Propagation effects and ultrafast optical switching in dense media

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We investigate ultrashort pulse propagation in a dense medium of two-level atoms. We numerically solve the coupled nonlinear Maxwell-Bloch equations by assuming that the fields are slowly varying in time only. We find that while intrinsic optical switching of the atomic inversion persists in extended media, self-phase-modulation effects, coherent energy transfer, and reflections can be large even for films that are only a small fraction of a wavelength thick. If the propagation distance is sufficiently large, a physical boundary that separates fully excited atoms from atoms in the ground state can be created within the medium. In turn, this leads to a rapid spatial modulation of the nonlinear polarization, and a deterioration of the intrinsic optical switching mechanism. We then show that, although the slowly varying envelope approximation in space may be preempted by the fast longitudinal variation of the field and atomic variables, the mean-field approximation remains valid in the regime of ultrafast optical switching, *provided* the film thickness is sufficiently small such that the medium is nearly uniformly excited. These results are generated by utilization of our calculational method that fully accounts for the longitudinal dynamics of the fields, including reflections.

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I. INTRODUCTION

Under suitable conditions, an electromagnetic wave impinging on a medium composed of two-level atoms or molecules can induce near-dipole-dipole (NDD) interactions [1–5]. If the density is such that on average there are many atoms per cubic resonance wavelength, then NDD interactions, or local-field effects, can no longer be ignored, as would be the case for more dilute media. NDD effects necessitate a correction to the macroscopic field that couples to an atom in terms of the macroscopic field and volume polarization. This is caused by the interaction with other atoms that reside within a volume of the order of a cubic wavelength. The field contribution from these near dipoles manifests itself in the renormalization of the field-atom frequency detuning by an amount proportional to the atomic inversion [4,5]. The importance of NDD interactions has been highlighted recently where, for instance, dense media have been shown to exhibit intrinsic optical bistability in the steady-state regime [4,5], invariant pulse propagation (solitons) [6], ultrafast intrinsic optical switching for ultrashort incident pulses [7,8], an order-of-magnitude index of refraction enhancement, piezophotonic switching and gain in systems that exhibit lasing without inversion [9]. Recently, intrinsic optical bistability has been demonstrated experimentally by Hehler *et al.* [5].

In a recent study it was shown that if an ultrashort pulse is allowed to interact with a thin film of optically dense two-level systems, in such a fashion that propagation effects can be ignored, medium response is characterized by a rapid switching effect [7]. This behavior is both quite uncharacteristic and more remarkable than the response of conventional two-level systems because the medium can only be found in one of two states: fully inverted, or in the ground state, depending on the initial conditions and the ratio between the peak field strength E_0 and the near dipole-dipole coupling strength ε, which we will define below. For values of E_0/ε smaller than 1, the medium returns to the initial state after the pulse has passed. If this ratio is near unity, the medium would be fully inverted if the initial state were the ground state. Larger values of this ratio would cause the final state of the inversion to switch between complete excitation or complete deexcitation, giving rise to a nearly square-wave pattern as a function of E_0/ε. However, the first instance of a full state of inversion always occurs when the ratio is nearly unity. This feature was found to be impervious to changes in pulse shape, and independent of pulse area [7]. This behavior is depicted in Fig. 1 (solid line), where the final steady-state value of the inversion is plot-

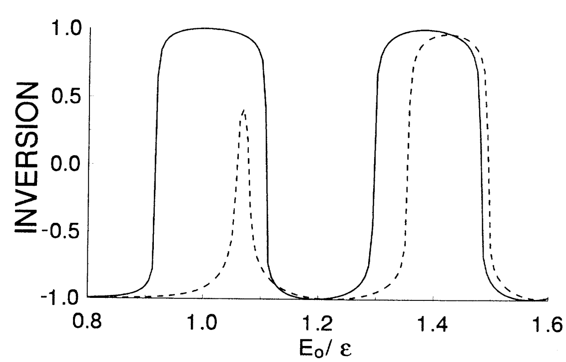

FIG. 1. Steady-state value of the inversion as a function of the ratio $\hat{E}_0/\hat{\varepsilon}$, without (solid) and with (dashed) a small positive static detuning. These values are recorded after the Gaussian pulse of Eq. (7) traverses the medium.

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ted as a function of the ratio E_0/ε.

Although the nature of the interaction had previously been studied in different regimes, this behavior had not been predicted and its appearance was quite surprising. In a later study, it was shown that the switching mechanism comes about as a result of an adiabatic following type of behavior, that causes the inversion to switch between the real roots of a quartic equation [8]. The stable analytical solutions for the final state of the inversion were found to be consistent with the numerical results of Ref. [7]. The behavior of the system was shown to be similar to that of adiabatic inversion by induced frequency chirp [10], except that in this case the frequency chirp, and hence adiabatic inversion, are intrinsic.

Medium response of this kind is important for several reasons. First, the phenomenon is ultrafast since ultrashort pulses are used to excite the medium. Second, the existence of only two final states suggests switchlike characteristics, with the welcome advantage that dissipative energy loss is virtually zero when compared to other switching mechanisms based on ordinary optical bistability. Third, the inversion characteristics are independent of pulse area, that is, exclusive use of π pulses in order to achieve full inversion of conventional two-level systems can be replaced by arbitrary pulses that require only that the ratio $E_0/\varepsilon \approx 1$. Finally, the NDD interaction alters the system's quantum coherence properties, leading to many new effects in quantum optics [4–9], such as modified lasing without inversion [9].

Previous studies were based on the assumption that propagation effects in a dense medium could be ignored [7,8]. Our goal in this paper is to examine the possibility of intrinsic optical switching and overall medium response under more realistic circumstances, namely, in media that are assumed to be finite in length. In reality, propagation always plays a role, and eventual comparisons with experimental results require that propagation by part of any theoretical investigation. Other workers have previously investigated propagation effects in dense media, and have highlighted the importance of reflections [11]. However, these studies were not performed in the limit of intrinsic optical switching, where field amplitude and ε are large and can be on the same order of magnitude. The mean-field approximation was also adopted in those studies, and phase-modulation effects of the field were ignored. We will show below that although intrinsic optical switching still occurs, a state of nearly full inversion throughout the medium can be sustained only if the film is a small fraction of a wavelength. We also find that the slowly varying envelope approximation in space for the Maxwell field cannot be used in these systems because rapid field amplitude variations, strong self-phase modulation effects, large reflections, and coherent energy transfer all occur well within a small fraction of a wavelength from the entrance of the medium. In general, then, the mean-field approximation cannot be used for these systems, *even* if the condition $L \ll \lambda$ is satisfied, where L is the length of the sample. However, we will show that if the medium can be excited uniformly in its entirety, and film thickness does not exceed $\frac{1}{100}$ of the incident wavelength, then the mean-field approximation remains a useful tool, although it is not generally applicable for thicker films. The results that we present in the sections that follow were briefly pointed out elsewhere [12], but here we expand the discussion to highlight the extremely complex and rich dynamics of the system.

II. MODEL

Our analysis of the dynamics of the Maxwell-Bloch equations is different in that an alternative beam propagation method is employed to solve the second-order propagation equation [13]. We consider the full dynamics of the fields along the longitudinal spatial coordinate. We only retain the slowly varying envelope approximation in time, since incident pulses are assumed to be on the order of 100 optical cycles or more. We assume the incident field is of the form $\Sigma = \frac{1}{2}[E(z,t)e^{i(kz-\omega t)} + \text{c.c.}]$, where E is the field envelope that varies slowly in time only. The modified optical Maxwell-Bloch equations take the form [4,5]

$$\frac{\partial W}{\partial t} = -\frac{i\mu}{\hbar}(E^* R - ER^*) , \tag{1}$$

$$\frac{\partial R}{\partial t} = -i\varepsilon WR - \frac{i\mu}{\hbar}\frac{EW}{2} , \tag{2}$$

$$\frac{\partial^2 E}{\partial z^2} + 2ik\frac{\partial E}{\partial z} + 2i\frac{\omega}{c^2}\frac{\partial E}{\partial t} = -\frac{4\pi\omega^2}{c^2}2N\mu R . \tag{3}$$

We have imposed the slowly varying amplitude and phase approximations in time (SVEAT) only. Here W is the atomic inversion, $P = 2N\mu R$ is the effective polarization of the medium, E is the complex macroscopic Maxwell field envelope, $\varepsilon = 4\pi\mu^2 N/3\hbar$ is the strength of the near dipole-dipole coupling, μ is the atom's dipole moment, $\hbar$ is Planck's constant divided by 2π, and N is the atomic density. We have taken the linear refractive index of the host medium to be unity, and chosen $k = \omega/c$. This choice implies that all phase modulation effects that ensue from propagation are contained in the field envelope function.

For simplicity, we have also assumed that the incident field is resonant with the single atom transition frequency, and that longitudinal and transverse relaxation rates can be ignored due to the ultrashort nature of the incident pulse. We now scale the time with respect of the optical period in the medium, that is, $\tau_p = \lambda/c$, such that $\tau = t/\tau_p$. If we scale the longitudinal coordinate such that $\xi = z/\lambda$, where λ is the wavelength of the radiation field in the host medium, we can then write

$$\frac{\partial W}{\partial \tau} = -i(\hat{E}^* R - \hat{E}R^*) , \tag{4}$$

$$\frac{\partial R}{\partial \tau} = -i\hat{\varepsilon}WR - i\frac{\hat{E}W}{2} , \tag{5}$$

$$\frac{\partial \hat{E}}{\partial \tau} = -\frac{\partial \hat{E}}{\partial \xi} + \frac{i}{4\pi}\frac{\partial^2 \hat{E}}{\partial \xi^2} + 6\pi i\hat{\varepsilon}R , \tag{6}$$

where $\hat{E} = \mu\tau_p E/\hbar$ is the scaled field, and $\hat{\varepsilon} = \varepsilon\tau_p$ is also scaled by τ_p. We note here that $\hat{\varepsilon}$ also appears in Eq. (6) as a propagation constant, and therefore its importance in this and many other systems cannot be overstated.

Equations (4), (5), and (6) are solved simultaneously for the field and the atomic variables. The method we employ is straightforward because one need only specify the location of an arbitrary number of boundaries, or a sudden density change as in our case, and not the boundary conditions associated with the field and its derivatives [13]. These conditions are embedded in the propagation equation and they are implicitly accounted for.

III. RESULTS AND DISCUSSION

We now specify a typical system by taking the incident wavelength to be $\lambda=10^{-6}$ m. Then a picosecond pulse corresponds to a pulse nearly 100 optical cycles (or wavelengths) at the waist. We also assume that the initial field is centered at $\xi=\xi_0$, and that for simplicity it is real and Gaussian in shape, that is,

$$\hat{E}(\xi,0)=\hat{E}_0 e^{-[(\xi-\xi_0)/25]^2} . \tag{7}$$

This field has a spatial spread of more than 100 optical cycles at the waist, and it is consistent with the slowly varying envelope in time assumption. We first integrate Eqs. (4)–(6) in the limit that the medium has essentially zero thickness, and for the conditions that the peak value of the field equals the dipole-dipole coupling coefficient, $\hat{E}_0=\hat{\varepsilon}=0.2$. We find that the medium is fully inverted after the pulse has passed, consistent with previous findings [7,8]. We assume that $\hat{\varepsilon}=0.2$ in what follows, unless we state otherwise.

A. Propagation in films of finite length

We now consider films of nonzero thickness. Our interest here is to investigate the highly nonlinear medium response that is coherently excited by a short pulse. When the pulse is still approaching the medium, the atoms are initially detuned by an amount $-\hat{\varepsilon}W$ at $\tau=0$, that is $+\hat{\varepsilon}$ for $W=-1$. This constitutes a static blueshift. Equation (5) then suggests that the evolution of the inversion leads to a time varying shift of the resonant frequency. This is also equivalent to an intrinsic time varying detuning that constitutes a redshift to saturation. If we ignore for the moment the coupling to the Maxwell field, these considerations lead to the intrinsic optical switching discussed previously [7,8]. The final state of the inversion is either the ground or the excited state, with the first occurrence of a periodic square wave of W vs $\hat{E}_0/\hat{\varepsilon}$ centered about $\hat{E}_0/\hat{\varepsilon}=1$.

Although the features of the square-wave pattern of W vs $\hat{E}_0/\hat{\varepsilon}$ were found to be stable to changes in pulse width and pulse shape, slight distortions in the shape of the exciting pulse, or a nonzero static detuning, could lead to a shift, a widening, or a shrinkage of the peaks [7,14]. This means that maintaining the ratio $\hat{E}_0/\hat{\varepsilon}=1$ does not guarantee a final full state of excitation of the medium. However, the medium can still be fully excited for a range of values whose ratio is higher, or lower, than 1, depending on the sign of the detuning. We show this effect in Fig. 1 (dashed line), where we plot the steady-state value of the inversion after a Gaussian pulse traverses a thin medium, with a small positive static detuning, $\delta=\omega-\omega_a$, where ω_a, is the single atom transition frequency. We see that although the mean features are unchanged, the first peak is reduced in magnitude. The first peak eventually disappears with increasing detuning, and an overall shift of the peaks to the right occurs. If the detuning were negative, the peaks would shift to the left [14]. This effect is explained in terms of the adiabatic following model of Ref. [8], and for further details on the effects of pulse distortions we refer the reader to Refs. [7,14].

If the Maxwell field is now allowed to couple into the dynamics, and the medium is initially in the ground state, the field effectively couples to a medium that is initially detuned by an amount $+\hat{\varepsilon}$ in this case as well. However, because the phase shift imparted to the field upon reflection is of order π, and because nonlinear dispersion and coherent energy transfer to and from the medium are significant, the pulse cannot simply pull the medium into resonance as in the case of thin films. That means that strong self-phase-modulation effects govern the dynamics of the system, leading to significant changes in the evolution of the Bloch vector. One can easily realize this if the Maxwell-Bloch equations are recast explicitly in terms of a general field amplitude and phase. That is, by writing the field as $\Sigma=\frac{1}{2}[|\hat{E}|e^{i[kz-\omega t-\phi(\xi,\tau)]}+\text{c.c.}]$, and the field envelope function as

$$\hat{E}=E_r+iE_i=|\hat{E}|e^{-i\phi(\xi,\tau)} , \tag{8}$$

where E_r and E_i are the real and imaginary components of the field, respectively, and performing the rotating-wave approximation [10], Eqs. (4) and (5) can be rewritten as

$$\dot{R}=i(\dot{\phi}-\hat{\varepsilon}W)R-i\frac{|\hat{E}|W}{2} , \tag{9}$$

$$\dot{W}=-i|\hat{E}|(R-R^*) . \tag{10}$$

Both amplitude and phase can still be taken to be slowly varying functions of time, but are allowed to vary rapidly in space. Equations (9) and (10) therefore suggest that self-phase-modulation effects can indeed contribute significantly to the evolution of the Bloch variables, by inducing a quite complicated spatiotemporal shift of the resonance. In fact, these effects may cause a dynamic shift of the peaks shown in Fig. 1, or may even preempt their formation. Full excitation of an extended medium can still be obtained, but the ratio $\hat{E}_0/\hat{\varepsilon}$ approximately unity may not be sufficient. Sample length is extremely important because it not only determines bulk reflection and transmission coefficients, along with the dynamics of the field inside the sample, but also the location, width, and perhaps the existence of the peaks of Fig. 1.

B. Quasiadiabatic inversion: field dynamics and material response

We now quantify our discussion by considering a film whose thickness is approximately 0.025λ. We observe that the *entire* medium can be inverted if $\hat{E}_0/\hat{\varepsilon}=1.1$. Now, the inversion is not stationary in time due to the inertia acquired by the Bloch vector and also in part due to radiation reaction that couples all atoms within the prescribed volume. This means that in contrast to the thin-film case, the medium cannot be sustained in the excited state for times that are much longer than the duration of the pulse, as our calculations show. Therefore, we define a final state of inversion with respect to the width of the incident pulse, i.e., we allow most of the pulse to exit the medium in order to determine that most of the population still resides in the excited state across all the sample.

Figure 2 depicts the time evolution of the inversion (solid line), real (dashed line), and imaginary (dotted line,) components of the polarization at the entrance of the medium. Everywhere inside the medium these dynamics are similar because the entire sample is excited to nearly the same level. Figure 3, on the other hand, shows the real (solid line) and imaginary (dashed line) components of the field that excite the medium response depicted in Fig. 2, which include both forward and backward propagating modes. While these figures suggest that the medium can still be said to undergo quasiadiabatic inversion [8], the temporal evolution of both quadratures of the field that is shown in Fig. 3 reveals the dynamics that in reality drive the interaction. The long-time behavior of the curves in Figs. 2 and 3 suggests that energy that is temporarily stored in the medium is slowly released back into the field. While the medium becomes almost completely inverted at first, photons are eventually reemitted. However, in this case the medium undergoes quasiadiabatic inversion, and it is able to store the energy provided by the input pulse for some time. In contrast, Figs. 2 and 3 should be compared to Figs. 4 and 5, where we show medium response and field dynamics, respectively, with $\hat{E}_0/\hat{\varepsilon}=1.05$. Although the medium initially becomes nearly fully excited in this case as well, the medium of Fig. 4 is not sustained in the excited state for any significant length of time. As a result of this coherent excitation and rapid deexcitation, photons are quickly reemitted and add to the total field, which in turn is strongly modulated, as Fig. 5 shows. The differences in the fields of Figs. 3 and 5 strongly hinge on self-phase-

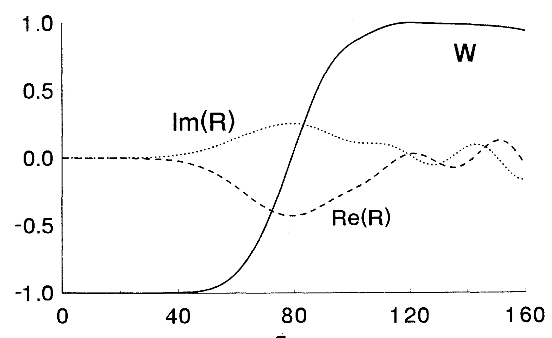

FIG. 2. Time evolution of the inversion (solid line), real (dashed line), and imaginary (dotted line) components of the polarization at the entrance and throughout the medium. $\hat{E}_0/\hat{\varepsilon}=1.1$, and the layer is 0.025λ thick. While the entire medium is inverted, the field changes little across the sample in this case. The time is in units of the optical period.

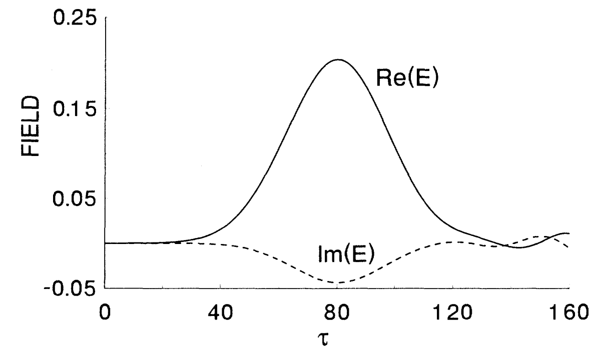

FIG. 3. Time evolution of the real (solid line) and imaginary (dashed line) components of the field that cause the medium response of Fig. 2.

modulation effects, and are both quantitative and qualitative, as a direct comparison shows. We will return to the discussion of self-phase-modulation effects below, and here we merely point out that the dynamics are extremely sensitive to small parameter variations (i.e., medium length, density, pulse width). Finally, we note that the magnitude of the imaginary component of the field is an indication of the size of reaction field that is generated within the medium. In fact, if the pulse were to propagate in free space, a real input pulse [such as that of Eq. (7) that we use] obviously could not generate the component in quadrature.

Another important result is that the medium dynamically induces a large reflected pulse in response to the pump pulse, even for extremely small film thicknesses. Figure 6 shows both input (solid curve) and scattered (dashed curve) pulse intensities that give rise to the dynamics of Fig. 2. From the figure we see that the reflected pulse is indeed significant. Coherent absorption and nonlinear refraction are also dominant mechanisms in the interaction. Moreover, if the propagation distance were sufficiently large, the field amplitude within the medium could rapidly decrease until the ratio $\hat{E}_0/\hat{\varepsilon}$ fell below the value necessary to sustain that part of the medium in the excited state [7]. As we will see below,

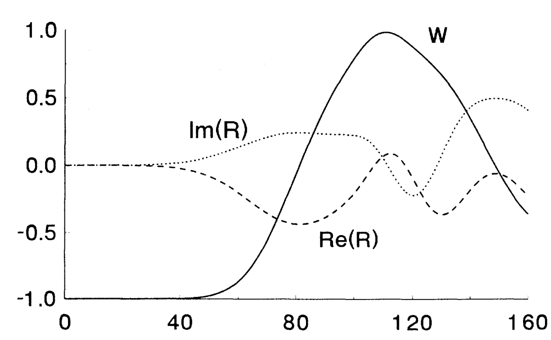

FIG. 4. Same as Fig. 2, but with $\hat{E}_0/\hat{\varepsilon}=1.05$. Note the larger oscillations in the nonlinear refractive index and absorption with respect to Fig. 2. This comes as a result of increased self-phase-modulation effects.

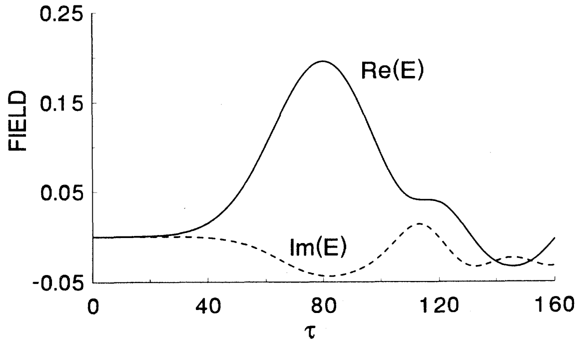

FIG. 5. Same as Fig. 3, but with $\hat{E}_0/\hat{\varepsilon}=1.05$.

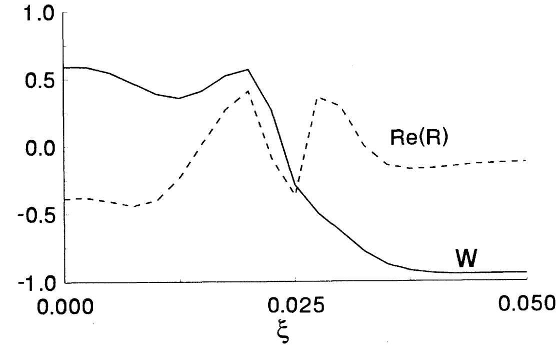

FIG. 7. Snapshot of the spatial variation of the inversion (solid line) and nonlinear refractive index (dashed line) for a sample of length 0.05λ and $\hat{E}_0/\hat{\varepsilon}=1.1$. At this stage the pulse interacts weakly with the medium. The nearly sudden drop that characterizes the inversion induces a spatial modulation in both nonlinear refraction and absorption. This longitudinal behavior of the inversion is caused by the loss of energy of the field as energy is coherently transferred to the medium. Pulse energy loss reduces the peak strength of the field, which in turn causes the medium to fully deexcite.

this can generate a physical boundary within the sample that separates atoms that are fully excited from atoms in the ground state, and a rapid spatial modulation of the polarization (i.e., nonlinear index of refraction as well as absorption) occurs as a result of conservation of the Bloch vector, in analogy to intrinsic optical bistability in media modeled by nonlinear classical oscillators [15]. In short, medium response cannot be predicted *a priori.* Even if the conditions at the input boundary were such that full excitation should occur there, this can no longer by guaranteed if the medium is sufficiently thick, and predicting the width of the inverted layer is a nearly impossible task. In general, the full longitudinal dynamics of the field must be included in any analysis of propagation effects, especially if we consider that the entire switching dynamics can occur within a very small fraction of a wavelength.

In Fig. 7 we plot the inversion (solid line), along with the nonlinear index of refraction (dashed line), as a function of distance inside a sample of thickness 0.05λ, after most of the pulse has passed. At this instant, the pulse is not interacting strongly with the medium. The parameters are the same as those used in Fig. 2, except for medium length. Note that only part of the medium is inverted, and that the fast longitudinal variation of the inversion (which occurs within a distance of approximately 0.01λ) causes the complex nonlinear refractive index to also follow, in order to satisfy conservation of the Bloch vector. This is in contrast to what we obtained in the case of Fig. 2, where the sample was only half as long. There, the Block variables became discontinuous *only* at the entrance and at the exit of the medium, since the entire length of the medium was inverted. In fact, it is the *discontinuity* of the nonlinear index of refraction at the medium entrance and exit that primarily gives rise to the coherently reflected pulse of Fig. 6, while in the case of Fig. 7 the situation is complicated by the additional spatial modulation of the nonlinear index that occurs inside the medium.

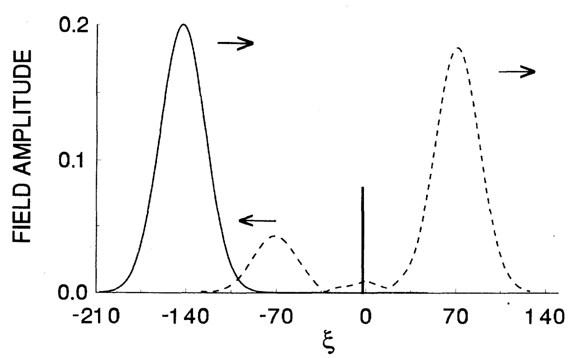

FIG. 6. Incident (solid line) and scattered (dashed line) pulses as a function of position. The medium is 0.025λ thick, $\hat{E}_0/\hat{\varepsilon}=1.1$, and is located near the origin. The longitudinal coordinate is scaled by the incident, vacuum wavelength.

As stated earlier, the dynamics described in the above paragraph are similar to what happens in some nonlinear oscillator media, where anharmonicities affect the motion of the electrons and cause intrinsic optical bistability [15]. There, physical longitudinal and transverse polarization boundaries [15(a)–15(c)] evolve within the medium, implying that the nonlinear index of refraction can take on two different values for a single incident field amplitude. As a result, a small reflected wave is induced [15(a)]. Then, in a similar manner as outlined in Ref. [15(a)], a thin film of dense two-level atoms is suitable to study four-wave mixing phenomena. For instance, if the boundary within the medium is probed with a copropagating pulse, scattering from the interface can give rise to a conjugate field that propagates in the opposite direction [15(a),16].

C. Phase-modulation effects

In order to highlight the importance of phase-modulation effects we now specifically address the question of how phase changes affect the dynamics. If we define a complex field envelope as in Eq. (8), that is, $|\hat{E}|e^{-i\phi(\xi,\tau)}=E_r+iE_i$, then after simple manipulations it follows that

$$\dot{\phi}=\frac{E_i\dot{E}_r-\dot{E}_iE_r}{|\hat{E}|^2} . \tag{11}$$

We now refer the reader to Figs. 8 and 9, where we plot the temporal evolution of $\dot{\phi}(\tau)$, $\hat{\varepsilon}W(\tau)$, and their relative difference, $\Delta(\tau)=\dot{\phi}(\tau)-\hat{\varepsilon}W(\tau)$, at the entrance of the medium for the cases of Figs. 2 and 4, respectively. Note that $\Delta(\tau)$ is now the new effective dynamic detuning as specified in Eq. (9). The figures show that in spite of phase-modulation effects, which can significantly change the effective detuning, quasiadiabatic inversion occurs in one case (Fig. 8), while it does not in the other (Fig. 9). In Fig. 10 we plot $\dot{\phi}(\tau)$ for $\hat{E}_0/\hat{\varepsilon}=1.1$ (solid line) and 1.05 (dashed line). We point out that this dynamical behavior repeats *throughout* the medium, and that the temporal evolution of the phase and all other quantities are only somewhat different at the output, a condition that is not met for thicker films. Figures 8–10 thus suggest that while the magnitude of $\dot{\phi}$ is comparable in all cases, the early onset of self-phase-modulation effects in one case causes the inversion to lose hold of the excited state. While we see that the importance of $\dot{\phi}$ cannot be overstated in describing the optical switching outlined above, it is also paramount from the point of view of sideband generation, especially since $\dot{\phi}$ can be large relative to $|\hat{\varepsilon}W|$. Although clearly this aspect of the dynamics is extremely important, it will be taken up separately later on because of the complexity of the problem.

For completeness, we point out that for films whose thickness is other than 0.025λ, quasiadiabatic inversion on the time scale of the incident pulse width occurs for different values of the ratio $\hat{E}_0/\hat{\varepsilon}$. Whether this value is greater or less than unity cannot be predicted *a priori*, and it will sensitively depend on medium length, pulse width, static detuning, the temporal evolution, and the sign of $\dot{\phi}$. However, we found that in the absence of a static detuning, an overall shift to the right of the peaks shown in Fig. 1 occurs. For example, we found that a sample of length $\approx 0.03125\lambda$ can also be inverted in its entirety if $\hat{E}_0/\hat{\varepsilon}\approx 1.12$, but the excited state is not as long

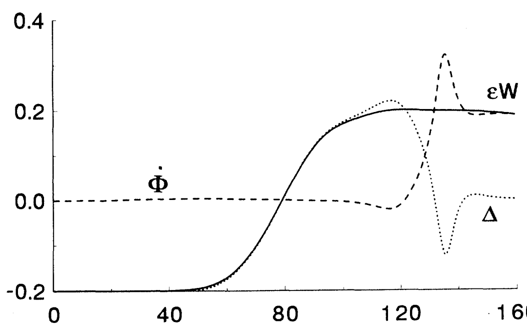

FIG. 8. Temporal evolution of $\hat{\varepsilon}W$ (solid line), $\dot{\phi}$ (dashed line), and $\Delta(\tau)=\hat{\varepsilon}W-\dot{\phi}$ (dotted line) at the entrance of the medium. Dynamics follow similar behavior inside the medium as well. In this case $\hat{E}_0/\hat{\varepsilon}=1.1$, and quasiadiabatic inversion persists because phase modulation does not become important until after the medium has nearly fully acquired the excited state.

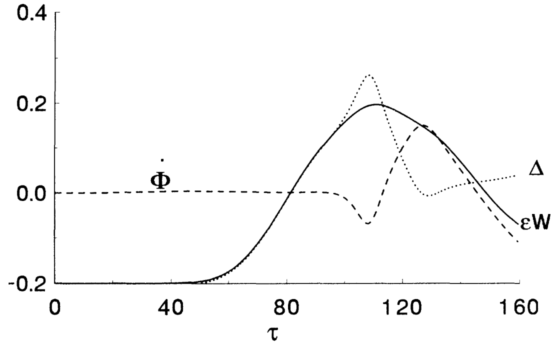

FIG. 9. Same as Fig. 8, but with $\hat{E}_0/\hat{\varepsilon}=1.05$. In this case the excited state is short lived. If Figs. 8 and 9 are compared, it is clear that self-phase-modulation determines the stability of the medium in the excited state.

lived as in the case we have examined above. We show this in Fig. 11, where we compare the inversion as a function of time at the entrance of the medium for sample lengths 0.025λ and 0.03125λ (solid and dashed lines, respectively). We have used the input field

$$\hat{E}(\xi,0)=\hat{E}_0e^{-[(\xi-\xi_0)/20]^2} , \tag{12}$$

which is slightly less then 100 optical cycles in duration at the waist, and $\hat{\varepsilon}=0.25$. Throughout the medium the inversion again follows almost the same temporal dynamics for both cases, although for the thicker sample the atoms are more strongly coupled by a larger reaction field and even stronger self-phase-modulation effects. The figure shows that most of the medium remains inverted until some time after most of the pulse has left the medium (i.e., $\tau\geq 100$). We point out that several values of the ratio $\hat{E}_0/\hat{\varepsilon}$ were investigated, and that the values that we have quoted above yielded the longest-lived excited states, although we did not seek to optimize our parameters further. Because we do not find a broad range of values for which the medium can still be found in the excited state after the pulse has passed, we predict an overall decrease of the width and a shift to the right of the peaks in Fig. 1, and a deterioration of the quasiadia-

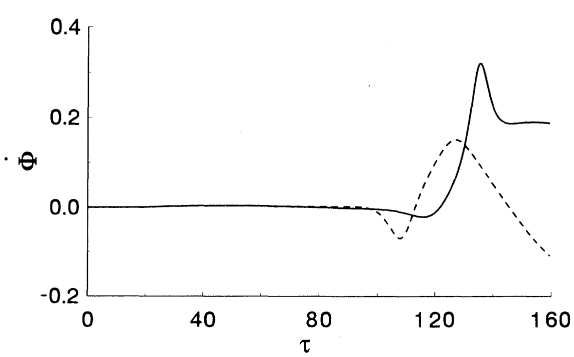

FIG. 10. Comparison of $\dot{\phi}$ in the cases of Figs. 8 and 9. This figure shows that the early onset of strong phase-modulation effects prevents quasiadiabatic inversion, thus highlighting the sensitivity of the dynamics on $\dot{\phi}$ and the effective detuning $\Delta(\tau)$.

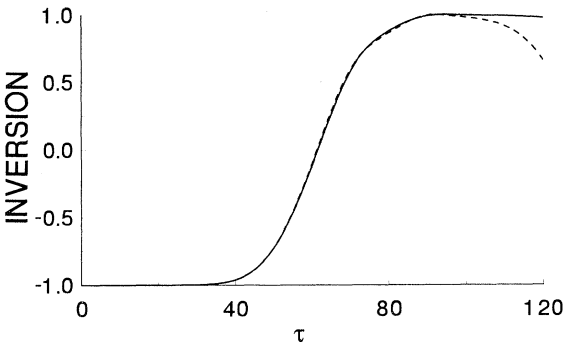

FIG. 11. Time evolution of the inversion at the entrance and throughout the medium for sample lengths 0.025λ (solid line, and $\hat{E}_0/\hat{\varepsilon}=1.1$) and 0.03125λ (dashed line, and $\hat{E}_0/\hat{\varepsilon}=1.12$), with $\varepsilon=0.25$. The medium can still be found in a state of inversion for some time after most of the pulse has excited the medium.

batic inversion mechanism solely as a result of propagation effects.

To conclude this section, we point out that the dynamics described above become even more complicated if the peak field value is chosen such that the ratio $\hat{E}_0/\hat{\varepsilon}$ is much greater than 1, i.e., it falls on the second, third, or even a higher cycle of the square-wave pattern of Fig. 1. In that case, several inverted layers alternating with layers that are in the ground state may be created within the medium, provided the sample is sufficiently thick. Then, the Bloch vector follows an extremely complex and even more unpredictable spatiotemporal dynamics. However, we will not dwell on this aspect of the dynamics here, and simply point out that the possibility of engineering a sample for a specific purpose should be evaluated on a case by case basis.

IV. THE MEAN-FIELD APPROXIMATION

In this section we explore the usefulness and the limits of validity of the mean-field approximation. As we have seen in the preceding section, the field and the atomic variables can vary rapidly in space, seemingly preempting the validity of the mean-field approximation. However, this may not be the case if quasiadiabatic inversion occurs, and if the film is sufficiently thin. If the medium is uniformly inverted in its entirety, the spatial variation of the field and atomic variables *within* the sample remains small. Then, assuming we can ignore the large phase shift that is imparted to the field upon reflection, and neglecting self-phase-modulation effects, the second-order spatial derivative of Eq. (6) can be discarded. This is equivalent to decoupling and therefore neglecting the reflected component of the field, which as we have seen in the preceding section can be quite substantial. Transforming to a retarded coordinate system such that

$$\eta=\xi-\tau\ , \tag{13}$$

a "mean field" can be derived for a sample of thickness L, and can be written as

$$\langle \hat{E} \rangle=\hat{E}_0+\frac{6\pi i\varepsilon L}{2}\langle R \rangle\ . \tag{14}$$

Here L is in units of the incident wavelength, and $\langle\ \rangle$ means spatial average. Equations (4)–(6) become

$$\frac{\partial\langle W \rangle}{\partial\tau}=-i(\hat{E}_0^*\langle R \rangle-\hat{E}_0\langle R \rangle^*)-\frac{1}{\tau_R}\langle |R| \rangle^2\ , \tag{15}$$

$$\frac{\partial\langle R \rangle}{\partial\tau}=-i\varepsilon\langle W \rangle\langle R \rangle-\frac{i}{2}\hat{E}_0\langle W \rangle+\frac{1}{4\tau_R}\langle W \rangle\langle R \rangle\ , \tag{16}$$

where $\hat{E}_0$ is now the incident field, and $1/\tau_R=6\pi\varepsilon L$. Therefore we see that if film thickness is nonzero, cooperativity is induced via coupling with the field resulting in the decay terms proportional to τ_R^{-1} in Eqs. (15) and (16). These terms correspond to superradiant and subradiant decays [4]. The condition that must be satisfied to maintain the validity of these equations is that the spatial variation of the field and atomic variables remain small inside the medium, i.e., film thickness should be no larger than $\frac{3}{100}$ of the incident wavelength. This condition is met in the case of quasiadiabatic inversion, even if reflections and phase-modulation effects are significant. Recall, however, that for thicker films only part of the medium can be inverted, and the atomic variables *do not* vary slowly in space. We also note that an additional restriction on medium length is imposed by the fact that both subradiant and superradiant decay rates should be taken to be much larger than longitudinal and transverse decay rates, which we have neglected for ultrashort pulses.

The analysis of Eqs. (15) and (16) is straightforward. In particular, we can see why the inversion can be sustained in the excited state for only a short time. In fact, while the medium can be nearly fully excited at first, it eventually coherently decays at a rate $1/\tau_R$. In Fig. 12 we plot the final state of the inversion as a function of $\hat{E}_0/\hat{\varepsilon}$ after the pulse of Eq. (7) has passed, with $\hat{\varepsilon}=0.2$,

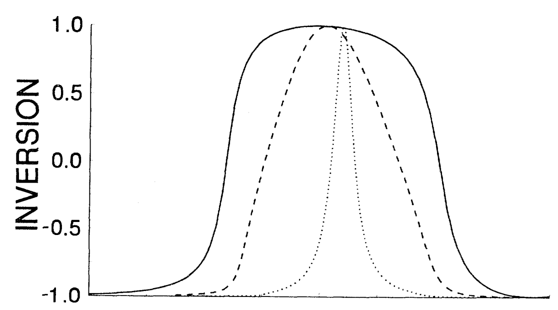

FIG. 12. Steady-state value of the inversion as a function of the ratio $\hat{E}_0/\hat{\varepsilon}$, with no static detuning, as in Fig. 1 (solid line), and with superradiant and subradiant decay (dashed line, $L=0.01\lambda$; dotted line, $L=0.025\lambda$). These values are recorded after a Gaussian pulse traverses the medium. The range of values of $\hat{E}_0/\hat{\varepsilon}$ for which the medium is inverted is clearly smaller as medium length is increased, and indicates a deterioration of intrinsic optical switching as a function of sample length.

and a film of zero thickness. The nearly square-wave pattern of Fig. 1 is recovered, but we show only the first half cycle (solid line). We now integrate Eqs. (15) and (16), and choose film thicknesses of 0.01λ and 0.025λ, respectively, so that we may compare with the results of our numerical integrations of the preceding section. The results are plotted in Fig. 12 (dashed and dotted lines). We can see that, while the widths of both peaks are significantly reduced, the peaks are centered at a value greater then unity, consistent with our numerical results, and a state of nearly full inversion persists for a much reduced range of values of the ratio $\hat{E}_0/\hat{\varepsilon}$. These effects come as a result of a change in the effective dynamic detuning, which no longer simply equals $\hat{\varepsilon}W$. The temporal evolution of the inversion is then very similar to the results of the preceding section, but for the thicker sample the location of the inversion peak does not agree with the results of our numerical simulations because this film thickness is essentially beyond the limit of validity of the mean-field approximation. In summary, the agreement with our full numerical solution is very good if medium length does not exceed $\frac{1}{100}$ of the incident wavelength. For thicker samples, the solution of Eqs. (15) and (16) approximates poorly the solution of Eqs. (4)–(6), and it may even give incorrect and misleading results. Therefore, although the mean-field approximation remains a useful tool within the limits outlined above, care must be exercised if it is applied outside the regime of intrinsic optical switching, or if reflections are important.

V. CONCLUSION

In conclusion, we have discussed the role of propagation effects in an extended medium of dense two-level atoms. We find that if film thickness is on the order of 0.03 of the incident wavelength or less, ultrafast intrinsic optical switching persists, and the *entire* film can be almost completely inverted. However, large self-phase-modulation effects induce an interesting and complicated dynamical shift of the atomic resonance. Because large reflections and energy transfer to and from the medium can occur even for film layers that are only a small fraction of a wavelength thick, the assumption of slowly varying envelope functions in space must be abandoned. For sufficiently thick films, only a portion of the medium may be inverted. In that case, a boundary that separates a state of excitation from one that is fully deexcited is created within the medium, a condition that leads to a strong spatial modulation of the nonlinear index of refraction, as well as absorption. We do not rule out the possibility of inverting in its entirety a film whose thickness is larger than 0.03λ, because the right combination of medium density, thickness, detuning, and perhaps pulse shape, may conspire to accomplish this. However, we predict a general deterioration of the square-wave pattern of Fig. 1 that exemplifies the switching dynamics, as a result of propagation effects. The range of values of the ratio $\hat{E}_0/\hat{\varepsilon}$ for which adiabatic inversion occurs may be significantly reduced, as shown in Fig. 12, unless medium thickness can be kept sufficiently small. Finally, we have determined that the mean-field approximation can describe Bloch-vector dynamics reasonably accurately in the case of quasiadiabatic inversion, *provided* the entire medium can be uniformly excited, and if sample length remains on the order of $\frac{1}{100}$ of the incident wavelength.

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