Dynamic Nonlinear X-waves for Femtosecond Pulse Propagation in Water

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Recent experiments on femtosecond pulses in water displayed long distance propagation analogous to that reported in air. We verify this phenomena numerically and show that the propagation is dynamic as opposed to self-guided. Furthermore, we demonstrate that the propagation can be interpreted as due to dynamic nonlinear X-waves whose robustness and role in long distance propagation is shown to follow from the interplay between nonlinearity and chromatic dispersion.

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The nonlinear Schrödinger equation (NLSE) in two or more dimensions is ubiquitous in physics as a model for weakly interacting nonlinear and dispersive waves, and arises in such diverse areas as Langmuir waves in plasmas, weakly interacting Bose-Einstein condensates, and optical propagation in nonlinear dielectrics [1]. The ubiquity of the NLSE means that new solutions or paradigms that arise in one area can extend into other areas. For example, previous experiments have shown that femtosecond (fs) pulses can propagate long distances though air while maintaining an almost constant fluence profile [2, 3, 4]. Although these results initially suggested a self-guiding mechanism, with self-focusing balanced by plasma defocusing, numerical simulations revealed that the propagation is highly dynamic, and this led us to the paradigm of dynamic spatial replenishment, whereby the propagating pulse collapses, the collapse is arrested, and the process is repeated several times as the collapse is replenished from spatially delocalized power [5]. One would then expect analogous phenomena in other fields, and indeed the dynamic spatial replenishment model in air has analogies with the Bose-Nova phenomenon in atomic gases [6].

Here our goal is to elucidate the physics underlying recent observations [7] of long distance propagation of fs pulses in water. Long distance propagation has previously been explored in glass [8] and there are clear differences with respect to air propagation. For example, in silica glass, and water also, normal group-velocity dispersion (NGVD) plays a much more dominant role in comparison to air, and this gives rise to nonlinear pulse-splitting [3, 10, 11, 12, 13]. Using numerical simulations we first verify the reported properties for long distance propagation in water, and we then perform diagnostic simulations to elucidate the underlying physics. In particular, we show that long distance propagation in water is given a natural explanation by combining the paradigms of nonlinear pulse-splitting and nonlinear X-waves. Nonlinear X-waves arise from the combination of diffraction, NGVD, and self-focusing, and have recently been introduced and examined theoretically [14] and experimentally [15]. Our main conclusion is that long distance propagation in water is best understood in terms of nonlinear X-waves.

Our model for fs pulse propagation in water is based on the propagation equation for the spectral amplitudes of the Bessel-beam expansion of the axially symmetric electric field:

\[
\frac{\partial E(\omega, k, z)}{\partial z} = i\sqrt{\frac{\omega^2e(\omega)}{c^2}} - k^2E(\omega, k, z) + \frac{i\omega}{2cn_b}P(\omega, k, z). 
\]

This is a scalar version of the Unidirectional Pulse Propagation Equation solved in the z-direction [16], and in the paraxial approximation becomes equivalent to the Nonlinear Envelope Equation of Brabec and Krausz [17]. We utilize a tabulated representation [18] of the complex frequency-dependent water permittivity \(\epsilon(\omega)\). In Eq. (1), \(k\) stands for the transverse wavenumber of each Bessel-beam component, and the nonlinear polarization \(P(\vec{r}, t) = \Delta\chi(\vec{r}, t)E(\vec{r}, t)\) is calculated in the real-space representation from the local nonlinear modification of the material susceptibility:

\[
\Delta\chi(\vec{r}, t) = 2n_2\omega I + \chi_{pl}(\rho) + \frac{n_2^2\beta^{(K)}}{k_0}I^{K-1} \quad (2)
\]

The first term represents the instantaneous optical Kerr effect with nonlinear coefficient \(n_2 = 2.7 \times 10^{-20} \text{ m}^2/\text{W}\), the second is the free-electron induced susceptibility change \(\chi_{pl}(\rho) = (i\epsilon_0\omega|\vec{m}_e\omega_0)/(1/\tau_e - i\omega_0)\), and the third term represents multi-photon ionization (MPI) energy losses, with MPI coefficient \(\beta^{(K)}\). The evolution of the free-electron density is described by the equation

\[
\frac{\partial \rho}{\partial t} = \frac{\sigma}{n_0^2E_g}\rho I + \frac{\beta^{(K)}}{K\hbar\omega_0}I^K - a\rho^2, \quad (3)
\]

where the three terms describe avalanche ionization with \(\sigma = (\epsilon_0^2\tau_e n_0/m_e\epsilon_0c)/(1 + \omega_0^2\tau_e^2)\) the cross-section for inverse bremsstrahlung, electron generation via MPI, and electron-ion recombination, respectively. The plasma model is parameterized by the collision time \(\tau_e = 10^{-14} \text{s}\) and recombination rate \(a = 2 \times 10^{-15} \text{ m}^3/\text{s}\) [19]. The MPI rate is calculated from the formula given in [20, 21] for \(E_g = 7 \text{ eV}\).

The simulation parameters were chosen to match the experiment of Ref. [7] as closely as possible for an incident pulse of center wavelength \(\lambda = 527 \text{ nm}\). The initial pulse amplitude within the water sample was chosen as a
focused Gaussian of spot size $w_0 = 99 \ \mu m$, pulse duration $\tau_p = 170 \ \text{fs}$, an initial radius of curvature corresponding to a lens of focal length $f = 5 \ \text{cm}$, and we varied the pulse input energy between 0.5 $\mu J$ and 2.5 $\mu J$. We note that, as in the experiment, the choice of initial focusing and spot size are essential for realizing long-distance propagation.

First we demonstrate that our simulations reproduce the basic features of the experiment in Ref. [7]. Figure 1 shows the maximum fluence (time-integrated intensity) over the transverse plane versus propagation distance $z$ in the water cell for several input pulse energies. An elevated and sustained fluence is taken as a sign that a light filament has formed. Similar to the experiment, at an input pulse energy 0.5 $\mu J$ a filament just starts to form but is terminated by diffractive spreading shortly after. At higher energies, we obtain filaments whose fluence fluctuates with $z$ but which persist over the scale of a couple of centimeters in quantitative agreement with the experiment. Furthermore, the transverse fluence profiles at different propagation distances reveal an almost constant filament diameter of around $\simeq 50 \ \mu m$ over the two centimeter range of the filament. This is shown in Fig. 2 where the $1/e^2$ diameter, obtained by approximating the spatial integral of the fluence profile with the on-axis fluence times $\pi d^2/8$ (corresponding to a gaussian), is shown versus propagation distance for several pulse energies. Thus, our simulation model agrees with the experiment of Ref. [7], in which the filaments also persist for around two centimeters and have diameters $\simeq 60 \ \mu m$.

The fluence maxima versus propagation distance in Fig. 1 suggest that multiple re-focusings occur. However, in contrast to propagation in air where the collapse is arrested dominantly by plasma defocusing [22], in water, and also in glass, NGVD is a key player. To diagnose this we have performed comparative simulations corresponding to Figs. 1 and 2 but with the plasma turned off, and the results (not shown), though clearly changed, are very similar in terms of the predicted 2 cm length scale for the filament and $\simeq 50 \ \mu m$ filament diameter. This indicates that the plasma is not essential to understanding this propagation regime: It somewhat slows down the dynamics and results in slightly thicker filament, because it helps to arrest the collapse, but the main collapse arresting mechanism is chromatic dispersion. It has previously been shown that a signature of collapse arrested by NGVD is nonlinear pulse splitting in the time domain [3, 10, 11, 12]. Figure 3 shows the results of our simulations, including the plasma, for the on-axis intensity as a function of local time for an input pulse energy 1.5 $\mu J$ and a variety of propagation distances after the filament first appears. The left-hand plot shows the pulse after the first pulse splitting, and the split daughter pulses are seen to move apart with distance. This figure is in keeping with the usual picture of pulse splitting [3, 10, 11, 12], and the evident asymmetries derive from inclusion of the plasma and the fact that we capture the spatio-temporal focusing terms to all orders and retain the full dispersion landscape as opposed to keeping only second-order GVD. We remark that the daughter pulses do not undergo subsequent cascade splittings [3], but rather after the first splitting, energy is replenished into the center of the local time domain around $\tau = 0$ which then grows, and it is this replenished center pulse that is subject to further pulse splitting. This is shown in the right-hand plot in Fig. 3. For larger propagation distances this process of pulse splitting, temporal replenishment of the on-axis pulse, followed by splitting of the new center pulse, can repeat itself several times, and this gives rise to the multi-peaked structures in Fig. 3 and the illusion that the filament is propagating in a self-guided manner. For comparative purposes we have repeated these simulations with the plasma turned off (results not shown) and we find that the same basic pulse splitting coupled with temporal replenishment picture emerges for the pulse propagation. The main difference is that the on-axis intensity profiles are more symmetric, have more structure and show evidence of shock phenomena since the smoothing action of the plasma is absent. This tells us that the
propagation is dominated by an interplay between dispersion and nonlinearity, in which the delayed nonlinear responses such as plasma do not play a crucial role.

Since the multiple pulse splittings are distinct from the cascade splitting predicted for a medium exhibiting NGVD, this begs the question as to what physics produces the temporal replenishment of the on-axis pulse? Our proposal here for the physical mechanism that produces the temporal replenishment is that it is due to dynamic nonlinear X-waves, and we shall make our case below. Nonlinear electromagnetic X-waves have only very recently been introduced theoretically [14] and produced experimentally [15] in the field on nonlinear optics. They are stationary (z-invariant) nonlinear solutions that result from the combination of linear diffraction, second-order NGVD, and nonlinear self-focusing. These X-waves propagate along z with unchanging intensity profile, and if one plots a space-time (local time) slice the resulting intensity profile has a central peak and arms that form a characteristic X shape (see Fig. 1 of Ref. [14]). Likewise, if one calculates the space-time Fourier-transform of a stationary X-wave it’s intensity spectrum in (k, ω)-space, k being the magnitude of the transverse wave vector and ω the frequency, likewise has an X-structure (see Fig. 3 of Ref. [14]). It has been shown that the X-shape in (k, ω)-space should follow the linear dispersion characteristics since they determine the relation between the propagation angle and frequency for phase-matched off-axis conical emission [14,23].

For the series of simulations presented here X-shaped textures can be discerned in the intensity profiles in both the space-time and (k,ω) domains past the first pulse splitting. The X-wave texture appears in the space-time domain in the region between the daughter pulses after splitting (not shown). This texture reflects optical energy that is transported from the off-axis regions and which serves as a reservoir for the replenishment of the pulse center as illustrated in Fig. 3 The X-wave signature for our pulse dynamic is much more evident in (k,ω)-space. This is shown in Fig. 4 which shows a representative example of a contour plot of the pulse spectrum at z = 1.7 cm (fully including plasma effects). The fine, “interference” structure in this figure results from the superposition of the spectra from both split daughter pulses. For longer propagation distances, after multiple pulse splittings the intensity spectra show similar features but become more complicated. The central X-shaped region where spectral energy concentration occurs is clearly visible. However, one quickly realizes that the X-shape has a slightly different angle compared to that expected for pure X-waves. Nevertheless, as we show next not only the spectrum in Fig. 4 can be explained using the paradigm of X-waves, but in the process we also gain further insight into the role of nonlinearity in X-wave propagation.

To proceed we make use of the three-wave mixing picture we recently employed to provide a qualitative explanation of supercontinuum generation in bulk media [24]. The nonlinear light-matter interaction creates a material response reflected in the change in the on-axis material susceptibility \(\Delta \chi \approx \Delta \chi(t - z/v_g, \tau_{slow})\) that propagates predominantly as a z-invariant shape modulated on a relatively slower time scale \(\tau_{slow}\), that we hereafter neglect. This response usually exhibits multiple peaks (roughly corresponding to multiple intensity maxima), which propagate with slightly different group velocities \(v_g\). New optical frequencies are generated through the scattering of incident fields of these “material waves” to produce a third wave. An incident optical wave at \((\omega, \vec{k}(\omega))\) will predominantly scatter into the wave component at \((\Omega, \vec{s}(\Omega))\) that satisfies the phase matching condition

\[
\sqrt{\frac{\Omega^2\varepsilon(\Omega)}{c^2}} - s_\perp^2 = \frac{(\Omega - \omega)}{v_g} + \sqrt{\frac{\omega^2\varepsilon(\omega)}{c^2} - k_\perp^2},
\]

\(s_\perp = k_\perp + m_\perp,\)  

(4)
where $m_\perp$ is the transverse Fourier component of the material wave. As explained in Ref. [24], this phase matching condition is not strictly enforced, but nevertheless, it is useful in identifying the loci in the spectral domain where the new spectral components are predominantly generated.

In the present case, despite strong white-light generation most of the spectral energy is concentrated in the vicinity of the original carrier frequency $\omega_0$ around zero transverse wavenumber. Therefore, we set $(\omega, \mathbf{k}(\omega)) = (\omega_0, k_0)$ for the incident optical field in Eq. (1) together with $m_\perp = 0$ to find the loci $(\Omega, \mathbf{s}(\Omega))$ where the three-wave mixing is near phase-matched and most effective. To solve Eq. (4) we need to estimate the group velocity $v_g$ for the strongest response peaks. We have obtained these estimates from the raw data for the on-axis value of the material response $\Delta \chi^{(2)}(r', t)$ by measuring the shift of the response peaks for both split daughter pulse with propagation distance. As a result we obtained two solutions to Eq. (4) corresponding to the scattering on the two strongest response peaks. The loci $(\Omega, \mathbf{s}(\Omega))$ where the three-wave mixing is most effective according to the above prescription are shown as dashed-lines in Fig. 4. The short-dashed line represents scattering from the material response generated by the leading daughter pulse, while the long-dashed line is due to trailing edge daughter pulse, and the phase-matching curves clearly coincide with the strong features of the spectrum. Thus, our phase matching argument captures the central X shape perfectly, including the curvature of its arms (due to nonparaxial effects), and it furthermore explains the occurrence of the low- and high-frequency “ridges” in the spectrum. An important point is that each half of the X is actually generated by one daughter pulse. If we used the group velocity $v_g(\omega_0)$ taken at the central wavelength instead of the actual $v_g$’s, we would obtain a single X-shaped locus expected for “normal” X-waves. This turns out to have the same center, and a small difference in the angles between the arms are due to $v_g \neq v_g(\omega_0)$. Thus, even after multiple pulse splitting, the split-off pulses “cooperate” and contribute to the intensity spectrum in the same X-shaped region in the $(\omega, \mathbf{k})$ space. This is the origin of the universality and robustness of the X-waves, namely, irrespective of the details of the temporal dynamics, they tend to concentrate their spectral energy around the locus that supports the $z$-invariant propagating waves. Though they may not form $z$-invariant light bullets, the X-wave character therefore clearly manifests itself in long-distance propagation in water. Finally, we note that the above argument doesn’t depend on details of the nonlinearity or of the linear dispersion. Thus, dynamic X-waves should be inherent to many systems, though whether their signature can be observed depends on the relative magnitudes of other competing effects and initial pulse conditions.

In summary, we have provided diagnostic numerical simulations to show that the recent experimental observations long distance propagation in water [1] may be interpreted by combining the paradigms of both nonlinear pulse splitting and dynamic X-waves that develop from the split pulses and replenish the pulse center. Our results therefore reveal intimate connections between long distance propagation in condensed matter and nonlinear electromagnetic X-waves.

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