, e.g., in Fourier transforms).

- A set of simple routines to find the minimum or maximum elements in an array, including the index or indexes of several of the maximum or minimum elements.

- A method to perform linear interpolation \[40\], accepting two arrays specifying the \((x_{in}, y_{in})\) coordinates of some data, as well as an array of new \(x_{out}\) values for which new values of \(y_{out}\) will be computed via linear interpolation.

- A method to perform cubic spline interpolation \[40\], accepting arguments similar to those accepted by the linear interpolation routine, and returning the smoothly interpolated points \(y_{out}\).

- A set of methods for performing fast Fourier transforms (FFT), based on the open-source FFTW library \[83\]. A set of helper methods is also
C.1. *QuantumBlackbox*

provided, which aid in doing the necessary interpolation and bookkeeping involved in computing a time-domain signal from a spectrum provided on a wavelength scale, and vice-versa. These methods are *not* used in the experiments done in this thesis, but provide a foundation for some other functionality that was written, but not used, in the program. These include routines to calculate the FROG (or XFROG) trace (Section 5.5.3) given some input field(s), part of the beginnings of an attempt to implement a genetic-search based FROG retrieval algorithm [84]. The use of the FFT to compute time, frequency and wavelength domain signals related to optical pulses is described in Section 5.4.1.

- A method to unwrap the phase in an array of points representing a phase function that has been wrapped modulo $2\pi$.

C.1.4 **JavaScript (ECMAScript) Engine and Editor**

Qt, and thus *QuantumBlackbox*, provide access to a JavaScript [85] engine. JavaScript is a scripting language often used in web pages, but the engine that actually runs the script is general and can be incorporated in any application. Qt wraps a JavaScript engine in a special way, such that methods from classes which inherit `QObject` (and are subsequently registered with the engine) can be called transparently.

In this way, all of the hardware device (proxy) classes and many of the numerical routines included in *QuantumBlackbox* are automatically made available via JavaScript. A particular experiment can be entirely coded in JavaScript, and will have full access to a wide variety of resources provided both by *QuantumBlackbox* and the LabVIEW suite.

C.1.5 **User Interface**

*QuantumBlackbox* runs natively as a windowed desktop application. Most of the graphical user interface provided is coded using web technologies. Then each window displayed to the user simply hosts an instance of WebKit [86] (included as part of the Qt library). Qt provides the necessary bindings which allow user actions on these web pages to be passed back to the C++ program, and vice-versa. The result is a very professional but easy-to-create interface.

In order to make parts of the interface that are hosted in WebKit, *QuantumBlackbox* makes use of the following open-source components:
• jQuery [87]. jQuery is an JavaScript library that enhances and simplifies that process of scripting elements on a web page.

• jQuery UI [88]. jQuery UI is an library that provides enhanced user-interface elements for use on a web page, and uses jQuery.

• CodeMirror [89]. CodeMirror is an interactive code editor that runs on a web page and uses JavaScript. QuantumBlackbox uses this component to expose a graphical user interface to the user where the JavaScripts that access lab hardware and run experiments can be edited.

The application also includes a module that can display any number of plots to the user. This module is accessed via JavaScript, and does not use WebKit, but rather makes use of an open-source library known as QCustomPlot [90].

C.1.6 Genetic Search Implementation Details

QuantumBlackbox uses GAlib [52], open-source collection of C++ classes that implement many of the common genetic algorithms and genetic operators. The genetic search component of QuantumBlackbox exposes a user interface which allows the user to choose values for many parameters provided by GAlib, including the population size, crossover and mutation rates, the replacement percentage, the selection scheme and scaling scheme. It also provides for the selection of the mapping function.

When GAlib is called on to run a genetic search, it repeated calls what it calls an “objective function.” This user-defined function is called by GAlib when it wants to receive an objective score for a particular genome. QuantumBlackbox sets this objective function to be a wrapper function that calls the user-provided JavaScript mapping function. This function is executed in an environment that has access to all same resources that the interactive JavaScript editor and engine (Section C.1.4) have access to. As such, the mapping function can basically do anything, including calling on the SLM, the spectrometer, and other laboratory equipment.

Each mapping function is actually defined as part of a JavaScript object, stored in a text file. As well as the map property which holds the mapping function, the object also defines a configure property. This latter property holds a function that returning the form of the genome which GAlib should use in the search. This entire object, originally loaded from the file, lives persistently in the JavaScript engine throughout the duration of the search.
Thus there exists a way to create very intelligent, flexible mapping functions that remember their state from one call to a next, if needed.

QuantumBlackbox provides a \textit{mixed} genome, meaning that searches can be conducted with any number of the following features:

- A binary string genome. This is simply a string of bits, the length requested by the configuration function.

- A decimal number genome. This genome provides a collection of decimal \textit{numbers}; each number in the genome will be represented by the requested number of bits, the number received in the mapping function taking values between some user-specified minimum and maximum. The number of bits in the representation will affect the performance of the search. The numbers can also, optionally, be Gray coded \cite{40, 52}. These numbers are also binary strings, as far as basic genetic operators are concerned.

- A real number allele genome. This provides another way to represent real numbers in a genome. The user may request one or more of: a real number within a specified range (internally represented differently than the decimal number genome), a set of numbers between lower and upper limits, with a given spacing, or a user-specified set of real numbers. This genome has its own special genetic operators associated with it; for example, mutation may change the number by a Gaussian-random amount, rather than flipping a bit.

The mapping function receives a copy of the genome, translated to a JavaScript-friendly form. When it is done it returns a JavaScript object containing the \textit{objective score}, and any other data that the user would like to store with this genome evaluation. The score along with all data are stored, in the text-based \textbf{JavaScript Object Notation} (JSON) \cite{91} format, for subsequent analysis and export.

\section*{C.2 Mathematica}

The numerical simulations related to the coherent control of two photon absorption in atomic rubidium (appearing in Chapter 7) use Mathematica routines that perform numerical principal value integrals (see also the reference in Section 3.2 of Appendix B).

To make these Mathematica routines callable from LabVIEW and QuantumBlackbox, a small module known as a dynamic-link library (DLL) was
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written. This DLL uses the interface known as *MathLink* to communicate directly with the Mathematica kernel. LabVIEW loads this DLL, which allows it to communicate with Mathematica. The TCP/IP channel between QuantumBlackbox and LabVIEW then allows the QuantumBlackbox direct access to numerical integration results returned from the Mathematica kernel.