

**Studies of Nonlinear Femtosecond Pulse Propagation in
Bulk Materials**

by

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Studies of Nonlinear Femtosecond Pulse Propagation in Bulk Materials

Thesis directed by Professor Stephen R. Leone

Femtosecond pulse lasers are finding widespread application in a variety of fields including medical research, optical switching and communications, plasma formation, high harmonic generation, and wavepacket formation and control. As the number of applications for femtosecond pulses increases, so does the need to fully understand the linear and nonlinear processes involved in propagating these pulses through materials under various conditions. Recent advances in pulse measurement techniques, such as frequency-resolved optical gating (FROG), allow measurement of the full electric field of the pulse and have made detailed investigations of short-pulse propagation effects feasible.

In this thesis, I present detailed experimental studies of my work involving nonlinear propagation of femtosecond pulses in bulk media. Studies of plane-wave propagation in fused silica extend the SHG form of FROG from a simple pulse diagnostic to a useful method of interrogating the nonlinear response of a material.

Studies of nonlinear propagation are also performed in a regime where temporal pulse splitting occurs. Experimental results are compared with a three-dimensional nonlinear Schrödinger equation. This comparison fuels the development of a more complete model for pulse splitting.

Experiments are also performed at peak input powers above those at which pulse splitting is observed. At these higher intensities, a broadband continuum is generated. This work presents a detailed study of continuum behavior and power loss as well as the first near-field spatial-spectral measurements of the generated continuum light. Nonlinear plane-wave propagation of short pulses in liquids is also investigated, and

a non-instantaneous nonlinearity with a surprisingly short response time of 10 fs is observed in methanol. Experiments in water confirm that this effect in methanol is indeed real. Possible explanations for the observed effect are discussed and several are experimentally rejected.

This thesis applies FROG as a powerful tool for science and not just a useful pulse diagnostic technique. Studies of three-dimensional propagation provide an in-depth understanding of the processes involved in femtosecond pulse splitting. In addition, the experimental investigations of continuum generation and pulse propagation in liquids provide new insights into the possible processes involved and should provide a useful comparison for developing theories.

“Truth gains more even by the errors of one who, with due study and preparation, thinks for himself than by the true opinions of those who only hold them because they do not suffer themselves to think.” –John Stuart Mill

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Contents

Chapter	
1	Introduction 1
2	Background 5
2.1	Short Pulses 5
2.2	Short Pulse Propagation 7
2.3	History of Short Pulse Measurement 15
3	Experiment 23
3.1	Laser System 23
3.2	Overview of Experimental Method 32
3.3	FROG 32
3.3.1	Some Practical Issues Concerning FROG 37
3.3.2	Ambiguities Inherent in FROG 38
3.3.3	PG FROG 43
3.3.4	SHG FROG 46
3.3.5	SHG FROG Data 50
4	Propagation in Fused Silica 60
4.1	Measuring the Nonlinear Index of Refraction 61
4.2	(1+1)-Dimensional Propagation 65

4.3	(3+1)-Dimensional Propagation	71
4.3.1	Experimental Results at 800 nm	72
4.3.2	Experimental Results at 1200 nm	76
4.3.3	Theory	80
4.3.4	Results	89
4.4	Summary	94
5	Continuum Generation	96
5.1	On the Verge of Continuum Experiments	103
5.2	Full Continuum Experiments	105
5.2.1	Qualitative Observations	108
5.2.2	Power Throughput as a Function of Intensity	110
5.2.3	Power Throughput as a Function of Time	112
5.2.4	Near-Field Spatial Spectrum	114
5.2.5	Discussion	121
6	One-Dimensional Propagation in Liquids	124
6.1	Methanol Studies	126
6.1.1	Stationary Cell Measurements	126
6.1.2	Flowing Methanol Measurements	131
6.1.3	Discussion	139
6.2	Water Studies	141
6.2.1	Data and Results	142
6.3	Summary	145
7	Summary	147
	Bibliography	153

Figures

Figure

2.1	Schematic diagram of the Stokes and anti-Stokes Raman scattering processes.	11
2.2	Change in frequency due to self-phase modulation	13
3.1	Schematic diagram of the amplified Ti:Sapphire laser system.	24
3.2	A typical pulse from the amplified Ti:sapphire laser system	26
3.3	Beam profile before and after traversing a calcite polarizer	27
3.4	Schematic diagram of the optical parametric amplifier	30
3.5	Normalized spectra of pulses from the OPA	30
3.6	Temporal intensity and phase of a pulse from the OPA	31
3.7	PG FROG traces of differently chirped pulses	40
3.8	Schematic diagram of PG FROG	44
3.9	Schematic diagram of SHG FROG	47
3.10	A measured SHG FROG trace and a FROG trace calculated from the corresponding retrieved field.	51
3.11	Measured and retrieved frequency and time marginals	52
3.12	A comparison of the measured and retrieved spectra for the pulse in Fig. 3.10	52

3.13	An example of the agreement between an independently measured autocorrelation of a pulse and the corresponding measured and reconstructed SHG FROG delay marginals.	54
3.14	A demonstration of the repeatability of SHG FROG measurements . . .	55
3.15	Phase matching efficiency curve for 50 μm BBO.	56
3.16	A demonstration of the retrieval of a temporally complicated pulse . . .	58
3.17	A comparison of measured and retrieved spectra corresponding to the FROG traces in Fig. 3.16	59
4.1	Temporal intensity and phase of a pulse after one-dimensional propagation in fused silica	64
4.2	Temporal field of an intense femtosecond pulse at propagation distances in fused silica of $z = 0, 1.27, \text{ and } 2.54 \text{ cm}$	67
4.3	Measured and calculated FWHM of the intensities of the fields presented in Fig. 4.2	69
4.4	Measured temporal and spectral fields before and after propagation through fused silica as a function of peak input power	73
4.5	Measured FROG traces and spectra after propagation of chirped input pulses	75
4.6	Temporal and spectral fields after propagation of chirped input pulses .	77
4.7	Measured fields before and after propagation at 1200 nm	79
4.8	Calculated intensity surface plots for a pulse after traversing a fused silica sample	81
4.9	Calculated fields after traversing a fused silica sample	83
4.10	Measured spatial FWHM of beam size as a function of input power . . .	85
4.11	A comparison of the measured and calculated fields after propagation .	90
4.12	A comparison of near- and far-field theory with the far-field measurement	92

4.13	A comparison of the measured and calculated spectra corresponding to the data in Fig. 4.12	93
5.1	Far-field image of continuum generated in sapphire.	99
5.2	Measured and calculated temporal fields after propagation at input powers on the verge of continuum generation	104
5.3	A comparison of measured and calculated spectra in the case of near-continuum generation	106
5.4	The percentage of power lost during propagation as a function of input power	111
5.5	Near-field spatial spectrum	116
5.6	On axis spectrum corresponding to Fig. 5.5.	117
5.7	Lineouts of spectral intensity from a spatial-spectral measurement of continuum	118
5.8	More lineouts of spectral intensity from a spatial-spectral measurement of continuum	119
5.9	On-axis spectrum of continuum	120
6.1	Temporal intensity and phase of a pulse prior to and after traversing a 1 cm sample of methanol	128
6.2	Calculated fields assuming an instantaneous and a non-instantaneous nonlinearity	130
6.3	Measured intensity and phase of a pulse before and after traversing 1.69 cm of methanol	132
6.4	Measured spectra before and after traversing 1.69 cm of methanol	134
6.5	Measured and calculated intensity and phase assuming three different values of τ	136

6.6	The difference between measured and calculated phase for the three calculations presented in Fig. 6.5	137
6.7	Measured and calculated spectra assuming $n_2 = 4 \times 10^{-16} \text{ cm}^2/\text{W}$ and $\tau = 10 \text{ fs}$	138
6.8	Measured intensity and phase along with calculated intensity and phase assuming Nibbering's reported values	140
6.9	Measured intensity and phase prior to and after propagation through 1.69 cm of water	143
6.10	A comparison of the measured and calculated fields after propagation through 1.69 cm of water	144

Tables

Table

3.1	Comparison of some common variations of FROG.	42
6.1	Measured and literature values of n_2 for water.	142