

Chapter 1

Introduction

The first mode-locked pulse laser was demonstrated in 1964 and produced nanosecond (10^{-9} s) pulses [1]. By 1987, pulses as short as six femtoseconds (6×10^{-15} s) had been achieved [2]. Today, commercial laser systems routinely produce sub-100 fs pulses, and these pulses are finding application in a variety of fields. Because of the high peak intensities of short pulses, femtosecond pulse propagation is a field rich in nonlinear phenomena. The short duration of the pulses have made possible the observation of many light-matter interactions that were inaccessible before the advent of short pulse lasers.

The availability of commercial femtosecond laser systems has led to many interesting and challenging problems involving short pulse propagation. First, there are many challenges associated with generating the pulses themselves. There is a demand for shorter pulses, more intense pulses, and pulses at wavelengths ranging from x-ray to infrared. In addition, designer pulses with a specific temporal shape or specific phase function are desired. Femtosecond pulses are currently finding application in many diverse areas of science and medicine. Propagation issues play a role not only in these current-day scientific endeavors, but will also be important for many as yet unforeseen applications.

One obvious application of short pulses is communications. When sending information packets through a fiber or air, shorter pulses mean higher information den-

sity and therefore faster transmission of the data. The high intensities associated with shorter pulses, however, can lead to nonlinear propagation effects and increased temporal spreading or pulse deformation during transmission. Temporal spreading limits the density of pulses that can be sent down a transmission line. It also lowers the peak intensity of the pulses such that, over a long distance, the signal needs to be amplified periodically. Minimizing pulse spreading minimizes the number of amplification stations needed as well as increases the throughput of the system.

The high intensities associated with short pulses can also benefit the communications industry. High peak intensities allow optical switches based on the nonlinear optical response of a material to be constructed. For example, a simple optical switch can consist of a short control beam, a nonlinear material, and a polarizing beam splitter that is crossed with the polarization of the signal beam. When both the temporally short control beam and the signal beam are present in the nonlinear material, a polarization rotation of the signal occurs, allowing it to proceed through the beam splitter. In this way, the signal beam can be switched on and off.

Another example of an application of femtosecond pulses is the generation of high harmonics of the central pulse frequency by focusing an intense pulse into a gas of atoms. Experiments by Zhou *et al.* have found that the duration of the pulse determines the efficiency of harmonic production and that the wavelength of the harmonics can be tuned by adjusting the chirp of the input pulse [3]. High harmonic generation is of interest as a method of generating short extreme-ultraviolet and soft x-ray pulses at a high repetition rate in the laboratory.

Femtosecond pulses are also finding application in ultrafast pump-probe experiments. The short duration of these pulses allows researchers to probe chemical reactions on a femtosecond time scale. Another area of current interest involves using designer femtosecond pulses to tailor atomic or molecular wavefunctions. The idea is to produce and probe coherent atomic wavepackets in a controlled and understandable

fashion.

Short pulses are also being used in many other areas. Some of these include the study of plasma generation and laser-plasma interactions, probing and treating diseased or damaged biological tissues, and the pursuit of new materials with larger nonlinear responses.

All of these applications of short pulses involve propagation through glass or crystal optics, air, other gases, fibers, or liquids such as laser dyes, solvents, or water. If one wishes to understand or control the interaction of a short pulse with any system, it is necessary to have a well characterized pulse, including knowledge of how that pulse changes as it propagates from the source to the sample to the detector. In addition, the nonlinear interactions of short pulses with materials result in complicated phenomena that are intriguing from the standpoint of basic scientific curiosity.

This thesis addresses the linear and nonlinear propagation of ~ 100 fs pulses in bulk media. Most experimental studies of pulse propagation prior to this work provided information on only one aspect of the electric field before and after propagation, usually the spectral or temporal intensity. Knowledge of only the spectral or temporal intensity, however, is not sufficient to fully elucidate propagation effects. Two pulses for example could have the same spectrum or the same intensity autocorrelation and be differently chirped. Only by knowing the phase of the two pulses can they be distinguished. The development of full-field measurement techniques have made possible investigations of pulse propagation involving accurate knowledge of both the amplitude and the phase of a femtosecond pulse. Our approach, therefore, is to measure the full electric field of a pulse before and after traversing the material of interest and thereby glean valuable information about the complex light-matter interactions involved in the propagation. We accomplish this full-field measurement with the second-harmonic generation form of frequency-resolved optical gating (FROG).

Some background information relevant to short pulses and their propagation is

discussed in Chapter 2. This chapter also provides a brief history of pulse measurement techniques leading up to the development of FROG and subsequent advances in technology.

The femtosecond laser system is described and an overview of the experimental procedure is given in Chapter 3. This chapter also presents a discussion of the FROG technique in general and the polarization gating and second-harmonic generation forms of FROG in detail. Specific emphasis is placed on those issues that are important for using FROG as a high dynamic range measurement technique for measuring broadband pulses. Experimental data illustrating the capabilities of FROG as a propagation technique are also presented.

Propagation in fused silica is investigated in Chapters 4 and 5. Propagation is investigated in both a one-dimensional, plane-wave regime and in a regime where self-focusing leads to complicated spatio-temporal dynamics including pulse-splitting and continuum generation. Experimental results are compared with a model based on the nonlinear Schrödinger equation. At the highest powers investigated, spectral bandwidths after propagation become so large that FROG measurements are no longer practical. In this case, we present observations of continuum behavior as a function of input power and time. This chapter also presents the first near-field spatial-spectral measurements of the continuum. Measurements in both chapters provide insight into the nature of short pulse propagation in fused silica and add significantly to the battery of experimental data available for comparison with advancing theories.

Propagation in liquid media is also investigated, and these results are reported in Chapter 6. Studies in methanol reveal the presence of a non-instantaneous nonlinearity with a very short response time. Studies in water lend credibility to the observed nonlinear response in methanol and provide a measure of the nonlinear index of refraction.

Finally, Chapter 7 provides a summary of the science presented in this thesis and discusses future directions.