Ultrafast Intrinsic Optical Switching in a Dense Medium of Two-Level Atoms

M. E. Crenshaw, M. Scalora, and C. M. Bowden

Weapons Sciences Directorate, AMSMI-RD-WS, Research, Development, and Engineering Center, U.S. Army Missile Command, Redstone Arsenal, Alabama 35898-5248

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We performed a numerical investigation of the dynamics of the Bloch equations, extended for dense media (media having many atoms within a cubic resonance wavelength), for optical pulses whose duration is much less than an induced-dipole dephasing time. We find a signature of near dipole-dipole interaction which may provide a useful method for validating the first-principles model, for measuring the strength of the interaction, and for coherent pumping. Further, we demonstrate a new and unique optical switching mechanism which may lead to the development of important devices.

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In dense media, of densities such that there are many atoms within a cubic atomic resonance wavelength, induced near dipole-dipole (NDD) interactions can cause a dynamic frequency chirp in the system [1-4]. In the steady-state limit, NDD interactions may cause bistability that is intrinsic to the material and does not require an external feedback mechanism [1]. Intrinsic optical bistability (IOB) can have important device applications in optical computing and optical data processing [5]. Other effects of NDD interactions which have been analyzed include conditions for self-induced transparency (SIT) in isotropic, homogeneously broadened media [2], and self-phase modulation in SIT [3], as well as linear and nonlinear shifts in the absorption spectrum [4]. Both linear and nonlinear spectral effects due to NDD interactions have recently been observed in reflectivity spectrum measurements using a sapphire window to form an interface with a dense potassium vapor [6].

In this Letter, we consider optical pulses, whose temporal duration is assumed to be much less than an induced-dipole dephasing time, incident upon a thin film of homogeneously broadened material composed of two-level atoms with NDD interactions. In this limit, the induced-dipole dephasing time and the population-decay time can be neglected. We consider the film thickness in the propagation direction to be much smaller than the atomic resonance wavelength so that propagation effects may be neglected.

Under these conditions, we find that the inversion w that remains after a pulse has passed has a nearly step-function response to the peak value of the time-varying field, largely independent of the pulse shape and pulse area. A peak Rabi frequency, $\Omega_0=\mu E_0/\hbar$, is associated with the peak amplitude E_0 of the field envelope, where μ is the matrix element of the transition dipole moment. The system, initially in the ground state, $w=-1$, is always returned to the ground state when the peak Rabi frequency is less than the strength of the NDD interaction ε. However, the final state of the inversion is the fully excited state, $w=1$, when Ω_0 is nearly equal to ε. This suggests that the strength of the NDD interaction can be measured by increasing the field amplitude and interrogating the system after the pulse has passed, in a pulse-probe scenario, to determine whether the system is in the ground or excited state. Furthermore, the nearly step-function response of the system to the field strength constitutes a new and unique optical switch, which, unlike optical bistability or IOB [1,7], does not have hysteresis and is independent of dissipation from incoherent effects, i.e., dephasing and spontaneous relaxation. Devices with subfemtojoule switching energies and picosecond switching times may be realized in condensed matter.

For a two-level system interacting, via an electric dipole transition, with an externally applied, classical, time-varying, coherent field $\frac{1}{2}[E(t)\exp(-i\omega t)+\text{c.c.}]$, the optical Bloch equations in the slowly varying amplitude variables, in the rotating-wave approximation, and extended for interaction with near dipoles take the form [1-4,8]

$$\frac{\partial u}{\partial t'}=-\varepsilon\tau_p w v\,, \tag{1}$$

$$\frac{\partial v}{\partial t'}=\varepsilon\tau_p w u+\Omega\tau_p w\,, \tag{2}$$

$$\frac{\partial w}{\partial t'}=-\Omega\tau_p v\,, \tag{3}$$

where the field carrier frequency ω is resonant with the atomic transition frequency ω_0. In writing Eqs. (1)-(3), we have assumed that the induced-dipole dephasing time T_2 and the population-decay time T_1 can be neglected. Differentiation is with respect to a dimensionless scaled time $t'=t/\tau_p$, where τ_p is a characteristic pulse width that must be much smaller than T_1 and T_2. The strength of the NDD interaction is given by $\varepsilon=(4\pi/3\hbar)\mu^2N$ [8], which must be much less than ω_0. Finally, N is the density of atoms and $\Omega(t)=\mu E(t)/\hbar$ is the instantaneous Rabi frequency, where the field envelope $E(t)$ varies slowly on the time scale of the period $2\pi/\omega$. The extended Bloch equations (1)-(3), like the ordinary optical Bloch equations [9], describe the trajectory of a vector, whose components are u, v, and w, on a unit sphere. Physically, u and v are the components of the polarization

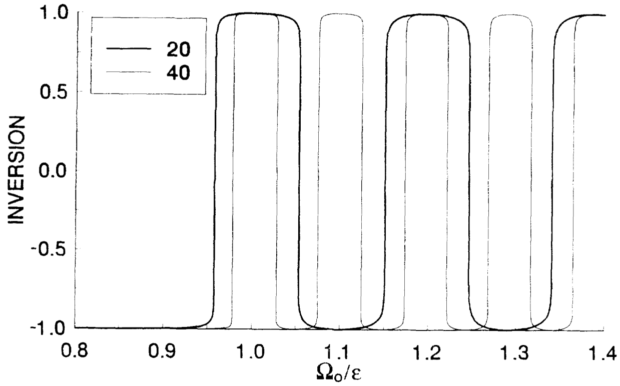

FIG. 1. The final state of the inversion as a function of the peak field strength E_0 of a Gaussian pulse for $\varepsilon\tau_p=20$ and for $\varepsilon\tau_p=40$. Although the abscissa is written in terms of $\Omega_0/\varepsilon=\mu E_0/\varepsilon\hbar$, only the peak field strength is being varied.

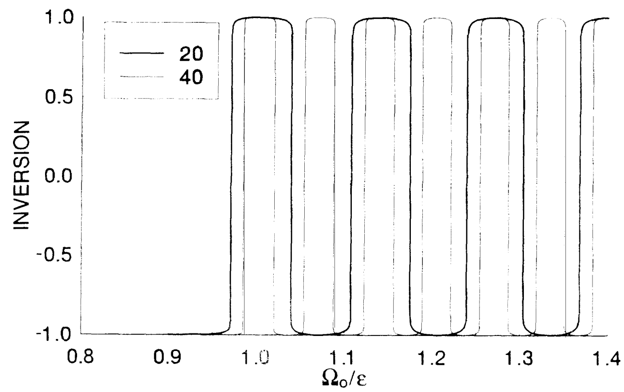

FIG. 2. The final state of the inversion as a function of the peak field strength E_0 of a hyperbolic secant pulse for $\varepsilon\tau_p=20$ and for $\varepsilon\tau_p=40$. Although the abscissa is written in terms of $\Omega_0/\varepsilon=\mu E_0/\varepsilon\hbar$, only the peak field strength is being varied.

in phase and in quadrature with the exciting field and w is the inversion, representing the difference between the population in the excited state and the ground state.

For times much shorter than the population-decay time T_1, Eq. (3) shows that the inversion that remains after a time-dependent field has passed, $w(t\rightarrow\infty)$, is constant. In the absence of NDD interactions, an ultrashort resonant optical pulse leaves the inversion in a state $w=-\cos\Theta$ that depends only on the pulse area Θ, where we have assumed ground-state initial conditions. The pulse area is the time integral of the instantaneous Rabi frequency. To our knowledge, no closed-form solution of the nonlinear differential equations (1)-(3) exists for fields of arbitrary temporal profile in the presence of NDD interactions. Therefore, we numerically solved the extended Bloch equations (1)-(3), using ground-state initial conditions, for the inversion that remains in the medium after a time-dependent field has passed. These calculations were performed for a wide range of pulse shapes and related parameters. Representative results are shown in Fig. 1 for a Gaussian pulse, $E_0\exp(-t^2/\tau_p^2)$, and in Fig. 2 for a hyperbolic secant pulse, $E_0\operatorname{sech}(t/\tau_p)$. These figures depict the final state of the inversion as a function of the peak field strength E_0, in terms of the ratio of the peak Rabi frequency to the strength of the NDD interaction, Ω_0/ε. Remarkably, there is a steplike transition in the character of the medium, from absorbing ($w<0$) to amplifying ($w>0$), as a function of the peak field of a transient field. In each case, the functional dependence of the final state of the inversion with respect to the peak field strength is approximately a translated periodic square wave. Equally remarkable is the fact that, for both pulse shapes, the system is left in a fully inverted condition, $w=1$, when $\Omega_0/\varepsilon=1$, *independent of pulse area*, and that, although not quite symmetric about this point, this is the approximate center of the first half cycle of the "square wave." Figure 3 summarizes the results of similar calculations performed for a range of $\varepsilon\tau_p$. This figure shows that the value of Ω_0/ε at which full inversion occurs can be much greater than unity if $\varepsilon\tau_p$ is small, but rapidly converges to unity as $\varepsilon\tau_p$ increases. This figure also shows that the first half cycle of the square wave narrows and becomes more centered as the frequency increases in proportion to $\varepsilon\tau_p$. We note that complete inversion of the system can be demonstrated analytically, in a particular case, by assuming, as suggested by numerical experiments, that the time development of the inversion is proportional to $\tanh(t/\tau_p)$ for a hyperbolic secant pulse. The assumed solution becomes consistent with ground-state initial conditions as $\varepsilon\tau_p\rightarrow\infty$ if and only if $\Omega_0/\varepsilon=1$.

The definition of pulse area yields a simple relationship between the final inversion as a function of E_0 and the final inversion as a function of pulse area for a given pulse shape. The pulse area corresponding to the first instance where complete inversion occurs is given by

$$\Theta=\varepsilon\int_{-\infty}^{\infty}\frac{\Omega(t)}{\Omega_0}dt\,, \tag{4}$$

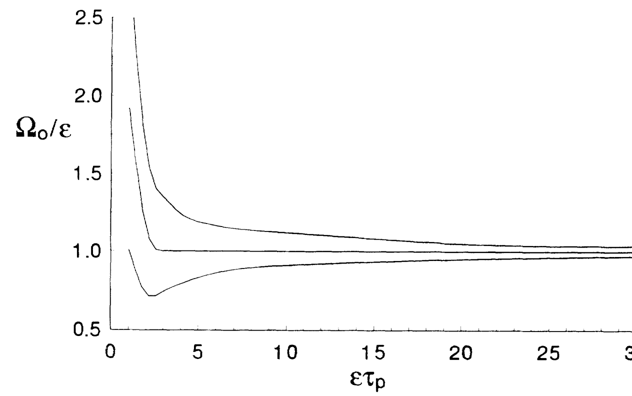

FIG. 3. The value of Ω_0/ε at the first occurrence of complete inversion and the values of Ω_0/ε that delineate the change of the system from absorbing to amplifying and back as functions of $\varepsilon\tau_p$ for a Gaussian pulse.

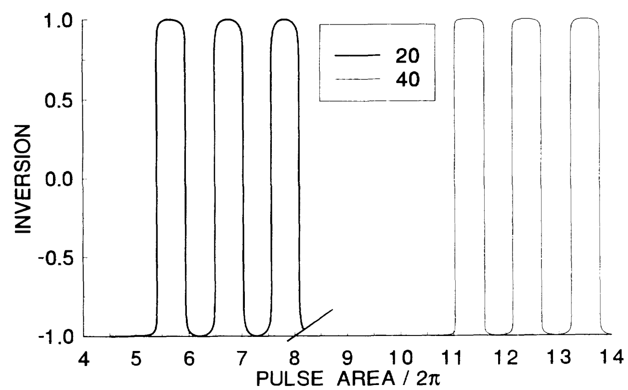

FIG. 4. The final state of the inversion as a function of the pulse area of a Gaussian pulse, as E_0 is varied, for $\varepsilon\tau_p=20$ and for $\varepsilon\tau_p=40$.

providing $\varepsilon\tau_p$ is sufficiently large. In Fig. 4, we have recast Fig. 1 in terms of pulse area. This figure shows that the first maximum of the final inversion occurs at a pulse area of $5.6\times 2\pi$ for $\varepsilon\tau_p=20$ and at $11.3\times 2\pi$ for $\varepsilon\tau_p=40$, in accordance with Eq. (4). If $\varepsilon=0$, the final inversion as a function of pulse area has the usual period of 2π. In Fig. 4, the final inversion as a function of pulse area is approximately periodic with a period slightly greater than 2π, each half cycle of the square wave having a width slightly greater than π. For Gaussian and hyperbolic secant pulses, we find the period to be a slowly increasing function of $\varepsilon\tau_p$. In these cases, the periodicity can be used to obtain a rough estimate of the width and spacing of the features in a graph of the final state of the inversion as a function of Ω_0/ε, e.g., Figs. 1 and 2, for an arbitrary $\varepsilon\tau_p$.

The foregoing results suggest a straightforward method for measuring the strength of the NDD coupling parameter ε by simply increasing the peak field strength (e.g., by removing neutral-density filters) from shot to shot and locating the points where the sharp transitions in the final state of the inversion occur. This can be accomplished by injecting a probe beam in order to determine the state of the medium. Probe amplification would indicate that the medium was left in the excited state, whereas probe depletion would indicate that the medium was left in the ground state. Assuming an appropriate choice of parameters, the midpoint between the peak Rabi frequencies at the first transition from absorbing to amplifying and the first transition back to an absorbing state is the approximate value of ε. The precision of the measurement clearly depends on the pulse width since, the increasing τ_p, these first transition Rabi frequencies converge to ε. However, these transitions may become more difficult to locate due to crowding from similar transitions at slightly higher Rabi frequencies and due to the increasingly narrow width of the full inversion region around ε. In addition, we remark that a rectangular pulse might also be used to measure ε [10]. In this case, an unstable steady-state solution, $w=0$, is obtained when $\varepsilon=2\Omega_0$. This unstable steady state separates two oscillatory regimes with markedly different characteristics. Because the time development of the inversion is oscillatory for the duration of the rectangular pulse, the final state of inversion depends on pulse area in a nontrivial way.

The kind of transitions displayed in Figs. 1 and 2 also suggest that a new kind of optical switch may be devised. The full inversion region for Rabi frequencies near ε can be interpreted as an "on" state and the ground-state region at values of Ω_0 well below ε can be regarded as the "off" state. Then there is a clear application of our results to a two-input AND gate with both inputs being identical pulses with peak Rabi frequencies of $\varepsilon/2$. Further, the ground-state regions and full-inversion regions at higher Rabi frequencies may be exploited. For instance, in the presence of a reference pulse with $\Omega_0=\varepsilon-\delta$, where δ is the width of the full-inversion region, we have an XOR (exclusive or) gate for a pair of input pulses with $\Omega_0=0,\delta$. A NOT gate would consist of an input at $\Omega_0=\varepsilon+(n-1)\delta$ and a reference pulse with $\Omega_0=\delta$. The extension of this reference-plus-inputs scheme to other logic operations is obvious. The kind of switching times and energies required can be calculated from the condition $\Omega_0=\varepsilon$. Restrictions on the parameters that must be satisfied are $\varepsilon\ll\omega_0$ and $\tau_p\ll T_1,T_2$. Further, our results indicate that it is reasonable to require $\varepsilon\tau_p>1$. Assuming a pulse width of 1 ps, a dipole moment of $10ea_0$, and an active device area of 1 μm^2, we determine the total pulse energy to be of the order of a femtojoule at optical frequencies.

Our results also suggest a method for coherent optical pumping since, under conditions described above, the medium is left in the excited state. Full inversion is obtained over a range of peak field strengths of the input field making this method potentially superior to optical pumping by a π pulse, especially for beams which have a transverse intensity profile.

The above results were presented in the context of simple, symmetric pulse shapes in order to maintain clarity in the exposition. Tests with other pulse shapes indicate that the important features of our results are reasonably robust with respect to pulse shape, providing the input field is not highly irregular, such as having multiple local maxima. Of course, this study was not exhaustive due to the enormous number of parameters involved. Figure 5, typical of these results, shows that, in the presence of pulse asymmetry, the wide, flat, full-inversion region near $\Omega_0=\varepsilon$ persists. Here, the periodicity of the square wave rapidly improves with increasing $\varepsilon\tau_p$.

In summary, the principal results of this paper are (i) the existence of a steplike transition in the character of a two-level medium with NDD interactions, from absorbing to amplifying, as a function of the peak field strength of a short optical pulse; (ii) the existence of a corresponding steplike transition from amplifying to absorbing; and (iii) the ability to predict the approximate position of these

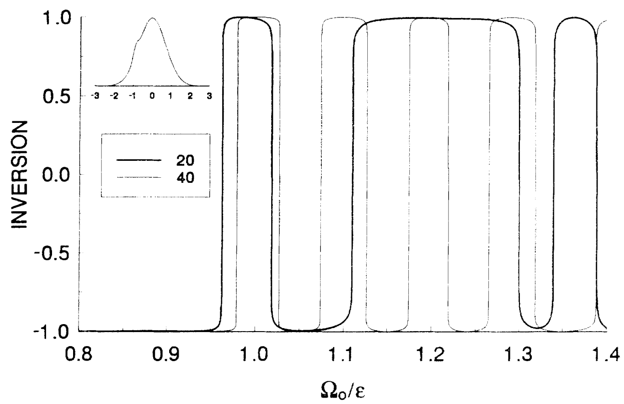

FIG. 5. The final state of the inversion as a function of the peak field strength E_0 of a compound Gaussian pulse, $E_0\{\exp(-t'^2)+0.143\exp[-25(t'+0.875)^2]\}$ for $\varepsilon\tau_p=20,40$. Inset: The temporal profile of the pulse.

transitions, providing $\varepsilon\tau_p$ is sufficiently large ($\varepsilon\tau_p > 1$). These results were obtained by a survey of the numerical solutions of the extended Bloch equations and could not have been predicted *a priori* from a cursory examination of the optical Bloch equations. These results can be used to measure the NDD coupling parameter and for a coherent optical pumping mechanism if propagation effects and medium relaxation times can be neglected, i.e., for thin films and short optical pulses. In addition, a fast, energy-efficient optical switching mechanism has been proposed. Materials in which these effects might be observed include oxygen ions in $KCl:O_2^-$ and bound I_2 excitons at doner sites in CdS single crystals [11].

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