

## Chapter 7

### Summary

Understanding the propagation of short pulses in a variety of media is of great importance to a number of applications spanning many fields of interest. By applying a full-field measurement technique to the study of short pulse propagation in several interesting materials and regimes, this work lends insight into the complicated combination of linear and nonlinear processes involved. This work also demonstrates the second-harmonic generation (SHG) form of frequency-resolved optical gating (FROG) as a powerful technique for resolving complicated propagation issues in the field of ultrafast science.

Experiments involving plane-wave propagation in fused silica reveal surprisingly large changes in temporal pulse width over relatively short propagation distances. These experiments show a broadening that is nearly 20 times larger than that predicted by group velocity dispersion (GVD) alone. These experiments, along with calculations based on a one-dimensional nonlinear Schrödinger equation (NLSE), illustrate that the effects of GVD and self-phase modulation (SPM) are interrelated and should not be treated separately as is commonly seen in the literature. This thesis presents the first full-field measurements detailing this interplay between GVD and SPM. The very good agreement between the experiment and the model validates FROG as a valuable technique for investigating short pulse propagation.

Propagation is also studied in a regime where conditions are such that the pulse

undergoes self-focusing. Self-focusing leads to much higher intensities and associated higher-order nonlinear effects that result in temporal pulse splitting. At lower input powers, a pulse splits into two sub-pulses, each shorter in duration than the parent pulse. With increasing input powers, multiple sub-pulses are observed. The overall phase across the entire split field indicates upchirp, with the leading pulse shifted to the red of the trailing pulse in frequency. The phase of the input pulse is also found to affect the propagation. An upchirped input gives a head-start to the propagation induced upchirp and results in split pulses that are broader in both the spectral and temporal domains. A downchirped input pulse, on the other hand, tends to offset the propagation induced upchirp and results in split pulses that are significantly narrower in both the temporal and spectral domains.

In this three-dimensional propagation regime, transverse changes to the beam can no longer be neglected, and the model must take into account propagation-induced spatial changes to the beam. A simple three-dimensional model based on the NLSE, however, is not sufficient to describe the observed pulse splitting. The observed splitting exhibits asymmetry in both intensity and phase that the simple model cannot reproduce. Observation of these asymmetries led to the development of a model including the effects of a Raman nonlinearity, linear and nonlinear shock, and third-order dispersion. The Raman nonlinearity preferentially amplifies red-shifted frequencies within the pulse and, with GVD, enhances the leading pulse of the split pair. Both linear and nonlinear shock enhance the trailing pulse during the pulse-splitting process. The very short-temporal duration trailing pulse then diffracts faster than the leading pulse and thus appears less intense at the exit face of the sample. The effects of the Raman nonlinearity are not sufficient to completely offset the effects of shock in the material, however it does temper them. Third-order dispersion also contributes to a larger leading pulse by increasing the dispersion of the blue components. It is, however, a smaller magnitude effect and plays a lesser role in determining the symmetry of the split pulses

than either Raman or shock. The paraxial assumption is still valid with this level of self-focusing.

This modified NLSE predicts asymmetric pulse splitting, however, it does not predict the multiply-split pulses observed in the experiment. In order to explain the multiple splittings, we have found that we must take into account propagation to the far-field. In fact, we have found that multiple splittings like those observed in experiment only occur in the far-field. These multiple pulses do not arise from subsequent splitting of the original split field, but rather result from a build-up of phase in the far-field. In general, pulse splitting occurs first in the far-field and then proceeds to the near-field. When fully split pulses are resolved in the near-field, the far-field is multiply-split. The modified NLSE including propagation to the far-field is in excellent agreement with the experiment at 800 nm for all of the peak input powers investigated.

We have also investigated propagation at higher powers where a continuum spectrum is generated. Continuum spectra have been used in parametric amplification, short pulse generation, and time-resolved spectroscopy and show potential for use in many evolving applications. Our studies near the power threshold for continuum generation indicate that the split temporal field begins to coalesce with the production of more spectral components. Above the threshold for continuum generation, full-field FROG measurements are no longer practical. In this regime, we present observations of continuum behavior as a function of both input power and elapsed time. The overall loss of power during the continuum generation process is an indication that multi-photon absorption occurs in the sample. The dependence of the continuum and power losses on the elapsed time indicates the presence of a saturable process and probable material damage. This thesis also presents the first near-field spatial-spectral measurements of the continuum spectrum. Comparisons with the model used in the pulse splitting experiments, modified to include four-photon absorption, reveal spectral asymmetries and initial power losses similar to those observed in the experiment. This theory, however,

is still not adequate to describe all aspects of the experimental data. These results uncover several of the basic processes involved in the generation of spectral continua and should provide a useful comparison for further theoretical development.

Propagation in liquid samples is also investigated in a one-dimensional propagation regime with the goal of providing a measurement of the nonlinear index of refraction. These measurements are of interest because an accurate determination of the nonlinear index is of particular importance in forecasting propagation effects. Measurements were performed in both methanol, a common organic solvent, and water which is of interest to medical and biological sciences. We find that full-field measurement techniques are ideally suited to these kinds of measurements, assuming one takes into account group velocity dispersion and limits the propagation to the plane-wave regime. Studies in methanol reveal contributions to the nonlinear index of refraction by a non-instantaneous nonlinearity with a surprisingly short response time of 10 fs. The exact mechanism responsible for this non-instantaneous nonlinearity is unknown. Electrostriction, molecular reorientation, molecular redistribution, rotation and libration should all occur on timescales longer than 10 fs and are therefore not responsible for the observed nonlinear response. The nonlinear index of refraction of water, on the other hand, results purely from the instantaneous electronic response. The nonlinear index of refraction of water is found to be in good agreement with previous literature values. Measurements in water thus validate both the observed non-instantaneous nonlinearity in methanol and the measurement technique.

The studies in this thesis elucidate important aspects of linear and nonlinear propagation under a variety of conditions. The science presented here is not only interesting from a fundamental standpoint, but it should also find applicability in a wide variety of fields. This work emphasizes the advantages of using a full-field measurement technique for investigating short pulse propagation and demonstrates the suitability of the SHG form of FROG for this purpose. These studies also exemplify the mutually ben-

eficial relationship that is possible between experiment and theory. These studies lend insight into several interesting and complicated short pulse propagation related issues and should have a bearing on other endeavors involving short, high-intensity pulses.

Of course, the experiments presented here suggest many opportunities for future investigations, some of which include measurements of the full spatial field of a propagating pulse, three-dimensional propagation in liquids, and studies of pulse splitting in materials that do not produce continuum. As discussed in Chapter 4, the pulse splitting experiments involve the spatial aspects of the field during propagation. It would thus be interesting to measure not only the on-axis field as we have done, but also to include a measure of spatial intensity and phase during propagation as well. A means of accomplishing this measurement is the technique of spectral interferometry.

Spectral interferometry consists of interfering an unknown pulse with a known reference pulse and recording the entire interferogram with a CCD [161, 40, 162]. A few requirements in spectral interferometry are as follows; there must exist a coherent phase relationship between the two pulses, the spectrum of the reference pulse must be greater than that of the pulse to be measured, and the reference pulse must be fully characterized. The reference pulse can be characterized using FROG. A single laser beam can be split to give both of the pulses thus ensuring a coherent phase relationship. The second pulse becomes “unknown” by traversing the sample of interest. In short pulse propagation, however, one must be careful either to limit the effects of self phase modulation in the sample or to spectrally broaden the reference pulse via SPM prior to its interaction with the unknown pulse to ensure that the second requirement is satisfied. The full field, including one spatial dimension, of the unknown pulse can be retrieved from the spectral interferogram.

We have performed spectral interferometry in a linear propagation regime and have accurately recovered the value of dispersion of BK-7 glass along the entire spatial dimension of the beam. We have also performed one preliminary experiment involving

nonlinear propagation and were able to observe the expected spatially dependent nonlinear index of refraction. If one could accurately determine the full field of a continuum reference pulse, spectral interferometry could be an invaluable tool for gathering much-needed information about the spatial phase of a pulse during continuum generation.

It would also be interesting to study three-dimensional propagation in liquids. We know empirically and from the literature that a continuum is generated in water. But, do the extra degrees of freedom afforded a liquid affect the continuum generation process? And if so, in what way? And, what effect does local boiling have on the observed continuum?

Investigating the literature tells us that the band gap of the material plays a role in determining the spectral broadening observed in continuum generation. There exist materials with a band gap that is too small for continuum to be generated [135]. It would be interesting to investigate pulse splitting in these materials at input peak powers above that where multiple splitting is first observed. In the absence of spectral superbroadening, it is improbable that the same pulse coalescence would be observed. Another experiment would also be interesting to perform. In materials with the smallest possible band gap above the threshold for continuum generation, where continuum would occur at lower input powers, a situation might arise where pulse-splitting never occurs. By investigating these extreme cases, one could potentially learn more about the processes involved.

In any case, propagation of short pulses should continue to present many fascinating and challenging problems to scientists for years to come.