Coherent control of two-photon transitions in Rubidium

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Abstract

Usually Doppler-reduced direct frequency comb spectroscopy is limited by the amount of Doppler-broadened background signal. To circumvent this limitation we have combined quantum coherent control with high-resolution Doppler-reduced frequency comb spectroscopy on two-photon absorption in a three-level system in Rubidium. We have shown a method to reduce Doppler-broadened background signal without sacrificing the Doppler-reduced signal by a smart choice of a spectral phase. To study coherent control of two-photon transitions enhanced by a single-photon transition near resonance the high spectral resolution of the frequency comb has been employed to separate different types of excitation. Single-sided excitation of Rubidium atoms was studied to gain an understanding of two-photon transitions, where an enhancement up to a factor 10 was possible in comparison with excitation with a transform-limited pulse. Furthermore, we have shown that from a coherent control perspective a counterpropagating beam alignment necessary for Doppler-reduced direct frequency comb spectroscopy is almost equivalent to two-photon Fourier spectroscopy with shaped pulses.
1. Introduction

Two-photon absorption by an atom is not as commonly known as single-photon absorption. To the experimenter it is much more interesting because the two photons required for the transition can be presented to the atom in all kinds of different geometries. A two photon transition is similar to a tandem of two single photon transitions within a short (ps) time delay. The atom is excited from an initial state to the final state, and the sum of the photon energies required to make the transition has to be the level separation. Therefore there is only a requirement on the sum frequency of the photons, not on the individual frequencies, providing an additional knob to turn.

Two-photon transitions were predicted by the theoretician Maria Göppert Mayer in 1929. Verification of her theory was not possible at the time because it requires a high number of available photons, and therefore an intense light beam. As the laser did not exist at the time, it was not possible to produce a beam that was intense enough to verify the prediction. More than thirty years later, with the invention of the laser in 1960, experimental verification using a pulsed laser was possible. A pulsed laser is a valuable tool for two-photon excitation since the photons are not only coherent, but they are also bunched in time. Therefore high number of photons are available in the pulse.

We will study coherent control of two-photon absorptions in Rubidium using a femtosecond frequency comb laser. Such a laser emits extremely short pulses of only a few quadrillionths of a second with a well defined repetition frequency and phase relation. Due to the short duration of the pulses their bandwidth is very broad, but within this broad spectrum there is a mode structure consisting of extremely narrow modes. Since the properties of these modes can be precisely controlled this makes them very suitable for spectroscopic applications. The broad bandwidth of ultrashort pulses makes them very suitable for pulse manipulation in the frequency domain.

To understand and manipulate two-photon transitions, two research fields will be combined that - other than that they are both laser science - do not seem to have anything in common with each other: coherent control and high precision frequency comb spectroscopy. Coherent control is usually employed to steer photochemical reaction processes, or study biological enzymes. High precision spectroscopy is the science to measure atomic transitions with an incredibly high precision, e.g. to test variation on fundamental constants or Quantum Electrodynamics. It could not be more distinguished from the study of biological enzymes.

Yet we are going to combine these fields. We will employ coherent control techniques to suppress unwanted background signals for high precision spectroscopy by pulse manipulation, and we will make use of the very high spectral precision of the frequency comb to switch on and off different paths of two-photon absorption to study coherent control of the transition.

In the next chapter properties of the frequency comb are discussed. In chapter 3 the theory of two-photon absorption is covered. The experiments are described in chapters 4-6. The experiments start with exciting atoms with a single pulse at a time, controlling the relative amount of excitation by manipulation of the characteristics of the pulses (chapter 4). Then we will switch to two pulses with a variable delay time. This leads to optical and quantum interferences in the atom (chapter 5). Lastly, we combine the knowledge from both the single pulse experiments and the double pulse experiments in a setup with counter-propagating shaped pulses, where coherent control meets high precision spectroscopy (chapter 6). The coherent control techniques developed in the previous experiments are employed to selectively excite the signal that is of interest. On the other hand, the high spectral resolution of the frequency comb allows us separate excitations where both photons originate from the same pulse from excitations where one photon originates from each counter-propagating pulse.
2. Mathematical description of ultrashort laser pulses and pulse shaping

2.1. Introduction

The leading principle of the research in this thesis is the effect of femtosecond laser pulse manipulation on two photon absorption (TPA) in atoms. Therefore a proper mathematical description of the relevant properties of laser light pulses is required. This chapter provides a mathematical background and discusses the setup required to manipulate femtosecond laser pulses.

Since the atom is mainly perturbed by the electric field of the light, the description will not cover the magnetic field. The pulses coming out of the laser are very short (on the order of femtoseconds), separated by the roundtrip time of the cavity. First we consider a description and methods to characterize and manipulate a single pulse. Then the effect of a pulse train of identical pulses on the spectrum is discussed.

2.2. Mathematical description of a single laser pulse

The most intuitive approach to describe a pulse might be to describe the electric field of the pulse as a function of time, \( E(t) \). However, due to the short duration, it is not possible to characterize and manipulate femtosecond pulses directly in the time domain. Therefore the description will be in the frequency domain, related by a Fourier transform to the time domain:

\[
E(\omega) = \int_{-\infty}^{\infty} E(t) e^{i\omega t} dt = \sqrt{I(\omega)} e^{i\Phi(\omega)} \tag{2.1}
\]

Where \( E(t) \) and \( E(\omega) \) are the electric as a function of time resp. frequency. \( I(\omega) \) is the spectrum of the pulse, and \( \Phi(\omega) \) is the spectral phase. By defining the spectral amplitude \( A(\omega) = \sqrt{I(\omega)} \) this equation can be written as

\[
E(\omega) = A(\omega) \exp(i\Phi(\omega)) \tag{2.2}
\]

Here both spectral amplitude and spectral phase are real functions. The shorter the duration of a pulse, the broader the bandwidth will be, also known as the Fourier limit. The spectral amplitude defines which frequencies are present in the pulse. The spectral phase contains the relative phases of the different frequency components.

The role of the spectral phase can be understood by expanding it in a Taylor series around the central frequency of the pulses \( \omega_c \).

\[
\Phi(\omega) = \phi(\omega_c) + \phi'(\omega_c)(\omega - \omega_c) + \frac{1}{2} \phi''(\omega_c)(\omega - \omega_c)^2 + \frac{1}{6} \phi'''(\omega_c)(\omega - \omega_c)^3 + \cdots \tag{2.3}
\]

Every order has a different effect on the pulses. The effects of the different orders of spectral phase in the time domain can be seen in figure 2.1. The zeroth order term does not change the pulse shape, it only leads to a phase difference in the carrier of the pulse. \( \phi'(\omega) \) is called the Group Delay (GD) and leads to a time delay. The delay is dependent on the steepness of the spectral phase. \( \phi''(\omega) \) is called Group Velocity Dispersion (GVD) and it can be interpreted as a different group delay for every frequency. The frequencies that are further off \( \omega_c \) will be shifted more, therefore stretching the pulse, and also causing that the carrier frequency of the pulse will change linearly in time (chirp). \( \phi'''(\omega) \) leads to pre- and postpulses, and is called Third-Order Dispersion (TOD). Higher-order modes will in general lead to more stretching of the pulses and pre- and postpulses.
2. Mathematical description of ultrashort laser pulses and pulse shaping

Pulses will be shortest when the spectral phase is flat, so there is no GVD or third- or higher order dispersion. If pulses have a flat spectral phase, they are called transform limited. In this case, there is a simple relation between pulse duration and bandwidth:

\[ \Delta t \Delta \nu = K \]  \hspace{1cm} (2.4)

Where \( \Delta t \) is the pulse duration and \( \Delta \nu \) is spectral bandwidth of the pulse. \( K \) is dependent on the shape of the spectrum. Our spectrum will mainly be rectangular, for which \( K = 0.88 \).

2.3. Femtosecond pulse shaping

Femtosecond laser pulses are very suitable for manipulation in the frequency domain because of their broad bandwidth. We use a 4\( f \)-pulse shaper. A nice introduction on shapers can be read in Monmayrant et al. 2000[1]. The setup works by focussing the different frequencies spatially dispersed in a plane called the Fourier plane. The properties of the pulse are then altered in this plane, and the pulse is transformed back again to the time domain[2, 3]. This is described in the next section.

The incoming beam is spatially dispersed onto a grating (g1 in figure 2.2). The different colors are propagating in different directions. The frequencies are focussed by a cylindrical mirror (c1). Since the different colors were propagating in different directions they are now focussed in different positions, in the Fourier plane (fp). Here the beam propagates through a Spatial Light Modulator (SLM). This is a nematic LCD that can apply a different phase retardation to every pixel. The phase retardation is accomplished by changing the index of refraction – and hence the light speed inside the pixel – of every pixel. Every pixel can be programmed individually. The maximum retardation that a pixel can apply is about 3\( \pi \). Steeper phases can be accomplished by wrapping a spectral phase around 2\( \pi \), as the spectral phase is insensitive to a wrapping around 2\( \pi \). This only works as long as the phase difference between two successive pixels is less than 2\( \pi \), otherwise aliasing will occur. Using a mirrored setup the pulse is transformed back into the time domain. Coarse amplitude shaping is possible in this setup by blocking a part of the beam in the Fourier plane. In this way the spectrum of the laser can be clipped.

Our setup employs only reflecting optics to minimize GVD and TOD due to the glass used in lenses and prisms. Furthermore, the setup is folded by using a flat mirror after the cylindrical mirror to allow...
2. Mathematical description of ultrashort laser pulses and pulse shaping

Figure 2.2. 4f-pulse shaper in folded geometry. The incoming laser beam is dispersed on the first grating (g1). The different frequencies are focused by a cylindrical mirror (c2) via a flat mirror (f1) into the Fourier plane (fp). In the Fourier plane a 640-pixel spatial light modulator can apply a different retardation to every pixel, and therefore a different retardation to the frequencies in the spectrum (orange box). The shaped light is reconstructed by a mirrored setup (g2, c2, and f2), to form the shaped outgoing pulse. By blocking a part of the beam in the Fourier plane, amplitude clipping is possible. The distance between all active optical elements (g and c, c and fp) is exactly the focal length of c, F.

This is a very sensitive optical setup. If the distance between the optical elements is not exactly the focal distance of the curved mirror $F$, the setup will cause GVD and TOD. In other fields such as optical chirped pulse amplification this is employed to create and compress chirped pulses[3], but in our case this is undesired behaviour. The pulses that come out of the shaper need to be transform-limited when no spectral phase is imprinted on the SLM. An interferometric autocorrelator was used to minimize GVD and TOD of the beam coming out of the shaper. An interferometric autocorrelator is a tool for pulse characterisation that is sensitive to the spectral phase. Two time delayed copies of the same pulse are focussed in a second harmonic generating crystal. A filter selects only the second harmonic light, and the intensity is measured using a PMT for a varying delay between the pulses. The signal is dependent on the spectral phase, but it does not easily allow for direct phase reconstruction. The distances in the 4f-shaper have been optimized on the autocorrelation signal. More information about interferometric autocorrelation can be found in Diels et al. 2006[4].

By minimizing the GVD of the shaper with the autocorrelator, any GVD that was present in the beam before it entered the shaper is also corrected. However, this comes at the expense of introducing spatial chirp. If the distance between the gratings is not exactly $4F$, the beam is not exactly focussed at the second grating. Therefore different colors enter g2 at slightly different positions. Therefore the spectrum of the beam coming out of the shaper can be dependent on the position in the beam. Also small errors of the vertical alignment will cause spatial chirp, or a setup that is not exactly mirrored around the Fourier plane. A procedure to align the shaper properly can be found in appendix A.

2.3.1. Typical phase profiles

Using the pulse shaper it is possible to imprint an arbitrary spectral phase on the pulses, provided that the desired spectral phase is not too steep. There are only three spectral phases that are used extensively in this thesis, and their most important features will be discussed here in the frequency domain as well as the time domain. They are shown in figure 2.3.
2. Mathematical description of ultrashort laser pulses and pulse shaping

\[ E(t) = A(\omega) \exp \left( \Phi(\omega) \right) \]

Figure 2.3. Spectral amplitude and phase, and the corresponding intensity and phase in the time domain for the most common spectral phases in this thesis. The left figure shows the pulse in the time domain, the right figure shows the corresponding spectral phase and amplitude (in corresponding colors). From top to bottom: A Transform-limited pulse (blue), a negative Vshape (green) and a Pi-step phase (red). The transform limited pulse shows a flat spectral phase. The Vshape splits the pulse in two subpulses, one containing all higher frequencies and one containing the lower frequencies, indicated with "low" and "high". To make this effect more visible, the central frequencies of the spectrum are suppressed. The Pi-step causes destructive interference instead of constructive interference at frequencies near the central frequency. Therefore at \( t = 0 \) there is no longer a maximum, and the pulse is stretched.
2. Mathematical description of ultrashort laser pulses and pulse shaping

**Transform limited pulse** A TL pulse is shown in the top row in figure 2.3, with a Gaussian pulse profile in the time and in the frequency domain. This is the pulse that comes out of the shaper when no phase is applied to the shaper. At \( T = 0 \) all the frequencies interfere constructively, thereby there is a maximum signal. For larger \( T \), they start to run out of phase. For large \( T \) they interfere destructively, leading to a short pulse in the time domain where the frequencies interfere constructively, and no signal for larger \( T \), since the frequencies interfere destructively.

**Vshape** A spectral Vshape is defined as follows:

\[
\Phi(\omega) = \tau |\omega - \omega_c|.
\]  

(2.5)

Where \( \omega_c \) is the step frequency. This is a more complicated version of a linear spectral phase, where the steepness is either \( \tau \) or \(-\tau\), depending on \( \omega \). Therefore this spectral phase splits the pulse in a subpulse containing all frequencies higher than \( \omega_c \) and a subpulse with all frequencies lower than \( \omega_c \) separated by \( 2\tau \). The sign of \( \tau \) determines the order of the pulses. For \( \tau > 0 \), the red subpulse arrives earlier than the blue subpulse, while for \( \tau < 0 \) this is reversed. Furthermore, the shape of the two subpulses is slightly different from the original pulse, since it contains only half the bandwidth and a spectrum that is clipped at \( \omega_c \).

**Pi-step** The last spectral phase to be considered is a Pi-step, a step function of height \( \pi \) at frequency \( \omega_\pi \).

\[
\Phi(\omega) = \begin{cases} 
0 & \omega < \omega_\pi \\
\pi & \omega > \omega_\pi
\end{cases}
\]  

(2.6)

The constructive interference at \( T = 0 \) that was discussed for the TL pulse is now lost since half of the pulse is \( \pi \) out of phase with the original pulse. Therefore the intensity at \( t = 0 \) will be reduced. Also the pulse will be stretched because the frequencies close to \( \omega_\pi \) now start to interfere constructively for larger \( T \) before finally going out of phase.

2.4. Pulse trains and Frequency Combs

So far, we just discussed the features of a single laser pulse. At the heart of our setup is a Kerr-lens mode-locked Ti:Sapph laser. It does not emit a single pulse, but it emits a series pulses separated by the roundtrip time \( T \). The envelope of the pulses is identical, but the carrier of the pulse may experience a pulse-to-pulse phase shift. For now, we’ll assume that the laser cavity is perfect: the roundtrip time and the pulse-to-pulse phase shifts do not change over time. Later on we will show that it is possible to actively control these properties. In this section we will discuss the effects of this repetitive pulse train on the spectrum. This section outlines only the basic properties. More information about Kerr-lens mode-locked lasers and frequency combs may be found in the Springer Handbook of Optics, Chapter 12[5]. A very readable introduction is also given by Witte 2007, chapter 2[6]. I will copy his notation.

Where previously the spectrum was continuous, the repetitive nature of the pulses alters the laser spectrum: The spectrum will consist of very narrow comb modes separated by a frequency that equals the repetition rate: \( f_{\text{rep}} = 1/T \). The relative height of the modes is given by the single-pulse spectral profile. The pulse-to-pulse phase shift will lead to a shift of the modes by \( f_{\text{ceo}} = \Delta\phi/(2\pi)f_{\text{rep}} \). Therefore the mode positions are described by

\[
f_n = f_{\text{ceo}} + nf_{\text{rep}} \quad n \in \mathbb{Z}.
\]  

(2.7)

This only holds as long as the cavity remains constant over time. Fortunately, both \( f_{\text{rep}} \) and \( f_{\text{ceo}} \) can be actively controlled. This is discussed in the next section.

**Locking the repetition frequency**

The repetition frequency of the laser light can be measured by diverting a part of the laser light onto a fast photodiode. The photodiode is insensitive to the carrier of the pulses, but it will measure a peak for every pulse, hence the repetition rate. Therefore, it’s frequency contains spikes at \( f_{\text{rep}}, 2f_{\text{rep}}, \ldots \) until
2. Mathematical description of ultrashort laser pulses and pulse shaping

Figure 2.4. A frequency comb in the time and frequency domain. A train of pulses separated by time $T$ in the time domain translates into a series of regularly spaced modes in the frequency domain. A pulse-to-pulse shift of the carrier $\Delta \phi$ translates to a frequency shift of the modes of the comb. The frequency of mode $n$ can be calculated using $f = nf_{\text{rep}} + f_{\text{ceo}}$. Graph courtesy of Dr. S. Witte.

the maximum frequency that the photodiode is able to measure. This signal can be difference frequency mixed with a reference frequency that is close to $n$ times the repetition frequency. By reflecting the beam inside the laser cavity over a mirror on a piezo translation stage, the cavity length and therefore the repetition frequency can be modified. Using a PID controller the repetition frequency can be locked to the reference frequency.

**Locking the carrier envelope offset**

Figure 2.5. f-to-2f method schematic. Low frequency modes from the comb are combined to nearly the double frequency, and difference frequency mixed with a high frequency mode $m = 2n$. The resulting signal contains $f_{\text{ceo}}$. Based on a figure by dr. S. Witte.

Locking the carrier envelope offset is much harder than locking the repetition frequency because the laser does not emit any radiation at this low frequency. This problem has only been solved in the year 2000 by the development of the f-to-2f technique[7, 8], where a beat note is created between a frequency-“doubled” low frequency mode and a high frequency mode of the comb. See figure 2.5. We will only explain the principle of the f-to-2f technique, but we will not go into detail on the experimental
2. Mathematical description of ultrashort laser pulses and pulse shaping

implementation in our setup. Interested readers may find the setup in Witte 2007, chapter 2[6].

Suppose that the spectrum of the laser spans more than a full octave. Therefore the frequency of
the highest mode is more than two times as high as the frequency from the lowest mode. Now the low
frequency comb modes are frequency “doubled” using sum-frequency generation. The frequency of such
a mode is then

\[ 2f_n = 2f_{ceo} + mf_{rep}. \] (2.8)

Where \( m = 2n \). This signal is difference frequency mixed with the 2n-th high frequency mode \( f_{2n} \). The
lowest difference frequency between the frequency doubled mode at frequency \( 2f_n \) and the mode at \( f_{2n} \)
is now

\[ 2f_n - f_{2n} = 2f_{ceo} + 2nf_{rep} - f_{ceo} - mf_{rep} = f_{ceo}. \] (2.9)

Of course, the beat signal between different modes is also available, giving a series of frequencies at
\( f_{ceo}, f_{rep} - f_{ceo}, f_{rep} + f_{ceo} \), etc. The first component can be filtered out with an electronic filter.

To perform feedback, the power of the pump laser can be modulated using an Acousto-Optic Modulator
(AOM). This results in a difference between phase velocity and group velocity of the pulses, therefore
changing the \( f_{ceo} \).

Therefore we now have a frequency comb with modes at \( nf_{rep} + f_{ceo} \), where it is possible to tune
both \( f_{rep} \) and \( f_{ceo} \). In the next chapter the interaction between the frequency comb light and an atom
is discussed.
3. Two-photon absorption in atoms

3.1. Introduction

This chapter explains the basics of two-photon excitation in a gas cell at room temperature. First an explanation is given on what two-photon transitions are. Then we consider excitation of a single stationary atom by a single pulse. We then expand the discussion until we arrive at frequency comb excitation of moving atoms in a gas at room temperature, taking into account Doppler broadening and Doppler shifts.

3.2. Two-photon transitions and transition probability amplitudes

Two-photon absorption (TPA) is similar to a tandem of two single photon transitions within a very short time delay. Two photons arrive more or less at the same time at an atom. The first photon excites the atom to a “virtual state”, and the second photon excites the atom to a final state, where the final state has an energy difference equal to the sum of the energy of the two photons. It should be stressed that the virtual state is not a real state of an atom. It is simply a superposition of wavefunctions.

Just like single photon transitions, there are selection rules for two-photon transitions. They are just the vectorial sum of two one photon transitions. So as the angular momentum quantum number \( l \) has to change by \( \pm 1 \) for a single photon transition, for a two-photon transition the change has to be \( \Delta l = \{-2, 0, 2\} \). So starting from an \( s \) state, it is possible to excite an atom either to a higher \( s \) state, or a \( d \) state, but not a \( p \) state. See figure 3.1(a) for an \( s \) to \( s \) transition, and figure 3.1(b) for an \( s \) to \( d \) transition. The relative amount of excitation can be measured because the atoms emit fluorescent light at 420 nm when decaying from the excited state.

\[
|g\rangle, 5S_{1/2} \rightarrow |i\rangle, 5P_{1/2} \rightarrow |f\rangle, 7S_{1/2}
\]

\[
|g\rangle, 5S_{1/2} \rightarrow |i\rangle, 5P_{3/2} \rightarrow |f\rangle, 5D_{3/2}
\]

\( \omega_{fg} - \omega_{ig} \)

\( 6P_{3/2,1/2} \)

\( 90 \text{ GHz} \)

\( 5D_{3/2}, 5D_{1/2} \)

Fluorescence

\[ \left| f \right> \times 7S_{1/2} \]

\[ \left| f \right> \times 5D \]

Figure 3.1. Energy level diagram for Rb showing the \( 5S \rightarrow 7S \) transition (a) and the \( 5S \rightarrow 5D \) transition (b). The ground state is called \( g \); the intermediate state is called \( i \) and the final state is called \( f \).

The broad spectrum of a short pulse means that there is not only a single combination of photons possible, but that there is a whole range of photon pairs that together match the energy difference. Not every pair of photons is equally probable. The transition is more likely to occur if the first photon is closer to a real state. If the atom was in an \( s \) state this will be a \( p \) state. Therefore there are three
3. Two-photon absorption in atoms

relevant energy levels: The ground state, the intermediate state and the final state, marked $|g\rangle$, $|i\rangle$ and $|f\rangle$. They have three associated frequencies corresponding to the difference energy between the levels, called $\omega_{g}, \omega_{f}$ and $\omega_{i} = \omega_{f} - \omega_{g}$. See figure 3.1.

There is another excitation path possible to excite an atom to the final state called resonant excitation. A single photon excitation can excite the atom to the intermediate state, and another single photon excitation can excite the atom from the intermediate state to the final state. This is different from two-photon excitation, as in this case the lifetime of the intermediate state is typically on the order of tens or hundreds of nanoseconds, and the excitation can therefore take place sequentially with a sizable time delay in between.

The transition probability can be calculated using second-order perturbation theory, calculating the overlap between the wavefunctions of the different eigenstates of the atom after a perturbation by the electric field of the light pulse. The complex two-photon transition amplitude between states $|g\rangle$ and $|f\rangle$ is given by: [9]

$$a_{fg} = 2\pi E(\omega_{ig})E(\omega_{fg} - \omega) + \int_{0}^{\infty} d\omega \frac{E(\omega)E(\omega_{fg} - \omega)}{\omega_{ig} - \omega}$$

(3.1)

To calculate the transition probability $a_{fg}$, should be modulus squared. $E(\omega)$ is the complex electric field of the exciting laser pulse, $\omega_{ig}$ is the frequency of the resonance level, and $\omega_{fg}$ is the frequency corresponding to the energy of the two-photon transition. The first term is the resonant excitation. The second term is the real two-photon transition, called the nonresonant excitation. The integral accounts for the different combinations of photon energies to reach the excited state. The weighting function accounts for the fact that not all photon pairs are equally likely to occur. It is plotted in figure 3.2. Consider a transform limited pulse. In this case $E(\omega) = A(\omega)$. The weighting function is antisymmetric around $\omega_{ig}$. Therefore an electric field that is symmetric around $\omega_{ig}$ will give rise to any nonresonant two-photon absorption, since the function to be integrated is perfectly antisymmetric. The only contribution will be from resonant excitation. An example is a transform-limited pulse. Intuitively one would think that transform limited pulses are optimal for TPA, because they yield the highest peak power, therefore during the peak the probability of two-photons coinciding with the right energy would be maximal[10]. This destructive quantum interference will be shown in section §4.3, by clipping a part of the spectrum, creating an enhancement of the TPA signal. A smarter way to enhance the relative TPA is by employing a spectral phase that makes use of the spectral response of the three level system. How this can be done using a $\pi$ step at $\omega_{ig}$ will be discussed in section §4.4.

3.2.1. Frequency comb effects

We will now extend the discussion from single pulse excitation to multi-pulse excitation of a single stationary atom by a frequency comb. In the frequency domain a single pulse corresponds with a continuous spectrum. As was discussed in section §2.4, a pulse train corresponds with a frequency comb with frequencies at specific modes $f_{n}$.

$$f_{n} = f_{ceo} + nf_{rep} \quad n \in \mathbb{Z}$$

The spectral intensity of a single pulse shows the distribution of the intensity of the different modes of the laser spectrum. To make a two-photon transition spectral intensity is required at $\omega$ as well as at $\omega_{fg} - \omega$. In this case all mode pairs can contribute to the transition, else there are no mode pairs that contribute to the transition. This leads to a constraint on the frequency of the laser modes determined by $f_{ceo}$ and $f_{rep}$. By scanning either the repetition rate or the carrier envelope offset, the frequency comb can be tuned in such a way that both frequencies are available, and TPA is possible. In this situation the frequency comb is in resonance with the two-photon transition. If the repetition rate is tuned such that no TPA is possible, the frequency comb is off resonance with the two-photon transition, and no TPA is possible. Since both $f_{rep}$ and $f_{ceo}$ are known and can be controlled, this is what makes a frequency comb such a versatile tool for high precision spectroscopy.

To measure an optical transition with high resolution, a method called Direct Frequency Comb Spectroscopy can be used. The repetition frequency is slowly scanned, and the TPA signal is measured. Changing the repetition frequency means adjusting the comb spacing. The frequency difference of a mode scales with the mode number. If the repetition frequency is changed by $\Delta f$, the frequency changes
3. Two-photon absorption in atoms

Figure 3.2. Weighting function as was derived in equation (3.1). The resonance at \( \omega = \omega_{ig} \) is shown by the dashed line. Calculating the transition probability involves integration over the weighting function times the electric field. The weighting function is antisymmetric with respect to \( \omega_{ig} \). Therefore an electric field that is symmetric with respect to \( \omega_{ig} \) will experience perfect destructive interference, and the net value of the integral is zero, as indicated by the blue shaded areas.
3. Two-photon absorption in atoms

by \( n \cdot \Delta f \) for the \( n \)th mode. For a certain \( \Delta f \), the frequency shift of the \( n \)th mode equals the mode spacing \( f_{\text{rep}} \). Then there is essentially no difference with the previous situation, only the \( n \)th mode was substituted with the \( n + 1 \)th mode. Therefore any signal that is dependent on the mode positions will repeat itself after scanning the repetition rate with \( f_{\text{rep}}/n \). Since the sum of the mode frequencies should add up to \( \omega_{fg} \), \( n \) corresponds with the mode number of the sum frequency, or in this case \( \omega_{fg} \). For example: The Rb 5\( S \) → 5\( D \) transition frequency is 770 THz. The mode spacing is approximately 127 Mhz, so \( n \) is approximately 6 million. A full repetition rate scan then corresponds to scanning the repetition rate by \( f_{\text{rep}}/n = 21 \) Hz.

3.3. Doppler Broadening

In a gas cell every atom can have a different speed and direction, governed by the Maxwell distribution. Therefore every atom experiences a different Doppler shift. Averaged over all the atoms it will cause a Doppler broadening that is more than the mode spacing of the frequency comb. This means that the frequency comb aspect of the laser is lost for a single beam exciting a gas of atoms. This is discussed in detail in section §3.3.1. While this is convenient to verify the model in equation (3.1), it is inconvenient to do spectroscopy. To be able to resolve the transition with frequency comb resolution, a technique will be used called Doppler-reduced frequency comb spectroscopy. For this technique two pulses are sent through the gas cell in a counter-propagating manner. In this way the Doppler broadening will only be proportional to the frequency difference of both photons that are required for the two photon transition. This is discussed in detail in section §3.3.2.

3.3.1. Single-sided Doppler broadening

So far we have discussed two-photon absorption of a single stationary atom excited by a frequency comb. In this section we show that for single sided excitation by a frequency comb it suffices to approximate it with single pulse excitation of a single atom. To do this we have to look at Doppler broadening of the TPA signal.

An atom moving with velocity \( \vec{v} \) through a laser beam that is propagating in the \( \vec{x} \) direction observes a Doppler shifted frequency \( \omega_{\text{obs}} \) of the photons in the beam:

\[
\omega_{\text{obs}} = \omega \left(1 - \frac{v_x}{c}\right)
\]

(3.2)

Where \( v_x \) is the velocity component of the atom in the direction of beam propagation and \( \omega \) is the photon frequency. This means that the frequencies of the frequency comb will appear to be shifted. A typical Doppler shift is on the order of a GHz, much more than the mode spacing of the laser. The Doppler shift is not the same for different frequencies, but since the spectrum of the exciting pulse is narrow compared to the central frequency of the pulse we can approximate that the Doppler shift is constant over the spectrum.

Suppose that the laser was in resonance with a stationary atom. If the atom starts moving, the modes will shift out of resonance, so the atom will not be excited any more. However, the laser can be in resonance once more if the velocity of the atom is such that the Doppler shift equals the mode spacing of the frequency comb. This is a condition on the velocity of the atoms. Only atoms that are within a certain velocity class can be excited. The most probable velocity of the atoms in the gas corresponds to a Doppler shift that is much more than the mode spacing of the frequency comb. For any position of the modes of the laser, there are a number of velocity classes that are excited. The TPA signal will therefore effectively be independent of the mode positions.

So the mode structure of the spectrum can be neglected. A spectrum without mode spacing corresponds with a single pulse. TPA by direct excitation of atoms in a gas at room temperature with a frequency comb can therefore be modeled as TPA by a single pulse on a stationary atom. Such an experiment can verify equation (3.1), but is disadvantageous for spectroscopy. A technique to reduce the Doppler broadening effect so that the modes (and therefore the precision) of the frequency comb are recovered is discussed in the next section.

3.3.2. Doppler broadening reduction by counter-propagating laser beams

To achieve frequency comb resolution in the experiments, a technique called Doppler-reduced spectroscopy developed by Theodor Hänsch et al. will be used[11]. It makes use of a counter-propagating
3. Two-photon absorption in atoms

Suppose that the two-photons that are required for the transition do not propagate in the same direction. Instead, they propagate in exactly the opposite direction. At the point where they coincide an atom at a certain velocity \(v_x\) along the first beam is excited by TPA. If the photons have frequencies \(\omega_1\) and \(\omega_2\), then the observed sum frequency \(\omega_{\text{sum}}\) by the atom only depends on the velocity by a factor that scales with the frequency difference between the two beams:

\[
\begin{align*}
\omega_{1,\text{obs}} &= \omega_1(1 - v_x/c) \\
\omega_{2,\text{obs}} &= \omega_2(1 + v_x/c) \\
\omega_{\text{sum}} &= \omega_{1,\text{obs}} + \omega_{2,\text{obs}} = \omega_1 + \omega_2 + v_x/c(\omega_2 - \omega_1).
\end{align*}
\]

Since an individual atom experiences a greatly reduced Doppler shift, an ensemble of atoms will have a greatly reduced Doppler broadening. The Doppler broadening due to the frequency difference is called the residual Doppler broadening. This is much less than the mode spacing of the laser, so the TPA signal will be sensitive to the frequency comb mode spectrum. Furthermore, the TPA signal will be much stronger because all velocity classes are excited, contrary to the single-sided excitation, where only a few velocity classes were excited. The residual Doppler broadening is dependent on the frequency difference between the two photons. Since the frequencies close to \(\omega_{ig}\) are most important, the residual Doppler broadening is dependent on the frequency difference between \(\omega_{fg}/2\) and \(\omega_{ig}\).

Experimentally this setup can be achieved by splitting the laser beam with a beam splitter, and aligning both beams through a gas cell. See figure 3.3. Two pulses enter the gas cell from opposite directions and meet in the middle of the gas cell. In the middle of the gas cell Doppler-reduced two-photon excitation may be observed.

For such a counter-propagating geometry single sided excitation of two photons by a single pulse is also possible. For spectroscopy this single-sided signal is disadvantageous, as it is independent on the mode structure. There are a number of techniques to reduce the amount of single-sided excitation while maintaining the Doppler reduced signal. They will be discussed in chapter 6. Lastly, there will also be a Doppler shifted signal. This is due to resonant excitation. For a given \(f_{\text{rep}}\) and \(f_{\text{coo}}\), there will always be excitation of a specific velocity class to the intermediate state. If the mode spacing is right for this velocity class, the atom can be excited to the final state. Since the first photon selects a velocity class, only atoms with a certain velocity are measured. This means that the peak will be shifted with \(v/c(\omega_2 - \omega_1)\) but not broadened. These peaks cannot be suppressed.
4. Single-sided excitation of Rb in a gas cell

4.1. Introduction

This chapter covers experiments on single-sided excitation of a two-photon transition. As was discussed in section §3.3.1, this can be seen as a single pulse exciting a single stationary atom. It provides a straightforward way to verify the theory of two-photon absorption discussed in section §3.2. Furthermore, the behaviour of TPA on excitation with different classes of spectral phases can be understood. For instance, as was discussed in section §3.2, transform limited pulses are far from optimal to excite a two-photon transition. We will discuss various ways to enhance the transition probability, thereby demonstrating this destructive quantum interference due to the resonance level at the intermediate state. Furthermore, an experiment is shown where interference between the resonant and nonresonant can be tuned. To aid in understanding the physics, the transition amplitude can be solved analytically for the experiments in this chapter, providing additional insight. For the later experiments in chapter 5 and chapter 6 only numerical solutions of equation (3.1) can be calculated.

The experiments will be done in a Rb gas cell, either exciting the Rb $5S \rightarrow 5D$ using the $5P_{3/2}$ or $5P_{1/2}$ state as an intermediate state. How the amount of TPA is measured is discussed in the next section. Sections 4.3 and 4.4 discuss optimization of TPA via amplitude clipping and spectral phase shaping, resp. Section 4.5 shows interference effects between resonant excitation and nonresonant excitation of single atoms.

4.2. Experimental Setup

The setup is shown in figure 4.1. The pulses from the Ti:Sapph modelocked laser are shaped by a pulse shaper discussed in section §2.3, and focussed into a gas cell. The gratings of the shaper are 1600 lines/mm, and the radius of curvature of the cylindrical mirrors is 50 cm. The SLM is a Jenoptik SLM-S 640/12, with a pixel size of 100 µm. This corresponds to a dispersion of 0.3 nm/pixel of the SLM. The gas cell is heated to about 70°C to increase the Rb gas cell pressure. The relative amount of TPA can be measured because the atoms decay from the excited state back to the ground state via the $6P$ state, emitting fluorescence light at 422 nm. The fluorescent light is selected by a filter and captured by a Photo Multiplier Tube (PMT), a sensitive detector for photons. It emits a pulse when a photon is detected. The pulses are counted using an Agilent 53132A Universal Frequency Counter (not shown). The counter collects the number of pulses over a certain time interval, called the gatetime. The number of counts in a gatetime is transmitted to the computer. A measurement of the relative TPA of a certain pulse manipulation consists of manipulating the pulse with the shaper, and recording the number of counts on the PMT for every manipulation.

The Rb $5S \rightarrow 5D$ transition will be studied using either the $5P_{1/2}$ or the $5P_{3/2}$ transition as an intermediate state. See figure 3.1(b). These two transitions are interesting because by using the $5P_{1/2}$ transition the resonance level is almost exactly at $\omega_{ig}/2$. In a counter-propagating geometry this is interesting because Doppler reduction will work better. An additional advantage is that due to the small frequency difference between photon pairs the setup is not very sensitive to spatial chirp. When using the $5P_{3/2}$ transition the photon pairs are a bit further apart.

4.3. Enhancing TPA by blocking a part of the spectrum

As was discussed in section §3.2, transform limited pulses are not optimal for nonresonant two-photon absorption, due to destructive quantum interference. Photon pairs with $\omega > \omega_{ig}$ interfere destructively with photon pairs with $\omega < \omega_{ig}$, due to the sign change of the weighting function at $\omega = \omega_{ig}$. Therefore the net value of the nonresonant term is almost zero. In this section this destructive interference will be demonstrated. By clipping the spectrum in such a way that there are no photon pairs for $\omega < \omega_{ig}$, only
4. Single-sided excitation of Rb in a gas cell

Figure 4.1. Experimental setup for single-sided excitation of Rb. The pulses from the laser are shaped in the Fourier plane of the spectral phase shaper setup, and then focussed into the gas cell, where the Photo Multiplier Tube (PMT) measures the amount of excitation. The pulses can be phase shaped by the Spatial Light Modulator (SLM), and the spectral bandwidth can be clipped by blocking a part of the beam in the Fourier plane of the spectral phase shaper setup (indicated by the black line).
4. Single-sided excitation of Rb in a gas cell

the photon pairs with $\omega > \omega_{ig}$ are left, enhancing the TPA by reduction of the destructive quantum interference. Even though the pulses have less power, and they are stretched in the time domain, the TPA rate is enhanced. In this experiment the SLM is only used for chirp cancellation in order to produce transform limited pulses.

![Intensity vs Frequency](image)

Figure 4.2. Amplitude clipping result for single pulse excitation in Rb. The left figure shows the procedure, the right the results. Left figure: The blue line indicates the spectrum of the laser. The laser light is cut below the cutoff frequency $\omega_c$, indicated by the black box. $\delta$ is indicated. In this case $\delta > 0$. In the right figure the TPA signal is shown for while scanning $\omega_c$. Red indicates a measurement, blue the analytical model. To correct for the imperfect resolution of the shaper, the model was convolved with a Gaussian of 0.2 nm width. The TPA fluorescence has a peak as the cutoff frequency approaches $\omega_{ig}$.

This effect has been shown by Dudovich et al. [9]. They did not show that in this case the transition amplitude can be solved analytically. This will provide additional insight as it shows that a transform-limited pulse with a symmetric spectral intensity around $\omega_{ig}$ does not yield any nonresonant transition amplitude. The TPA transition amplitude can be calculated in the following way: assume that the pulses have a flat spectral phase, and a constant spectral intensity, i.e. a locally flat spectrum. Since we only use about 5-10% of the laser bandwidth for the experiment this is a valid approximation. All the frequencies below $\omega_{ig} - \delta$ are clipped (see left figure in figure 4.2). In this case $E(\omega)$ can be simplified to:

$$ E(\omega) = \begin{cases} 1 & \omega > \omega_{ig} - \delta \\ 0 & \omega < \omega_{ig} - \delta \end{cases} \quad (4.1) $$

This can be inserted in equation (3.1). Since the $E(\omega) = 0$ when $\omega < \omega_{ig} - \delta$, the integration boundaries can be simplified to $\omega_{min} = \omega_{ig} - \delta$ and $\omega_{max} = \omega_{fg} - \omega_{ig} + \delta$. The upper limit of integration is the limit where the complementary frequency is below $\omega_{ig} - \delta$. Furthermore, the resonant term is only present as long as there is spectral intensity at $\omega_{ig}$.

$$ a_{fg} = \frac{\tau}{\pi} \frac{1}{2} (1 + \text{sign } \delta) + \int_{\omega_{ig} - \delta}^{\omega_{fg} - \omega_{ig} + \delta} \frac{1}{\omega_{ig} - \omega} d\omega \quad (4.2) $$

Here $\frac{\tau}{\pi} (1 + \text{sign } \delta)$ is just a function to return $\tau/\pi$ when $\delta > 0$. The integral in equation (3.1) can be split in a part that is symmetric around $\omega_{ig}$ and an asymmetric part that only extends to higher frequencies (the lower frequencies are clipped).
4. Single-sided excitation of Rb in a gas cell

\[ a_{fg} = i \pi \frac{1}{2} (1 + \text{sign } \delta) + \int_{\omega_{ig} - \delta}^{\omega_{ig} + \delta} \frac{1}{\omega_{fg} - \omega} d\omega + \int_{\omega_{ig} + \delta}^{\omega_{fg} - \omega + \delta} \frac{1}{\omega_{fg} - \omega} d\omega \]  

(4.3)

\[ = i \pi \frac{1}{2} (1 + \text{sign } \delta) - \int_{\omega_{fg} - 2\omega_{ig} - \delta}^{\omega_{fg} - \omega + \delta} \frac{1}{x} dx \quad (x = \omega - \omega_{ig}) \]  

(4.4)

\[ = i \pi \frac{1}{2} (1 + \text{sign } \delta) - \log \left| \frac{\delta}{\omega_{fg} - \omega_{ig} + \delta} \right|. \]  

(4.5)

The first integral is zero due to perfect destructive interference around \( \omega_{ig} \). This means that a symmetric spectrum around \( \omega_{ig} \) will have complete destructive interference of the nonresonant excitation.

In equation (4.4) we change variables, note that the boundaries change too. The last term shows an enhancement as \( \delta \) approaches zero. This is the expected resonance when the spectrum is clipped at \( \omega_{ig} \), and the destructive interference is reduced due absence of photons pairs with \( \omega < \omega_{ig} \). In the limit when \( \delta \to 0 \), the TPA would go to infinity, which is not very realistic. For this result we assumed that the clipping has an infinite resolution. If this is not the case the peak value will not reach infinity. The actual resolution of our setup is on the order of 0.2 nm, or about 100 GHz.

Scanning \( \delta \), we expect that when the cutoff frequency is far from resonance, or \( \delta >> 0 \), there will only be resonant excitation as the nonresonant excitation interferes destructively. When \( \delta \) approaches 0, there will be a peak in the nonresonant integral. When \( \delta < 0 \), resonant excitation is no longer possible, and when the cutoff frequency approaches \( \omega_{fg}/2 \) no excitation is possible anymore as there are no photon pairs possible anymore.

The experimental results are shown in figure 4.2. To allow for the finite resolution of the shaper the model – which assumes a step function with infinite resolution – has been convolved with a Gaussian of 0.2 nm bandwidth. The experiment confirms the peak enhancement at \( \omega = \omega_{ig} \), and a reduction of TPA signal for pulses with a broader bandwidth caused by destructive interference of photon pairs.

4.4. Nonresonant TPA enhancement by spectral phase shaping

![Figure 4.3. Spectral width separation in a part that is symmetric around \( \omega_{ig} \) and a part that is asymmetric around \( \omega_{ig} \). The symmetrical part with bandwidth \( 2BW \) is indicated in rose, and the asymmetric part with bandwidth \( b \) is indicated in blue. The spectral phase is shown as a green line, with a step frequency at \( \omega_{fg} + \delta \).](image)

In the last section enhancement of TPA was shown by clipping the spectrum at the resonance level, reducing the destructive interference. Even more enhancement of the TPA can also be achieved by spectral phase manipulation. By shaping the pulse in a way that exploits the spectral response of TPA around the resonance, the destructive interference can be turned into constructive interference, enhancing the TPA probability by up to a factor 10. First this spectral phase will be introduced, and an analytical expression for the TPA probability will be derived, and then the model will be compared with the theory.
4. Single-sided excitation of Rb in a gas cell

Destructive interference is caused by the sign change of the weighting function at $\omega = \omega_{ig}$. In the last section this was compensated by clipping a part of the spectrum. Another way to enhance the transition probability is by changing the sign of $E(\omega)$ at $\omega = \omega_{ig}$. Using Euler’s identity, this can be done by employing Pi-step, where the step frequency written in terms of the detuning with respect to $\omega_{ig}$.

$$\Phi(\omega) = \begin{cases} 0 & \omega < \omega_{ig} + \delta \\ \pi & \omega > \omega_{ig} + \delta \end{cases}, \quad E(\omega) = A(\omega)e^{i\Phi(\omega)} = \begin{cases} A(\omega) & \omega < \omega_{ig} + \delta \\ -A(\omega) & \omega > \omega_{ig} + \delta \end{cases} \quad (4.6)$$

Where we once more assume a flat spectral intensity profile, so $A(\omega) = 1$. The step frequency at $\omega_{ig} + \delta$ is scanned.

This experiment has been performed on the Rb $5S \rightarrow 5D$ transition using the $5P_{3/2}$ transition as an intermediate state (resonance level at 795 nm). The difference between $\omega_{fg} - \omega_{ig}$ and $\omega_{ig}$ is about 30 nm. The spectrum is clipped such that there are two boxes of about 10 nm around the resonance level and the complementary resonance level $\omega_{ig}$ and $\omega_{fg} - \omega_{ig}$. Since they are sufficiently far from $\omega_{fg}/2$ we can assume that the contributions for $\omega > \omega_{fg}/2$ are far enough from resonance that they can be discarded. Due to small alignment errors in the clipping the center of the boxes might not be exactly centered at the resonance level. To incorporate this in the model the boxes are split into a box that is symmetric around $\omega_{ig}$ with bandwidth $2BW$, and a box that only extends to higher frequencies, called $b$, see figure 4.3. This distinction is important because the spectral amplitude in $b$ will always give a constant contribution if $|\delta| < BW$. For our experiments $b$ is about 10% of $B$.

![Figure 4.4. Experimental and theoretical results for the two-photon transition using a Pi-step as a spectral phase. The TPA signal is relative to the TPA signal of a transform limited pulse. The enhancement at $\omega = \omega_{ig}$ and $\omega = \omega_{fg} - \omega_{ig}$ can be seen, allowing for an increase of up to a factor 12 in signal compared to a transform limited pulse.](image)

By splitting the nonresonant integral in equation (3.1) at $\omega_{ig} - \delta$ and $\omega_{ig} + \delta$, the integral can be solved. This leads to the following expression:

$$a_{fg} = \pi \text{sign}(-\delta) + \begin{cases} 2\ln \left| \frac{\delta}{BW} \right| + \ln \left| \frac{BW}{BW + \delta} \right| & |\delta| < BW \\ \ln \left| \frac{BW}{BW + \delta} \right| & BW < |\delta| < BW + b \\ \ln \left| \frac{BW}{BW + b} \right| & |\delta| > BW + b \land |\delta| < -BW \end{cases} \quad (4.7)$$
4. Single-sided excitation of Rb in a gas cell

Figure 4.5. \( \pi/2 \) box spectral phase. The spectral phase consists of a box of height ±\( \pi/2 \) with a width of \( \omega_{fg} - \omega_{iq} \), so that it can occupy the region between \( \omega_{iq} \) and \( \omega_{fg} - \omega_{iq} \). The center frequency of the box at \( \omega_c \) can be varied. \( \delta \) is the detuning of the central frequency of the box with respect to \( \omega_{fg}/2 \).

A derivation can be found in section §B of the appendix. This solution is only valid when the step position is below \( \omega_{fg}/2 \). When the step position is above \( \omega_{fg}/2 \) the solution can still be used by mirroring the step position around \( \omega_{fg}/2 \). We see that the resonant part changes sign when the step position crosses \( \omega_{iq} \), but since it is purely imaginary and the nonresonant term is purely real this does not affect the transition probability.

The experimental results are shown in figure 4.4. A peak is visible at \( \omega_{iq} \) and \( \omega_{fg} - \omega_{iq} \). There seems to be a slight decrease of the TPA signal at a step frequency of 392 THz, near \( \omega_{fg} - \omega_{iq} \). This might be due to misalignment of the shaper. The enhancement is much more than for the amplitude clipping because instead of clipping a part of the spectrum the whole spectrum now contributes to the pulse.

4.5. Symmetry breaking with a \( \pi/2 \) box

The last two sections mainly focused on optimizing the nonresonant term of the transition amplitude. No attention had to be paid to the resonant term since it was purely imaginary and the nonresonant term was purely real. In this section interference between resonant and nonresonant paths will be shown. This leads to symmetry breaking with respect to a TPA experiment where the exciting spectrum is far off resonance so resonant contributions can be ignored. In that case the atom is insensitive to a sign change of the spectral phase[10]. In this experiment this symmetry is broken due to interference between the nonresonant and resonant paths. The spectral phase that will be used to demonstrate this effect has been used before by Dudovich et al., but there it was used to optimize the TPA, like the previous section.[9] They did not notice the symmetry breaking.

Until now the nonresonant path and the resonant path were orthogonal in the complex plane. To create interference we will use a spectral phase function called a \( \pi/2 \) phase box. In a region of the width between the resonance frequency and the complementary resonance frequency there will be a constant phase retardation of ±\( \pi/2 \). The central wavelength of the box can be scanned. See figure 4.5. If the center is exactly at \( \omega_{fg}/2 \), the nonresonant term will be enhanced, since all the photon pairs are shifted twice by \( \pi/2 \), hence a \( \pi \) phase shift, just like the previous section. When \( \omega_c \neq \omega_{fg}/2 \), this spectral phase will lead to interference between resonant and nonresonant terms. In this case only one of the terms of the electric field of the nonresonant interaction is shifted by \( \pi/2 \).
4. Single-sided excitation of Rb in a gas cell

\[ \mp E(\omega_{ig})E(\omega_{fg} - \omega_{ig}) = \begin{cases} \mp \pi & \delta \neq 0 \\ -i\pi & \delta = 0 \end{cases} \quad (4.8) \]

Where \( \delta \) is the detuning from the center of the window to \( \omega_{fg}/2 \), and the sign is dependent on the sign of the phase step height. So except for \( \delta = 0 \), the resonant term will be a real value, but the sign is dependent on the sign of the phase step height.

The nonresonant integral will also be affected when the phase is shifted by \( \pi/2 \) for a region. To look at the effect the analytical expression for the transition probability has been derived. Once more the usual approximations are made that the spectral intensity is constant and that the resolution of the shaper is infinite. Furthermore, we assume that the bandwidth of the laser is exactly three times \( \omega_{fi}/2 \), e.g., the bandwidth is three times the frequency difference of the resonant transition with the complementary frequency. For other widths the derivation is possible, but more involved.

\[
a_{fg} = \mp \pi + 2\ln \left| \frac{\delta}{BW} \right| + \ln \left| \frac{2BW - \delta}{BW} \right| - \ln \left| \frac{3BW}{2BW + \delta} \right| \\
\pm i\ln \left| \frac{2BW - \delta}{2BW + \delta} \right|, \quad \text{for } |\delta| < \omega_{fg} - \omega_{ig} \quad (4.9)
\]

This is only valid as long as the center of the phase window is within \( \omega_{ig} \) and \( \omega_{fg} - \omega_{ig} \), and (purely because of the resonant term) as long as \( \delta \neq 0 \). Near resonance, when \( \delta << BW \), the most important contribution is the \( 2\ln \left| \frac{\delta}{BW} \right| \), much bigger than the imaginary last term of the solution. So a slightly detuned phase box will rotate the nonresonant contribution only slightly in the complex plane. Therefore interference between the resonant term and the nonresonant term is possible.

We expect a peak at \( \delta = 0 \). The sign of the phase box step determines whether there will be constructive or destructive interference between the resonant and nonresonant paths, and therefore the width of the peak. When the resonant contribution has the same sign as the nonresonant contribution, the peak will be broader than when the resonant contribution counteracts the nonresonant contribution. If the sign of the phase box step is positive, the nonresonant contribution has the same sign as the resonant contribution. Therefore constructive interference takes place. If the sign of the phase box step is negative, however, the nonresonant contribution has the opposite sign of the resonant contribution, leading to destructive interference.

Our experimental results confirm this model. In figure 4.6 two scans of the TPA signal as a function of the central wavelength of the \( \pi/2 \) box are shown for the Rb \( 5S \to 5D \) transition, one with a positive phase box step, and one with a negative phase box step. The signal is scaled to the TPA signal for a transform limited pulse. The top graph shows a broad central peak, and two small side peaks at \( \omega_{fg} - \omega_{ig} \) and \( \omega_{ig} \), whereas the lower graph shows a very narrow peak at \( \omega_{fg}/2 \), but two big side peaks. The totally different response is caused by either constructive or destructive interference. The width of the central peak for the positive spectral phase can be attributed to imperfect resolution of the phase shaper. The height of the peak agrees well. The reduction of the signal at about 1 THz from resonance in the negative spectral phase can be attributed to the destructive interference once more. In this regime, the sign of the nonresonant contribution is still opposite to the sign of the resonant contribution, but they are of the same order of magnitude, leading to an overall destruction of the transition amplitude. The side peaks are caused by slight errors in the width of the phase box. When one of the edges of the phase box is close to a resonance frequency there will be an enhancement, just like the Pi-step. To incorporate this into the model, equation (3.1) was integrated numerically where the width of the phase window was fitted. The spectral phase width was off by 0.3 nm. This corresponds to one pixel of the SLM.
Figure 4.6. Symmetry breaking between a positive and a negative $\pi/2$ box. See figure 4.5 for the definition of the $\pi/2$ phase box. The central wavelength of the phase window is scanned and the TPA signal is plotted. The top graph is a measurement for a box height $\pi/2$, bottom graph is a measurement with height $-\pi/2$. The difference in the signal is caused by either constructive or destructive interference between the nonresonant path and the resonant path. Because the width of the phase window was not exactly $\omega_{fg} - 2\omega_{ig}$, there are two side peaks visible in the top measurement. They are caused by an imperfect width of the spectral phase window. To incorporate this in the model, the width of the phase window was fitted.
5. Two-pulse experiments with shaped pulses on a resonant two-photon transition

5.1. Introduction

We now extend from our single pulse experiment to a two-pulse experiment. Instead of exciting the atom with a single laser pulse, two identical copies of the same pulse are used to excite the atom. There are two ways to combine two pulses: both pulses can either travel in the same direction (co-propagating pulses) or the pulses can travel in opposite directions (counter-propagating pulses). The use of co-propagating pulses has the advantage of precise control of the delay between the pulses. This chapter will cover experiments with such a setup. The next chapter will be about counter-propagating pulses.

Two co-propagating pulses can be created by employing a Michelson interferometer. The delay between the pulses $T$ can be achieved by controlling the difference between the length of both arms. This is a standard setup for Fourier Spectroscopy, only with femtosecond laser pulses. Scanning the delay time $T$ oscillations of the signal at the characteristic frequencies of the transition are observed. The envelope of the signal can be manipulated by employing an appropriate spectral phase.

Similar experiments have been done on two-photon transitions where the existence of an intermediate state could be ignored because the laser spectrum was far off resonance[12, 13]. Due to the intermediate state the time delay $T$ plays a very different role in our experiment than in previous experiments. Previous work showed a clear distinction between time delays that are longer than the pulse duration and time delays that are shorter than the pulse duration. This distinction will not be possible in this case.

In this chapter we will first explain the experimental setup. The expected signal will be discussed in the theory section, and finally the results will be compared with the theory per spectral phase chosen.

5.2. Experimental setup

For the experiments described in this chapter we need shaped pulse pairs. The shaping is done in the same way as in chapter 4. After the pulse shaper a Michelson interferometer is inserted, to produce two identical pulses with a time delay $T$. The resulting beam is focused in the gas cell, and the TPA signal is measured once more with the PMT. See figure 5.1. The Michelson interferometer consists of a stable mirror and a moveable mirror. The moveable mirror is set on a piezo stage with high precision but a limited travel of 500 $\mu$m. The piezo stage is mounted on a translation stage with a much bigger range (cm), but limited precision. Precise control of $T$ is done with the piezo stage while the range can be extended with the translation stage. See the yellow inset in figure 5.1.

A scan consists of moving one arm of the interferometer at a constant speed over the whole range of interest with the piezo stage. While the piezo stage is moving, the counter is read out as fast as possible. To achieve a high resolution scan, the gate time should be as short as possible, but there should also be sufficient counts within the gate time. Therefore the PMT gain was set as high as possible without saturating the PMT. The advantage of moving the piezo stage at constant velocity is that the measurement time is short. The disadvantage is that the measurement points do not have to be equidistant.
5. FTS with spectrally shaped pulses

Figure 5.1. Experimental setup for the double-pulse experiment. The shaped pulses enter a Michelson interferometer. One arm of the interferometer can be moved on a translation stage (TS). The two copies of the pulse separated by time $T$ enter the Rb vapour cell.

Yellow inset: Schematic of the stacked translation stage. The moveable mirror position can be coarsely adjusted using the Manual translation stage, and the fine control is done using the Piezo stage connected to the computer.

5.3. Theory

Calculating the transition amplitude for two pulses with a time delay $T$ is very similar to calculating the transition amplitude for a single pulse. We can use equation (3.1) once more.

$$a_{fg} = \frac{\pi}{2} E(\omega_{ig}) E(\omega_{fg} - \omega_{ig}) + \int_0^{\infty} d\omega E(\omega) \frac{E(\omega_{fg} - \omega)}{\omega_{fg} - \omega}$$ \hspace{1cm} \text{(5.1)}$$

In this case $E$ is not the electric field for a single laser pulse, but it is the electric field for a pulse pair with a time delay $T$. Such a pulse pair can be written as the sum of two identical copies of a pulse with a time delay $T$.\footnote{In principle it should be half of that, since the generating pulse was split in two. But since the model has to be scaled anyway we can discard the factor 2.} In the frequency domain such a time delay corresponds to a linear phase.

$$E_{dp}(\omega, T) = E(\omega) + E(\omega)e^{i\omega T} = \left(1 + e^{i\omega T}\right) E(\omega).$$ \hspace{1cm} \text{(5.2)}$$

Inserting this into equation (5.1) leads to:

$$a_{fg}(T) \propto \frac{\pi}{2} E(\omega_{ig}) E(\omega_{fg} - \omega_{ig}) \left[ 1 + e^{i\omega_{fg} T} + e^{i(\omega_{fg} - \omega) T} + e^{i\omega T} \right] A$$

$$+ \int_0^{\infty} d\omega E(\omega) \frac{E(\omega_{fg} - \omega)}{\omega_{fg} - \omega} \left[ 1 + e^{i\omega_{fg} T} + e^{i(\omega_{fg} - \omega) T} + e^{i\omega T} \right] B$$ \hspace{1cm} \text{(5.3)}$$

We still see the same structure as in equation (5.1): There is a resonant and a nonresonant excitation path. There is simply an additional factor between the square brackets due to the two pulses. These factors are due to the different combination of pulses that are possible to make a two-photon transition.
The two photons that are needed for the transition can either come from a single pulse or one photon can come from each pulse. The terms marked A correspond to absorbing two photons from the same pulse. The terms marked B correspond to absorbing one atom from both pulses of the pulse pair.

The terms marked A are independent on $\omega$. It is just a time-dependent modulation of the single sided excitation signal. The modulation is caused by the fact that there are two pulses that can excite the two-photon transition, and these transition amplitudes interfere. So from the single-sided excitation constant background and a modulation at $\omega_{fg}$ is expected, but other than that the signal will not show any spatial structure. Part B, on the other hand, corresponds to absorbing one photon from each pulse. Hence it is dependent on $\omega$, and it can be dependent on the spatial structure.

We would like to predict what the frequencies are of the oscillations in the signal when $T$ is scanned. This is a little bit harder than just looking at the previous equation, as we have to bear in mind that $a_{fg}$ is only the transition amplitude. The transition probability is the square modulus of the transition amplitude. As $a_{fg}$ contains trigonometric functions, the frequencies of the transition probability need not be the same as for the transition amplitude. Unfortunately we are unable to take the square modulus of equation (5.3). We can take the square modulus of only the resonant part of the equation. This will not be the same as for the transition amplitude. Unfortunately we are unable to take the square modulus of equation (5.3). We can take the square modulus of only the resonant part of the equation. This will only give some information about the different frequencies that might be encountered, but not about the relative amplitude of the different components. By noting $|z|^2 = z^*z$, for any complex number $z$, we get

$$|E(\omega_{ig})E(\omega_{fg} - \omega_{ig})|^2 = (5.4)$$

So the signal will contain four different frequencies: $\omega_{fg}$, $\omega_{ig}$, $\omega_{fg} - \omega_{ig}$, and $\omega_{fg} - 2\omega_{ig}$. The first three were already visible in equation (5.3). $\omega_{fg} - 2\omega_{ig}$ is also the difference frequency of $\omega_{ig}$ and $\omega_{fg} - \omega_{ig}$.

5.4. Results

A scan of the PMT signal versus the time delay was done for three different spectral phases: A flat phase, a Vshape and a Pi-step. The measurements were done for two transitions in Rubidium: The Rb 5S $\rightarrow$ 5D and the Rb 5S $\rightarrow$ 7S transition. See figure 3.1 for the energy level diagrams. The Rb 5S $\rightarrow$ 7S transition was analyzed because there is no fine structure splitting between s-levels. The Rb 5S $\rightarrow$ 5D transition has an intermediate state that is very close to $\omega_{fg}$. This means that it is easier to see the different frequency components in the time domain. The results will be discussed per spectral phase chosen.

5.4.1. Transform Limited Pulses

Figure 5.2 shows a scan of the delay time with transform limited pulses, therefore a flat spectral phase for the Rb 5S $\rightarrow$ 5D transition and the 5S $\rightarrow$ 7S transition. The measurement and the corresponding simulation are plotted above each other, where blue indicates a simulation and red a measurement. The lower graph shows a detailed part of the Rb 5S $\rightarrow$ 7S measurement. It is not apparent from the signal whether there is temporal overlap of the pulses or not. This is a remarkable difference with previous experiments with an intermediate state far off resonance, where this is visible[13]. Due to the presence of the intermediate state, the delay between the first photon and the second photon can be much larger.

The signal shows oscillations at frequencies $\omega_{fg}$, best visible in the lower right graph. There is a beat signal visible in the 5S $\rightarrow$ 5D measurement. The beat corresponds to the frequency difference between $\omega_{fg}$ and $\omega_{fg} - \omega_{ig}$. In this case these frequencies are very close to each other, therefore the period of the beat signal is large. For the 5S $\rightarrow$ 7S measurement, the difference between $\omega_{fg} - \omega_{ig}$ and $\omega_{ig}$ seventeen times more. The corresponding beat signal is therefore seventeen times faster, which is too fast to be visible in the top graph. The lower right graph shows a zoomed in part of the Rb 5S $\rightarrow$ 7S measurement where the beat signal is visible.
5. FTS with spectrally shaped pulses

Figure 5.2. Fluorescence signal plotted against the delay in picoseconds of the pulse pair for the Rb $5S \rightarrow 5D$ transition (left column) and for the Rb $5S \rightarrow 7S$ transition (right column). The dashed lines indicate the range of a single subscan. The model is plotted on the top row in blue. The experimental data are plotted on the next row in red. The lower left plot is a longer scan of the Rb $5S \rightarrow 5D$ transition. The lower right plot shows a detailed part of Rb $5S \rightarrow 7S$ measurement, showing that also in that case there is a beat signal visible in the envelope.
5. FTS with spectrally shaped pulses

Figure 5.3. Fourier transform of a single subscan of the Rb $5S \rightarrow 7S$ measurement indicated by the dashed rectangle in figure 5.2. The four frequency components at $\omega_{fg}$, $\omega_{fg} - \omega_{ig}$, $\omega_{ig}$ and $\omega_{fg} - 2\omega_{ig}$ are indicated by arrows. The insets show a zoomed in part of the graph around $\omega_{fg} - 2\omega_{ig}$ (left inset) and $\omega_{fg}$ (right inset).

The spectrum of central part of the Rb $5S \rightarrow 5D$ measurement is shown in the lower left three graphs. The spectrum is zero everywhere except for three regions around $\omega_{fg}$, $\omega_{ig}$ and $\omega_{fg} - 2\omega_{ig}$. Only these parts are shown in the graphs. The frequencies at $\omega_{ig}$ ($2\pi \times 384.2$ THz), $\omega_{fg} - \omega_{ig}$ ($2\pi \times 386.3$ THz) and $\omega_{fg}$ ($2\pi \times 770.5$ THz) are clearly visible. This is exactly what was estimated in the theory section. The part of the data that was used to compute the spectrum is indicated by the black dashed rectangle in figure 5.2.

Lastly, there is a peak centered around $T = 0$ in the Rb $5S \rightarrow 7S$ measurement. This peak might be due to residual chirp in the spectral phase. If the pulses that come out of the shaper are not perfectly transform limited, but they have a very small temporal chirp, there will be an enhancement near $T = 0$. Because the difference frequency between $\omega_{ig}$ and $\omega_{fg} - \omega_{ig}$ corresponds to almost 70 nm, the setup is very sensitive even to a small amount of chirp.

5.4.2. Vshape

We now discuss a spectral Vshape with a central frequency at $\omega_{fg}/2$.

$$\Phi(\omega) = \tau |\omega - \omega_{fg}/2|$$

As was discussed in section §2.3.1 this Vshape splits the pulse into two subpulses, one containing all frequencies higher than $\omega_{fg}/2$ and one containing all frequencies below $\omega_{fg}/2$. They are separated by $2\tau$, and the order is determined by the sign of the $\tau$. After such a shaped pulse has passed the interferometer we will have four pulses: Two copies of the shaped pulses separated by time $T$ – called the pulse pair – and these shaped pulses consist of a blue subpulse and a red subpulse separated by time $2\tau$. See also the top figure in figure 5.4.

Because single pulse excitation by a Vshape has not been studied in experimentally in the previous chapter, we will first consider a theoretical single pulse experiment. The transition probability for a Vshape while scanning $\tau$ is shown in figure 5.5. The excitation probability is dependent on the sign of $\tau$. When $\tau << 0$, there is no excitation at all. The sign of $\tau$ determines the order of the subpulses, so
5. FTS with spectrally shaped pulses

Figure 5.4. Graphical representation of the distinction between the situation where the subpulse separation is smaller than the pulse separation ($T > 2 \tau$, top image) and the situation where $T < 2 \tau$, bottom image. In the latter case the first first subpulse from the later pulse comes before or simultaneously with the last subpulse from the first pulse.

Figure 5.5. Single-pulse transition amplitude for a V-shape of varying steepness. The transition probability is scaled to the transition probability of a transform-limited pulse. The yellow insets shows a diagram for the temporal profile of the pulses when $t < 0$ (left inset) and when $t > 0$ (right inset). $R$ and $B$ indicate the subpulse with all the lower resp. all the higher frequencies, showing that the order is reversed for a different sign of $\tau$. On the right the energy level diagram for the Rb $5S \rightarrow 5D$ transition is shown, with a red arrow indicating the lower frequency $\omega_{fg} - \omega_{ig}$ and a blue arrow indicating the higher frequency $\omega_{fg}$. 

Relative intensity [a.u.]

$\tau \begin{cases} < 0 \\
> 0 \end{cases}$

$\tau = 0$

$\omega_{fg}$

$\omega_{ig}$

$\omega_{fg} - \omega_{ig}$

Fluorescence

$|f >, 5D^2 \}$

$5D_{3/2}$

$5D_{1/2}$

$6P_{3/2,1/2}$

$90 \text{ GHz}$
the amount of excitation is dependent on the order of the subpulses. The order is indicated in the graph by the two yellow insets. This is similar to using a quadratic spectral phase (chirp), if we approximate the quadratic spectral phase by two straight lines at $\omega_{ig}$ and $\omega_{fg} - \omega_{ig}$. Other experiments have shown that in this case the sign of the chirp determines the transition probability. [14, 15]

First consider the case where $\tau > 0$. In this case the red subpulse arrives earlier than the blue subpulse. Therefore the electrons are excited to the intermediate state. Then the blue subpulse passes by, exciting the electrons to the final state. This is similar to exciting with upchirp. In the other case, when $\tau < 0$, the blue subpulse arrives earlier than the red subpulse. The blue subpulse is only capable of exciting an electron to the final state when it is already in the intermediate state. As the red subpulse has not passed by yet there are no electrons in the intermediate state, hence there is no excitation possible. This is similar to exciting with downchirp.

Now we turn to the two pulse experiment. Here there is a distinction between the region where the pulse separation is smaller than the subpulse separation ($T < 2 \tau$) and the region where the pulse separation is bigger than the subpulse separation ($T > 2 \tau$). See figure 5.4. The sign of $\tau$ will lead to a very different behaviour in the region where $T < 2 \tau$.

When $\tau < 0$ a single pulse is unable to make the transition as the order of the subpulses is wrong. The only way to excite the atom is by pulse pair excitation: The atom can be excited when the electron is excited to a virtual level close to the resonance level by the last subpulse from the first pulse, and then is excited to the final state by the first subpulse from the last pulse. If single pulse excitation is not possible, the term marked $A$ in equation (5.3) can be discarded. Both subpulses from the first pulse have to pass before the second pulse arrives. Therefore $T$ has to be greater than $2 \tau$ for any excitation to take place. This is confirmed in our measurement, shown in figure 5.6. In the region where $T < 2\tau$, indicated by the dashed lines, there is almost no excitation visible, whereas for the region where $T > 2\tau$, the excitation is visible once more. There beating due to fine structure splitting is visible once more, just like the transform limited pulse. The Rb $5S \rightarrow 7S$ signal decays a little bit less in the central region. This is probably caused by the steepness of the Vshape. The model shows rather strange behaviour when $T > 2.1$ ps. This is due to a numerical overflow encountered because the large value of $T$ leads to a highly oscillatory integrand in equation (5.3).

![Graph 1](image1.png)

(a) $5S \rightarrow 5D$, Negative Vshape ($t = -700$ fs)

![Graph 2](image2.png)

(b) $5S \rightarrow 7S$, Negative Vshape ($t = -700$ fs)

Figure 5.6. Negative Vshape transitions. The dashed lines indicate the subpulse separation. The left column is a measurement of the Rb $5S \rightarrow 5D$ transition, the right column is a measurement of the Rb $5S \rightarrow 7S$ transition. The top row shows the theory, the bottom row the experimental data. In the middle part the signal is much smaller than at the edges. This is because a single pulse of the pulse train cannot excite the atom at all.
5. FTS with spectrally shaped pulses

When \( \tau \) is positive the order of the subpulses is right to make an excitation with a single pulse. When \( T < 2\tau \), it is possible to excite the atom with the first subpulse of the last pulse with the last subpulse of the first pulse. When \( T > 2\tau \), this is no longer possible. Therefore we expect a relative enhancement when \( T < 2\tau \). Furthermore, the average signal should be more than for the negative Vshape. Both of these properties are confirmed in our experiment shown in figure 5.7.

![Graphs showing PMT signal vs. time for different transitions](image)

(a) Rb 5\( S \rightarrow 5D \), Vshape +700 fs
(b) Rb 5\( S \rightarrow 7S \), Vshape +700 fs

Figure 5.7. Scan for a Vshape spectral phase of +700 fs. Left column is for the Rb 5\( S \rightarrow 5D \) transition, right column is for the Rb 5\( S \rightarrow 7S \) transition. The dashed lines indicate the subpulse separation. Between the dashed lines \( T < 2\tau \). A clear enhancement is seen in this region. The PMT signal has been scaled with a factor 0.5 to allow a fair comparison with figure 5.6.

5.4.3. Pi-step

The last spectral phase that will be discussed is a Pi-step. In section §4.4 this has been defined as follows:

\[
\Phi(\omega) = \begin{cases} 
0 & \omega < \omega_{ig} + \delta \\
\pi & \omega > \omega_{ig} + \delta 
\end{cases} \quad (5.6)
\]

Where \( \delta \) is the detuning from resonance. In this experiment, the Pi-step is optimized for maximum signal, to get it as close as possible to \( \delta = 0 \). In the numerical model this is not possible, as the integral for the single-sided excitation would be infinite everywhere. Therefore \( \delta \) was set to 1 Mhz, which is small compared to the resolution of the shaper (on the order of a GHz). We expect an increase of the average signal. This is not shown in the plot as the increase was so much that the gain voltage of the PMT had to be adjusted to prevent damaging the PMT. The measurement is shown in figure 5.8(a). There is a big peak visible in the measurement that is not visible in the simulation. This is due to an assumption that we make about the resolution of the shaper. By taking the spectral phase as a real step function, the resolution of the shaper is implicitly assumed to be infinite. In reality, pixellation in the SLM and the finite focal spot size of the different frequencies in the Fourier plane cause the abrupt transition from 0 to \( \pi \) to be smoothed. Previous spectral phases did not have such an abrupt transition, therefore we did not see this effect so clearly. A better way to describe the spectral phase might be:

\[
\Phi_{dp}(\omega) = \frac{1}{2} \pi \left( 1 + \tanh \left( \frac{\omega - \omega_{ig}}{\Delta \omega} \right) \right) \quad (5.7)
\]

32
There is no physical meaning to the hyperbolic tangent. It is merely a function that exhibits the desired behaviour. There is a rather smooth transition from zero to one for arguments from -2 to 2, and that it looks like \( \text{sign } x \) for larger values. The width is determined by the focal spot size in the Fourier plane. The focal spot size in the Fourier plane is 50µm. This translates to \( \Delta \omega = 7 \cdot 10^{10} \) Hz. Using this spectral phase, we can once more calculate the model. In figure 5.8(b) a new comparison is shown with the theory. In this case the central peak is explained better by the theory. The lower side of the envelope seems to rise for larger values of \( T \) in the model, however. In the experiment this behaviour is not shown. This might be due to the exact function that was used for the spectral phase.

Figure 5.8. Scan for a Pi-step spectral phase. All images are the Rb 5S \( \rightarrow \) 5D transition. The bottom row shows two copies of the same experimental data. The top row shows the model for two versions of the Pi-step. Left image: Abrupt Pi-step. Right image: Smoothed Pi-step. The two models are not to the same scale.
6. Spatial Excitation with counter-propagating Frequency Comb Pulses

6.1. Introduction

This chapter will cover an experiment that is similar to the two-pulse experiment described in the last chapter, but using a different experimental setup. It is able to separate single-pulse excitation and two-pulse excitation. The new setup recovers the frequency comb resolution and it allows for single shot measurements instead of having to scan a stage. Instead of creating two pulses with a time delay using a Michelson interferometer, the laser beam is split and the beams are recombined traveling in opposite directions. This is the same as a two-pulse experiment with a position-dependent time delay between the pulses (figure 6.1): in the center of the gas cell the pulses coincide and the time delay is zero. At a distance $z$ from the center (along the beam), one pulse arrives earlier by $z/c$ and the other one is delayed by $z/c$. Therefore the delay is $T = 2z/c$. As was discussed in the previous chapter a two-pulse signal will be dependent on the time delay between the pulses. In this case, the signal will therefore be dependent on the position. By recording the spatial excitation profile with a position sensitive detector (a camera), it is possible to take a single shot measurement, instead of having to scan a stage.

The photons of the two-pulse excitation are counter-propagating, since both pulses that contribute with a single photon are also counter-propagating. As was discussed in section §3.3.2, counter-propagating pulses lead to a Doppler reduced signal, recovering frequency comb resolution. The two-pulse signal is therefore sensitive to the mode frequencies, making Direct Frequency Comb Spectroscopy (DFCS) possible. What differentiates our setup from a standard DFCS setup is the ability to arbitrarily shape the pulses. In normal DFCS, a lot of mode structure independent background signal is created by single-sided single-pulse excitation of both beams. Using the shaper we are able to control the relative amount of single sided excitation by applying a spectral phase. Unwanted background signals can be suppressed, almost without sacrificing the counter-propagating signal that we’re interested in. This combination of spatial and spectral coherent control has been employed in this lab before to measure the Rb $5S \rightarrow 7S$ transition frequencies with KHz accuracy[16].

In this chapter we will first demonstrate the frequency comb resolution of the two-pulse excitation. Then we take a more coherent control approach and we isolate the two-pulse excitation profile in a way that is independent of the spectral phase and look at the effect of various spectral phases.

6.2. Experimental setup

As was discussed in the introduction, two shaped pulses are needed that counterpropagate through the gas cell. The pulses have to overlap throughout the gas cell, otherwise no excitation is visible for larger $T$ due to misalignment of the beams. To achieve this the beam is split in two arms by a 50% beamsplitter.

![Figure 6.1. Schematic representation of the counter-propagating beam geometry inside the gas cell. Three example positions are indicated by blue dots. In reality the whole gas cell contains atoms so there will be a continuous excitation profile. Two pulses $l$ and $r$ travel in counter-propagating directions. In the center they coincide, so the time delay is 0. At $z = a$, the right pulse arrives earlier by $a/c$, and the left pulse arrives later by $a/c$. Therefore the time delay is $2a/c$. In general $T = \frac{2z}{c}$.](#)
Both arms are focussed into the gas cell, and the length of both arms is kept at the same distance by a retroreflector mounted on a translation stage. The resulting excitation profile can be measured using a PMT or a Camera, with a magnification of 2:1. Control of the mode structure is achieved by locking $f_{\text{rep}}$ to 10 MHz, and locking a multiple of the repetition frequency to a frequency generator that can be computer controlled.

Even though the setup does not look too complicated, it is very sensitive to the alignment. The field of view of the camera is 1.5 mm. Therefore the length of both arms should be equal to within 1.5 mm, while the length of the arms is about 1.1 meter. If the length does not match, no counter-propagating signal will be visible. Furthermore, the two arms have to overlap exactly throughout the gas cell, or there will be no counter-propagating signal visible or the signal will not be symmetric.

![Figure 6.2. Experimental setup for counter-propagating measurements. The spectral phase shaper applies a spectral phase to the pulses (a Vshape in this example). After the shaper, the pulses are split with a 50% beamsplitter and focussed in the gas cell in a counter-propagating fashion. The excitation profile can be measured with a PMT or with a Camera. The repetition frequency can be controlled by the computer. To minimize GVD, the lenses in the beam were later changed to cylindrical mirrors.](image)

### 6.3. Theory: Modelling the spatial excitation profile

As was discussed in the introduction, this experiment is similar to a scan of the double pulse experiment where the position corresponds with a time delay $T = 2z/c$. The excitation profile of a double pulse experiment was already modelled in the previous chapter:

$$
\begin{align*}
\alpha_f(T) & \propto i\pi E(\omega_{fg})E(\omega_{fg} - \omega_{ig}) & \\
& \left[1 + e^{i\omega_{fg}T} + e^{i(\omega_{fg} - \omega_{ig})T} + e^{i\omega_{ig}T}\right]
\end{align*}
+ \int_{0}^{\infty} \frac{E(\omega)E(\omega_{fg} - \omega)}{\omega_{fg} - \omega} & \\
& \left[1 + e^{i\omega_{fg}T} + e^{i(\omega_{fg} - \omega)T} + e^{i\omega T}\right].
\end{align*}

(6.1)
Here A were the contributions with both photons coming from a single pulse. Therefore these photons are co-propagating. This is just the single-sided excitation that was studied before. The signal is therefore dependent on the spectral phase, Doppler broadened, and independent on the position in the gas cell. The rapid fringes at frequency $\omega_{fg}$ are washed out by the distance the atom travels in the 6P state before decaying$^1$. It will be called the co-propagating signal, even though it’s the photons that copropagate, not the TPA signal.

Part B is the two-pulse excitation, where each pulse contributes with a single photon. In this setup the photons are counter-propagating. Therefore this signal is Doppler reduced, and dependent on the mode structure of the laser. Furthermore, this signal is dependent on the time delay between the pulses, hence on the position in the gas cell. We will call this signal the counter-propagating signal. The amount of counter-propagating signal is dependent on the spectral phase chosen, the amount of overlap between the beams and the mode structure.

For these experiments, the counter-propagating signal is much more interesting than the co-propagating signal: It is dependent on the repetition frequency and it is spatially dependent. The counter-propagating signal may be extracted by taking two measurements. For the first measurement the repetition frequency is tuned to be in resonance with the transition, thereby recording the co-propagating signal and the counter-propagating signal. For the next measurement the repetition frequency is tuned such that the modes are out of resonance with the transition, recording only the co-propagating signal. By subtracting these two images only the counter-propagating signal is extracted. This means that the single pulse contributions in equation (6.1) can be neglected. Therefore the excitation profile can then be expressed as

$$a_{fg}(T) \propto \pi E(\omega_{ig})E(\omega_{fg} - \omega_{ig}) \left[ e^{i(\omega_{fg} - \omega_{ig})T} + e^{i\omega_{ig}T} \right] +$$

$$\int_{0}^{\infty} \frac{d\omega}{\omega_{ig} - \omega} E(\omega_{fg} - \omega) \left[ e^{i(\omega_{fg} - \omega)T} + e^{i\omega T} \right].$$

with $T = 2z/c$. To account for the effects of the imaging of the system, the model should be convolved with a Gaussian of the proper width.

### 6.4. Results

First we show that it is possible to separate the co-propagating signal from the counter-propagating signal by scanning the repetition frequency of the frequency comb. Then the effect of various spectral phases are discussed and compared with the two-pulse excitation.

First the separation of counter-propagating and co-propagating signal is shown. Then we show the dependence of the counter-propagating signal on the mode structure of the laser spectrum, and lastly we present a way to separate the co-propagating from the counter-propagating signal that is not dependent on the spectral phase.

#### 6.4.1. Extracting counter-propagating signal

counter-propagating signal can be extracted in two ways. The first method is by choosing a spectral phase that eliminates single sided excitation. This more suitable for spectroscopy, since the co-propagating signal is directly removed, but it requires a certain type of spectral phase. The second method employs the sensitivity of the counter-propagating signal to the mode structure. An image where the laser is off resonance is subtracted from an image where the laser is in resonance. The difference image is the counter-propagating signal. This method does not impose a type of spectral phase, but it is not that suitable for spectroscopy since the single sided signal is not physically removed. First the spectroscopy method will be discussed, and then the coherent control approach.

An example of a spectral phase that eliminates the co-propagating signal while maintaining the counter-propagating signal is a negative Vshape. As was discussed in chapter 5, figure 5.4, the order of

---

$^1$ In principle this signal contains an oscillatory signal with frequency $\omega_{fg}$. Using $T = 2z/c$, this corresponds to an oscillation in the spatial excitation of $cT/\omega_{fg} = 1.2 \mu m$. The lifetime of the 6P state is about 88 ns [17]. During this time, the atom moves about $22 \mu m$. Since this corresponds to roughly 20 wavelengths, the fringes are washed out.
6. Spatial Excitation with counter-propagating Frequency Comb Pulses

Spatial excitation profiles

<table>
<thead>
<tr>
<th>VSP</th>
<th>In resonance</th>
<th>(a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VSP</td>
<td>Off resonance</td>
<td>(b)</td>
</tr>
<tr>
<td>VSP</td>
<td>Difference</td>
<td>(c)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>VSN</th>
<th>In resonance</th>
<th>(d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VSN</td>
<td>Off resonance</td>
<td>(e)</td>
</tr>
<tr>
<td>VSN</td>
<td>Difference</td>
<td>(f)</td>
</tr>
</tbody>
</table>

1 \[ f_{\text{rep}} \rightleftharpoons \omega_{fg} \]

Repetition frequency scan

Figure 6.3. Clockwise from right top to bottom: (a): Spatial excitation profile for negative Vshape, with the laser in resonance with the transition. (b): Spatial excitation profile for negative Vshape with the laser out of resonance with the transition. (c): Difference between (a) and (b). (d, e, f): Same as (a, b, c), but using a positive Vshape. (g): Repetition frequency scan showing the PMT signal for scanning the repetition frequency while employing a negative spectral Vshape. All images are for the Rb 5S \( \rightarrow \) 7S transition.

(a, b, c) Represent one way to extract the counter-propagating signal by computing the difference between an image with counter-propagating signal and an image without counter-propagating signal. (d, e, f) Represent a reduction of the counter-propagating signal by employing a spectral phase that almost eliminates the single-sided excitation. (g) Shows the dependence on the mode spacing of the counter-propagating signal. The repetition frequencies at which the images were acquired are indicated. The small peaks in the signal are caused by Doppler shifted excitation.
6. Spatial Excitation with counter-propagating Frequency Comb Pulses

the subpulses is wrong to excite the transition with a single pulse. The only way to excite the atom is by two-pulse excitation. In the new geometry, this means that there will be no co-propagating signal (= single pulse excitation), but only counter-propagating signal (= two pulse excitation). This signal is dependent on the mode structure. The right top image in figure 6.3 displays a spatial excitation profile where the mode spacing is optimized for signal. The signal shows a clear spatial dependence. The splitting between the two regions of interest is the subpulse separation $2\tau$. The image below, marked 2 shows the same image but minimized for signal. The signal is still spatially dependent. This is due to the fact that all possible transitions are mapped onto one repetition frequency scan. Therefore there may not be a single repetition frequency for which there is no transition probability amplitude. In this case the repetition frequency will have to be changed to a more suitable repetition frequency. The dependence on the repetition frequency is shown in the lower graph, where the repetition frequency is scanned and the position independent TPA signal is recorded. The PMT is very sensitive to the background signal, since it’s field of view is bigger than the field of view of the camera. Image 1 is recorded at the maximum of the PMT signal, while image 2 is recorded at the minimum. The small peaks in the repetition frequency scan are caused by Doppler shifted excitation.

This method is suitable for spectroscopy, since the single sided excitation can be suppressed, but it imposes a spectral phase that suppresses single-sided excitation. Therefore it is not suitable to study the behaviour of the counter-propagating signal due to different spectral phases. To study the behaviour for any spectral phase two images can be acquired, one with maximum counter-propagating signal and one with minimum counter-propagating signal. Both images have the same background due to co-propagating Doppler-broadened signal. The counter-propagating signal can be extracted by computing the difference between the two images. This works even if the image out of resonance still contains some counter-propagating signal, since only the amplitude of the counter-propagating signal depends on the repetition frequency, not the spatial structure. Therefore when the image off resonance still contains counter-propagating signal the contrast will be reduced, but the shape of the excitation profile is not different.

This procedure is shown for a positive Vshape ($t=-700\ \text{fs}$) in the left image column. In this case there is a lot of co-propagating signal, since the order of the subpulses is correct for TPA. In the center there is an additional excitation path available (see section §5.4). Therefore only in the center there will be counter-propagating excitation, which is shown in the figure.

6.4.2. Spatial Excitation profiles

To study the behaviour of the spatial excitation profiles on the spectral phase, the difference image is computed as was discussed in the previous section. The central part of the excitation profiles has been averaged for comparison with the theory. This is plotted directly below the images. The results will once more be compared with theory per spectral phase chosen.

Transform Limited pulse The transform limited excitation profile is shown in figure 6.5(a) and the figure 6.5(d) for the Rb $5S \rightarrow 5D$ excitation by a transform-limited pulse. Acquired by blocking one of the beams. The averaged signal in the vertical direction is shown in the plot directly below. The averaging area is indicated by the two yellow lines. The signal decays towards the edges of the image. This is due to the combined effect of the increased beam diameter towards the edges of the image and the imaging setup. It is fitted with a Gaussian, shown as the blue line. All subsequent models have been multiplied by the same Gaussian function.

Figure 6.4. Single sided excitation profile for Rb $5S \rightarrow 5D$ excitation by a transform-limited pulse. Acquired by blocking one of the beams. The averaged signal in the vertical direction is shown in the plot directly below. The averaging area is indicated by the two yellow lines. The signal decays towards the edges of the image. This is due to the combined effect of the increased beam diameter towards the edges of the image and the imaging setup. It is fitted with a Gaussian, shown as the blue line. All subsequent models have been multiplied by the same Gaussian function.

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6. Spatial Excitation with counter-propagating Frequency Comb Pulses

the imaging of the setup and the focussing of the beams might cause a apparent decay of the signal for positions further out of the center. To check whether this is indeed the cause, an image was acquired with one of the beams blocked. Therefore only single sided excitation is present. As can be seen in figure 6.4 it shows indeed a decay towards the edges. This decay can be modeled with a Gaussian. However the rate of decay does not match the decay rate of the counter-propagating signal. The same decay function is shown in green in figure 6.5(a). The counter-propagating signal clearly decays faster than the decay that can be attributed to the imaging, so the decay is not only caused by imaging and focussing effects.

Another source of the decay might be the beam overlap. If the beams overlap in the center, but they are at a slight angle, the overlap will be less further away from the center. Therefore the signal will decay near the edges of the signal. It is unsure whether this is the real cause of the decay towards the edges, but the signal has been fitted with a second Gaussian to incorporate the decay. This is shown in blue, and it seems to fit reasonably well. Nevertheless, we are unsure what the physical reason for this decay is. The decay is independent on the spectral phase if it is indeed caused by the beam overlap. However, if there is a bit of spatial chirp in the beam coming out of the shaper, or chromatic aberration in the focus, the decay can be dependent on the spectrum that is used, and therefore on the transition that is measured. Therefore the decay time was computed for both the Rb 5S → 7S and the Rb 5S → 5D transition. All subsequent models have been multiplied by this same decay.

Positive Vshape  

The positive Vshape is shown in figure 6.5(b) and figure 6.5(e). The model has been multiplied by the same Gaussian decay function as the Transform limited decay for the corresponding transition. This decay seems to fit quite well. There is a sharp increase in signal in the central region where the delay is less than the subpulse separation (|T| < τ). In the central region there are two counter-propagating excitation paths possible, and outside the central region there is only one path available. Therefore we intuitively expect an increase in signal for small T, which is what we see.

Negative Vshape  

The negative Vshape is shown in figure 6.5(c) and figure 6.5(f). Single sided excitation is suppressed with this spectral phase since the order of the subpulses is wrong. Only for |T| > τ excitation is possible. This is shown by the dip in the center of the image. In the Rb 5S → 7S profile the excitation profile is not perfectly symmetric due to imperfect alignment of the beams.
Figure 6.5. Counter-propagating signal for different spectral phases. Only the counter-propagating signal is shown by computing the difference image between an image with the laser in resonance and an image off resonance with the transition frequency. The central part of the images is averaged in the vertical direction. The theoretical model is shown in blue. It has been convolved with the resolution of the imaging. The theoretical model includes an experimentally observed Gaussian decay for larger $T$, of which the physical origin is not yet known (see text). The decay of the signal due to the combined effect of the imaging in the setup and the Rayleigh length of the focus is shown in figure (a) in green.
We’ve seen a beautiful synthesis of coherent control by spectral phase manipulation and by spectral manipulation of the modes of the laser, especially in the last chapter. The coherent control can aid high resolution spectroscopy by appropriately shaping the pulses, selectively eliminating Doppler-broadened excitation without eliminating the counter-propagating signal. On the other hand, the precise control over the spectrum of the frequency comb can be employed to better understand the response of counter-propagating pulse behaviour for arbitrary spectral phase manipulation.

We’ve seen that the counter-propagating excitation is very similar to a two-pulse experiment. The relative excitation probability between single-pulse and two-pulse excitation can be manipulated with the spectral phase just like the counter-propagating experiment. The spectral control that is available for the counter-propagating measurements is not available for the two-pulse experiment, therefore this feature is a little less apparent. The resolution in terms of pulse delay is by far superior, however.

Lastly, we’ve demonstrated interference between resonant excitation and nonresonant excitation by single sided excitation, making the transition sensitive to the sign of the spectral phase. Furthermore, we’ve demonstrated that the single-sided transition probability can be enhanced by spectral phase manipulation.

Of course, there are still a few open issues. First of all the reason for the decay of the counter-propagating excitation continues to be an open question. The first possibility would be a residual misalignment of the two beams. The setup can be made less sensitive to beam overlap by changing the imaging to a microscope objective. In this case the spectral phases can be made less steep. Furthermore, an attempt can be made to separate the Doppler-shifted excitation from the Doppler-broadened excitation. From the experiments it is clear that a counter-propagating pulse setup is very suitable for high precision spectroscopy. Care must be taken for the case of Rb $5S \rightarrow 5D$ spectroscopy because all hyperfine transitions are mapped onto a single repetition frequency interval. A GHz comb would be preferable, because it has a greater repetition frequency interval, and hence the different transitions can be more easily separated.
Twee-foton absorptie in een atoom is niet zo bekend als absorptie van een enkel foton. Twee-foton absorptie biedt echter veel meer mogelijkheden voor fysische experimenten omdat de twee fotonen op allerlei verschillende manieren kunnen worden aangeboden. Het heeft verwantschap met twee enkelfoton transities die tegelijkertijd of heel kort (± picoseconden) na elkaar plaatsvinden. Het grote verschil is de vrijheid van de fotonenenergie. Bij een enkelfoton transitie moet het foton de juiste energie hebben om de overgang te maken. Bij een twee-foton transitie moet de som van de energie van de fotonen de juiste energie hebben. Dit betekent dat er allerlei verschillende combinaties mogelijk zijn van verschillende energieën van fotonen. Dit biedt extra ruimte voor experimenten.

Om een twee-foton overgang aan te slaan heb je een hoge intensiteit licht nodig, want de twee fotonen moeten op hetzelfde moment op dezelfde plek zijn. Daarvoor gebruiken wij een frequentiekam laser. Dit is een laser die hele korte pulsen maakt (femtoseconden) met steeds precies dezelfde tijd ertussen. Doordat de pulsen zo kort zijn, is het spectrum erg breed. Er worden dus fotonen met veel verschillende frequenties uitgezonden. Als je heel precies naar het spectrum kijkt blijkt het te bestaan uit hele smalle "modes", met een hoge spectrale intensiteit, en er tussen zit niets. De frequentie van de modes zijn bij een frequentiekam heel precies bepaald. Daarom wordt zo'n laser vaak gebruikt voor hoge precisie spectroscopie.

Om twee-foton overgangen te begrijpen worden twee onderzoeksfelden gecombineerd die op het eerste gezicht niets met elkaar te maken lijken te hebben. Het eerste is coherent control. Dit is het aanpassen van de spectrale eigenschappen van de puls om de transitiewaarschijnlijkheid te beïnvloeden. Het wordt meestal gebruikt om fotochemische reacties te sturen, of voor microscopie. Het andere onderzoeksveld is hoge precisie spectroscopie. Dit is het extreem precies meten van overgangsfrequencies van atomen om bijvoorbeeld een mogelijke variatie van fundamentele natuurconstanten te meten. Het heeft erg weinig te maken met het manipuleren van fotochemische reacties.

Bij het masterproject hebben we de twee eigenschappen echter gecombineerd, op twee manieren. Aan de ene kant gebruiken we coherent control technieken om achtergrondsignalen te onderdrukken die normaliter optreden bij frequentiekam spectroscopie. Aan de andere kant hebben we de hoge spectrale resolutie van de frequentiekam gebruikt om verschillende manieren van excitatie aan en uit te zetten, om de coherent control eigenschappen beter te kunnen begrijpen.

Om het effect van spectrale manipulatie goed te begrijpen, hebben we eerst gekeken naar de effecten van spectrale manipulatie van een enkele puls op de overgangswaarschijnlijkheid. Hierna is naar het effect van twee pulsen met een bepaalde tijd er tussen gekeken, en de effecten van spectrale manipulatie van beide pulsen tegelijk als functie van de tijd tussen de pulsen. Uiteindelijk combineren we de kennis van twee-puls excitatie met coherent control in een opstelling met pulsen die tegen elkaar in lopen. De twee fotonen kunnen afkomstig zijn van één puls, enkelpuls excitatie, of iedere puls kan één foton bijdragen, tweepuls excitatie. Dit laatste signaal bevat informatie over de overgangsfrequentie. Een groot gedeelte is echter achtergrondsinaal van enkelpuls excitatie. Coherent control kan hier worden gebruikt om dit achtergrondsinaal te onderdrukken. Andersom kan ook: Coherent control van tweepuls excitatie is interessanter dan coherent control van enkelpuls excitatie. De hoge spectrale resolutie van de frequentiekam kan worden gebruikt om het tweepuls excitatie signaal te isoleren van het enkelpuls signaal.
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Bibliography


Bibliography


A. Aligning the shaper

The key aspect of all experimental setups is the shaper. The shaper is by far the hardest part of the setup to align. A number of papers have been written on how to align the shaper.[1] The main difficulty is that the degrees of freedom are coupled in the shaper. To try to help future prospective experimenters, the procedure that was used to align the shaper is discussed in detail. A copy of figure 2.2 is shown in figure A.1, and the abbreviations used in that figure will be used in the following text.

A.1. Main artefacts due to imperfect shaper alignment

If the shaper is not well aligned, there are a number of artefacts that will occur. The main artefact is spatial chirp. Spatial chirp means that different parts of the outgoing beam will have a different spectrum. When focussing a beam with spatial chirp, the position of the focus will be different for every frequency. This will greatly reduce the signal. Another artefact that will occur is temporal chirp, due to second order dispersion, so that the pulses that come out are stretched a lot. Improper alignment of the telescope C1–C2 will lead to astigmatism that is different for every frequency.

The main difficulty is that all these properties are coupled. For instance, when changing the distance of the gratings to C1 and C2, the height of the beam at C1 will change. When compensating for that, it will mean that all the other components will have to be realigned as well.

A.2. Alignment procedure shaper

Because all the properties of the shaper are coupled, it is very important to start out with a configuration that is as close to the ideal configuration as possible. So make a careful design, and make the vertical angle that B1 has to make as small as possible, to prevent too many artefacts due to the tilting of C1.

The biggest decoupling in the shaper that is possible is the decoupling between the \( \vec{x} \) direction and the \( \vec{y} \) direction (the height). The \( \vec{x} \) direction controls the central frequency of the shaper, and spatial chirp in the \( \vec{x} \) direction, and the \( \vec{y} \) direction controls the height, and therefore the spatial chirp in the \( \vec{y} \) direction.

To align the shaper, the following procedure was used:

1. When starting, select the frequency that will be used as a central frequency with an interference filter. Make sure that the incoming beam is collimated, and that the beam is going horizontal at the entrance. If the beam has to travel via a periscope, make sure that the beam is going straight up, otherwise the polarisation will be different. Put a pinhole at the entrance of the shaper.
2. Mark the center of every optical element. Put G1, C1 and F1 exactly on one line, just like G2, C2 and F2. Start aligning the optical components one by one, making sure that the different optical elements are set at the right distance, and that the central frequency is above the center of the elements. Especially make sure that the distances between the same elements before and after the SLM are exactly equal. Make sure that the beam coming from G1 goes back over the center of C2. Put a pinhole at the exit of the shaper.
3. Remove the interference filter, as it will divert the beam a bit. After aligning the \( \vec{x} \), align the heights in such a way that the height of G1 and G2 are the same, and that the height of C1, C2, F1, and F2 are the same. Make sure that the flat mirrors are exactly vertical.
4. Reinsert the interference filter. Check again that the central frequency goes through exactly the center of the beam.
5. Remove the interference filter. Probably the beam will not be horizontal in the Fourier plane. This will be fixed later. Using an optical spectrum analyzer, check for spatial chirp by putting the fiber in the outcoming beam in a way that in can be controlled both in the \( \vec{x} \) and \( \vec{y} \) direction. If the initial alignment was done properly, there should be quite some spatial chirp in the \( \vec{x} \) direction.
A. Aligning the shaper

Figure A.1. 4f-pulse shaper in folded geometry. The incoming laser beam is dispersed on the first grating (g1). The different frequencies are focussed by a cylindrical mirror (c2) via a flat mirror (f1) into the Fourier plane (fp). In the Fourier plane a 640-pixel spatial light modulator can apply a different retardation to every pixel, and therefore a different retardation to the frequencies in the spectrum (orange box). The shaped light is reconstructed by a mirrored setup (g2, c2, and f2), to form the shaped outgoing pulse. By blocking a part of the beam in the Fourier plane, amplitude clipping is possible. The distance between all active optical elements (g and c, c and fp) is exactly the focal length of c, F.

because the distances are not yet optimized, but there will not be much spatial chirp in the $\mathbf{\hat{y}}$ direction. If the spatial chirp in the $\mathbf{\hat{y}}$ direction is very bad, probably the heights are not exactly equal.

6. Spatial chirp in the $\mathbf{\hat{x}}$ direction is due to the fact that the different colors are not focussed at G2. Use the translation stages of G1 and G2 to correct for the spatial chirp. Mark the initial position of the screws and move G1 and G2 by the same amount every time.

7. Check the height of the beam again, and repeat steps 3 and 6 as necessary.

8. It could be that the lines of the gratings are not at the right angle. To check this, reflect the outgoing beam back into the shaper. If the gratings are not aligned perfectly, there will be an angle between the two beams. Rotate G2 and G1 until the outgoing and the incoming beam overlap at C1. After this realignment, check for spatial chirp in any direction.

9. Attenuate the beam until it is possible to focus the outcoming beam into the imaging chip of a camera. Look at the focus for any astigmatism. Use the translation stages of C1 and C2 to compensate astigmatism, but note that it is only possible to compensate astigmatism in the $\mathbf{\hat{x}}$ direction. Repeat steps 3 through 8 and look again at the focus, repeating as often as necessary.

10. Now check for temporal chirp. Analyze the outgoing beam with an autocorrelator. (It might be a good idea to first align the autocorrelator with the incoming beam, provided that the beam does not have too much spatial chirp.) This is the hard part. Mark the screws of the translation stages that G1 and G2 are on. Rotate them by the same amount until a pulse is visible at the autocorrelator. Of course, the spatial chirp will now become worse. To optimize both spatial and temporal chirp, it is possible to change only G2. Play with both options, moving G1 or G1 and G2, but keep track of the original position where there was a pulse. There are a lot of possibilities to compress the pulse and compensate for spatial chirp, but unless the setup is perfect, the third-order dispersion will create a lot of unwanted pre- and postpulses. After aligning, repeat steps 3 and 4 again.

After this procedure, the alignment should at least be acceptable. In principle, to realign the shaper, it is only necessary to make sure that the bundle goes through the entrance and the exit pinhole, and it should be properly aligned. Be warned, however, that when changing the telescope usually the bundle divergence will also change, and this will mean that the shaper will have to be realigned.
B. Analytical solution of the TPA equation for a Pi-step

For many classes of spectral phases, it is possible to derive an analytical solution for the nonresonant term of the TPA expression. This section shows how to solve for the Pi-step for single sided excitation. A similar approach can be employed for the $\pi/2$ step.

The spectral phase is defined as follows:

$$\Phi(\omega) = \begin{cases} \pi & \omega > \omega_{ig} + \delta \\ 0 & \omega < \omega_{ig} + \delta \end{cases}$$

(B.1)

Because $E(\omega) = A(\omega) \exp(i\Phi(\omega))$, $E(\omega)$ can be reduced to

$$E(\omega) = \begin{cases} 1 & \omega > \omega_{ig} + \delta \\ -1 & \omega < \omega_{ig} + \delta \end{cases}$$

(B.2)

Now we need to solve

$$a_{fg} = \pi E(\omega_{ig}) E(\omega_{fg} - \omega_{ig}) + \int_0^\infty d\omega E(\omega) E(\omega_{fg} - \omega)$$

(B.3)

We’ll make the assumption that we can truncate the integral at $\omega_{fg}/2$. This is not necessary, but it is easier to evaluate and for a typical transition the error is less than 1%. Furthermore, we’ll assume the spectrum described in figure 4.3.

Truncating the integral at $\omega_{fg}/2$, we get:

$$a_{fg} = \pi E(\omega_{ig}) E(\omega_{fg} - \omega_{ig}) + \int_{\omega_{ig} - BW}^{\omega_{ig} + BW} d\omega E(\omega) E(\omega_{fg} - \omega)$$

(B.4)

$$= \int_{\omega_{ig} - \delta}^{\omega_{ig} + \delta} d\omega E(\omega) E(\omega_{fg} - \omega)$$

(B.5)

$$= \int_{\omega_{ig} - \delta}^{\omega_{ig} + \delta} \frac{1}{\omega_{ig} - \omega} d\omega + \int_{\omega_{ig} + \delta}^{\omega_{ig} + BW} \frac{1}{\omega_{ig} - \omega} d\omega$$

(B.6)

$$= \int_{\omega_{ig} - BW}^{\omega_{ig} + BW} d\omega - \int_{\omega_{ig} - \delta}^{\omega_{ig} + \delta} \frac{1}{\omega_{ig} - \omega} d\omega + \int_{\omega_{ig} + \delta}^{\omega_{ig} + BW} \frac{1}{\omega_{ig} - \omega} d\omega$$

(B.7)

The integral around the resonance is zero, as the contributions above $\omega_{ig}$ will exactly cancel the contributions under $\omega = \omega_{ig}$.

This will lead to:

$$a_{fg} = \pi E(\omega) E(\omega_{fg})$$

(B.9)

$$= \int_{\omega_{ig} - \delta}^{\omega_{ig} + \delta} \frac{1}{\omega_{ig} - \omega} d\omega - \int_{\omega_{ig} + \delta}^{\omega_{ig} + BW} \frac{1}{\omega_{ig} - \omega} d\omega$$

(B.10)

$$= 2 \ln BW - 2 \ln |\delta|$$

(B.11)

So

$$a_{fg} = \pi E(\omega) E(\omega_{fg} - \omega) + 2 \ln \left|\frac{BW}{\delta}\right|.$$