Sub- and Superluminal Propagation of Intense Pulses in Media with Saturated and Reverse Absorption

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We develop models for the propagation of intense pulses in solid state media which can have either saturated absorption or reverse absorption. We model subluminal propagation in ruby and superluminal propagation in alexandrite as three and four level systems, respectively, coupled to Maxwell's equations. We present results well beyond the traditional pump-probe approach and explain the experiments of Bigelow *et al.* [Phys. Rev. Lett. **90**, 113903 (2003); Science **301**, 200 (2003)] on solid state materials.

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Since the discovery of ultraslow light with a group velocity 17 m/s in a Bose condensate by Hau *et al.* [1–3], many experiments have reported slow light in varieties of media [4–6]. Kash *et al.* [4] demonstrated light propagation with a group velocity of 90 m/s at room temperature in Rb vapor. Using Zeeman coherences, Budker *et al.* [5] reported slow light with group velocity 8 m/s in Rb vapor. Hemmer *et al.* [6] first reported slow light in solid state material, Y_2SiO_5 doped with Pr, maintained at a cryogenic temperature of 5 K. The slow light ideas have been successfully used in storage and retrieval of light pulses [7,8]. The understanding of storage and retrieval of light pulses has been provided by Dey and Agarwal [9], using the adiabaton theory of Grobe, Hioe, and Eberly [10].

Work on pulse propagation continues to produce interesting results [11–14]. Recently, Bigelow *et al.* [15] showed the propagation of light pulses in ruby at a group velocity of 57.5 m/s. This experiment differs considerably from all earlier experiments which were based on electromagnetically induced transparency [16]. Bigelow *et al.* recognize that a two level system driven by a strong field and a probe gives rise to a hole in the probe response function with a width of the order of $1/T_1$, where T_1 is the longitudinal relaxation time [17]. Note that in a material such as ruby the transverse relaxation time $T_2 \ll T_1$. These authors also discovered that they need not use separate pump and probe fields. A field with a peak power of the order of saturation intensity could be slowed down considerably to about 57.5 m/s. Further, Bigelow *et al.* [18] extended their work to a material such as alexandrite, where they reported superluminal propagation. There have been several earlier reports of superluminal propagation in solid state systems [19] and in vapors [20,21].

The purpose of this Letter is to study the propagation of intense pulses in a homogeneously broadened medium, such as ruby, which can exhibit saturated absorption or a medium such as alexandrite, which can exhibit reverse absorption. Note that the pulse propagation in a nonlinear transparent medium has been extensively studied [22]. The systems studied here differ from the previous studies as our systems possess very strong transverse and longitudinal relaxation effects. In order to model the experiments, we model ruby as a three level system and alexandrite as a four level system. We solve the system of coupled equations numerically to delineate various aspects of pulse propagation. We do not make any approximation on the strength of the pulses so that we can model experimental observations on strong pulses. We calculate group velocity from the relative delay or advancement between the reference pulse and the output pulse. We present numerical results on the propagation of Gaussian and modulated pulses and show qualitative agreement with the experimental data of Refs. [15,18]. The experiments of Bigelow *et al.* fall into two categories. One consists of a weak probe and a strong coherent cw field. These were explained in terms of the response to a probe field of a two level medium pumped by a coherent field [23]. The other category consists of the self-delay of a strong pulse. The latter requires solutions of the coupled Maxwell-Bloch equations and this is the case on which we concentrate.

Media with saturated absorption.—For pulse propagation in ruby, we represent the ground state as $|g\rangle$, the $4F_2$ absorption band as $|e_1\rangle$, and the levels $2\bar{A}$ and $\bar{E}$ as $|e_2\rangle$. The system is modeled as a three level system [24] in ruby, one has very rapid decay of the level $|e_1\rangle$ to $|e_2\rangle$, and as a result some of the coherences become irrelevant on experimental time scale. The density matrix equations for the model of Fig. 1 are

$$\begin{aligned}\dot{\rho}_{gg} &= 2\Gamma_2\rho_{22} + i\frac{\Omega}{2}(\rho_{1g} - \rho_{g1}),\\ \dot{\rho}_{22} &= 2\Gamma_1\rho_{11} - 2\Gamma_2\rho_{22},\\ \dot{\rho}_{1g} &= -\Gamma_1\rho_{1g} + i\frac{\Omega}{2}(\rho_{gg} - \rho_{11}),\\ \rho_{gg} + \rho_{11} + \rho_{22} &= 1, \end{aligned} \quad (1)$$

where $\rho_{ij} = \langle e_i|\rho|e_j\rangle$, $i, j = 1, 2$. The Rabi frequency Ω is defined by $\Omega(z, t) = 2\vec{d}_{1g} \cdot \vec{\mathcal{E}}(z, t)/\hbar$, where $\vec{d}_{1g}$ is the dipole matrix element and $\vec{\mathcal{E}}(z, t)$ is the envelop of the pulse.

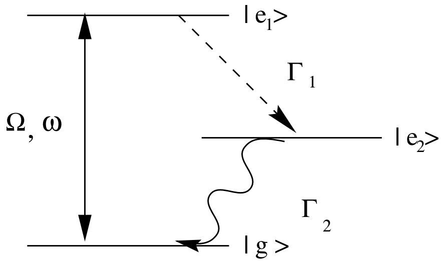

FIG. 1. Three level model for ruby crystal.

We assume that the carrier frequency, ω, is on resonance with the frequency of the $|e_1\rangle \leftrightarrow |g\rangle$ transition. Under the approximations, $\Gamma_1 \gg \Gamma_2, \Omega$; $\dot{\rho}_{1g} \sim 0$, we derive the approximate equation for the evolution of the ground state population as

$$\dot{\rho}_{gg} = 2\Gamma_2(1 - \rho_{gg}) - \frac{\Omega^2}{2\Gamma_1}\rho_{gg}. \tag{2}$$

Note that we can prove that $\dot{\rho}_{11} \approx 0$, if $\Gamma_1 \gg \Gamma_2, \Omega$. Under the same conditions and the slowly varying envelop approximation, the evolution equation for the Rabi frequency of the field is governed by

$$\frac{\partial\tilde{\Omega}}{\partial z} = -\frac{\alpha_0}{2}\tilde{\Omega}\rho_{gg}, \qquad \tilde{\Omega} = \Omega/\Omega_{\text{sat}}, \tag{3}$$

where $\alpha_0 = 4\mathcal{N}\pi\omega|d_{1g}|^2/c\hbar\Gamma_1$ and $\Omega_{\text{sat}} = 2\sqrt{\Gamma_1\Gamma_2}$. In Eqs. (2) and (3) we have used the pulse coordinates; i.e., $t - z/c, z$. The time derivative in Eq. (2) is with respect to $(t - z/c)$. The time t can be expressed in units of $1/2\Gamma_2$. For numerical computation, we consider two types of input pulses, viz. a Gaussian pulse with a temporal width $\gtrsim 1/\Gamma_2$:

$$\tilde{\Omega}_{\text{in}} = \tilde{\Omega}^0 e^{[-t^2/2\sigma^2]}, \tag{4}$$

and amplitude modulated pulse,

$$\tilde{\Omega}^2_{\text{in}}(t) = I = I_0(1 + m\cos[\Delta t]). \tag{5}$$

Equations (2)–(5) are our working equations. We use these for numerical computations. We calculate the evolution of pulse for arbitrary values of $\tilde{\Omega}^0$ or I_0. Some typical results for the Gaussian pulses are shown in the Fig. 2. We get group velocities in the range 50 m/s for $\Omega/\Omega_{\text{sat}} \backsim 1$ and the transmission is rather small. In Fig. 3, we exhibit the behavior of v_g and transmission as a function of the input intensity. These results, in general, are in agreement with the experimental findings of transmission in the range 0.1%. In Fig. 3, we also show for comparison the results of the group velocity and the transmission for the propagation of an intense pulse through a two level system described by the traditional Bloch equations. The coupled Maxwell-Bloch equations under the approximation $T_1 \gg T_2$ are given by

$$\dot{\rho}_{gg}T_1 = (1 - \rho_{gg}) - \frac{|\tilde{\Omega}|^2}{2}(2\rho_{gg} - 1), \tag{6}$$

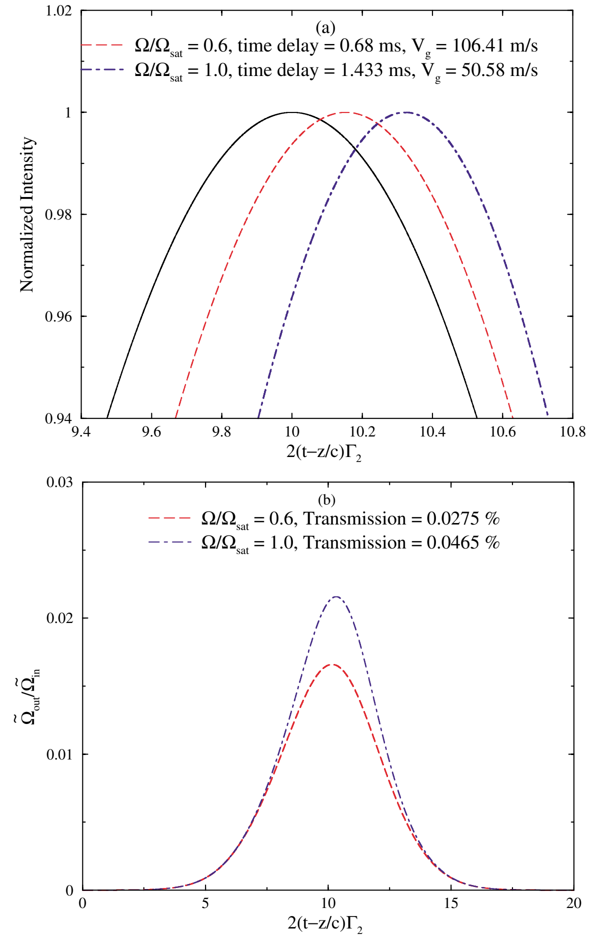

FIG. 2 (color online). (a) The solid curve shows light pulse propagating at speed c through 7.25 cm in vacuum. The long dashed and dot-dashed curves show light pulses propagating through a medium of length 7.25 cm at different input amplitudes. The temporal width σ of the Gaussian pulse is 20 ms and $1/2\Gamma_2 = 4.45$ ms. Part (b) gives the amplitudes of the output pulse normalized to the input amplitudes. The transmission increases with increasing the input field intensity.

$$\frac{\partial\tilde{\Omega}}{\partial z} = -\frac{\alpha_0}{2}\tilde{\Omega}(2\rho_{gg} - 1), \tag{7}$$

where $\tilde{\Omega} = \Omega\sqrt{T_1T_2}$ and the dot denotes $\partial/\partial(t - z/c)$. As seen from Fig. 3, there are substantial differences in the propagation of pulses in two level and three level media. Note that the time T_1 is equal to $1/2\Gamma_2$. We believe that, in the light of the energy level diagram of ruby, it is more appropriate to model it as a three level system. The two level model misses the interesting physics, as in the effective two level model there would be a field induced transition from $|e_2\rangle$ to $|g\rangle$, whereas in the three level scheme this does not occur. As a result, there are important differences in the way the two level and the three level systems saturate.

We next consider the input pulse as a modulated pulse given in Eq. (5). The output pulse is modulated with a phase shift (time delay). We show this time delay as a

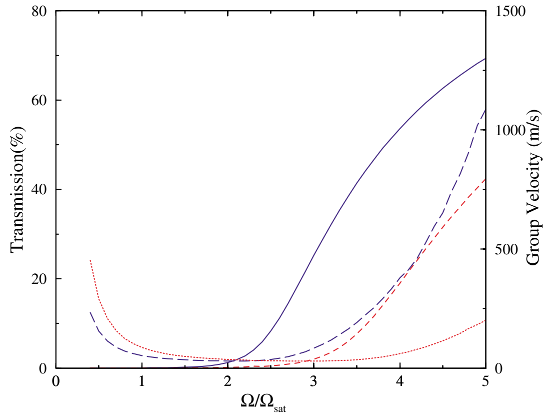

FIG. 3 (color online). Variation of transmissions and group velocities as a function of the input amplitude of the light pulse. The solid (dashed) curve gives the intensity transmission of the pulse for the three (two) level model of the medium. The corresponding group velocities are given by the dotted curve (two level model) and the long dashed curve (three level model). The light pulse is propagating through the medium of length 7.25 cm.

function of modulation frequency for three different pump powers in Fig. 4 [25]. While our results in Figs. 2–4 are in broad agreement with the experimental data, we do not make a precise comparison due to the sensitive dependence on pump powers, focusing of the pump and the possible uncertainty in the known value of the saturation power for the conditions of the experiment.

Media with reverse saturation.—For the superluminal propagation in alexandrite, Bigelow *et al.* recognized how the reverse saturation mechanism [26] can be at work in a material such as $BeAl_2O_4$ doped with Cr^{3+} ions and some Cr^{3+} ions replaced by Al^{3+}. The reverse saturation produces an antihole in the susceptibility for the probe in the presence of a pump field. This antihole can result in the superluminal propagation. In what follows, we show how the measurement can follow by modeling the system as a four level system to account for reverse absorption. The model is shown in Fig. 5, where state $4A_2$ as $|g\rangle$, the absorption bands $4T_2$ and $4T_1$ as $|e_1\rangle$, and the level $2E$ as $|e_2\rangle$. The density matrix equations are now given by

$$\begin{aligned} \dot{\rho}_{gg} &= 2\Gamma_2\rho_{22} + i\Omega(\rho_{1g} - \rho_{g1})/2, \\ \dot{\rho}_{22} &= 2\Gamma_1\rho_{11} - 2\Gamma_2\rho_{22} + 2\Gamma_3\rho_{33} \\ &\quad + i\Omega(\rho_{32} - \rho_{23})/2, \\ \dot{\rho}_{33} &= -2\Gamma_3\rho_{33} + i\Omega(\rho_{23} - \rho_{32})/2, \\ \dot{\rho}_{32} &= -\Gamma_3\rho_{32} + i\Omega(\rho_{22} - \rho_{33})/2, \\ \dot{\rho}_{1g} &= -\Gamma_1\rho_{1g} + i\Omega(\rho_{gg} - \rho_{11})/2, \\ \rho_{gg} &+ \rho_{11} + \rho_{22} + \rho_{33} = 1. \end{aligned} \tag{8}$$

Following the same procedure as in the case of ruby, we have derived the working equations

$$\frac{\dot{\rho}_{gg}}{2\Gamma_2} = (1 - \rho_{gg}) - |\tilde{\Omega}^2|\rho_{gg}, \tag{9}$$

$$\frac{\partial\tilde{\Omega}}{\partial z} = -\frac{\alpha_0}{2}\tilde{\Omega}\rho_{gg} - \frac{\tilde{\alpha}_0}{2}\tilde{\Omega}(1 - \rho_{gg}), \tag{10}$$

where $\tilde{\alpha}_0$ gives the reverse saturation. Following the experimental data of Bigelow *et al.* [18], we estimate $(\tilde{\alpha}_0/\alpha_0) \approx 4$. Equations (9) and (10) are numerically integrated for the input Gaussian pulse given by Eq. (4). A representative set of results is shown in Fig. 6. This figure also shows how the group velocity and net transmission depend on the peak intensity of the Gaussian pulses. It should be borne in mind that, in the range of the intensities of Fig. 6, no perturbation theory can be used. One has to study the full nonlinear behavior. We also notice that the input pulses get distorted in shape. The distortion becomes more pronounced as the nonlinearity of the medium becomes more pronounced.

In conclusion, we have shown how to model the propagation of an intense pulse in solid state media with very strong relaxation effects. The media can exhibit either saturated absorption or reverse absorption. Our modeling goes well beyond the traditional pump-probe approach.

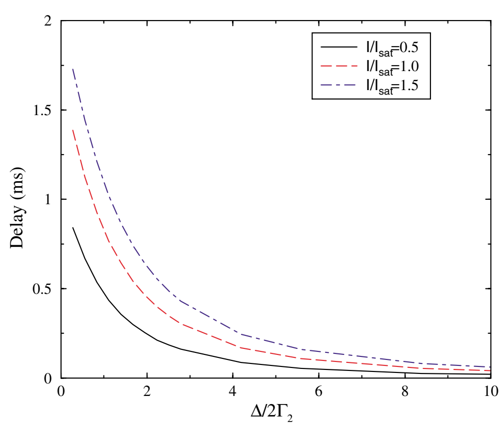

FIG. 4 (color online). Time delay of the light pulse as a function of modulation frequency for three different input powers for modulation index, $m = 0.06$. Note that $\Delta/2\Gamma_2 \sim 1$ corresponds to a modulation frequency ~35 Hz.

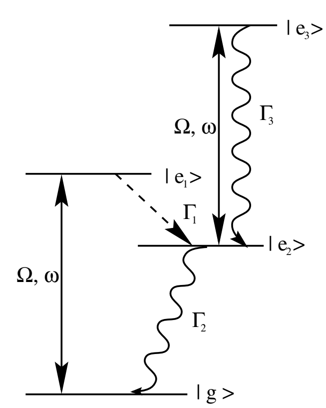

FIG. 5. Four level model for alexandrite crystal.

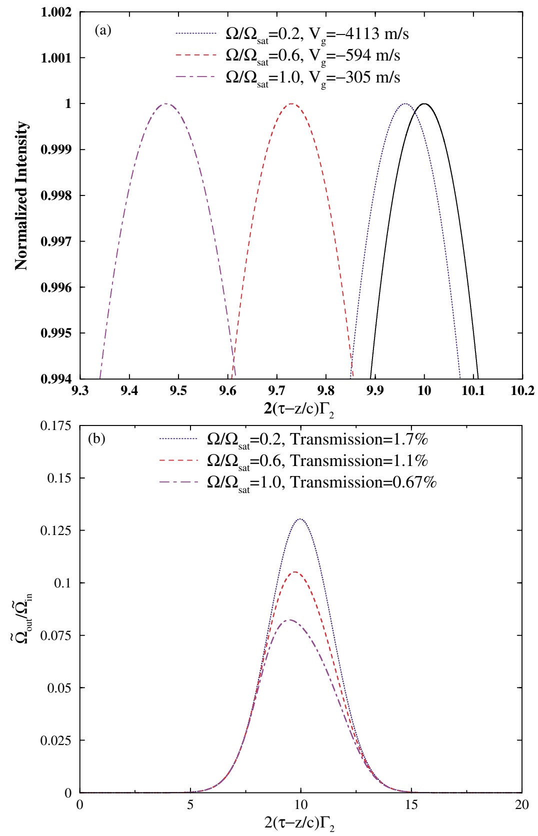

FIG. 6 (color online). The solid curve of (a) shows light pulse propagating at speed c through a distance of 4 cm in vacuum. The dotted, long dashed, and dot-dashed curves depict light pulse propagating through a medium of length 4 cm at different input amplitudes. The pulse width σ is 500 μs, whereas $1/2\Gamma_2 = 250$ μs. Part (b) shows the amplitude of the output pulse normalized with input amplitude. The transmission is decreased with an increase in the input field intensity.

We specifically present results on the propagation of pulses in ruby and alexandrite. Our model would also be applicable to other systems where reverse absorption could be dominant.

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