

**STUDY OF THE NONLINEAR
PROPAGATION OF FEMTOSECOND
LASER PULSES**

Thesis by

Martin Centurion

In Partial Fulfillment of the Requirements for the

Degree of

Doctor of Philosophy



CALIFORNIA INSTITUTE OF TECHNOLOGY

Pasadena, California

2005

(Defended May 11, 2005)

© 2005

Martin Centurion

All Rights Reserved

ACKNOWLEDGEMENTS

In my five years as a student at Caltech I have learned a great deal about both science and life. The journey has been as rewarding as challenging. I am very grateful to my advisor Demetri Psaltis for the many lessons he taught me about optics, research and life as a scientist. I will always be indebted to him for accepting me in his group (even though I did not know the first thing about optics), where I have greatly benefited from his creativity and vision. His trust and support have been instrumental in my accomplishments.

I would like to thank my mentor in the lab and good friend, Zhiwen Liu. It was a pleasure and a great opportunity to work with such a brilliant mind, who also had the patience to teach me. I am indebted to George Panotopoulos and Jose Mumbru for their guidance during my first years. George was an endless source of judicious advice and a dear friend. Jose was a reference on experimental skills and also provided a touch of humor when it was most needed. Hung-te Hsieh was my companion in the bumpy road to graduation, it was refreshing to share an office with him as he always brought plenty of excitement. Very special thanks to Lucinda Acosta for the administrative support, but more importantly for her moral support and friendship. She provided comfort in the times of frustration and helped me maintain a positive attitude.

I greatly benefited from collaborating with Ye Pu, a very good experimentalist and electronics wizard with whom I shared countless hours in the lab, Mankei Tsang, who helped me with the theoretical work, and Karsten Buse, who gave me excellent comments and advice during his visits from Germany. For their help and support, I would also like to thank Yayun Liu, David Erickson, Chris Moser, Greg Steckman, Greg Billock, Wenhui Liu, Manos Fittrakis, Todd Meyrath, Zhenyu Li, Hua Long, Jim Adleman, Eric Ostby, David McKeen and Bayiang Li. My friends, both here and at home have allowed me to keep a healthy balance between work and play.

Caltech was also the place where I met my beautiful wife Lucia, who became my muse and endless source of happiness. Her love is truly a blessing. My older brother has been a positive influence in my life, and I know he will always be there for me. All of my family has supported me in this adventure and surrounded me with their affection. Finally, I would like to dedicate this work to my father and my mother. Every success is a direct consequence of their influence in my life and their love.

ABSTRACT

This work presents a comprehensive study of the propagation of femtosecond pulses and the formation and evolution of spatial solitons. The first half (Chapters 2-3) is devoted to the implementation of a novel ultrafast holographic system to capture the nonlinear propagation of laser pulses with femtosecond resolution. Femtosecond pulses are used to record holograms of the ultrafast changes in the material properties. Amplitude and phase changes of the laser beam inside the medium are reconstructed numerically. The strength of the nonlinear material response and the density of free electrons can be recovered from the phase information in the hologram. A single hologram can be captured with fine spatial resolution, or a time-sequence of holograms can be captured in a single shot with reduced spatial resolution. We have observed dramatic differences in the light propagation depending on the material properties.

The second part of the thesis (Chapters 4-5) covers the formation and evolution of spatial solitons in a Kerr medium. We have measured the evolution of the beam profile as a function of pulse energy and propagation length. The optical beam breaks up into a pattern of connected lines (constellation) and self-focused spots (solitons). The solitons self-focus to a minimum diameter and release their excess energy through conical emission, which in turn overlaps with the background constellation and seeds the formation of new solitons. The solitons also show a collective self-organizing behavior caused by their mutual interactions. The evolution of 1-D arrays of solitons was captured using Femtosecond Time-resolved Optical Polarigraphy, a technique that measures the transient birefringence

induced by the pulses in the medium. When the array was generated in an unstable configuration, the solitons re-arranged themselves into an array with a (larger) more stable period. A transition to a chaotic state is observed when the input power is increased above a threshold level. A time-averaged pulse propagation equation was used to numerically solve for evolution of the beam. There was good agreement between the experimental results and the computer simulation.

TABLE OF CONTENTS

Acknowledgements	iii
Abstract	v
Table of Contents	vii
List of Figures	x
List of Video Clips	xiii
1. Introduction	1
1.1. Motivation and background	1
1.1.1. Research with femtosecond laser pulses.....	1
1.1.2. Ultrafast cameras	2
1.1.2. Visualization of the propagation of femtosecond pulses.....	3
1.2. Laser system and diagnostics	4
1.3. Numerical simulations.....	7
1.4. Thesis outline.....	9
References	10
2. Analysis of on-axis holography with femtosecond pulses	11
2.1. Capture of nonlinear pulse propagation with pulsed-holography	11
2.2. Induced nonlinear index changes.....	12
2.2.1. Kerr effect.....	12
2.2.2. Index change due to plasma generation.....	15
2.3. Interaction and overlap of femtosecond pulses.....	16
2.4. Digital recording and reconstruction of on-axis holograms.....	19
2.4.1. On-axis holograms and the twin image problem.....	19
2.4.2. Numerical reconstruction for small objects.....	21

2.5. Removal of the twin image.	23
2.6. Reconstruction of index changes.	30
2.7. Resolution limits of femtosecond holography.	30
References	34
3. Holographic capture of femtosecond pulse propagation	36
3.1. Introduction.....	36
3.2. Experimental setup.	37
3.2.1. Single-frame capture.	37
3.2.2. Multiple-frame capture (holographic movie).	38
3.3. Experimental results with single-frame capture.	40
3.3.1. Digital recording and background subtraction	40
3.3.2. Pulse propagation in air, water and CS2.....	41
3.3.3. Comparison of the holographic phase reconstruction with an interferometric phase measurement	48
3.3.4. Reconstruction of 3-D information.....	50
3.4. Experimental results with multiple-frame (movie) setup.....	51
3.4.1. Recording spatially multiplexed holograms.	51
3.4.2. Propagation dynamics in liquids: water and CS2.....	53
3.4.3. Pulse propagation in LiNbO ₃	56
References	61
4. Dynamics of filament formation in a Kerr medium	63
4.1. Introduction.....	63
4.2. Experimental setup.	64
4.3. Constellation formation and filamentation.	66
4.3.1. Experimental results.	66
4.3.2. Numerical results and discussion.	70
4.4. Phase transition in the rate of filamentation as a function of input energy.....	75
4.5. Conical emission, filament spatial splitting and fusion.....	77

4.5.1. Experimental results	77
4.5.2. Numerical results and discussion.....	78
4.6. Summary.....	82
References	83
5. Self-organization of spatial solitons	86
5.1. Introduction.....	86
5.2. Experimental setup.	87
5.3. Experimental results.	89
5.3.1. Beam profile and instabilities as a function of pulse energy.....	89
5.3.2. Spatial evolution of the beam and self-organization.	93
5.3.3. Periodic arrays of filaments.....	98
5.4. Numerical simulations.....	100
5.5. Summary.....	105
References	106
Appendix A: Derivation of the nonlinear Schrodinger equation	108

LIST OF FIGURES

1.1. Schematic diagram of the laser system.....	5
1.2. Screen capture of the FROG trace and reconstruction of the pulses from the seed laser (Mira).	6
1.3. Screen capture of the FROG trace and reconstruction of the pulses from the laser amplifier (TSA).....	7
2.1. Sketch of holographic capture.....	16
2.2. Overlap region for two ultrashort pulses.	18
2.3. Recording and reconstruction of on-axis holograms.....	21
2.4. Simulation of phase reconstruction from on-axis holograms.	23
2.5. Simulation of object reconstruction.....	27
2.6. Amplitude and phase reconstructions with iterative algorithm.	28
2.7. Cross sectional plots of simulated object, initial reconstruction and reconstruction after applying the correction algorithm.	28
2.8. Phase reconstruction of an experimentally recorded hologram.	29
2.9. Resolution limit.	33
3.1. Experimental setup for single hologram recording.	38
3.2. Experimental setup for holographic movie recording.	39
3.3. Background subtraction to remove artifacts due to spatial noise on the optical beam.....	40
3.4. Laser induced discharge in air.	43
3.5. Amplitude and phase reconstructions of femtosecond pulse propagation in liquids.	47
3.6. Formation of bubbles and hot spots in water.....	48
3.7. Comparison of on-axis reconstruction with interferometric phase measurement.	49
3.8. Three depth slices of the object reconstructed from a single hologram.	50

3.9. Time sequence of the pulse propagation in water in a single frame of the CCD camera.	53
3.10. Holographic amplitude and phase reconstructions of femtosecond pulse propagation in water.	55
3.11. Holographic amplitude and phase reconstructions of femtosecond pulse propagation in CS_2	56
3.12. Holographic amplitude and phase reconstructions of femtosecond pulse propagation in a LiNbO_3 crystal.	58
3.13. Shot to shot differences in the spatial pattern of a pulse in a LiNbO_3 crystal.	59
4.1. Experimental setup.	65
4.2. Beam profile and 2-D FFT before and after traversing 10 mm of CS_2	67
4.3. Beam breakup and filamentation as a function of propagation distance and energy.	69
4.4. Comparison of numerical and experimental results.	73
4.5. Numerical simulation of beam propagation.	74
4.6. Filamentation statistics as a function of input pulse energy.	76
4.7. Conical emission and seeding of new filaments.	76
4.8. Behavior of individual filaments.	78
4.9. Simulation of the propagation of a single filament over 10 mm.	80
4.10. Simulated filament propagation in the presence of a noisy background.	80
5.1. FTOP setup.	87
5.2. Beam profile of the pump pulse at the output of the CS_2 cell.	89
5.3. Beam profile inside the cell as a function of power.	92
5.4. Pulse trajectories and 1-D Fourier transforms.	96
5.5. Cross sectional plots of filament formation for $P = 390 P_{\text{cr}}$	97
5.6. Interactions between filaments from 3.5 mm to 4.2 mm from the cell entrance for an input pulse power of $1200 P_{\text{cr}}$	98

5.7. Propagation of a pulse with a periodic beam profile.....	99
5.8. Formation of a 2-D array of filaments.....	100
5.9. Comparison of experimental and numerical results of beam profile as a function of pulse energy.....	103
5.10. 1-D Fourier transforms for numerically calculated beam propagation.	104

LIST OF VIDEO CLIPS

5.1. Movie of changes in the beam profile as a result of fluctuations in the pulse energy for $P = 170 P_{\text{cr}}$	91
5.2. Movie of changes in the beam profile as a result of fluctuations in the pulse energy for $P = 390 P_{\text{cr}}$	91
5.3. Pulse propagation inside CS_2 from 2 mm to 4 mm from the cell entrance for a pulse power of $390 P_{\text{cr}}$	94