Observation of Optical Precursors in Water

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We observe the formation of optical precursors while propagating 540 fs pulses through 700 mm of deionized water. The launched pulses were strongly chirped to give them a bandwidth of approximately 60 nm to more readily excite the precursors. The precursors attenuated nonexponentially with distance.

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Introduction.—Precursors (or forerunners) were first discussed by Sommerfeld and Brillouin in 1914 [1]. They performed a thorough theoretical study of pulse propagation in a linear, causal, dielectric medium. The purpose of their study was to clarify if the group velocity could exceed the speed of light (which it can). Several groups have improved on the early analysis of Sommerfeld and Brillouin [2–4]; however, the main result still stands—that energy is shed from the main pulse, creating new pulses with different properties. Despite the name "precursor" the Brillouin pulses may arrive ahead of, or behind the main pulse, depending on the exact nature of the medium [5]. The precursor property which is the main motivator for our work is that they do not attenuate exponentially. In particular, the peak amplitude point of the Sommerfeld precursor attenuates as $\sim\frac{1}{z}$ while the peak amplitude point of the Brillouin precursor attenuates as $\sim 1/\sqrt{z}$ with propagation distance z. After propagation through 10 m of water this translates into 8 orders of magnitude lower attenuation for a Brillouin precursor than a conventional pulse.

Despite the numerous theoretical papers [6] on the subject, very little has been done experimentally. Pleshko and Palócz [7] performed a classic experiment in the microwave regime where they clearly showed the existence of precursors. In the optical regime there have, to our knowledge, been only two papers purporting to have observed precursors, both of them in connection with pulse propagation through semiconductors close to excitonic resonances [8,9]. Neither of the previous optical experiments measured pulse attenuation with distance nor did they rule out possible nonlinear interactions.

In this paper we report the observation of precursors in deionized water. We chose to use deionized water because of the abundance of data which exists for its complex refractive index as a function of wavelength [10] and because of its reproducibility.

Theory.—Using the integral representation for the scalar wave field of the propagating pulse through a distance z in the medium, we find the wave field as

$$A(z,t) = \frac{1}{2\pi}\mathrm{Re}\left[\int_{ia-\infty}^{ia+\infty} \tilde{u}(\omega-\omega_c)\, e^{(i[k(\omega)z-\omega t])}\, d\omega\right], \qquad (1)$$

where $\tilde{u}(\omega-\omega_c)$ is the spectrum of the launched pulse. Solving this integral for a typical dielectric medium is very difficult. In the case of a narrow band pulse we can do a Taylor expansion around the carrier frequency ω_c and retain up to the second term. For a broad band pulse we can use an asymptotic method which relies, yet again, on a Taylor expansion; however this time we expand, to second order, around the saddle-point frequencies associated with the phase function $\Phi(z,t) = i[k(\omega)z - \omega t]$ where $\omega = \omega' + i\omega''$ [6]. The launched pulse can break up into as many pulses as there are saddle points. The saddle points evolve with time and space and a pulse (precursor) will coalesce for a period in time and space rather than exist unchanged for all times. A closer look at the saddle points [5] reveals that the Sommerfeld precursor is derived from the higher frequencies in the pulse and the Brillouin precursor is related to the lower frequencies.

Rewriting the phase function according to Oughstun [11], we obtain

$$\Phi(z,t) = \phi(\omega,\theta) = i\omega[n(\omega) - \theta], \qquad (2)$$

where $\theta = ct/z$,

$$n(\omega) = \sqrt{1 - \sum_i [(f_i w_p^2)/(\omega^2 - \omega_{0i}^2 + 2i\gamma_i\omega)]},$$

f_i are the oscillator strengths, ω_p is the plasma frequency, the ω_{0i} are the undamped resonance frequencies, and γ_i are the damping constants. The asymptotic representation of the propagated field (as $z \to \infty$) can now be written as

$$A(z,t) \sim A_{\mathrm{main}}(z,t) + A_S(z,t) + A_B(z,t), \qquad (3)$$

where

$$A_B(z,t) \sim \sqrt{\frac{c}{2\pi|\phi''(\omega_{\mathrm{sp}})|z}} \cdot \mathrm{Re}[\tilde{u}(\omega_{\mathrm{sp}} - \omega_c)e^{(z/c)\phi(\omega_{\mathrm{sp}},\theta)}]. \qquad (4)$$

In this expression ω_{sp} is the complex angular frequency related to the Brillouin precursor. From Eq. (4) we can clearly see the $\sim 1/\sqrt{z}$ dependence with distance. The strength of the attenuation is proportional to the magnitude of the second derivative of the phase function $\phi(\omega,\theta)$ with respect to ω at the saddle point (i.e., $\phi'(\omega_{\mathrm{sp}},\theta) = 0$. The regions where the phase function is flat as a function of ω contribute the most to the propagated field.

It should be noted that $|\phi''(\omega_{\mathrm{sp}})|$ is a function of z and therefore the specific z dependence of the peak amplitude of the Brillouin precursor is different from $\sim 1/\sqrt{z}$ for short distances. However, since $|\phi''(\omega_{\mathrm{sp}})| \rightarrow$ const value asymptotically, the peak amplitude of the Brillouin precursor is, asymptotically, approaching $\sim 1/\sqrt{z}$.

Although it is difficult to estimate the validity of the asymptotic expression a rule of thumb is that it can be used safely at distances longer than one absorption depth (the distance for which the amplitude is reduced to $1/e$ of its original value). For the two carrier wavelengths used in this study (700 and 780 nm) the $1/e$ distances are 1660 and 440 mm, respectively.

Unfortunately, there is very poor agreement between measured data of the complex refractive index and a linear combination of Lorentz-Lorenz expressions [12], and therefore no asymptotic analysis exists for water.

However, these pulse breakups are all due to *linear* interactions. From a simulation point of view this is fortunate since it allows us to use a straightforward Fourier analysis to model [13] the propagation through deionized water. We use experimental data for the real and imaginary parts of the refractive index with no fitting parameters. This analysis shows that contrary to earlier discussions [8] it is not necessary to have a short pulse with a steep leading (or trailing) edge to excite precursors. Instead, we found that the most important features in a pulse for exciting precursors in water are a broad bandwidth (>10 nm) and a positive chirp.

Experiments and results.—Our measurements were performed using a Spectra-Physics Mai-Tai 100 fs laser, tunable between 730–870 nm, with an average output power between 0.5–1 W.

Since precursors are a linear phenomena we spent considerable effort characterizing the water for nonlinear optical effects using both temporal and spectral measurements. The two nonlinearities we were most concerned with were two-photon absorption (TPA) and self-phase modulation (SPM). The nonlinearity with the lowest threshold is TPA. We measured the TPA coefficient to be $\beta = 10^{-8}$ cm/W at 780 nm. SPM was eliminated by constantly comparing the spectrum before and after propagation through the water to ensure that no new frequencies had been created. As a final test, after we had observed the precursors (see Fig. 2), we varied the input power up to a factor of 5 without any measurable change in relative peak amplitudes. To maintain a high level of power after propagating through the water and still avoid TPA we expanded the beam to a 1 cm radius ($1/e$) limiting the peak intensity to less than 1 kW/cm^2. This also eliminated thermal blooming.

The first measurement we made was of the integrated pulse energy as a function of spectral width and distance. We coupled the laser into a holey fiber, generating a continuum with a bandwidth $\Delta\lambda > 500$ nm. We then used a series of Gaussian profile bandpass filters centered at 700 nm to control the width of the input spectrum. The center wavelength of 700 nm was chosen to maximize the signal at the detector. For $\Delta\lambda < 5$ nm we have complete exponential decay as a function of propagation distance with an absorption coefficient of 0.005 (1/cm) which is within 8% of published data [10]. As we increased the bandwidth, the amplitude decreased in a nonexponential decay. With $\Delta\lambda \approx 80$ nm, for distances larger than ~ 1 m the amplitude is decreasing less rapidly than a pure exponential, as shown in Fig. 1.

We investigated several possible explanations for this behavior. The first was that as more strongly absorbing wavelengths were removed, the pulse attenuation was shifting towards the attenuation for the least absorbing wavelength within our pulse. To gauge that effect, we plotted (Fig. 1) the expected attenuation for the extreme wavelengths of our pulse. Since at long distances our measured points have a lower slope than the exponential decay at 640 nm, we concluded that this was not the dominant effect. We then fitted the attenuation for the Brillouin precursor in the same graph and observed that our measured points have the same behavior as the precursor for large distances. The fitting was done using only one independent variable, namely, $|\phi''(\omega_{\mathrm{sp}})|$. For the fit in Fig. 1 the value of $|\phi''(\omega_{\mathrm{sp}})| \approx 239$ (s).

To further clarify the behavior, we performed a number of temporal measurements with distance. For this experiment we sent $\sim 70\%$ of our pulse energy through a single-mode fiber ($\lambda_{\mathrm{cutoff}} \sim 630$ nm). Our pulse was approximately 100 fs long and centered around 780 nm (for maximum output power) with a bandwidth of 10 nm. After propagation through ~ 30 cm of fiber it was ~ 540 fs with a bandwidth of 60 nm and a chirp which was linear over $\sim 85\%$–90% of the pulse (estimated from the noncompressed wings using a grating compressor). We

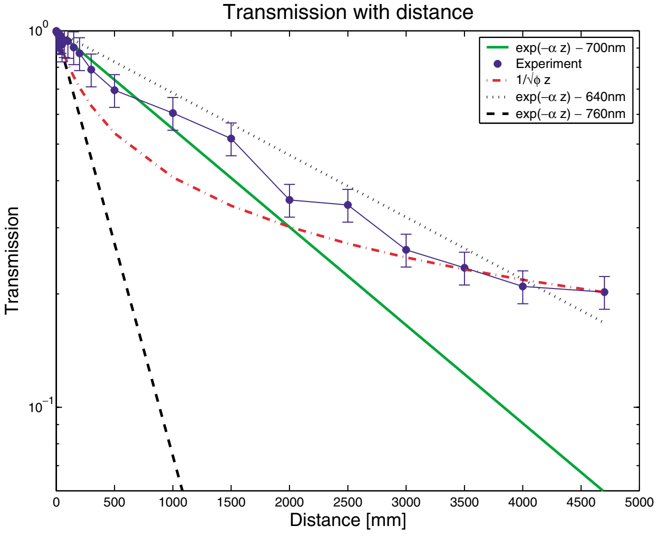

FIG. 1 (color online). Transmission with distance for a 80 nm wide pulse centered around 700 nm.

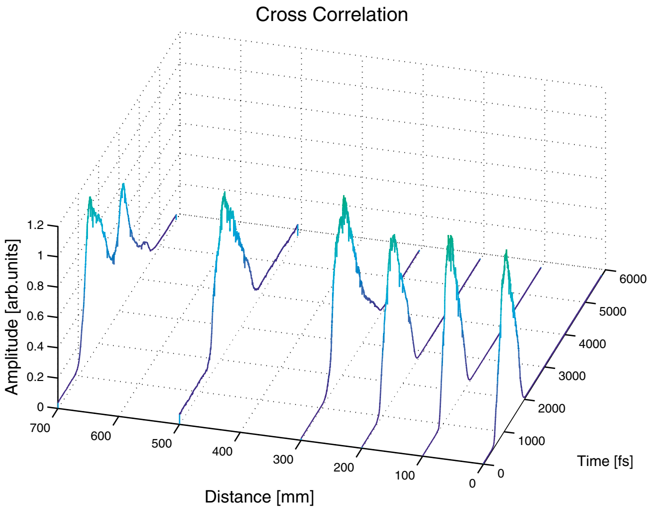

FIG. 2 (color online). Cross correlation of pulses, normalized, traveling through 100–700 mm of deionized water.

used the remaining 30% of laser energy to make a cross correlation with the pulse that had propagated through the water. Since the cross correlation is a nonlinear measurement we were able only to get temporal profiles for distances up to 700 mm. The first set of temporal measurements is shown in Fig. 2, where the lengths were determined by available water tubes. From 300 mm and onwards the pulse is clearly broadening asymmetrically and at 700 mm we clearly have pulse breakup. The amplitudes are normalized to show the pulse evolution.

To probe the distances between 500–700 mm we made a new set of tubes. The results from these measurements are shown in Fig. 3. We see complicated pulse dynamics which eventually lead to the two peaks at 700 mm. The data in Figs. 2 and 3 are from different runs, and curves from 500 and 700 mm are indicative of the reproducibility of the behavior.

The main peak for all of these measurements follows an exponential decay while the lagging peak is seen to be at first slowly decaying and then growing (relative to the main pulse) with distance. We suggest that the main reason for the complicated dependence on distance for the precursor peak is due to the fact that it is receiving energy from the original pulse and attenuated by the medium simultaneously.

Conclusion.—We observe pulse breakup in a linear energy regime for 540 fs long pulses with a bandwidth of 60 nm bandwidth propagating through 700 mm of deionized water. Since the peak power is well below the threshold for any nonlinearities to occur, we attribute the pulse breakup to the formation of optical precursors. This is further supported by subexponential attenuation with distance for the new peak as well as approximate $\sim 1/\sqrt{z}$ attenuation at distances exceeding 3.5 m. Experiments to characterize the spectral decay at longer distances are underway.

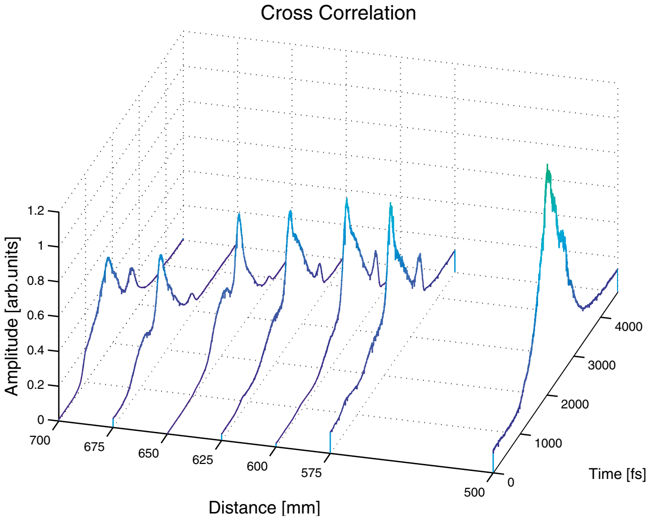

FIG. 3 (color online). Cross correlation of pulses, traveling through 500–700 mm of deionized water.

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