Absorption of ultrashort optical pulses in water

Yoshitomo Okawachi,* Aaron D. Slepkov, Imad H. Agha, David F. Geraghty, and Alexander L. Gaeta

School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853, USA
**Corresponding author: yo22@cornell.edu*

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We investigate the linear propagation of 800 and 1530 nm ultrashort optical pulses in water. For all pulse repetition rates studied, we observe pure exponential decay down to a transmission of 2.5×10^{-5}. We further demonstrate that previous observations of nonmonoexponential decay and pulse splitup in broadband pulses are consistent with Beer's law in the purely linear regime. © 2007 Optical Society of America

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1. INTRODUCTION

Underwater communications has been an active area of research for decades. Because of relatively large absorption coefficients, electromagnetic waves cannot propagate over long distances in water, and the primary means of underwater communications has been limited to acoustic and radio waves [1]. Recent results offer renewed prospects for underwater optical communications by reporting subexponential decay of ultrashort ~800 nm pulses [2–4]. For example, measurements in [2] suggest the possibility of subexponential decay in water owing to the presence of Brillouin–Sommerfeld optical precursors [5], which have been an area of extensive research [6,7]. The observation of deviations from Beer's law and subexponential decay of electromagnetic waves in water would also have a significant impact on applications other than underwater communications, such as medical imaging [8].

Choi and Österberg [2] report nonexponential attenuation and pulse breakup of broadband femtosecond pulses in water, which they attribute to precursor formation. Their findings have quickly led to further analysis offering alternative explanations [9,10]. More recently, Fox and Österberg [4] propose that the observed nonexponential decay depends not only on pulse duration but also on the pulse repetition rate; they report the observation of nonexponential decay for a 1 kHz repetition rate, 60 fs pulse train, but do not observe such behavior for an 80 MHz repetition rate, 900 fs pulse train. Fox and Österberg suggest that this deviation from exponential decay is due to the time scale of relaxation processes in water, where the pulse duration must be short enough to avoid vibrational transitions and the repetition rate must be low enough to allow water to reach equilibrium prior to the arrival of the subsequent pulse [4].

In contrast, Li *et al.* [11] report that the bandwidth, not the repetition rate, is the primary cause of deviations from a single-exponential decay. In their experiments they use spectrally broad 100 nm pulses centered at 800 nm that vary from 10 to 250 fs in duration at a repetition rate of 75 MHz to demonstrate deviation from single-exponential decay independent of pulse duration. Li *et al.* show that the subexponential decay occurs only when the pulse bandwidth is high (~100 nm) and that for smaller bandwidths (<30 nm) the decay is monoexponential, which is in disagreement with the results reported by Fox and Österberg [4], who observed subexponential decay with 20 nm bandwidth pulses. However, Li *et al.* used 300 fs pulses for investigating propagation dynamics at a 1 kHz repetition rate. This pulse duration is insufficiently short to compare with the claims made in [4] for observing subexponential decay.

In this paper, we report our investigation of femtosecond pulse propagation in water. The body of the paper is divided into two sections. First, we perform numerical simulations using various bandwidths and center wavelengths to determine to what extent the absorption dynamics depend on these parameters. Our simulations show that both nonmonoexponential decay and temporal pulse breakup can be obtained within the framework of Beer's law for the broad-bandwidth pulses used by Choi and Österberg in [2]. Next, we perform experiments to determine the dependence of repetition rate and pulse duration on pulse absorption, using four different laser systems at 800 and 1530 nm. The experimental parameters we use here are similar to those used in [4], where the bandwidth should not affect deviations from a monoexponential decay. In all four experimental regimes, we observe strictly monoexponential decay, confirming that propagation of femtosecond pulses in water obeys the Beer–Lambert law.

2. THEORY

To verify the dependence of the pulse bandwidth and center wavelength on the absorption dynamics, we perform a numerical simulation of pulse propagation through water. We assume a Gaussian input spectrum,

$$I_0(\lambda) = \exp\left[-\frac{(\lambda-\lambda_0)^2}{2\sigma^2}\right], \tag{1}$$

where $\lambda_{\mathrm{FWHM}}=\sqrt{8\ln 2}\sigma$ is the pulse bandwidth (FWHM). We use bandwidths of 20–60 nm and propagate the pulses through different path lengths in water according

to the Beer–Lambert relationship,

$$I(\lambda) = I_0(\lambda)e^{-\alpha(\lambda)z}, \tag{2}$$

where $I(\lambda)$ and $I_0(\lambda)$ are the output and the input pulse spectra, respectively; $\alpha(\lambda)$ is the absorption spectrum of water in inverse centimeters, and z is the path length in centimeters. In our modeling, we use sources similar to those used by Li *et al.* [11,12] and Choi and Österberg [2,13] for the frequency-dependent absorption spectrum of pure water. To simulate the temporal dynamics, we use the following empirical approximation for the index of refraction that has been shown to be accurate in the visible and the near IR [14,15]:

$$n(\lambda) = 1.128 + 15.76\lambda^{-1} - 4382\lambda^{-2} + 1145500\lambda^{-3}, \tag{3}$$

where λ is the wavelength in nanometers. Temporally, we use Gaussian pulses given by

$$I_0(t) = \exp\left[-\frac{(t-t_0)^2}{2\tau^2}\right], \tag{4}$$

where τ is related to the FWHM pulse width according to $\tau_{\mathrm{FWHM}} = \sqrt{8\ln 2}\tau$. In order to model the pulses used by Choi and Österberg [2], we use the FWHM wavelength and pulse-width values of 20–60 nm and 540 fs, respectively, and add a linear chirp given by

$$\lambda(t) = t\frac{\sigma}{\tau} + \lambda_0, \tag{5}$$

where λ_0 is the pulse center wavelength of 780 nm. Finally, we propagate these pulses through (linearly dispersive) water, which yields the arrival time for all wavelength components of the pulse,

$$t_{\mathrm{arrive}}(\lambda) = t_0 - \left[t(\lambda) - \frac{n(\lambda)z}{c}\right], \tag{6}$$

where t_0 is the arrival time of the center wavelength [$t_0 = t_{\mathrm{arrive}}(780\ \mathrm{nm})$] and $t(\lambda)$ is the inverse of $\lambda(t)$.

Our findings show that the absorption spectrum of water acts as a filter that spectrally reshapes the pulse and thus yields nonmonoexponential energy decay [4,10,11], While a large spectral bandwidth is a major factor in determining the deviation from monoexponential decay [11], we also find that this behavior strongly depends on the pulse central wavelength. This leads to complex decay dynamics. Figure 1(a) shows the transmission as a function of propagation length for various center-wavelength pulses with a spectral bandwidth of 20 and 50 nm (FWHM). We see that for a 760 nm pulse, a bandwidth of 20 nm yields strictly monoexponential decay, whereas an increase in bandwidth to 50 nm is sufficient to introduce large deviations from monoexponential behavior, in agreement with Li *et al.* [11]. However, Fig. 1(a) also shows that the decay dynamics changes significantly over a narrow range of center wavelengths; a 760±50 nm pulse displays considerable curvature away from monoexponential behavior, a 780±50 nm pulse is considerably straighter, and a 800±50 nm pulse nearly decays monoexponentially. It is interesting to note that the difference between these three wavelengths is nearly equal to the difference between the 50 and 20 nm bandwidths of the aforementioned 760 nm pulses. Thus, we find that in addition to bandwidth, the position of the pulse center wavelength within the absorption spectrum is a crucial factor in the absorption dynamics of broadband pulses in realistic dielectric media.

In addition to reproducing the curvature in the absorption dynamics seen by Choi and Österberg [2], our modeling of their linearly chirped pulses further reproduces their observed pulse splitting. Figure 1(b) displays the effects of both the absorptive spectral filtering [$\alpha(\lambda)$] and the linear dispersion [$n(\lambda)$] of the water on the temporal profile of the propagating pulse. As seen in Fig. 1, an initially narrow 540 fs pulse is considerably broadened after propagating through 250 cm of water and appears to split in time, yielding two peaks separated by ≈15 ps. This pulse splitting is nearly identical to that experimentally demonstrated by Choi and Österberg [2], who presented it

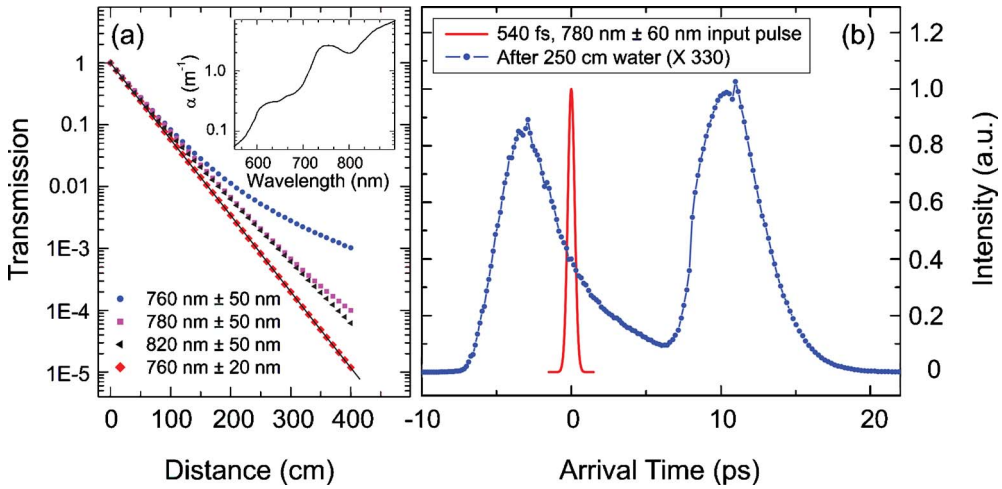

Fig. 1. (Color online) Theoretical analysis of linear propagation of broadband pulses in water. (a) Transmitted power as a function of propagation distance for Gaussian pulses with a spectral width of 50 and 20 nm (FWHM) for three different center wavelengths. Inset, absorption spectrum of water in the visible and NIR range [12,13]. Solid line, fit to monoexponential decay for the 760±20 nm pulse. (b) Temporal pulse splitup of linearly chirped 540 fs, 780±60 nm pulses after traveling through 250 cm of water. Note that this pulse breakup is obtained solely from wavelength-dependent linear absorption (Beer's law) and dispersion.

as strong evidence of the presence of optical precursors. The simulations presented here, however, show that this temporal splitting can be entirely explained within the framework of Beer–Lambert (linear) absorption by accounting for spectral filtering and normal dispersion in water.

3. EXPERIMENT

As presented above, our simulations account for nonmonoexponential decay dynamics in broad-bandwidth pulses. However, they do not address previous observations of nonmonoexponential decay dynamics in narrow-bandwidth pulses that were attributed by Fox and Österberg [4] to pulse duration and repetition rate. Thus, we look to study these conditions experimentally. First, we investigate the absorption characteristics of water near 800 nm using a Ti:sapphire oscillator with a pulse duration of 66 fs and a repetition rate of 80 MHz. The spectrum is centered at 792 nm with a bandwidth of 18 nm FWHM, similar to that used in [4]. We use a multipass cell and a pick-off mirror on a translation stage to vary the number of mirror bounces. A photoschematic of the cell is shown in Fig. 2. We propagate the pulse train through the water cell and measure the transmission by using a high-sensitivity powermeter (Thorlabs PM130). The total path length is varied from 60 ± 1 to 480 ± 1 cm. The losses due to the mirrors in water are estimated by comparing two power measurements. First, we measure the transmission straight through the water cell. We then position a mirror in the center of the cell and take a reflection measurement. This way, both paths are equidistant in water and include the same number of passes through the windows; the only difference between them is the reflection from the single dielectric mirror. From these measurements, the reflectivity of the mirror underwater is estimated to be 98.5%. Figure 3(a) presents the transmission measurements for 800 nm pulses, for which pure exponential attenuation with propagation distance is observed. Thus, our results indicate that short pulse durations do not contribute to deviations from monoexponential decay.

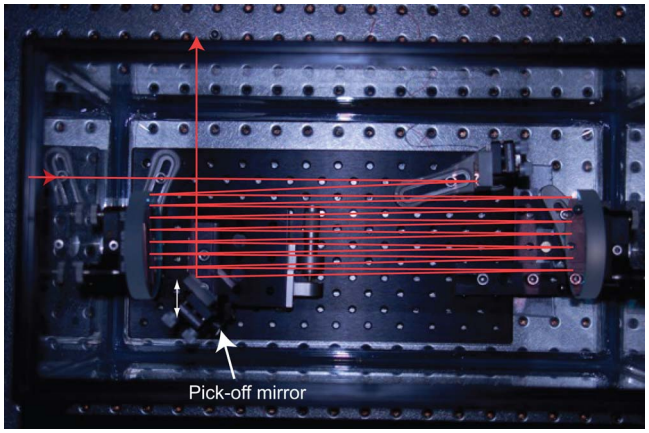

Fig. 2. (Color online) Experimental setup of water cell for transmission measurements at 800 nm. The path length was varied by moving the pick-off mirror to change the total number of bounces.

It has been suggested that observation of subexponential absorption of optical pulses in water is dependent on both the pulse duration and the repetition rate [4]. To investigate whether the combination of short pulse duration and low repetition rate influences the absorption, we use an ultrafast amplifier (Quantronix Integra-E), which generates 84 fs pulses at a repetition rate of 1 kHz. The pulse spectrum is centered at 804.7 nm with a bandwidth of 12.8 nm FWHM, similar to parameters used in [4]. Using the water cell described above, we measure the transmitted power by using the same high-sensitivity powermeter. For the amplified pulses, we work at lower average powers and with peak intensities well below those for which nonlinearities such as multiphoton absorption could be observed. As a result, the average power is much lower than in the 80 MHz case, which makes detection over a large dynamic range more difficult. It is important to operate above the noise threshold of the detector at longer cell lengths to avoid a dc bias in our measurements. As shown in Fig. 3(b), 800 nm ultrashort pulses at a repetition rate of 1 kHz also follow a strictly monoexponential absorption with path length. Thus, we conclude that the pulse duration and the repetition rate do not contribute to subexponential absorption of optical pulses in our experiments.

To determine whether this behavior is dependent on a particular wavelength regime, we also investigate water absorption at telecommunication wavelengths. Since the absorption coefficient is much larger in this wavelength regime, we designed a water cell with a much shorter path length. We investigate absorption for two different repetition rates. We use an optical parametric amplifier (OPA), which outputs 100 fs pulses at a repetition rate of 1 kHz. The pulse spectrum of the OPA is centered at 1530 nm, with a bandwidth of 30 nm FWHM. In addition, we use an optical parametric oscillator (OPO), which outputs 270 fs pulses at a repetition rate of 80 MHz. The spectrum of the OPO is centered at 1532 nm with a bandwidth of 24.5 nm. The pulses are sent into a water cell, and the transmission spectrum is measured by using an optical spectrum analyzer. For both cases, we perform the measurements with the spectral window of ~200 nm centered on the pulse spectrum.

Figure 4 shows the relative power output as a function of propagation distance for the two different repetition rates. For the measurement performed using the OPO, the spectral window over which we perform the measurement is critical. The spectrum directly from the OPO has an artifact at 1361 nm, which is 32 dB lower than the main peak at 1532 nm. However, this artifact becomes dominant at longer cell lengths (>6.0 mm), effectively acting as a background in our measurement that leads to deviations from a monoexponential decay. This indicates that when taking such measurements it is essential to choose the correct spectral window to measure the absorption characteristics of the pulse spectrum and that careful attention must be paid to ensure the complete extinction of spurious wavelengths. The outcome of the measurement depends strongly on the quality of the bandpass filter used, since a poor extinction can lead to high transmission at the tails of the transmission band, leading to higher overall transmission. This will be espe-

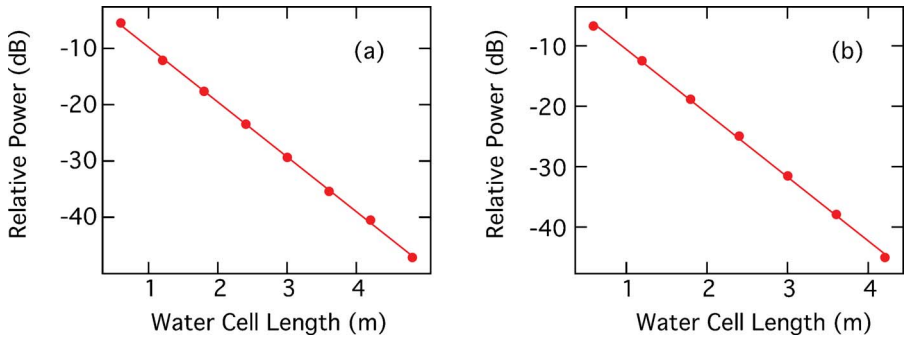

Fig. 3. (Color online) (a) Plot of transmitted power through distilled water as a function of path length for a 60 fs pulse train with an 80 MHz repetition rate centered at 795 nm. The solid line is a monoexponential fit to the data. (b) Plot of transmitted power through distilled water as a function of path length for an 84 fs pulse train with a 1 kHz repetition rate centered at 805 nm. The solid line is a monoexponential fit to the data.

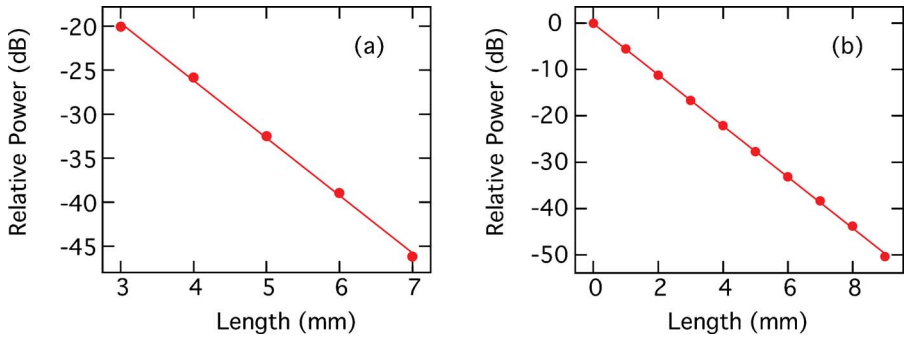

Fig. 4. (Color online) Plot of transmitted power through distilled water as a function of path length for (a) a 100 fs pulse train with a 1 kHz repetition rate centered at 1530 nm and (b) a 270 fs pulse train with an 80 MHz repetition rate centered at 1532 nm. Solid lines are monoexponential fits to the data.

cially important when filtering a small component from a broadband spectrum. This offers a possible explanation in the discrepancy between our results and that of [4], since they use a bandpass filter to shape their input spectrum going into the water cell. The measured loss is exponential down to values of –45 dB for both cases, which shows that in our experiment pulse duration and the repetition rate do not affect linear absorption in water.

4. CONCLUSION

We conducted measurements of the absorption dynamics of femtosecond pulses in water at two different wavelengths and repetition rates, and we do not observe deviations from exponential decay down to nominal transmissions of 2.5×10^{-5}. Simulations show that broadbandwidth pulses can lead to deviations from monoexponential decay and to spectral pulse breakup within the strict framework of linear absorption and are thus not an indication of any deviation from Beer's law. The frequency components of the pulse corresponding to regimes in the water spectrum having a smaller absorption coefficient lead to higher transmission. Thus, in order to use optical wavelengths for underwater communications, it is necessary to center the carrier frequency of the pulses in a regime where the water is more transparent, rather than relying on the wings of the pulse spectrum to propagate through the medium.

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