Linear and nonlinear optical precursors in inhomogeneously broadened two-level media

Robert Marskar* and Ulf L. Österberg†

Department of Electronics and Telecommunication, Norwegian University of Science and Technology, N-7491 Trondheim, Norway

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By solving the two-level Maxwell-Bloch equations for resonant pulses numerically, we observe precursors in a regime where the area theorem is fulfilled. The precursors are 0π pulses traveling at a velocity close to the speed of light, and the main signal is a self-induced transparency (SIT) 2π soliton traveling at a much lower velocity. The manifestation of the precursors is strongest when the input pulse duration is on the same order as the inhomogeneous lifetime and the input area is close to π. Depending on the relationship between the input pulse duration and the inhomogeneous lifetime, the precursors can interact linearly or nonlinearly with the material. Experimental confirmation of these results should offer a direct and reasonably straightforward way of measuring optical precursors.

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Brillouin and Sommerfeld, in their century-old theoretical study of superluminal group velocity in causal, linear dielectrics [1], showed that a precursor (or forerunner) can significantly precede a main signal (in their studies, they used a single resonance Lorentz medium and a step pulse). Although the theoretical literature on optical precursors has been improved over the years and is now quite extensive [2], only a handful of experimental observations have been made: Aaviksoo and co-workers [3] used one-sided exponential pulses on a narrow excitation line in GaAs, Jeong *et al.* [4] observed precursors by the propagation of steplike pulses close to a single Lorentz resonance, Du *et al.* [5] observed precursors at the biphoton level using slow light, and Wei *et al.* [6] performed precursor experiments with electromagnetically induced transparency in a cloud of laser-cooled rubidium atoms. There have also been reports of precursors in water [7], although their existence has been contested [8,9]. Recently, Macke and Ségard have suggested the existence of precursors with self-induced transparency (SIT) in a homogeneously broadened absorber [10]. This was also investigated by Crisp in his theoretical study of propagation of step-function pulses in two-level absorbers and amplifiers [11]. We also mention the work by Diels and Hahn [12], who showed that precursorlike phase-modulated pulses occur for off-resonant pulse propagation within the absorption line in a ruby rod. For comparison, in this paper we suggest the excitation of precursors by the application of a resonant pulse whose spectrum can be broader or narrower than the inhomogeneous absorption line. We show that self-induced transparency is responsible for a large temporal separation between the precursor and the 2π soliton main pulse. For on-resonance input pulses that have no phase modulation, the pulse area is well defined and we show that our precursors are 0π pulses. These precursors are also free of chirp, and can interact linearly or nonlinearly with the material.

We write the real-space electric field as $E(t,z) = \mathcal{E}(t,z)e^{-i\omega_0(t-z/c)} + \text{c.c.}$, where $\mathcal{E}(t,z)$ is the complex pulse envelope. If the spectral bandwidth of $\mathcal{E}(t,z)$ is sufficiently narrow to interact with only a single resonance in a material, the interaction between the field and the material is described by the Maxwell-Bloch model, which can be summarized as [13]

$$i\partial_\tau \rho = [H,\rho], \tag{1a}$$

$$\partial_z \Omega = i\mu\langle\rho_{21}\rangle_\Delta, \tag{1b}$$

$$H = \begin{pmatrix} 0 & -\frac{1}{2}\Omega^* \\ -\frac{1}{2}\Omega & \Delta \end{pmatrix}, \tag{1c}$$

where we have made the rotating-wave approximation (RWA) and the slowly varying envelope and phase approximation (SVEA). The 2×2 density matrix of the system in the frame rotating at ω_0 is denoted by $\rho = \rho(t,z,\Delta)$, and the Rabi frequency is denoted $\Omega(\tau,z) = \frac{2d}{\hbar}\mathcal{E}(\tau,z)$, where d is the transition dipole moment. The transition frequency is denoted by ω_0, τ is the local time $t - z/c$, and Δ is the atomic detuning. The brackets are a shorthand notation for the sum over all detunings such that $\langle(\cdots)\rangle_\Delta = \int g(\Delta)(\cdots)d\Delta$, where $g(\Delta)d\Delta$ is the fraction of atoms that are detuned an amount Δ from the line center. For a Gaussian velocity distribution, $g(\Delta)$ can be taken as $g(\Delta) = \frac{T_2^*}{\sqrt{2\pi}}\exp[-(\Delta T_2^*)^2/2]$, where $\Delta = \omega_0 v/c$ is the Doppler shift for an atom moving at a velocity v, and T_2^* is the inhomogeneous relaxation time of the material. The coupling constant μ is equal to $Nd^2\omega_0/(\hbar c\epsilon_0)$, where N is the atomic number density. Homogeneous broadening is disregarded in Eq. (1) on the grounds that it occurs on a time scale much slower than the ones we consider here.

Self-induced transparency, the phenomenon where a pulse is strong enough to make itself transparent to a medium, was first discovered by McCall and Hahn [14], who showed that resonant hyperbolic secant (h.s.) pulses that had areas $\theta(z) = \int_{-\infty}^{\infty} d\tau\,\Omega(\tau,z) = 2\pi$ propagated through the medium without loss. SIT was later experimentally observed by Gibbs and Slusher [15]. The general solution to Eq. (1) was later derived by Ablowitz *et al.* [16] by use of the inverse scattering transform (IST). They showed that the inhomogeneously broadened medium acts as a filter for the scattering eigenvalues of the initial pulse. In this regard, a soliton is never created by the medium but is already present in the initial pulse, and appears only when the inhomogeneously broadened medium has peeled away all the parts of the initial pulse that is not a soliton. An inhomogeneous linewidth that is relatively broad

*robert.marskar@iet.ntnu.no
†ulf.osterberg@iet.ntnu.no

compared to the pulse spectrum is crucial to this reshaping. We also point out that the linearization of Eq. (1) (including homogeneous relaxations) is precisely the equation set that has been traditionally used for the study of optical precursors [17,18]. The work by Ablowitz *et al.* [16], Diels and Hahn [12], and Kaup [19] has largely been disregarded in the context of precursors, and only recently has the search for precursors been extended to the nonlinear regime [10,20].

The solutions in Refs. [16] and [19] are difficult to make use of in the present study because the evolution of the continuous eigenvalue spectrum cannot be calculated in general. Nonetheless, the conditions under which a soliton and a precursor can coexist in a two-level medium are fairly simple to understand on an intuitive level. First of all, the precursor pulse and the soliton should be temporally separated, and also be the only two pulses that propagate in the medium. Second, to be comparable with the linear theory, the precursor must eventually be absorbed by the material and can therefore not itself be a soliton. Third, we are forced to consider only input pulses that are not already SIT pulses, as it is only the continuous eigenvalue spectrum that manifests itself as a precursor. Therefore, we only consider input pulses with an initial area between π and 3π that are not h.s. pulses. Kaup [19] has shown that for a Gaussian input pulse of a given duration, the amplitude of the emerging soliton increases monotonically with increasing input area. The duration of the soliton also increases monotonically with increasing input pulse duration, when the input area is fixed. The same is true for box-shaped input pulses, or raised Gaussians. It is also known, even for other nonlinear wave equations such as the Korteweg–de Vries or the nonlinear Schrödinger equation, that the amplitude, duration, and velocity of nonlinear wave forms are coupled. A larger amplitude soliton is narrower and travels faster. Thus, a slow soliton, and consequently a large separation of the precursor and the main signal, can be achieved by considering an input pulse with area close to, but larger than, π.

In the following, we solve Eq. (1) numerically by using a predictor-corrector method. For the physical parameters, we take the transition wavelength $\lambda = 800$ nm, the inhomogeneous lifetime $T_2^* = 0.5$ ns, and a number density $N = 10^{17}$ m^{-3}. The dipole moment is taken as $d = 2.75 \times 10^{-29}$ C m, corresponding to a Beer's length $\alpha^{-1} \approx 2.5$ mm, where the absorption coefficent is $\alpha = \pi\mu g(0)$ [13]. The length of our material is $L = 8$ cm, giving an optical density of $\alpha L \approx 32$, and we assume that all atoms are in their ground state before the pulse enters. As input pulses, we first use transform-limited Gaussians $\mathcal{E}(\tau,0) = E_0 \exp[-\tau^2/(2\tau_0^2)]$ with area 1.1π, and we perform computer simulations where τ_0/T_2^* varies in the range of 0.15–2. Our Gaussian pulses are abruptly turned on at $\tau = -10\tau_0$ and therefore have very small turn-on step values.

At first, we keep the area constant at $\theta_0 = 1.1\pi$ and change the pulse duration from $\tau_0 = 0.15T_2^*$ to $\tau_0 = 2T_2^*$. Since the area is proportional to the field spectrum value on resonance, $\theta(z) = \frac{2d}{\hbar}\int_{-\infty}^{\infty}\mathcal{E}(t,z)dt = \frac{d}{\hbar}E(\omega_0,z)$, decreasing the pulse duration while keeping the area constant means that we are increasing the energy of the spectral wings of the input pulse (the energy of the pulse as a whole increases too). The rows in Fig. 1 show the exit pulses after propagating through the material, and also the phase $\psi = \arctan[\mathcal{E}_I(t,z)/\mathcal{E}_R(t,z)]$,

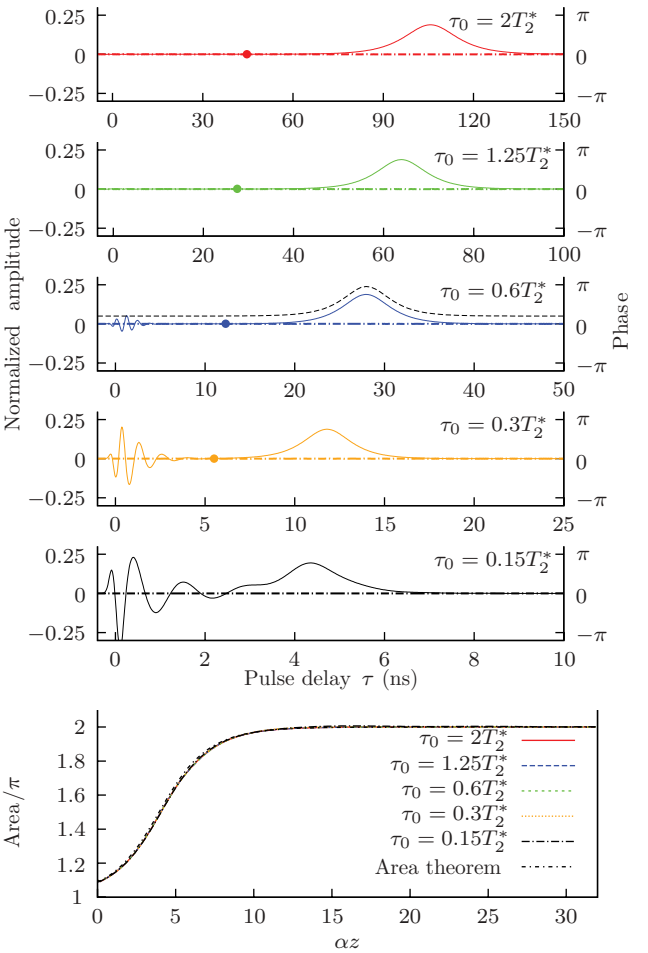

FIG. 1. (Color online) Top five figures: Amplitude (solid line) and phase (dashed-dotted line) of the exit pulses for input pulses with durations $\tau_0 = 0.15T_2^*$, $0.3T_2^*$, $0.6T_2^*$, $1.25T_2^*$, and $2T_2^*$. The dashed line for $\tau_0 = 0.6T_2^*$ shows a h.s. fit to the main pulse, and is displaced along the y axis for readability. The y axis is normalized against the individual input peak amplitudes. Bottom plot: Area evolution of the pulses for the various input pulse durations (input area fixed at 1.1π).

where $\mathcal{E}(t,z) = \mathcal{E}_R(t,z) + i\mathcal{E}_I(t,z)$, which shows that the exit pulses are not phase modulated. We have verified that this is true in all of our simulations. As noted by Hopf and Scully [21], the absence of phase modulation is readily understood from the formal solution for $\rho_{21}(t,z,\Delta)$:

$$\rho_{21}(\tau,z,\Delta) = \frac{i}{2}\int_{-\infty}^{\tau} d\tau'\Omega(\tau',z)e^{-i\Delta(\tau-\tau')} \times [\rho_{22}(\tau',z,\Delta) - \rho_{11}(\tau',z,\Delta)]. \quad (2)$$

Under the conditions of exact resonance, no initial chirp, and symmetric $g(\Delta)$, then the in-phase component of the polarization vanishes after taking the ensemble average $\langle\cdot\rangle_\Delta$ of Eq. (2). Insertion of this result into Eq. (1b) then shows that the field propagates without acquiring a modulated phase. The envelopes plotted in Fig. 1 are normalized to the individual peak amplitudes $E_0 = 1.1\pi\hbar/(2\sqrt{2\pi}d\tau_0)$, and show that the magnitude of the precursor decreases when the input pulse duration increases. For the longest pulses there are no immediate signs of a precursor and only the main

signal can be clearly seen (although it is not discernible in Fig. 1, there is an oscillating front with a relative peak amplitude 2.6×10^{-4} for $\tau_0 = 2T_2^*$, and a front with a relative peak amplitude 2.5×10^{-3} for $\tau_0 = 1.25T_2^*$). By curve fitting a hyperbolic secant shape $\mathcal{E}(t) = A\mathrm{sech}[(t - t_0)/t_p]$ to the numerical results, we have confirmed that the main pulses (apart from $\tau_0 = 0.15T_2^*$) are h.s. pulses. The parameters that give the elevated black dashed curve for $\tau_0 = 0.6T_2^*$ in Fig. 1 are $A = 0.189E_0 \approx 1.7$ kV/m, $t_0 = 28.0$ ns, and $t_p = 2.3$ ns, and gives a pulse velocity $u \approx c/106$ and area $\theta \approx 2\pi$. The pulse velocity is estimated from the peak delay τ_d and is given by $u = c/(1 + c\tau_d/L)$. As the initial pulse duration decreases, there are much clearer signs of a pulse propagating at a velocity close to c with the trailing main pulse propagating at a lower velocity. For the shortest of these pulses ($\tau_0 = 0.15T_2^*$), we cannot yet distinguish between the main pulse and the precursor. A comparison of the area evolution of these pulses to the area theorem is shown in the bottom plot in Fig. 1. It is not surprising that the area theorem is fulfilled, even though the conditions under which the area theorem was first derived are clearly violated. As Kaup [19] has shown, the proper generalization of the area theorem for unchirped pulses is

$$\partial_z \ln\tan\frac{1}{2}\theta(z) = \mp\left(\frac{1}{2}\alpha + \frac{\mu}{2i}\mathrm{P}\int_{-\infty}^{\infty} g(\Delta)\frac{d\Delta}{\Delta}\right), \qquad (3)$$

where P indicates the Cauchy principal value integral. When $g(\Delta)$ is symmetric then the integral vanishes, leaving only the integrated form of the McCall-Hahn area theorem

$$\tan\tfrac{1}{2}\theta(z) = e^{\mp\alpha z/2}\tan\tfrac{1}{2}\theta_0. \qquad (4)$$

Thus, the area theorem holds whenever the initial pulse is unchirped and resonant, and the inhomogeneous broadening line is symmetric. We also mention a rederivation of the area theorem by Eberly [22], who showed that the area theorem holds even for chirped pulses, provided that the inequality $T_2^* \ll \tau_0$ is satisfied. The results in Fig. 1 are similar to the behavior of optical precursors in the linear theory, in the qualitative sense that they are both highly dependent on the placement of the initial pulse spectrum relative to the absorption band of the material.

To investigate linearity, spectrum, and area of the precursors in Fig. 1, we perform a frequency analysis of these pulses. We split the field into two contributions, $\mathcal{E}(\tau, L) = \mathcal{E}_p(\tau, L) + \mathcal{E}_m(\tau, L)$, where $\mathcal{E}_p(\tau, L)$ is the precursor pulse, defined as $\mathcal{E}(\tau, L)$ up to the breakpoints shown in Fig. 1 and zero otherwise. The main pulse is denoted by $\mathcal{E}_m(\tau, L)$ and is zero for all times before the breakpoint, and equal to $\mathcal{E}(\tau, L)$ for times after. This decomposition of the electric field is artificial and can only be performed once the separation between $\mathcal{E}_p$ and $\mathcal{E}_m$ is clear. The separation occurs in all of our computer simulations except when the input pulse duration becomes much shorter than T_2^*. For example, there is no clear distinction between the precursor and the main signal for $\tau_0 = 0.15T_2^*$ in Fig. 1 after 32 Beer lengths, but we have verified that there is after a distance of 60 Beer lengths.

The two plots in Fig. 2 show the reshaping of the input pulse with $\tau_0 = 0.6T_2^*$, and the spectrum of this pulse at the exit of the material. After a propagation depth of $\alpha z \sim 16$, the two pulses are clearly separated. The spectral amplitude of the precursor at ω_0 is clearly zero, and it is therefore a 0π

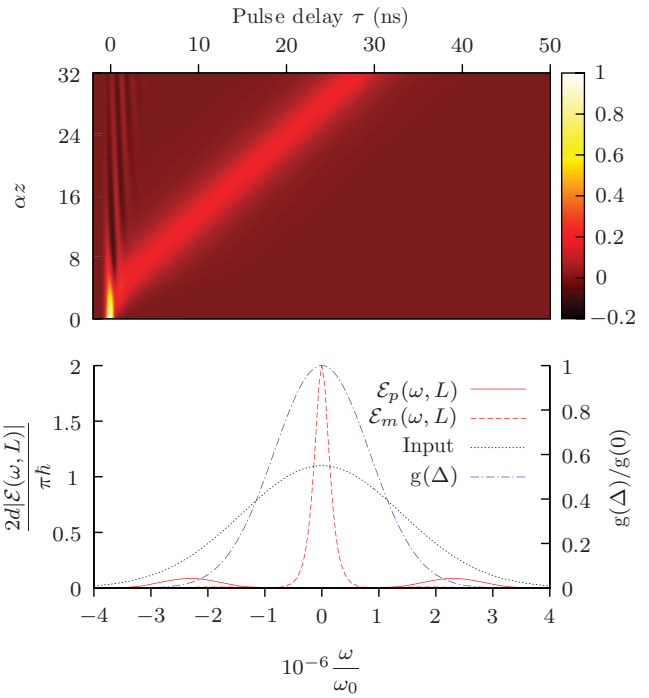

FIG. 2. (Color online) Top: Reshaping of a Gaussian input pulse with duration $\tau_0 = 0.6T_2^*$ and initial area 1.1π. The color coding shows the normalized amplitude of the field. Bottom: Spectral magnitude of the exit pulse after propagating a distance $\alpha L = 32$. The solid line shows the spectrum of the precursor, the dashed line shows the spectrum of the soliton, and the dotted line shows the spectrum of the Gaussian input pulse. The dashed-dotted line shows the inhomogeneous absorption line of the medium.

pulse (the area is given by the dc value in Fig. 2). Moreover, the energy of this pulse is located around the wings of the absorption band of the material, which is the reason why the precursor decays very slowly over the length of the material. The temporal beats of the precursor in Fig. 2 are occurring more rapidly with increasing propagation depth. The physical explanation of this is that the absorption is stronger closer to the line center, which causes the two spectral bumps of the precursor in Fig. 2 to move farther apart when the pulse propagates deeper into the material. This precursor is similar to the 0π pulse first derived by Crisp [17] (later observed experimentally by Rothenberg [23]); it is linear, located around the wings of the absorption band, shows temporal beats, is slowly decaying, and has a 0π area. In fact, the connection between the traditional linear optical precursor and Crisp's 0π pulse has already been established (see, e.g., Ref. [18] and references therein). The area of the main signal in Fig. 2 is clearly 2π, and it is therefore a 2π h.s. soliton. We have also verified that after the precursor and the main signal begin to separate around $\alpha z \sim 8$, the maximum population transfer caused by the precursor is less than 2.5% and occurs for atoms that are detuned an amount $\Delta/\omega_0 \approx \pm 1.8 \times 10^{-6}$ (i.e., where the spectral amplitude of the precursor is largest). Hence, this precursor approximately interacts linearly with the material. This is not surprising since the amplitude of the precursor is in the linear regime, which has been previously discussed by both Crisp [11] and Macke and Ségard [10]. Eventually,

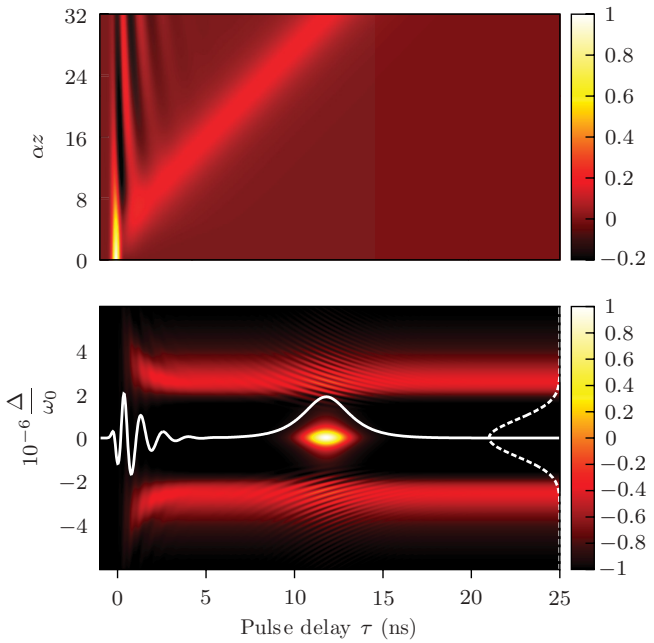

FIG. 3. (Color online) Top: Reshaping of a pulse with input pulse duration $\tau_0 = 0.3T_2^*$ and initial area 1.1π. The color coding shows the normalized amplitude of the field. Bottom: Excitation of the absorption line at $\alpha L = 32$. The color coding shows the inversion $\rho_{22}(\tau,L,\Delta) - \rho_{11}(\tau,L,\Delta)$, the solid line shows the exit pulse, and the dashed line shows $g(\Delta)$ (the height of both lines are in arbitrary units).

for a sufficiently thick material, the precursor is completely absorbed by the detuned atoms, and they are left in an excited state. However, due to the 0π area of the precursor, the atoms on the line center are essentially unaffected by the precursor.

We have confirmed that when the input pulse duration becomes longer than $\tau_0 = 0.6T_2^*$, less population is transferred during the precursor passage, and the linear approximation is even better. However, when the input pulse duration becomes shorter than $\tau_0 = 0.6T_2^*$, the interaction between the precursor and the material becomes nonlinear. Figure 3 shows as much as 40% population transfer for $\tau_0 = 0.3T_2^*$ (for atoms that are detuned an amount $\Delta/\omega_0 \sim \pm 3 \times 10^{-6}$), and we have observed higher transfers for even shorter input pulse durations. Nonetheless, we have also verified that the precursor retains most of its other properties; its spectrum still lies in the wings of the absorption bands, it decays slowly over the length scale α^{-1}, has a 0π area, and is not phase modulated.

Finally, we perform numerical simulations with boxcar input pulses

$$\Omega(\tau,0) = \begin{cases} 0, & |\tau| > \tau_p/2, \\ \Omega_0, & |\tau| \leqslant \tau_p/2. \end{cases} \tag{5}$$

The pulse duration is $\tau_p = T_2^*$ and the input area is chosen to be 1.1π such that $\Omega_0 = 1.1\pi/\tau_p$. The boxcar pulse spectrum has several sidelobes (see Fig. 4) that are themselves strong enough to nonlinearly interact with the absorption wings of the medium. Figure 4 shows the reshaping of the boxcar pulse as it propagates through the medium. Similarly to the smooth Gaussian input pulse, a precursor starts to separate from the main pulse around $\alpha z \sim 8$ and the two are temporally

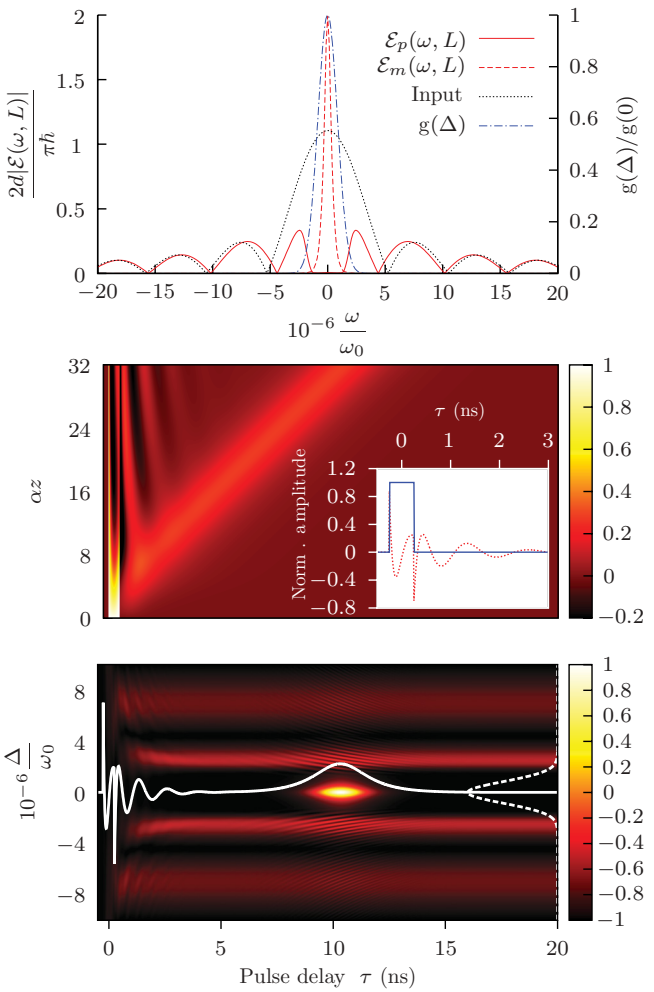

FIG. 4. (Color online) Pulse evolution of a boxcar input pulse with duration $\tau_0 = T_2^*$ and area 1.1π. Top: Spectral magnitude of the precursor and main pulse at the exit face. The dotted line shows the input pulse spectrum, and the dashed-dotted line shows the absorption line of the medium. The solid line shows the spectrum of the main pulse and the dashed line shows the spectrum of the precursor. Middle: Spatiotemporal reshaping of the pulse. The inset shows the precursor at the exit face (dotted line), and the input pulse (solid line). Bottom: Excitation of the absorption line at the exit face. The solid line shows the exit pulse, and the dashed line shows the absorption line of the medium (the heights of both lines are in arbitrary units). The color coding shows the inversion $\rho_{22}(t,L,\Delta) - \rho_{11}(t,L,\Delta)$.

well separated at the exit face of the medium. There are two rapid oscillations on $\tau = \pm\tau_p/2$ that were not present for the Gaussian input pulse. These two oscillations coincide with the steps of the input pulse, showing that the pulse fronts propagate at the speed of light in vacuum. In most other aspects, this precursor shows similar qualitative behavior as for the Gaussian input pulse: The main pulse is a hyperbolic secant with area 2π, the precursor has 0π area, the frequency pushing of the lobes closest to resonance persists, and neither the precursor or the main pulse are phase modulated. Figure 4 shows that the precursor excites detuned atoms $\Delta/\omega_0 \sim \pm 3 \times 10^{-6}$ and $\Delta/\omega_0 \sim \pm 8 \times 10^{-6}$, which coincides with the two closest lobes in $\mathcal{E}_p(\omega,L)$. However, the low atomic density at the detuning $\Delta/\omega_0 \sim \pm 8 \times 10^{-6}$ ensures that these atoms

contribute little or nothing to the pulse evolution. We also remark that the boxcar pulse produces much larger precursors than the Gaussian. In particular, our simulations show that using a boxcar pulse with duration $\tau_0 = 2T_2^*$ and area 1.1π produces a precursor with a peak amplitude $\sim 0.95E_0$, while for a Gaussian pulse the precursor amplitude was negligible (see Fig. 1).

In this study, pulse energy loss through spontaneous emission (occurring at a rate $1/T_1$) has not been taken into account. Typically, the emerging 2π soliton is not stable if it does not exit the material well within a time T_1, and can collapse to a 0π pulse [24]. This effect could be included with an additional complexity, but does not provide further insight into the physics. Nevertheless, it places restrictions on the materials and pulses one can use for the experimental verification of the above results. For example, an input area of 1.1π may result in a soliton that is so slow that it loses most of its energy through spontaneous emission before it exits the material.

In summary, by solving the Maxwell-Bloch equations numerically, we predict the existence of 0π area precursors in inhomogeneously broadened two-level materials for resonant pulses, where the main pulse is a 2π SIT soliton. These results show several interesting features: The soliton and its precursor are temporally separated, and this separation can be quite large due to the reduced velocity of the soliton. Depending on the relationship between the input pulse duration and the inhomogeneous lifetime, the precursor can interact linearly or nonlinearly with the material. Due to the symmetry of the input pulse and absorption line around the line center, neither the precursor nor the soliton are phase modulated. Lastly, although boxcar pulses produce very strong precursors, even smooth Gaussian pulses can yield precursors of relatively large amplitudes. We envision experimental confirmation of these results in a low-pressure warm vapor of one of the alkali-metal atoms. An exact resonance condition can be established using a magnetic field (which also enhances the two-level approximation), and the desired relationship between the pulse duration and the Doppler lifetime can be achieved by adjusting the pressure and temperature of the gas cell. Finally, the results in this paper are generic to the two-level atom and it is therefore interesting to speculate if they can be derived from the general solution [16,19] (i.e., can the shape and decay rate of the precursor be found from the continuous spectrum of the eigenvalue problem?). Such a study is interesting in its own right and has not, to the best of our knowledge, been performed to date.

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