CHAPTER 1

Introduction

1.1 Historical

In this thesis, we demonstrate a laser system that can produce amplified shaped pulses. The details of design and construction of this system will be described. The applications of such a pulse shaping system include communications, laser selective chemistry, and biomedical imaging. The application discussed in this work is adiabatic rapid passage, which we introduce in this section.

The concept of controlling a single atomic state once seemed impossible, but in fact it has been demonstrated recently. In atomic force microscopy, researchers can isolate a single atom and place it where they want to. This is evidenced in the famous IBM logo that was spelled out with atoms. Recently, researchers from the same group demonstrated an atomic stadium. In this experiment, they took atoms and arranged them in a circular pattern. The resultant energy level structure created a standing wave pattern that could be observed with the AFM [1, 2].

In this thesis, we will discuss results that, although preliminary, demonstrate another intriguing way to control atoms. If an atom is placed in a very high electric field, such as that produced by an ultrafast laser, the dynamics of the atom will be altered. The shaped laser pulses that the laser pulse shaping system produces will not only generate the necessary electric fields, but they will have the right time dependence to excite the atom in a unique way. To understand the time dependence of the shaped laser pulse, it is necessary to trace the line of thought that led to this experiment.

The Bloch equations [3] describe the response of an atom in a magnetic field to an applied rf (radio frequency) pulse. These equations are essential for understanding the result of NMR (nuclear magnetic resonance) experiments. One particularly interesting solution of the Bloch equations is known as adiabatic rapid passage. In general, if we have a system of dipoles with two energy levels, at most 50% of them can be excited to the upper level. This fact is familiar to those who work with lasers; lasers will generally have three or four levels so that the population is excited to the excited state, and then rapidly falls to a lower state. However, under certain conditions, it will be possible to completely invert a two-level system. One way to describe adiabatic rapid passage is through the dressed state picture. The atoms will start in the lower state, which will slowly change its energy as the external electric field is applied. After the pulse is completed, all of the atoms will now reside in the upper state.

In 1957, R.F. Feynman [4] extended the Bloch equations to show that they actually apply to any two level quantum system. The Bloch equations, which were developed for the case of a dipole in a magnetic field, were shown to be quite general and applicable to a wide range of physical phenomena.

One application of the Bloch equations is to a two-level atomic system. An example of such a system would be the $5S_{1/2} \rightarrow 5P_{1/2}$ transition of Rubidium. Studying such a system is particularly interesting to physicists and chemists because it is so simple, and it is a component of more complex systems such as molecules. If we can advance our understanding and our ability to control such a two-level system, there should be significant ramifications in many diverse areas.

There is an important distinction to be made in the case of optical excitation of a two-level system in an optically dense medium. In the case of NMR, since the RF wavelength is generally longer than the sample size, the beam will not be significantly affected as it propagates. However, in the optical domain, as the beam propagates, it will be reshaped. This reshaping is described in Maxwell's equations for light propagation. The full set of equations is known as the Maxwell-Bloch equations [3], and they must be solved numerically. As the light propagates, it affects the atoms, and the atoms will then affect the light.

Much work has been done in studying the Maxwell-Bloch equations. Work by S.L. McCall and E.L. Hahn [5] predicted an effect known as self-induced transparency (SIT). This refers to the somewhat startling fact that for certain pulse shapes, a pulse will propagate through a medium without being absorbed. Normally, one would expect an atomic system to absorb light at resonance. However, in this case, the atom will reradiate the light in exactly the right way as to leave the pulse unchanged. Creating a pulse with exactly the right shape to demonstrate SIT may be difficult. Fortunately, it is found that regardless of the input pulse shape to a system, as it propagates, the pulse will reshape itself to this SIT shape.

Another interesting study of the Maxwell-Bloch equations was made by W.S. Warren and F.C. Spano [6] in 1988. In this work, it is shown that, for the special case of the hyperbolic secant pulse with hyperbolic tangent frequency sweep (which we abbreviate as the sec pulse), the pulse will not become reshaped as it propagates through the medium.

For the general work of the SIT pulse, the pulse is required to have a special area, where area is defined as the integral of the electric field with respect to time (another parameter often used is the Rabi frequency, which is related to the pulse area by a constant). However, for the case of the sec pulse, the area is not critical. In later work (solving just the Bloch equations), D. Goswami and W.S. Warren [7] showed that the sec pulse would have other useful properties. One property is robustness, which means that it will be able to invert the system over a wide range of pulse areas. Another is frequency selectivity. This refers to the fact that the sec pulse will invert the pulse over a certain range of optical wavelengths, but outside this range, there will be no inversion. In an experiment in 1993, adiabatic rapid passage was demonstrated with chirped pulses [8]. It was shown that the system could be completely inverted.

In this work, we experimentally test the predictions of [6], and study the effect of the sec pulse on the rubidium system. We observed pulse reshaping not predicted by the theory. One important aspect is that our laser linewidth was much broader than the atomic linewidth. The opposite case was studied in [6]. Using the model of [6], we were able to theoretically explain our experimental observations. Our experimental results show many interesting phenomena, including pulse reshaping, evidence of inversion, and the effect of different pulse shapes. This experiment increases our understanding of optical pulse propagation, and the excitation of two level systems.

1.2 General Outline

In this thesis, we describe the development of a femtosecond pulse shaping system [9], and an experiment that was conducted with this system [10].

Chapter 2 discusses technologies that have previously been developed. We will discuss the STRUT [11] (spectrally and time resolved upconversion technique), a technique which is used to give both the phase and amplitude of the pulse. This technique was particularly useful in characterizing amplified shaped pulses. We will give the details of the amplification system [12]. This Ti:sapphire amplification system gives an output of 200μJ/pulse, at a rate of 1kHz. The pulse width is approximately 150fs, the center wavelength is 800nm, and the bandwidth is 10nm. The amplification system consists of a Ti:sapphire laser, a stretcher, a regenerative amplifier pumped by a Nd:YAG laser, and a compressor. We also describe previous work in pulse shaping, which has been investigated with many different approaches [13-18].

In Chapter 3, we describe the construction of the amplified pulse shaping system, and give details on how to construct such a pulse shaping system, including techniques that allow for precise alignment of system parameters such as the grating angles. This pulse shaper has been designed so that it should be easy to insert into an standard pulse amplification system. Such amplification systems are commercially available and used in many research labs.

The experimental data from the pulse shaper demonstrates that we are able to produce high resolution, high contrast pulses. We show modulation in the amplitude domain with over 40 bits of data. We will also show phase modulation, which is detected by the STRUT. This phase modulation allows us to produce complex shaped pulses, such as the secant hyperbolic pulse with hyperbolic tangent frequency sweep. The STRUT data is fit to theoretical curves, showing that we have full control over the pulse shape.

Having developed this amplified pulse shaping system, we next applied it to a two-level system. Chapter 4 will present the Maxwell-Bloch equations [3]. From these equations, phenomena such as self-induced transparency, and adiabatic rapid passage are derived. The secant hyperbolic pulse mentioned above is particularly useful for the adiabatic rapid passage experiment. The physical system that we chose to study was the rubidium atom, which has a transition that can be approximated as a two-level transition at the D_1 line [19]. Shaped pulses were used to excite this atomic transition. Our experimental data show a variety of intriguing results, including stimulated emission, and rapid adiabatic passage. The experimental data consists of pump-probe experiments and pulse reshaping experiments.

Using the Maxwell-Bloch equations, a computer model [6] is used to simulate our data. The theoretical results agree quite well with the experimental results. We think that these initial results will lead to a wide variety of experiments in chemistry and physics, demonstrating quantum control.

1.2 References

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