Ultrashort Laser Pulse Propagation in Water

George W. Kattawar and Alexei V. Sokolov
Dept. of Physics
Texas A&M University
College Station, TX 77843-4242
phone: (979) 845-1180 fax: (979) 845-2590 email: kattawar@tamu.edu
phone: (979) 845-7733 fax: (979) 845-2590 email sokol@physics.tamu.edu

Award Number: N000140810037
http://people.physics.tamu.edu/trouble/work.html

LONG-TERM GOALS

The major objectives of this proposal are twofold. The first objective is to perform both an experimental and theoretical study of the factors affecting the propagation distance behavior of ultrashort (femtosecond) laser pulses in water. This study will be conducted in the so-called linear regime which involves laser intensities that are below the threshold where nonlinear effects set in. A fundamental problem, which will be resolved by this research, is whether or not the temporal width and spacing of short pulses affects its absorption spectrum. If one simply knows the spectral composition of the input pulse and the absorption spectrum of the water, is this sufficient to predict the temporal evolution of the pulse? If such is not the case, then we will have to consider the way the system responds to pulse widths and pulse spacing which are short compared to vibrational relaxation times in water.

The second phase of the research will be to explore the nonlinear regime where dramatic changes to the temporal, spatial, and spectral properties of the medium occur. The primary processes being self-focusing and self-phase modulation due to the Kerr effect (also called the quadratic electro-optic effect which was discovered by John Kerr in 1875). Self-focusing can lead to an enormous increase in the peak intensity where long filaments can occur and in some cases lead to supercontinuum generation or "white light" generation first discovered in 1970.

OBJECTIVES

The propagation of an ultrashort pulse of light through a linear and absorptive medium such as water, is of great fundamental importance for several reasons. One of the most important of which is that it may be possible to transmit information over much greater distances using ultrashort pulses compared to propagation distances achieved by using pulses with long time durations, including CW (continuous waves). The first prediction of optical precursors was given by both Sommerfeld and Brillouin[1] in 1914 using an asymptotic method now called steepest descent. Their analysis was based on a step-modulated field propagating through a Lorentz dielectric which is nothing more than a collection of damped harmonic oscillators. Later refinements to their conclusions were made by Oughstun and Sherman[2]. The first measurements which claimed to observe optical precursors in deionized water were made by Choi and Österberg[3] where they found that the precursors were attenuated non-exponentially with distance. What was extremely significant about this work was that they found the

Report Documentation Page

Form Approved
OMB No. 0704-0188

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED
2010		**00-00-2010 to 00-00-2010**

4. TITLE AND SUBTITLE	5a. CONTRACT NUMBER
Ultrashort Laser Pulse Propagation in Water	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **Texas A&M University,Dept. of Physics,College Station,TX,77843-4242**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			
unclassified	**unclassified**	**unclassified**	**Same as Report (SAR)**	**16**	

pulse energy detected at a distance of 5 m in water with a 60 fs pulse was one hundred times greater than the signal obtained using a 900 fs pulse (having the same fundamental wavelength) which had the usual Lambert-Beer behavior (It should be noted that the so called Lambert-Beer law was first discovered by Pierre Bouguer in his published work in 1729 titled *Essai d'optique sur la gradation de la lumiére* and we will hereafter refer to it as the Bouguer-Lambert-Beer Law or BLB. The law is valid for monochromatic sources but unfortunately researchers have mistakenly applied it to pulse propagation where the bandwidth covers changes in the absorption coefficient). Their conclusions were brought into serious doubt by Alfano[4] et al. who claimed that their bandwidth was not wide enough to cross the 760 nm absorption band of water and therefore that their conclusions were questionable.. In a later paper, Fox and Österberg[5] found deviation from BLB when they used pulses with temporal widths of 60 fs and a repetition rate of 1 kHz where they observed more than two orders of magnitude less absorption after propagation through 6 m of water when compared to BLB. They also found that BLB was not violated for pulses with varying bandwidth with temporal widths of 900 fs (pulses centered at 800 nm) and repetition rates of 80 MHz. Li et al[6] have performed a more recent measurement for liquid water and they found deviations from the BLB law after the pulse had propagated only 1.5 m and the deviation continued to increase out to 3.5 m which was as far as they could detect the signal. The shortest pulse they used was 10 fs with a repetition rate of 75 MHz which gives a pulse spacing of 13.3 ns. They showed that deviation from BLB behavior only occurred when the pulse bandwidth was of the order of 100 nm but for shorter bandwidths (< 30 nm) BLB behavior was verified. It is important to note that this pulse spacing is much longer than the vibrational relaxation times in liquid water which are typically of the order of a picosecond. The most recent experiments by Okawachi et al.[7] used four different laser systems centered at 800 and 1530 nm to show the dependence of pulse duration and repetition rate on pulse absorption. Their bandwidths ranged from 20-60 nm and they showed that BLB behavior was obeyed in every case. It is now clear that there is a real enigma associated with the entire phenomena of ultrashort laser propagation in water.

APPROACH

Experimental:

There are three basic improvements over what has been done in the past that we are proposing:

(a) Improved input pulse shapes

The laser system that we will use in this experiment is the one we currently use for femtosecond CARS spectroscopy of dipicolinic acid and bacterial endospores[8-11]. We have an amplified femtosecond laser (Mira + Legend, Coherent) whereby we obtain up to 1 mJ per 35 fs pulse (at 1 kHz repetition rate) at the fundamental 800 nm wavelength. We can control the chirp (positive or negative) of these 800 nm pulses over a wide range (increasing the pulse duration by up to two orders of magnitude, to over 3 ps, while keeping the spectrum unchanged) very easily, and very precisely by adjusting our pulse compressor. Thus we will be able to study propagation of laser pulses of different length but having identical spectra. Fig. 1 shows the experimentally measured pulse shapes obtained by this method and measured by our home-made variation of frequency-resolved optical gating (FROG).

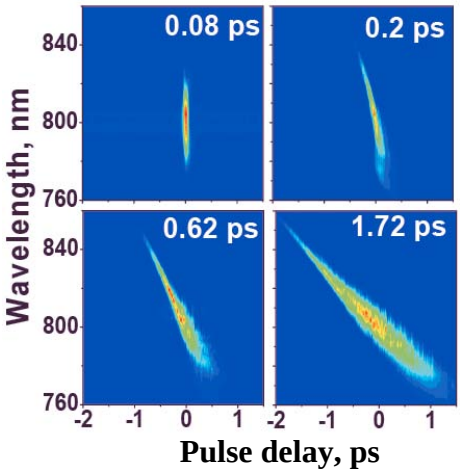

Fig. 1. Chirped pulses obtained by adjusting the pulse compressor and measured by a modified frequency-resolved optical gating technique. The numbers in the upper right corner of each plot give the pulse duration; the spectra of all four pulses are identical.

Alternatively, we can modify the pulse spectral intensities while keeping the spectral phases fixed. Our home-made pulse shaper allows us to make the spectral width up to 100 times narrower and the pulse length correspondingly longer without introducing any chirp. Moreover, our pulse shaper allows us to fine-tune the precise frequency of these narrow-band pulses. We will use such spectrally-modified pulses for the detailed study of light propagation through water. In addition, we have the capability to produce some more complex pulse shapes, should it become necessary or interesting. Our recent paper[12] gives a colorful illustration of potential possibilities, and shows an example of the use of pulse shaping for coherent Raman spectroscopy. More complex pulse shapes will be particularly important for the studies of nonlinear pulse propagation. In the second year of the project, we plan to purchase and use a commercially available computer-programmable pulse shaper for fast (and potentially adaptive) shaping of the laser pulses propagating through water.

Further into the project, we will expand our work into other wavelength regions, and use our two computer-controlled optical parametric amplifiers (OPerA, Coherent), pumped by our amplified femtosecond laser system (described above). Signal and idler pulses obtained from the two OPAs can be frequency-doubled or mixed with the fundamental to produce up to 20 μJ per 50 fs pulse at tunable visible wavelengths. In addition, we may use un-amplified light from the fs oscillator (at 85 MHz), for fast-repetition-rate low-peak-power experiments.

While the previous experimental work on optical precursors suffered from limited choice of laser sources, our laser system will allow great flexibility and precise control over the pulse shapes, for a broad range of well-controlled measurements.

(b) Spectrally-resolved highly-sensitive measurement at the output

We will measure the transmitted spectral intensities (and not just the total output power). We will use a state-of-the-art CCD-equipped spectrometer which will count photons at each wavelength separately. A second CCD-equipped near-infrared spectrometer will allow us to extend our overall working wavelength range. What has been used in many of the past experiments were photomultiplier tubes (PMT's). A PMT is a "bucket" detector that just counts the total number of photons and does not

provide spectral resolution (unless it is combined with a spectrometer, in which case a tedious scanning is needed). PMT's are virtually unsurpassed in sensitivity; however, a good cooled CCD can be almost as sensitive. Highly sensitive signal detection over a large dynamic range is essential for the success of the proposed project, since the intensity of light transmitted through many meters of water will vary over many orders of magnitude as a function of propagation length. Our group has a great deal of experience in performing measurements where the signal varies up to over nine decades. As an example, we reproduce here a figure[13] [Fig. 2] which shows signal varying over seven orders of magnitude. That particular set of data was obtained without spectral resolution. Currently we are capable of achieving similar or even higher sensitivity and dynamic range with spectral resolution, which is afforded by the use of a spectrometer equipped with a cooled CCD. Our measurements on pulse propagation through water will be done with spectral resolution.

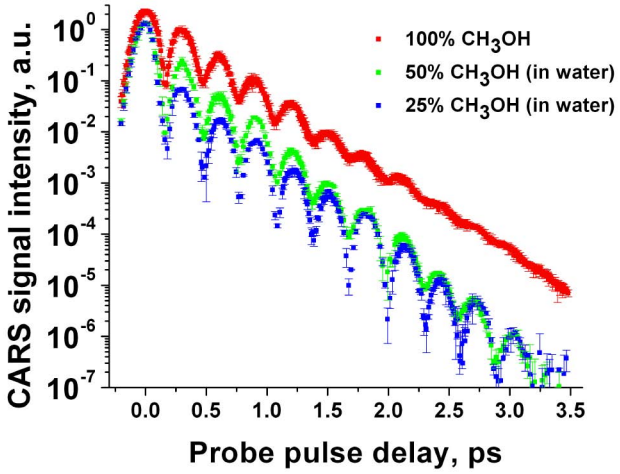

Fig. 2. An example from our previous work, where we used three ultrashort pulses at different wavelength (810 nm, 660 nm, and 575 nm) and measured femtosecond coherent anti-Stokes Raman scattering (CARS) signal (measured in methanol-water solutions) varying in magnitude over many decades[15].

At a further stage of the project we plan to measure the transmitted pulse shapes. For relatively small propagation distances (several meters) we will use a commercial autocorrelator (presently available in our lab), and a FROG apparatus (that we intend to purchase). For large propagation distances this measurement will become challenging since the output power will get very low. For the low-power measurements, we will set up a cross-correlation measurement, such as the one that has produced pictures shown in Fig. 1. Another good option for direct pulse shape measurement is an ultrafast streak camera. We have recently installed and tested such a state-of-the-art streak-camera (Hamamatsu) in our lab, and we will use it to measure the duration of pulses propagated through over 10 meters of water.

(c) Water cell design

We have designed and already constructed our own water cell. Our focus in designing the new cell was to minimize the number of mirror reflections, and to be able to increase or decrease the path lengths continuously and not just in steps of 0.5 meters.

In previous experiments, the maximum path length through the water was 6 meters. In our water cell, we will be able to run experiments at a maximum path length of 18 meters. A schematic of a simplified experimental set up is provided in Fig. 3. We currently have the equipment installed (as shown) and can have the complete setup within a short period of time.

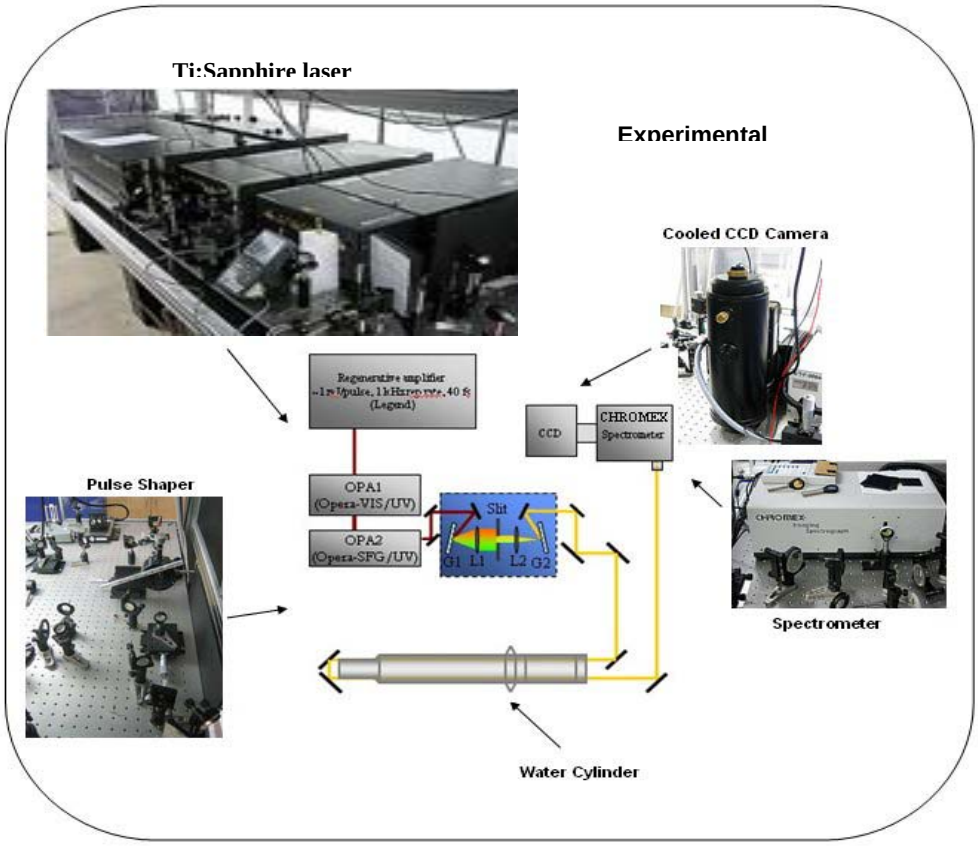

Fig. 3. Experimental setup schematics.

Theoretical:

In order to be able to model the complete pulse propagation process, we must be able to handle the actual pulse shape that will be used in our experiments along with the best data we have on the actual absorption spectra of pure water. In fact the water we will use will be prepared by the same method as in the absorption measurements done by Pope and Fry[14]. Since we have a great deal of experience with the finite difference time domain (FDTD) method, we will have to substantially rewrite parts of it to handle the actual dispersion of the refractive index of water, particularly around the overtones of the OH-stretch/scissor coupled modes. The regions around the sharp slope changes in refractive index are precisely the regions where the interesting precursors will appear. This type of approach has never been performed but is the *sine qua non* for successful modeling. The basis of our method is to take the two Maxwell curl equations in Rationalized MKSA units and assuming no currents; namely,

$$\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t}$$
$$\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \qquad (1)$$

and then discretize them over a spatial grid. We also need the relation between the displacement current and the electric field; namely, $\vec{D} = \varepsilon(\omega)\vec{E}$ where $\varepsilon(\omega)$ is the frequency dependent permittivity and is related to the refractive index *n* by $n^2 = \varepsilon(\omega)$. In the many papers we have published using the FDTD method to solve scattering from irregularly shaped particles (see references 15-26), we have worked only at a single frequency since the incoming radiation was assumed to be monochromatic. Now when using short pulses, there are a plethora of frequencies involved and so then we must be able to use the actual dispersion relation for the medium involved. In the case of water in the near IR (our pulses are centered around 800 nm) we will use the actual measured absorption data of either Kuo or Segelstein in order to apply the FDTD method to this problem, we need to convert the frequency dependent permittivity to a time dependent one. In particular, the temporal characteristic of the water permittivity should be modeled around the overtones of the OH-stretch/scissor coupled modes. These are the regions where the sharp changes in the slope of the absorption coefficient, denoted by α, occur. Another very interesting aspect of water occurs when it is deuterated to form either HDO or D_2O. These forms will have radically different absorption spectra and we can further test our analyses by studying these different forms of water.

If we denote the refractive index of water by $n=n_R-in_I$ and the permittivity by $\varepsilon=\varepsilon_R-i\varepsilon_I$, where the subscripts R and I denote real and imaginary parts respectively, then it is easy to show that $\varepsilon_R = n_R^2 - n_I^2$ and $\varepsilon_I = 2n_R n_I$. The absorption coefficient α is related to n_I by $\alpha = 4\pi n_I / \lambda_{vac}$ where λ_{vac} denotes the wavelength of the radiation in vacuo.

In order to get the time dependence of water permittivity, we must Fourier transform $\varepsilon(\omega)$. Actually it is more physical to Fourier transform $\varepsilon(\omega)$-1 and to obtain the susceptibility kernel $G(\tau)$ defined by (see Jackson[27]):

$$G(\tau) = \frac{1}{2\pi}\int_{-\infty}^{\infty}[\varepsilon(\omega)-1]e^{-i\omega\tau}d\omega \qquad (3)$$

then

$$\begin{aligned}\vec{D}(\vec{x},t) &= \vec{E}(\vec{x},t) + \int_{-\infty}^{\infty} G(\tau)\vec{E}(\vec{x},t-\tau)d\tau \\ &= \vec{E}(\vec{x},t) + \sum_{\tau=0}^{t} G(\tau)\vec{E}(\vec{x},t-\tau)\Delta\tau\end{aligned} \qquad (4)$$

Thus we see that the displacement current at a certain instant in time, depends on the electric field from earlier times. It should also be noted that if ε is independent of frequency, then Eq. (4) reduces to $\vec{D} = \varepsilon\vec{E}$ which is what our present FDTD code now uses.

Once we have established the fact that the FDTD method is a viable one for solving this problem, then we would like to next explore the possibility of using the pseudospectral time domain (PSTD) method which we feel will run much faster than the conventional FDTD method. We have tested this method on some canonical scattering problems and found substantial reductions in computation time when compared to the conventional FDTD method. With this method we will actually be able to monitor the complete temporal evolution of the pulse shape.

WORK COMPLETED

a) We have demonstrated, for the first time, precursor-like propagation through a bulk medium composed of an organic dye solution. This is an important first step toward precursor formation and observation in various other bulk media.

b) We simulated the propagation of a reconstructed electric field through the above organic dye solution to demonstrate the formation of precursor features; theoretical simulations and experimental results were in good agreement.

c) In order to characterize the optical pulses involved in precursor formation, we have developed a novel and accurate pulse retrieval algorithm. This algorithm is a hybrid between the popular graduated optimization algorithm (GOA) and the genetic algorithm (GA) approach; it is able to characterize sharp perturbations in the spectral phase of propagating pulses more effectively than other popular methods, and so is ideal in describing the propagation of optical precursors through optically dispersive media.

d) We have demonstrated, for the first time, energy coupling between optical filament-forming pulsed laser beams in bulk media. This demonstrates that filament formation and propagation can be controlled after the initial formation of the filament and has important consequences in atmospheric and oceanic remote sensing, communications, and spectroscopy.

RESULTS

We first present the results obtained from observation of precursor propagation through a bulk medium composed of an organic dye. The dye itself (NP800, Exciton) was selected for its sharp absorption resonance in the near infrared portion of the electromagnetic spectrum, with the peak absorption occurring around 800 nm with a full width at half maximum (FWHM) of about 100 nm. The calculated refractive index of the dye, obtained via the Kramers-Kronig relationship, shows strong anomalous dispersion in the region around 800 nm. This strongly peaked absorption and anomalous dispersion are requirements for precursor formation.

The dye was dissolved in 1-1-2-2-tetrachloroethane, with a final dye concentration of 10 g/L. A thin layer of the solution was sandwiched between two microscope cover slips, each about 190 um in thickness; surface tension held the dye solution between the slips, and the whole was placed in the path of the beam. The laser pulses were generated by a femtosecond laser system (Rainbow, Femtolasers) and had a central wavelength of 800 nm, a temporal duration of 9 fs FWHM, and pulse energy of 4 nJ. The low energy ensures that our experiments were completed within the linear regime of the light-matter interaction.

We first measured the autocorrelation trace of the initial pulse; comparison with the autocorrelation pulse after propagation through the organic dye solution thus provided sufficient information for the

identification of precursor formation. Reconstruction of the pulse electric field from the autocorrelation traces was completed with the hybrid genetic algorithm described below; the initial electric field and the electric field after propagation through the organic dye solution are shown in Figs. 4 and 5, respectively.

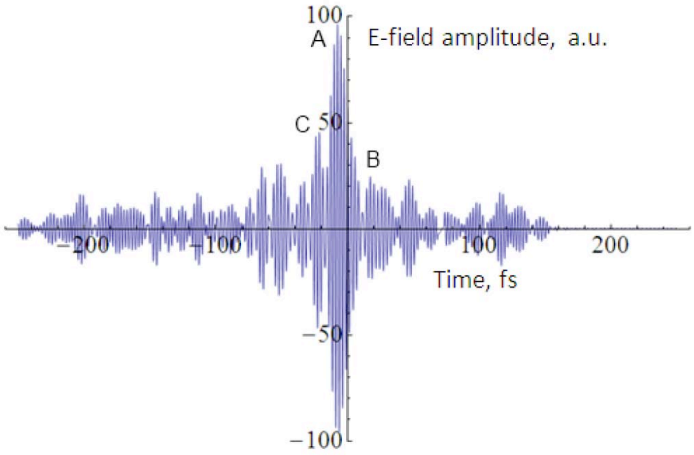

Fig. 4 The reconstructed electric field of the initial femtosecond laser pulse as provided by the hybrid genetic algorithm. Reference peaks A, B, and C are labeled.

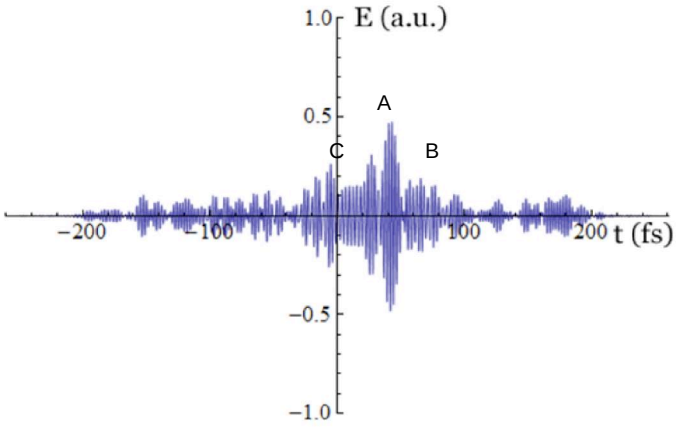

Fig. 5 The reconstructed electric field of the femtosecond laser pulse after propagation through the organic dye solution as provided by the hybrid genetic algorithm. Reference peaks A, B, and C are labeled. Notice that magnitude of the peaks have attenuated at differing rates as a result of propagation through the dye solution.

The simulated autocorrelation traces using the reconstructed electric fields were in good agreement with the measured traces. Theoretical numercial simulations propagating the intitial electric field were then completed via the generalized pulse propagation equation to visualize the shape of pulse features

as a function of propagation distance. The peak magnitude of the pulse features labeled as A, B, and C in Figs 4 and 5 were tracked over propagation distance *x* to the thickness of the organic dye solution *d*. The results are shown in Fig. 6. One can see the differential attenuation of these pulse features as a function of propagation distance. Such behavior is characteristic of precursors.

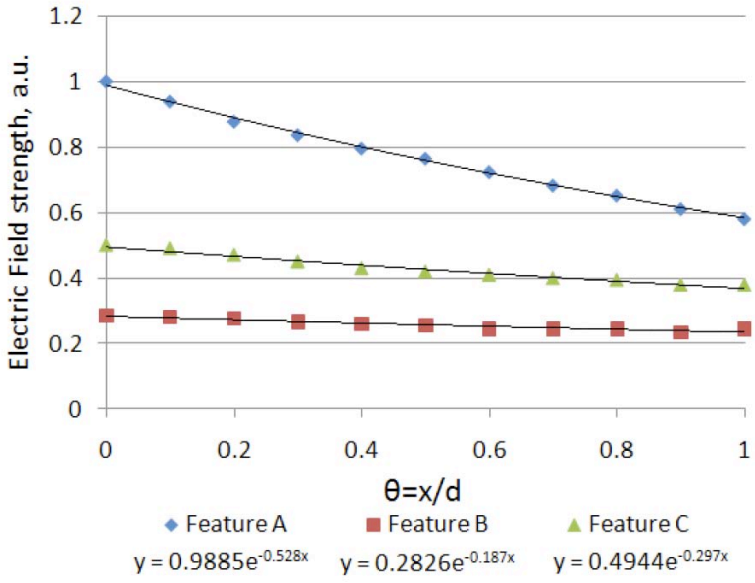

Fig. 6 Simulated attenuation behavior of pulse features A, B, and C from Figs. 1 and 2 over the thickness of the organic dye solution. Notice the varying attenuation coefficients; such behavior is characteristic of precursor propagation.

Understanding of precursor formation and propagation in bulk media is important in realizing sophisticated methods of utilizing pulsed laser beams for communication and remote sensing. In addition, precursors themselves may perhaps find applications in characterization of material properties.

We now briefly present the hybrid genetic algorithm developed to retrieve the above electric fields from the autocorrelation traces.

The graduate optimization algorithm (GOA) was able to scan the chirp parameters of the spectral phase of ultrashort pulses. Combined with the measured spectral intensity, spectral phase can be coarsely retrieved by matching retrieved and measured second harmonic interferometric autocorrelation (SHIAC) within a calculation time of seven seconds.

The genetic algorithm (GA) was then incorporated into the retrieval and employed to make fine adjustments of both individual spectral phases and the chirped parameters to further reduce the difference between retrieved and measured SHIAC. The calculation took several minutes or more than ten minutes depending on how many iterations were used in the calculation.

Fig. 7 shows the GOA+GA combined algorithm retrieved a spectral phase that matches with measured SHIAC perfectly. (a) shows the retrieved SHIAC from GOA only. (b) shows the retrieved SHIAC from GOA+GA. There is a perfect match between retrieved and measured SHIAC. (c) shows the retrieved phase from GOA (blue curve) and GOA+GA (red curve). (d) shows the evolution of rms errors between retrieved and measured SHIAC.

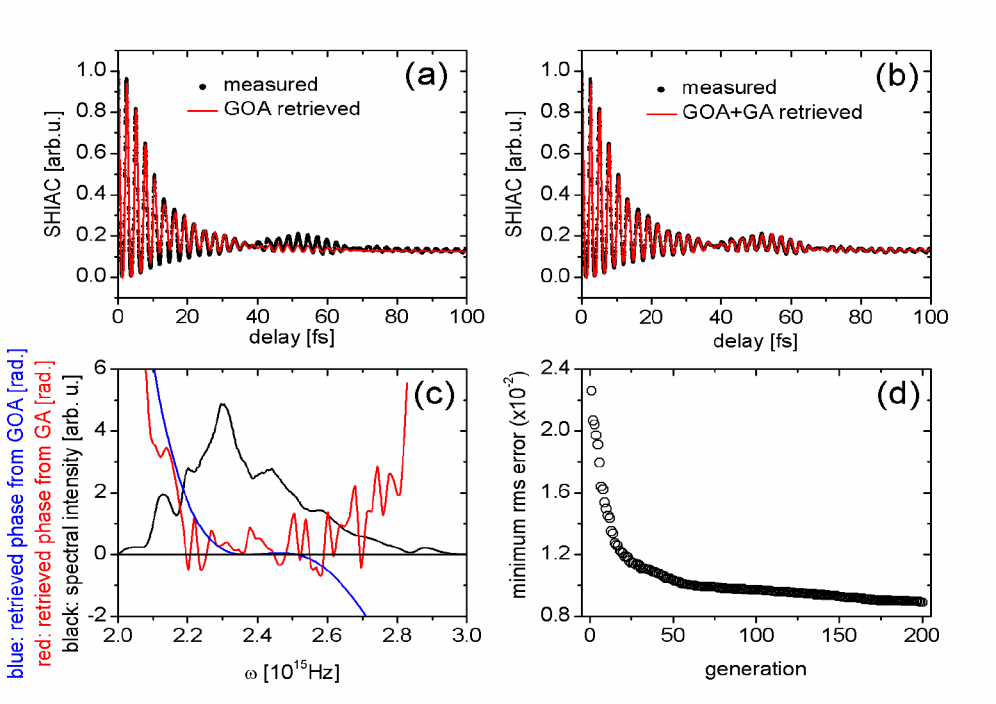

Fig 7. (a) Retrieved SHIAC from GOA only. (b) Retrieved SHIAC from GOA+GA. (c) Retrieved phase from GOA (blue curve) and GOA+GA (red curve). (d) Evolution of rms errors between retrieved and measured SHIAC.

Fourier transform of the above spectrum with corresponding spectral phase yields the pulse shown in Fig. 8.

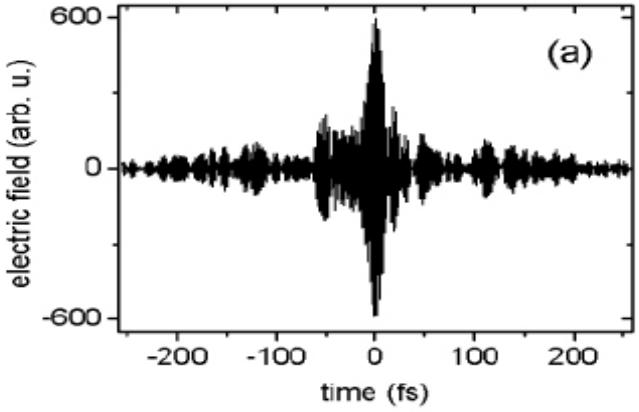

Fig. 8 Retrieved electric field from the hybrid genetic algorithm shown in Fig. 7.

GOA+GA also works well with more complex pulse shapes, as shown in Fig. 9. (a) shows the retrieved SHIAC from GOA only. (b) shows the retrieved SHIAC from GOA+GA. There is a very good match between retrieved and measured SHIAC. (c) shows the retrieved phase from GOA (blue curve) and GOA+GA (red curve). (d) shows the evolution of rms errors between retrieved and measured SHIAC.

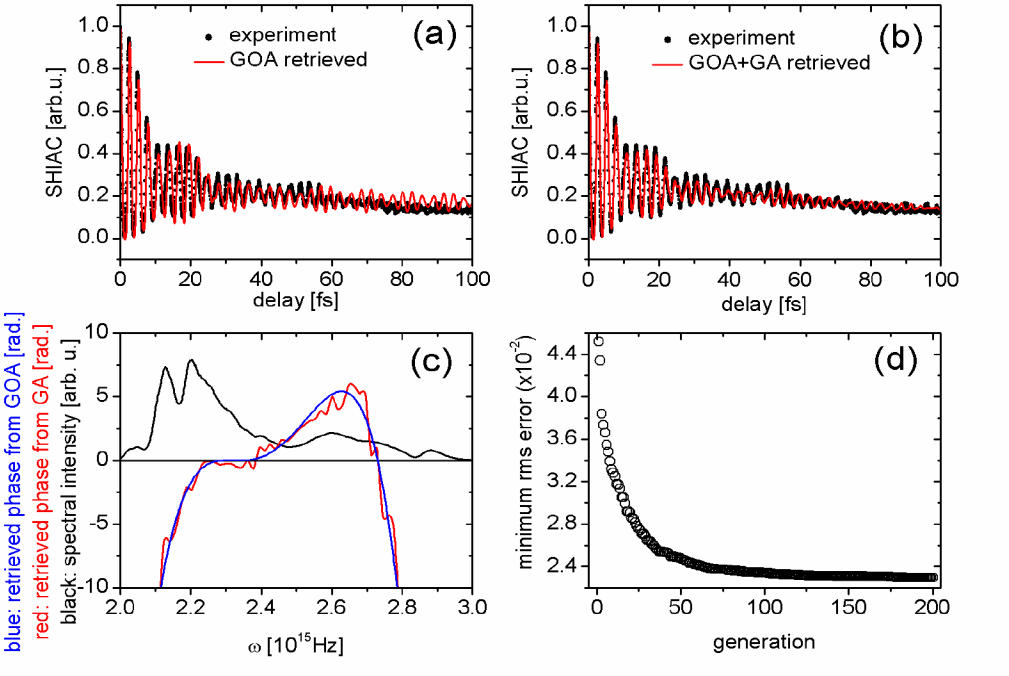

Fig. 9 (a) Retrieved SHIAC from GOA only. (b) Retrieved SHIAC from GOA+GA. (c) Retrieved phase from GOA (blue curve) and GOA+GA (red curve). (d) Evolution of rms errors between retrieved and measured SHIAC.

Fourier transform of the spectrum and corresponding spectral phase yields the pulse shown in Fig. 10.

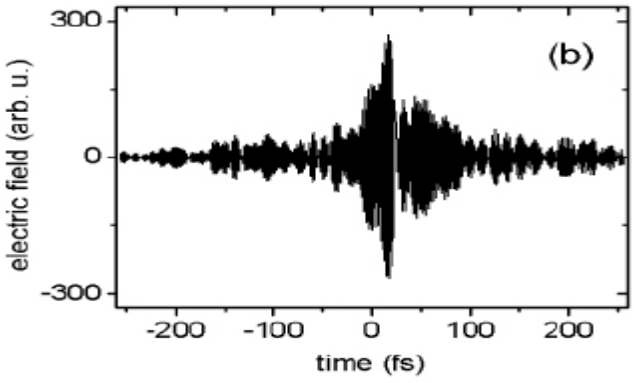

Fig. 10 Retrieved electric field from the hybrid genetic algorithm shown in Fig. 6.

Hence we have shown that the developed genetic algorithm is able to accurately reconstruct pulses which possess complex and sharply varying spectral phases, and thus enjoys an advantage over other less intensive approaches. Reconstruction of complex pulse shapes is important in understanding their propagation through media which possess highly dispersive and/or absorptive refractive indices, and the developed algorithm is therefore a valuable tool in studying their pulse propagation behavior.

Having reported investigations into linear phenomena, we now present experimental results involving nonlinear phenomena. Optical filaments are complex light pulses resulting from intensity-dependent Kerr lensing and self-focusing; the Kerr focusing is balanced by plasma defocusing effects, thus allowing an ultrashort laser pulse to propagate large distances without diverging. They have promising applications in sophisticated techniques for atmospheric and oceanic remote sensing, imaging, communication, and spectroscopy.

One important parameter in these applications is control of the propagating filament. This may be achieved through beam-coupling between two filament-forming pulsed beams. Such coupling has already been demonstrated in air (A.C. Bernstein *et al*. (2009), Y. Liu *et al*. (2010)); we have demonstrated, however, the first instance of energy coupling between filament-forming beams in bulk media. Moreover, we report an undocumented phenomenon in which the energy transfer oscillates on an increment in relative pulse delay equal to the time period of the incident radiation.

We used a Ti:sapphire based laser system consisting of an oscillator (Mira, Coherent) and an amplifier (Legend, Coherent: 800 nm center wavelength, 35 fs transform-limited pulse duration, 1 kHz repetition rate, 1 mJ pulse energy). Two identical chirped pulses were generated and crossed at a small angle within a glass cell containing methanol. We chose methanol as the bulk medium because of its transparency at the wavelengths used; it is also an important first step toward realizing energy coupling in other media. The energy of the pulses was about 2 uJ, enough to form filaments after passing through a lens with a focal length of 5 cm. The pulse energy was then varied as an experimental parameter. The resulting filament energies were then measured with a spectrometer (USB2000, Ocean Optics). As we used chirped pulses, the relative delay between the pulses determined the direction and amount of energy transfer; this was controlled by passing one beam through a translation stage (ESP301, Newport). We refer to the two beams as the fixed and translation stage beams, respectively.

Results are shown in Fig. 11. Six curves are shown for various pulse energies. It is important to notice that lower pulse energies show energy transfer consistent with A.C. Bernstein *et al*. (2009), while higher pulse energies show energy transfer consistent with Y. Liu *et al*. (2010). This is interesting in that behaviors documented for filament energy transfer in air are also present in bulk media.

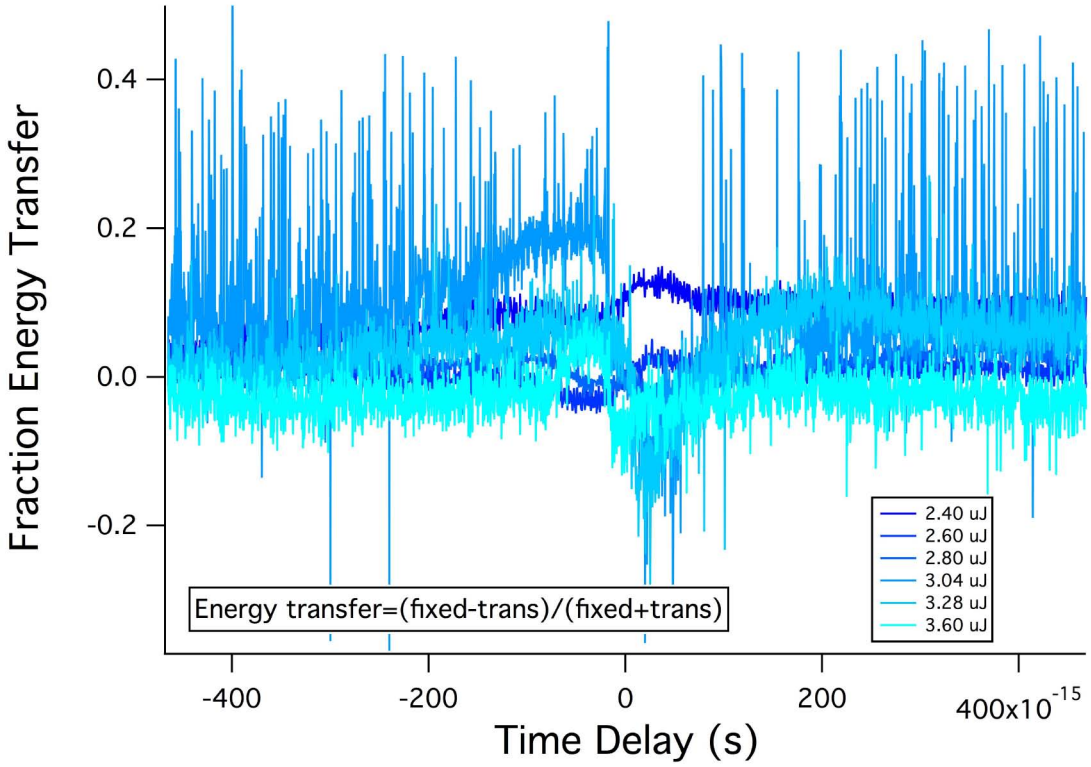

Fig 11. Energy transfer curves for various beam powers in methanol. Lower powers show energy transfer consistent with A.C. Bernstein et al. (2009), while higher power levels show energy transfer consistent with Y. Liu et al. (2010).

The curves shown in Fig. 8 appear "noisy"; however, it can be shown that the above curves actually oscillate at the same frequency as the incident radiation. This is shown in Fig. 12.

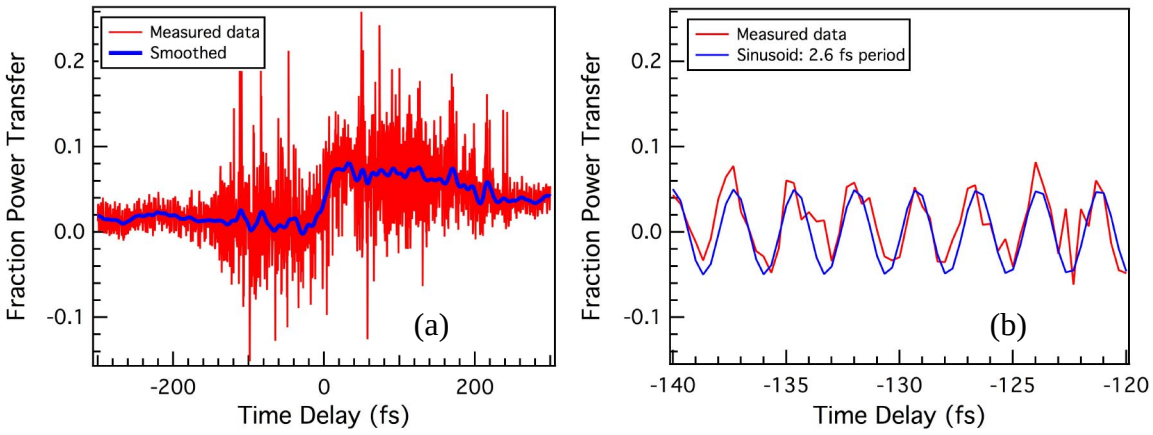

Fig. 12 (a) Energy transfer curve for filament-forming pulses propagating in methanol. (b) Zoomed in region of the same curve in (a) with superimposed sinusoid having time period of 2.6 fs, the same as the incident radiation.

(a) shows an energy transfer curve for two filament forming beams in methanol, while (b) shows a zoomed-in region of the same curve with a superimposed sinusoid have the same time period as the incident radiation.

These results indicate that very fine control of filaments may be achieved through two-beam coupling. Many more applications for these results may soon be realized.

IMPACT/APPLICATION

The work we are doing on nonlinear filament formation may lead to a very new and novel way to do hyperspectral remote sensing and underwater communication.

TRANSITIONS

If the results of this research uncover novel ways to propagate acoustic signals, then we will have a scheme to measure the speed of sound in water very quickly and accurately from helicopters or aircraft covering large regions of the ocean.

RELATED PROJECTS

We use the results from our research on the RaDyO project to give us the IOP's for water we use in our analysis for short pulses.

REFERENCES

1. L. Brillouin, *Wave Propagation and Group Velocity* (Academic Press, New York, 1960).
2. K. E. Oughstun and G.C. Sherman, "Propagation of electromagnetic pulses in a linear dispersive medium with absorption (the Lorentz medium), JOSA B, 817-849, (1988)
3. S-H Choi and U. Österberg, "Observation of Optical Precursors in Water", PRL, 92, 193903-193903-3, (2004)
4. R. Alfano, J. L. Birman, X. Ni, M. Alrubaiee, and B.B. Das, Comment on "Observation of Optical Precursors in Water", PRL, 94, 239401-1, (2005)
5. A. E. Fox and U. Österberg, "Observation of non-exponential absorption of ultra-fast pulses in Water", Opt. Express, 14, 3688-3693, (2006)
6. J.C. Li, D.R. Alexander, H.F. Zhang, U. Parali, D.W. Doerr, J.C. Bruce III, H. Wang "Propagation of ultrashort laser pulses through water" Opt. Express 15, 1939-1945 (2007)
7. Y. Okawachi, A. D. Slepkov, I. H. Agha, D. F. Geraghty, and A. L. Gaeta, "Absorption of ultrashort optical pulses in water", JOSA A, 10, 3343-3347
8. D. Pestov, M. Zhi, Z. E. Sariyanni, R. Murawski, A. V. Sokolov, N. G. Kalugin, Y. V. Rostovtsev, V. A. Sautenkov, A. A. Kolomenskii, H. Schuessler, G. G. Paulus, G. R. Welch, T. Siebert, D. A. Akimov, S. Graefe, W. Kiefer, and M. O. Scully, "Visible and UV Coherent Raman Spectroscopy of Dipicolinic Acid", Proc. Natl. Acad. Sci. USA 102 (42), 14976-14981 (2005)

9. Yu Huang, A. Dogariu, Y. Avitzour, R. K. Murawski, D. Pestov, M. Zhi, A. V. Sokolov, and M. O. Scully, "Discrimination of dipicolinic acid and its interferents by femtosecond CARS", J. Appl. Phys. 100, 124912-124916 (2006)

10. M. Zhi, D. Pestov, X. Wang, R. K. Murawski, Y. V. Rostovtsev, Z. E. Sariyanni, V. A. Sautenkov, N. G. Kalugin, and A. V. Sokolov, "Concentration dependence of femtosecond CARS in the presence of strong absorption", JOSA B 24, 1181-1186(2007)

11. D. Pestov, R. K. Murawski, G. O. Ariunbold, X. Wang, M. Zhi, A. V. Sokolov, V. A. Sautenkov, Y. V. Rostovtsev, A. Dogariu, Y. Huang, and M. O. Scully, "Optimizing the laser-pulse configuration for coherent Raman spectroscopy", *Science* 316**,** 265-268 (2007)

12. D. Pestov, G. O. Ariunbold, R. K. Murawski, V. A. Sautenkov, A. V. Sokolov, and M. O. Scully, "Coherent versus incoherent Raman scattering: molecular coherence excitation and measurement", Opt. Lett. 32, 1725-1727 (2007)

13. D. Pestov, M. Zhi, Z. Sariyanni, N. G. Kalugin, A. Kolomenskii, R. Murawski, Y. V. Rostovtsev, V. A. Sautenkov, A. V. Sokolov and M. O. Scully, "Femtosecond CARS of Methanol-Water Mixtures", J. Raman Spectr. 37, 392-396 (2006)

14. R. M. Pope and E. S. Fry, "Absorption spectrum (380-700 nm) of pure water. II. Integrating cavity measurements," Appl. Opt. 36, 8710-8723 (1997)

15. P. Yang, H. Wei, G. W. Kattawar, Y. Hu, D. Winker, C. Hostetler, and B. Baum, "Sensivity of backscattering Mueller Matrix to particle shape and thermodynamic phase", Appl. Opt. 42, 4389-4395, (2003)

16. Z. Zhang, P. Yang, G. W. Kattawar, S. –C. Tsay, B. A. Baum, H.-L Huang, Y. X. Hu, A. J. Heymsfield, and J. Reichardt,"Geometric optics solution to light scattering by droxtal ice crystals Appl. Opt. 43, 1-10, (2004)

17. P. Yang, G. W. Kattawar, K.N. Liou, and J. Q. Lu, "Comparison of Cartesian grid configurations for application of the finite-difference time domain method to electromagnetic scattering by dielectric particles", Appl. Opt.,43, 4611-4624 (2004)

18. C. Li, G. W. Kattawar, and P. Yang "A new algorithm to achieve rapid field convergence in the frequency domain when using FDTD", J. of Electromagnetic Waves and Applications, 18, 797-808 (2004)

19. P. Zhai, G. W. Kattawar, and P. Yang "Implementing the near to far field transformation in the finite-difference time domain method", Appl. Opt. 43, 3738-3746, (2004)

20. Ping Yang, George W. Kattawar, and Warren J. Wiscombe "Effect of particle asphericity on single-scattering parameters: Comparison between Platonic solids and spheres", Appl. Opt. 43, 4427-4435, (2004)

21. C. Li, G. W. Kattawar, and P. Yang "Effects of Surface Roughness on Light Scattering by Small Particles", JQSRT, 89, 123-131 (2004)

22. P. Zhai, G. W. Kattawar, P. Yang, and C. Li "Application of the Symplectic FDTD Method to Light Scattering by Small Particles", Appl. Opt , 44,1650-1656, (2005)

23. C. Li, G. W. Kattawar, Pengwang Zhai, and P. Yang "Electric and magnetic energy density distributions inside and outside dielectric particles illuminated by a plane electromagnetic wave", Optics Express, 13, 4554-4559, (2005)

24. P. Yang, H. Wei, , H-L. Huang, B. A. Baum. Y. X. Hu, G. W. Kattawar, M. I. Mischenko, and Q. Fu, "Scattering and absorption property database for nonspherical ice particles in the near-through-far-infrared spectral region", Appl. Opt. 44, 5512-5523, (2005).

25. G. Chen, P. Yang, G. W. Kattawar, and M.I. Mishchenko, "Scattering phase functions of horizontally oriented hexagonal ice crystals", JQSRT, 100, 91-102, (2006).

26. Y. Xie, P. Yang, B-C Gao, G. W. Kattawar, and M. I. Mishchenko, "Effect of ice crystal shape and effective size on snow bidirectional reflectance", JQSRT, 100, 457-469, (2006).

27. J. D. Jackson, *Classical Electrodynamics, 2nd ed.* (Wiley, New York, 1975)

28. L.M. Naveira, B.D. Strycker, J. Wang, G.O. Ariunbold, A.V. Sokolov, and G.W. Kattawar, "Propagation of femtosecond laser pulses through water in the linear absorption regime," Appl. Opt. 48, 10, 1828-1836 (2009)

PUBLICATIONS

1. Sokolov, Alexei V., Lucas M. Naveira, Milan P. Poudel1, James Strohaber, Cynthia S. Trendafilova, William C. Buck, Jieyu Wang, Benjamin D. Strycker, Chao Wang, Hans Schuessler, Alexandre Kolomenskii, and George W. Kattawar, "Propagation of ultrashort laser pulses in water: linear absorption and onset of nonlinear spectral transformation", Applied Optics, 49, 513-519, 2010 (published, refereed)

2. Wenlong Yang, Matthew Springer, James Strohaber, Alexandre Kolomenski, Hans Schuessler, George Kattawar and Alexei Sokolov, "Spectral Phase Retrieval from Interferometric Autocorrelation by a Combination of Graduated Optimization and Genetic Algorithms", Optics Express, Vol. 18, No. 14 / Optics Express, 15028-15038, 2010 (published, refereed)