Highly Noninstantaneous Solitons in Liquid-Core Photonic Crystal Fibers

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The nonlinear propagation of pulses in liquid-filled photonic crystal fibers is considered. Because of the slow reorientational nonlinearity of some molecular liquids, the nonlinear modes propagating inside such structures can be approximated, for pulse durations much shorter than the molecular relaxation time, by temporally highly nonlocal solitons, analytical solutions of a linear Schrödinger equation. The physical relevance of these novel solitons is discussed.

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Introduction.—Highly nonlocal solitons were originally introduced as an ''accessible'' toy-model for describing self-trapped optical beams [1]; but this was followed by experimental demonstrations [2], also driven by early works on plasma physics, Bose-Einstein condensation and dissipative systems [3–10], which unveiled the fundamental role of nonlocality in spatially self-trapped waves. Indeed, nonlocality allows stabilization with respect to collapse and the existence of a rich class of propagation-invariant waves [11,12]; in addition, applications such as light-steering and all-optical logic gates [13] and in soft-matter and thermal liquids [14–16] have been demonstrated. Recently, insights from spatially nonlocal nonlinear waves also emerged in the temporal domain [17,18]. However, the relevance of temporal nonlocality is largely limited by the unavoidable instantaneous Kerr effect, as, e.g., for silica glass in fiber optics, where ''nonlocal'' Raman-like terms, although leading to important consequences as the Raman self-frequency shift (RSFS) of solitons [19], can be considered as small perturbations. Recent fabrication advances, however, open up innovative perspectives. Indeed, microstructured photonic crystal fibers (PCFs, see Ref. [20]) may be fabricated with a central hole filled by a material displaying noninstantaneous response as, e.g., molecular liquids with large reorientational effects [21,22], with instantaneous nonlinearities acting as small perturbations: exactly the opposite situation of the silica glass.

In this Letter we show that a completely novel class of solitary waves exists in these new fibers, which are described by an essentially linear model. These localized waves are shown to display truly remarkable properties, such as robustness with respect to noise perturbations and collisions, and emission of phase-matched dispersive waves—phenomena that are typically associated to purely nonlinear temporal waves only. In addition they are shown to be not affected by the RSFS if the response time of the molecular reorientation is slow enough. As a result, these highly noninstantaneous fibers can be used in a new variety of optical devices and applications, also including quantum information processing.

Reorientational nonlinearities.—Suitable liquids with large reorientational nonlinearities are composed by small molecules, as spectroscopy solvents, available in extremely high purity with large transparency windows (from 0.5 to 3 μm) [23]. An in-depth survey shows that best potential candidates are nitrobenzene ($C_6H_5NO_2$), toluene (C_7H_8), and carbon disulfide (CS_2), which all possess small cigar-shaped molecules with low Kerr nonlinearity ($C_6H_5NO_2$: 671×10^{-16} cm^2/W, C_7H_8: 168×10^{-16} cm^2/W, CS_2: 340×10^{-16} cm^2/W; all at 1064 nm [24]) and refractive indices larger than silica. $C_6H_5NO_2$ and C_7H_8 have low vapor pressure at room temperature and are easy to handle. CS_2, in contrast, is volatile, but has the best transparency due to its simpler composition. We have performed calculations for CS_2, whose physical properties are well documented [25], and which has been recently considered for enhanced supercontinuum generation in PCFs [21,26]. We stress that our results are not restricted to one specific liquid.

Theory.—The dimensionless generalized nonlinear Schrödinger equation (GNLSE) is written as:

$$i\partial_z A + \frac{1}{2} s \partial_t^2 A + A \int_{-\infty}^{+\infty} R(t-t')|A(t')|^2 dt' = 0, \quad (1)$$

where $R(t) \equiv \Theta(t)h(t)$ is the response function, with $h(t) \equiv e^{-t/T}/T$ the CS_2 reorientational nonlinearity, $Tt_0 = 1.68$ ps [25], $\Theta(t)$ the Heaviside function, $A(z, t)$ the field envelope, $s = +1$ ($s = -1$) denotes anomalous (normal) group velocity dispersion (GVD), z the propagation coordinate (scaled with the second-order dispersion length of the fiber, see Ref. [19]), and t is time (scaled with the pulse duration t_0). $R(t)$ must be normalized such that $\int_{-\infty}^{+\infty} R(t)dt = 1$. In the following, we will consider the case $s = +1$; when $s = -1$ interesting effects such as wave-breaking in the highly nonlocal limit can be investigated and will be reported elsewhere. The shock term is not considered in Eq. (1) since the GVD will always completely dominate the self-steepening effect for a pulse duration $t_0 > \lambda/c$, which is also the condition of validity of the slowly varying envelope approximation [19].

Because of the large transparency bandwidth of CS_2, we can safely neglect linear losses [23].

For a response time much shorter that the pulse width, $T \equiv \int_{-\infty}^{+\infty} tR(t)dt \ll 1$, [19] one can use the property $\int_{-\infty}^{+\infty} R(t-t')|A(t')|^2dt' \equiv \int_{-\infty}^{+\infty} R(t')|A(t-t')|^2dt'$ to expand the envelope in a Taylor series: $|A(t-t')|^2 \simeq |A(t)|^2 - t'\partial_t|A(t)|^2 + \cdots$. By Eq. (1), we obtain the well-known Raman NLSE: $i\partial_z A + \frac{1}{2}s\partial_t^2 A + |A|^2A - TA\partial_t|A|^2 = 0$, where the last term models the Raman effects on long pulses. In silica fibers this approximation is quite good for ps pulses, since $Tt_0 \sim 2$ fs. A completely different scenario occurs when $T \gg 1$, i.e., when the pulse duration is much shorter than the response time of the medium and one expands in a Taylor series the response function. This is a more delicate procedure since $R(t)$ is discontinuous in $t = 0$ due to causality. At the first order, $\int_{-\infty}^{+\infty} R(t-t')|A(t')|^2dt' = \int_{-\infty}^{t} h(t-t')|A(t')|^2dt' \simeq \int_{-\infty}^{t}[h(t) - t'(\partial h/\partial t)]|A(t')|^2dt' = h(t)\mathcal{E}(t) - (\partial h/\partial t)\mathcal{A}(t)$, where $\mathcal{E}(t) \equiv \int_{-\infty}^{t}|A(t')|^2dt'$ is the partial pulse energy and $\mathcal{A}(t) \equiv \int_{-\infty}^{t} t'|A(t')|^2dt'$ is the partial pulse temporal asymmetry. We stress that, despite the presence of the Heaviside function in $R(t)$, all the integrals above are continuous and derivable $\forall\ t$. For each derivative of $h(t)$ in the Taylor series there is a factor $1/T$: higher-order derivative terms become increasingly negligible for $T \to \infty$, and always continuous in $t = 0$. Maintaining only the 0th order term, for $T \gg 1$, the integral in Eq. (1) tends asymptotically to $\mathcal{E}R(t)A$ (discontinuous in $t = 0$) and we obtain

$$i\partial_z A + \frac{1}{2}s\partial_t^2 A + \mathcal{E}R(t)A = 0, \tag{2}$$

where $\mathcal{E} \equiv \mathcal{E}(t \to +\infty)$ is proportional to the total number of photons launched into the fiber. In the highly noninstantaneous limit, the GNLSE (1) is equivalent to a linear Schrödinger equation (with time and space interchanged with respect to quantum mechanics): the response time is so long that the system "remembers" the total injected energy, while the pulse is propagating. As a result, the pulse feels the liquid exponentially decaying response as a linear potential, whose depth is determined by the energy $\mathcal{E}$. Note that, however, the superposition principle is not valid for Eq. (2) in the general case.

Localized states.—Being the exponential well among the solvable potentials, the solution of (2) is known from standard quantum mechanics. Solitonlike bound states are found by solving the equation for $t < 0$ and $t > 0$, and then by imposing continuity of the envelope and its derivative at $t = 0$; this provides the wave number $\beta > 0$. Letting $A(z,t) = a(t)\exp(i\beta z)$ leads to $(t > 0)$

$$\frac{1}{2}\partial_t^2 a + \frac{\mathcal{E}}{T}\exp(-t/T)a = \beta a. \tag{3}$$

For $t < 0$ the localized solution is $a = \mathcal{N}\exp(\sqrt{2\beta}t)$ where $\mathcal{N}(\mathcal{E})$ is a normalization constant. Considering the solutions of (3) vanishing as $t \to \infty$ leads to

$$a = \mathcal{N}\frac{J_\nu(\sqrt{8\mathcal{E}T}e^{-t/2T})}{J_\nu(\sqrt{8\mathcal{E}T})} \tag{4}$$

with $\nu = \sqrt{8\beta}T$. For large t, a decays as $\exp(-\nu t/2T)$. The allowed eigenvalues β are implicitly given by

$${}_0F_1(;\sqrt{8\beta}T; -2\mathcal{E}T) = 0, \tag{5}$$

where ${}_0F_1(;a;z) = \sum_{n=0}^{\infty} z^n/[n!(a)_n]$ is the confluent hypergeometric limit function, and $(a)_n \equiv \Gamma(a+n)/\Gamma(a)$ is the rising factorial. Equation (5), which provides the dispersion relation $\beta(\mathcal{E},T)$, can only be solved numerically. The constant $\mathcal{N}$ cannot be written explicitly, and is found by requiring that the total soliton energy is $\mathcal{E}$. Solutions can be classified by counting the number of nodes m. We shall call the "fundamental" state the solution with $m = 0$, which has the largest β and the shortest pulse duration. For small $\mathcal{E}$, Eq. (5) can be expanded to give $\beta \simeq \mathcal{E}^2/2$, which shows that $\beta = \mathcal{E} = 0$ is also a solution. For large values of $\mathcal{E}$, when $m = 0$, one has $\beta \simeq (2/\pi)^2\mathcal{E}/T$. "Moving" solitons can be found by the solutions of Eq. (3) with $2\beta \to 2\beta - v^2$, and by multiplying the fields by $e^{-iv t}$, where v is an extra parameter associated with the soliton velocity $(\beta > v^2/2)$. For $|t|$ large, the field decays as $a \sim \exp(-\sqrt{2\beta}|t|)$, this means that the original approximation for Eq. (2) is verified *a posteriori* only if $\beta \gg 1/(2T^2)$, or $\mathcal{E} \gg \pi^2/(8T)$.

Figure 1(a) shows the profiles of some analytical solutions of Eq. (2). Figures 1(b) and 1(c) show the dependence of β on $\mathcal{E}$ and T for fixed T and $\mathcal{E}$ respectively, as calculated by solving Eq. (5) numerically. Approximate expressions in terms of $\mathcal{E}$ and T are given below.

RSFS.—The solutions of the model (2), being linear, are stable against small noise fluctuations, as shown in Fig. 2(a) where we report, e.g., the propagation of the higher-order soliton in Fig. 1(a) with 5% noise. If the

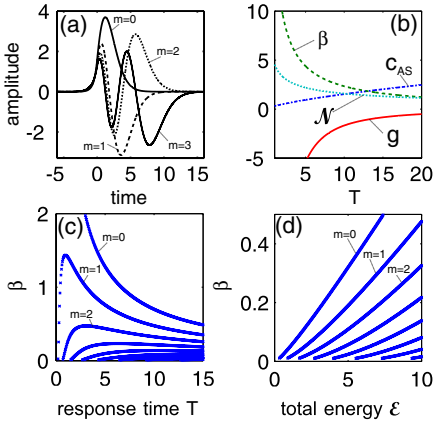

FIG. 1 (color online). (a) Localized solutions of Eq. (2) for $T = 10$, $\mathcal{E} = 30$, for $\beta = 2.33614$ $(m = 0)$, 1.82551 $(m = 1)$, 1.45665 $(m = 2)$ and 1.16691 $(m = 3)$, calculated by using Eq. (5). (b) $\mathcal{N}$, c_{AS}, g, β as functions of T, for $\mathcal{E} = 30$. (c) $\beta(T)$ for fixed $\mathcal{E} = 10$. (d) $\beta(\mathcal{E})$ for fixed $T = 10$.

same solution is propagated inside the full model (1), we observe that the pulse is subject to a Raman shift, which is not described by Eq. (2). We developed a general theory of RSFS of solitons for Eq. (1) by writing Eq. (1) as $i\partial_z A + \frac{1}{2}s\partial_t^2 A + \mathcal{E}R(t)A + [\int R(t-t')|A(t')|^2 dt' - \mathcal{E}R(t)]A = 0$ and treating the term under square brackets as a perturbation of the stationary states. One finds that the soliton central frequency shifts according to $\Omega(z) = -(z/\mathcal{E}) \times \int dt|A(t)|^2 \partial_t[\int R(t-t')|A(t')|^2 dt' - \mathcal{E}R(t)] \simeq c_{\rm AS} z \int dt[\partial_t^2 |A(t)|^2]R(t)$, where $c_{\rm AS} \equiv \mathcal{A}/\mathcal{E}$ is the asymmetry coefficient. The final result is ($x \equiv \sqrt{8\mathcal{E}T}$):

$$\Omega = \frac{c_{\rm AS} z \mathcal{N}^2}{J_\nu(x)^2 T} \int_0^\infty e^{-t/T} \partial_t^2 J_\nu^2(x e^{-t/2T}) dt \cong -\frac{32}{\pi^7}\frac{\mathcal{E}^2}{T} z, \quad (6)$$

which shows that the RSFS of these solitons depends on the asymmetry of their temporal profiles. The last expression in (6) is valid for large $\mathcal{E}$ and T (where $\mathcal{N}^2 \cong 2\mathcal{E}/\pi^2 T$ and $c_{\rm AS} \cong T/\pi^2$), and illustrates how, as the degree of nonlocality T grows, the Raman shift is inhibited, while also growing with the energy. The rate of RSFS $g \equiv \Omega/z$ and the asymmetry coefficient $c_{\rm AS}$ of the stationary states are shown in Fig. 2 in terms of T and $\mathcal{E}$ for the fundamental soliton solutions.

Dispersive resonant radiation.—In the presence of higher-order dispersion terms in Eq. (2), the highly noninstantaneous solitons can resonantly emit dispersive radiation at well-defined frequencies, analogously to what occurs for Schrödinger solitons in solid-core PCFs [27]. By substituting $A(z,t) = [F(t) + f(z,t)]e^{i\beta z}$ into Eq. (2), where $F(t)$ is the localized state profile and f is the small dispersive radiation amplitude, assuming that the response time T is large in comparison with the localized state duration, and keeping only the first order term in f, we have: $[i\partial_z - \beta + \hat{D}(i\partial_t) + \mathcal{E}R(t)]f = -\hat{D}_H(i\partial_t)F$, where $\hat{D}(i\partial_t) \equiv \frac{1}{2}\partial_t^2 + i\alpha\partial_t^3$ and $\hat{D}_H \equiv i\alpha\partial_t^3$, and α is the third-order dispersion coefficient, the only one that we include here. At phase matching, the radiation and soliton have the same β and $f(\omega) \simeq S(\omega)/[D(\omega) + \mathcal{E}R(\omega) - \beta]$, where $S(\omega)$ is the Fourier transform of the source term $-\hat{D}_H(i\partial_t)F$. This yields the condition $D(\omega) + \mathcal{E}R(\omega) \simeq \beta$, which determines the resonant frequency ω_R. The energy-dependent part is an extra contribution to the resonant condition that is unique for highly noninstantaneous solitons, and allows us to tune the frequency position of the emitted radiation by adjusting the total input pulse energy.

Kerr nonlinearity.—An important issue concerns the effect of a residual instantaneous Kerr nonlinearity. This may be due to the Kerr effect of the fiber cladding, or of the liquid itself. As shown in panels 3(a) and 3(b) for a sechlike input pulse (i.e., an input that is not matched with the profile of the fundamental state), in the absence of the Kerr effect [Fig. 3(a)], the propagation largely resembles a standard fundamental soliton. Conversely, increasing the contribution of the Kerr effect, measured by a coefficient r, which is the relative weight of the instantaneous versus the noninstantaneous part of the nonlinearity (of the order of ~0.1 in liquids) [Fig. 3(b)], induces a characteristic oscillation of the central position of the pulse in the temporal and spectral domains. This is due to the coupling between higher-order localized states excited in the fiber, as induced by the Kerr nonlinearity perturbation. Details will be given elsewhere.

Liquid-filled PCF.—CS_2 has a relatively large refractive index around 1.6–1.7. If used in a standard silica step-index fiber, the field would be considerably localized in the liquid, and our calculations show that it will be not possible to obtain anomalous dispersion in the optical region. By a

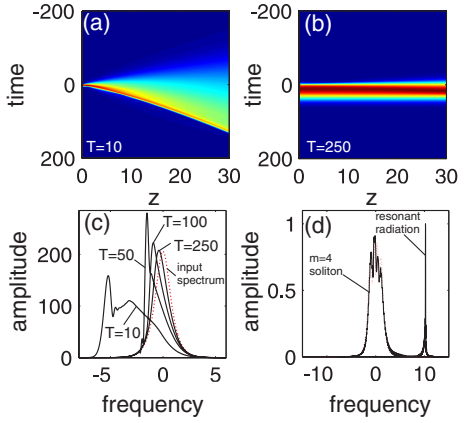

FIG. 2 (color online). (a) Propagation of 1-soliton in Eq. (1), $T = 10$, $\mathcal{E} = 30$, $\beta = 2.33614$. (b) Same as (a) but for $T = 250$, $\mathcal{E} = 30$, $\beta = 0.109846$. (c) Various final spectra showing reduction of RSFS ($\mathcal{E} = 32$, $T = 10$, 50, 100, 250). (d) Resonant radiation in reduced model for $\alpha = 0.05$ (for $m = 4$ state).

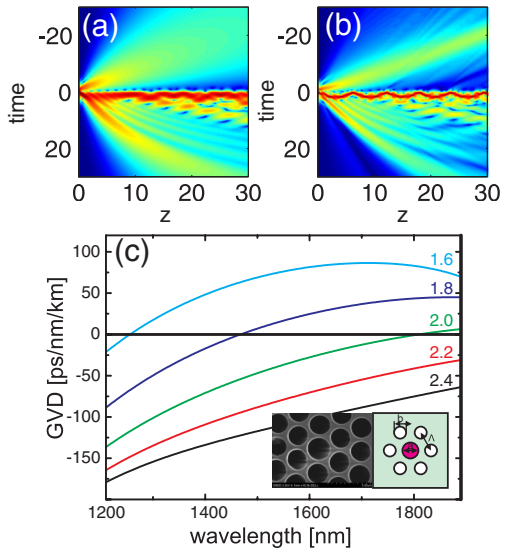

FIG. 3 (color online). (a),(b) Soliton oscillations due to a small Kerr term [$r = 0$ in (a) and $r = 0.11$ in (b), $T = 10$, input pulse $A = N\text{sech}(t)$ with $N = 5$], in Eq. (2). (c) GVD of the fundamental mode (HE_{11}) of a CS_2-filled PCF. Fiber parameters are with $a = 1.6$ μm, $b = 1.2$ μm (see inset). Each curve refers to different hole spacing Λ expressed in μm.

single ring of air holes in the silica cladding, however, the zero-GVD point can be largely shifted in the region where experiments are most easily carried out (i.e., $\lambda < 2\ \mu$m). In Fig. 3(c) we report the GVD of various CS_2-filled core silica PCFs (with a single ring of holes arranged in a triangular cladding lattice as shown in the inset) for different values of the pitch Λ. Signatures of the existence of highly noninstantaneous solitons are the Kerr-induced oscillations shown in Fig. 3(b), and the reduction of the Raman self-frequency shift when gradually decreasing the pulse-duration t_0 from the ps to the fs regime, opposite to what happens in silica fibers.

Conclusions.—We predict the existence of a novel class of temporally localized waves propagating inside microstructured fibers with a central core filled by nonlinear liquids with a slow reorientational nonlinearity. Surprisingly, these waves behave very much like solitons—albeit being the solution of a linearized equation—and are sustained by a strongly nonlocal temporal response, due to a pronounced Raman-like effect induced by reorientational nonlinearity of cigar-shaped molecules. By borrowing concepts from nonlocal spatial solitons, we find that these objects are very robust with respect to noise, caused by (for example) amplified spontaneous emission. Highly noninstantaneous solitons may thus support the development of novel classes of lightwave fiber systems and novel soliton based lasers, as well as opening up a new route towards quantum solitons and multidimensional solitary waves sustained by a highly noninstantaneous nonlinearity. In addition, by exploiting the nonlinear coupling between highly noninstantaneous solitons, we believe that it will be possible to control and enhance supercontinuum generation in novel and unexpected ways.

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