Slow light with persistent hole burning

R. N. Shakhmuratov,^{1,2,3} A. Rebane,⁴ P. Mégret,⁵ and J. Odeurs¹

¹Instituut voor Kern- en Stralingsfysica, Katholieke Universiteit Leuven, Čelestijnenlaan 200 D, B-3001 Leuven, Belgium

²Optique Nonlinéaire Théorique, Université Libre de Bruxelles, Campus Plaine CP 231, B-1050 Bruxelles, Belgium

³Kazan Physical Technical Institute, Russian Academy of Sciences, 10/7 Sibirsky trakt, Kazan 420029 Russia

⁴Physics Department, Montana State University, Bozeman, Montana 59717, USA

⁵Faculté Polytechnique de Mons, Boulevard Dolez 31, B-7000 Mons, Belgium

(Received 1 February 2005; published 24 May 2005)

We consider the propagation of a Gaussian probe pulse in an absorptive, optically dense, two-level medium if a deep, persistent hole is created in advance by another pump field in the inhomogeneously broadened absorption spectrum of this medium. Both fields are well separated in time and the lifetime of the hole is assumed to be long with respect to the delay time between the pump and probe pulses. We show that the group velocity of the Gaussian probe pulse reduces several orders in magnitude, similarly to the reduction of the group velocity for the probe field in electromagnetically induced transparency (EIT) phenomena. In contrast to EIT, the width of the transparency window can be made very wide because of the saturation broadening of the hole.

DOI: 10.1103/PhysRevA.71.053811

PACS number(s): 42.50.Gy, 42.50.Md, 42.25.Bs

I. INTRODUCTION

The propagation of light pulses with extremely low group velocity is a phenomenon that attracts continuously growing attention [1-5]. This is because slow light allows for high-efficiency nonlinear conversion with substantially alleviated phase matching problems. Some other applications are also possible. We mention just a few: controllable optical delay lines, storage of light or information it contains, high precision spectroscopy and magnetometery, etc.

Slow light can be produced by using electromagnetically induced transparency (EIT) [6-10]. The EIT proposal was stimulated by the observation of the dark resonances in sodium vapor [11], which were explained by the population trapping in the dark, nonabsorbing state [9,12]. EIT is achieved in a three-level atom with the help of a coupling field producing together with a probe field a two-quantum coherence with a long lifetime. This coherence is due to the population trapping in the dark state. The atom in this state cannot be excited because of destructive interference of two transition paths to the excited state. The creation of the dark state coherence is revealed as a narrow hole in the absorption line for the probe. Since the width of the hole is usually not greater than the Rabi frequency induced by the coupling field, one cannot produce a very broad EIT hole. A typical value of the broadest hole produced by an extended cavity diode laser is about 15 MHz [13]. There are two exceptions when powerful lasers were used. This is a dye laser, pumped by a Nd:YAG laser, with intensity 15 MW/cm², which produced a Rabi frequency ≈ 30 GHz in Sr vapor [7]. Another example is the Nd:YAG laser with intensity $\approx 9 \text{ MW/cm}^2$, which produced a Rabi frequency ≈ 300 GHz in Pb vapor [8]. The limitation of the EIT bandwidth by the value of the induced Rabi frequency sets a constraint for the application of EIT for telecommunication and fast transient phenomena performed with semiconductor lasers integrated into a microelectronic circuit. However, EIT has many advantages in high-resolution spectroscopy and frequency stabilization, since the width of the EIT hole can be made very narrow, of the order of 10 kHz [5].

Recently, two proposals to produce slow light in a twolevel medium have been published by Bigelow et al. [14] and Agarwal et al. [15]. Actually, the first proposal, Ref. [14], addresses the three-level system. However, in this scheme two fields excite the same transition e-g and the third level (a trapping state t coupled with the excited state e by the fast nonradiative decay process $e \rightarrow t$) is introduced to have a slow decay of the population difference of the levels e and g coupled by the fields, where g is the ground state. The first field, the pump, excites the broad absorption line and the second field, the probe, is slightly detuned from the frequency of the pump, ω_{pump} . Such a case can be realized by a small amplitude modulation of the pump with frequency ω_m , which produces sidebands $\omega_{\text{prob}} = \omega_{\text{pump}} \pm \omega_m$. The simultaneous interaction of the probe and the pump with the medium results in a narrow hole in the absorption of the probe when $\omega_{\text{prob}} \rightarrow \omega_{\text{pump}}$ or $\omega_m \rightarrow 0$. The width of this hole is defined by the value $1/T_1$, where T_1 is the lifetime of the population difference of the ground g and excited e states. In case of Ref. [14], T_1 is mostly defined by the slow radiative decay of the trapping state to the ground state, $t \rightarrow g$. This is the so-called T_1 hole, which is actually a coherent hole, i.e., the hole is present only when the pump is on and this hole vanishes immediately after the pump is switched off; see, for example, Refs. [16–18]. The T_1 hole appears due to the coherent process of the population oscillation on the beat frequency $\omega_m = |\omega_{\text{pump}} - \omega_{\text{prob}}|$. The disadvantage of the production of slow light with the help of a T_1 hole is the small frequency difference between the pump and probe fields.

In the second proposal, Ref. [15], a similar idea of producing slow light is employed. In this scheme, Dopplerbroadened two-level systems (without any third, trapping state) are exposed to the probe and pump, acting simultaneously such that a slightly modulated probe experiences the influence of the counterpropagating cw pump. The modulation of the probe at the output of the medium has a delay with respect to the modulation of the field, which would travel the same distance in free space. The delay shows that the group velocity of the probe field inside the absorber reduces many times.

The difference between these two proposals, described in Refs. [14,15], is that the hole produced by the pump in the Doppler-broadened absorption spectrum of the two-level systems is much broader. This is because in gases T_1 is not long and usually we have $T_1 = T_2/2$, where T_1 and T_2 are the lifetimes of the population difference and of the coherence, respectively (here T_2 describes a homogeneous dephasing process). It is well known that the half width of the hole seen by the probe broadens due to the saturation by the pump as $T_2^{-1} + \sqrt{T_2^{-2}} + \Omega_{\text{pump}}^2 T_1 / T_2$, where Ω_{pump} is the Rabi frequency (the coupling parameter) of the pump; see, for example Ref. [19]. Therefore the half width of the narrowest hole in a Doppler-broadened absorption line, which a probe can see, is $2/T_2$. This hole has coherent and incoherent contributions. The coherent part originates from the oscillations induced by the pump and probe fields acting simultaneously. The incoherent part is caused by the saturation of the population difference of the resonant levels by the pump or, in other words, due to the hole burning. The lifetime of the incoherent part of the hole is defined by T_1 [19].

In this paper we propose to produce slow light in solids doped by impurity ions or in polymers doped by dye molecules where the incoherent hole has a very long lifetime. In our scheme, in a first preparative stage, a pump pulse creates a hole in the absorption spectrum of the sample. Then after some delay time shorter than T_1 but longer than T_2 , the probe is applied. We show that the probe propagates with slow group velocity. In this case the pump and probe are applied at different times. The hole can be made as narrow as $2/T_2$ by a weak pump $(\Omega_{pump}^2 T_1 T_2 \ll 1)$, or it can be made very broad by a strong saturating pump $(\Omega_{pump}^2 T_1 T_2 \gg 1)$ because for solids $T_1 \ge T_2$ and on the condition of strong saturation, the half width of the hole approaches $\Omega_{\text{pump}}\sqrt{T_1/T_2}$. Narrow and broad holes allow work with long and short probe pulses, respectively. Several holes can be burnt in different parts of the inhomogeneous absorption line. Therefore several probe pulses with frequencies coinciding with the frequency positions of these holes can be applied simultaneously. They will all have slow group velocities. This opens wide perspectives for the construction of multichannel optical storage devices (not discussed in this paper).

Our consideration can be applied to the case of a true two-level system with long lifetime of the excited state or to the case of an effective two level system having a long relaxation rate of the population difference due to the trapping state. For ions T_1 can be as long as several milliseconds, as, for example, in the case of the R_1 line of Cr^{3+} in ruby (true two-level system). For some organic dye molecules, this time can be even much longer because of photochemical reactions, which alter the whole chemical structure of the molecule, see, for example, Refs. [20,21]. The latter is due to the trapping state *t* having almost infinite lifetime. There are also trapping states in solids doped with Sm²⁺ ions. In these solids it is possible to obtain persistent spectral hole burning even at room temperature, see, for example, Refs. [22,23].

The paper is organized as follows. In Sec. II we give a brief introduction to the pump-probe spectroscopy, showing

how the hole is burnt by the pump and how the probe interacts with atoms prepared in the nonequilibrium state created by the hole burning. In Sec. III we consider the propagation of the probe in a thick absorptive medium if a deep hole is burnt in the inhomogeneous absorption spectrum. In Sec. IV we compare EIT and hole burning from the viewpoint of the population trapping in the nonabsorbing state.

II. HOLE BURNING

We consider a solid at low temperature with impurities (ions or molecules) whose resonant frequencies ω_n are distributed around some central frequency ω_c according to $N_c(\omega_n)$ due to crystal imperfections. For simplicity we model an individual impurity by a two-level system (TLS) with resonant frequency ω_n . For each TLS we assume that the relaxation time T_1 of the population difference is much longer than the dephasing time T_2 of the induced coherence. In a first, hole burning stage, such an ensemble of TLS is prepared by the pump pulse, which is sufficiently strong and long to saturate those TLS whose frequencies are close to the frequency of the pump, ω_{pump} . When the pump is switched off, for example, abruptly, the induced polarization of the TLS decays with time T_2 , i.e., much faster than the population difference modified by the pulse. Then, after some delay time τ_d , which is much longer than T_2 but shorter than T_1 , we apply the probe pulse. If the frequency of the probe is in resonance with those TLS whose population difference is still saturated (i.e., nearly equalized due to the pump), the medium becomes transparent for the probe. If the frequency of the probe is tuned far from ω_{pump} but still inside the contour of the inhomogeneous line, the probe field is absorbed.

All three regimes, i.e., pump on, pump off and probe on, are described by the matter equations for the *n*th TLS,

$$\dot{\sigma}_n = (i\Delta_n - \Gamma)\sigma_n + i\frac{\chi}{2}w_n, \qquad (1)$$

$$\dot{w}_n = i\chi(\sigma_n - \sigma_n^*) - \gamma(w_n - 1), \qquad (2)$$

where $\sigma_n(z,t) = \rho_{eg}(z,t) \exp(i\omega t - i\mathbf{kr})$ is the slowly varying amplitude of the nondiagonal component of the TLS density matrix $\hat{\rho}$ and $w_n = \rho_{gg} - \rho_{ee}$ is the population difference of the ground g and excited e states. The decay rates of the coherence and of the population difference are defined as Γ $= 1/T_2$ and $\gamma = 1/T_1$, respectively. ω is the pulse frequency ($\omega = \omega_{pump}$ for the pump and $\omega = \omega_{prob}$ for the probe), **k** and **r** are the wave vector and the propagation distance of the field inside the medium, respectively; $\Delta_n = \omega - \omega_n$ is the resonant detuning for the *n*th TLS, and $\chi = 2\mathbf{d}_{eg}\mathbf{E}/\hbar$ is the Rabi frequency ($\chi = \Omega_{pump}$ for the pump and $\chi = \Omega_{prob}$ for the probe), where **E** is the amplitude of the pump or probe pulse, which is in general time dependent, and \mathbf{d}_{eg} is the dipole transition matrix element (assumed to be real, i.e., $\mathbf{d}_{eg} = \mathbf{d}_{eg}$).

For simplicity we assume that the duration of the pump is long enough and that it has a rectangular shape, so that the TLS reaches a steady-state saturation at the end of the pulse. This state is described by the population difference [19]

$$w_{\rm st}(\Delta_n) = 1 - \frac{\Omega_{\rm pump}^2 T_1 T_2}{1 + \Delta_n^2 T_2^2 + \Omega_{\rm pump}^2 T_1 T_2}.$$
 (3)

We also neglect the relaxation of the population during the delay time τ_d between the pump and probe pulses. Therefore the initial condition in Eqs. (1) and (2) for the probe pulse is $w_n = w_{st}(\Delta_n)$, $\sigma_n = 0$. We assume that the probe pulse is sufficiently weak so that the linear-response approximation is applicable for the solution of the matter equations. Then one can consider only Eq. (1), neglecting the change of the population difference w_n , which is supposed to remain $w_n = w_{st}(\Delta_n)$. By means of the Fourier transform

$$F(\nu) = \int_{-\infty}^{+\infty} f(t)e^{i\nu t}dt,$$
(4)

Eq. (1) is reduced to a simple algebraic equation. Its solution is

$$\sigma_n(\nu) = \frac{i\Omega_{\text{prob}}(\nu)w_{\text{st}}(\Delta_n)}{2[\Gamma - i(\nu + \Delta_n)]},$$
(5)

where $\Omega_{\text{prob}}(\nu)$ is the Fourier transform of $\Omega_{\text{prob}}(t)$ and it is assumed that $\lim_{t\to\pm\infty} [\Omega_{\text{prob}}(t)]=0$, $\lim_{t\to\pm\infty} [\sigma_n(t)]=0$. For simplicity we consider the case when the frequencies of the pump and probe fields are the same, $\omega_{\text{pump}}=\omega_{\text{prob}}$. Therefore $w_{\text{st}}(\Delta_n)$ in Eq. (3) contains the same detuning parameter Δ_n as $\sigma_n(\nu)$ in Eq. (5).

The average response of the TLS ensemble is given by

$$\langle \sigma(\nu) \rangle = \frac{1}{N_0} \int \sigma_n(\nu) N_c(\omega_n) d\omega_n,$$
 (6)

where $N_c(\omega_n)$ is the partial concentration of the TLS with frequency ω_n in the sample and N_0 is the overall concentration: $N_0 = \int N_c(\omega_n) d\omega_n$. The function $N_c(\omega_n)$ is usually a Gaussian distribution. Due to the complexity in the integration with the Gaussian distribution, we approximate it with a Lorentzian function,

$$N_{c}(\omega_{n}) = \frac{1}{\pi T_{2}^{*}} \frac{N_{0}}{(\omega_{n} - \omega_{c})^{2} + \left(\frac{1}{T_{2}^{*}}\right)^{2}},$$
(7)

where $1/T_2^* = \Gamma_{\text{inh}}$ is the half width of the inhomogeneous absorption line. This is a conventional approximation, which leads to a fairly simple form of the inhomogeneously broadened susceptibility, allowing for a detailed analytical treatment (see, for example, Ref. [24]). Below we consider the case if pump and probe are tuned to the center of the inhomogeneous line ω_c , i.e., their carrier frequencies satisfy the condition $\omega_{\text{pump}} = \omega_{\text{prob}} = \omega_c$.

The polarization induced by the probe field in the ensemble of two-level atoms is described by the equation

$$\mathbf{P}(\nu) = N_0 \mathbf{d}_{eg} \langle \sigma(\nu) \rangle. \tag{8}$$

Performing the integration, we obtain

$$\mathbf{P}(\nu) = [\chi'(\nu) + i\chi''(\nu)]\mathbf{E}_{\text{prob}}(\nu), \qquad (9)$$

where $\mathbf{E}_{\text{prob}}(\nu)$ is the Fourier transform of the probe field envelope, $\mathbf{E}_{\text{prob}}(t)$, and



FIG. 1. (a) and (b) Frequency dependence of the imaginary and real parts of the effective linear susceptibility of the TLS ensemble if the pump field is applied in advance, $\chi''(\nu)$ and $\chi'(\nu)$ (bold lines), and if not, $\chi''_0(\nu)$ and $\chi'_0(\nu)$ (thin lines). Both plots are normalized to $\chi''_0(0)$. The frequency scale is in units of the homogeneous half width, ν/Γ .

$$\chi'(\nu) = \chi_0'(\nu) \left\{ 1 - \frac{p^2}{(\Gamma_{\text{inh}} + \Delta_h)\Delta_h} \left[1 + \frac{(\Gamma_0 + \Delta_{hp})\Gamma_{\text{inh}}}{\nu^2 + \Delta_{hp}^2} \right] \right\},\tag{10}$$

$$\chi''(\nu) = \chi_0''(\nu) \Biggl\{ 1 - \frac{p^2}{(\Gamma_{\rm inh} + \Delta_h) \Gamma_0 \Delta_h} \Biggr[\Gamma + \frac{(\Gamma_0 + \Delta_{hp}) \Gamma_{\rm inh} \Delta_{hp}}{\nu^2 + \Delta_{hp}^2} \Biggr] \Biggr\}.$$
 (11)

Here $p = \Omega_{\text{pump}} \sqrt{\Gamma/\gamma}$, $\Delta_h = \sqrt{p^2 + \Gamma^2}$ is the half width of the dip in the function $w_{\text{st}}(\Delta_n)$, and $\Delta_{hp} = \Delta_h + \Gamma$ is a half width of the hole seen by the probe. $\chi'_0(\nu)$ and $\chi''_0(\nu)$ are the real and imaginary parts of the effective linear susceptibility of the TLS ensemble if the pump is not applied in advance (p=0), i.e.,

$$\chi_0'(\nu) = -\frac{N_0 |d_{eg}|^2}{\hbar} \frac{\nu}{\nu^2 + \Gamma_0^2},$$
(12)

$$\chi_0''(\nu) = \frac{N_0 |d_{eg}|^2}{\hbar} \frac{\Gamma_0}{\nu^2 + \Gamma_0^2},$$
(13)

and $\Gamma_0 = \Gamma_{inh} + \Gamma$ is the half width of the absorption line seen by the probe if the TLS ensemble is in the equilibrium state and not perturbed in advance. Figure 1 shows the frequency dependence of $\chi''(\nu), \chi''_0(\nu)$ (a) and $\chi'(\nu), \chi'_0(\nu)$ (b). The parameters are $\Gamma_{inh} = 100\Gamma$ and $\Delta_h = 10\Gamma$. The presence of a narrow hole is clearly seen at the center of the absorption line. The dispersion curve, Fig. 1(b), exhibits a steep variation in the same frequency domain where the dip in the absorption line is present.



FIG. 2. Excitation scheme of a thin slab by the pump E_{pump} and probe E_{prob} fields. The length of the slab is *l* and its thickness is $d(l \ge d)$.

Such a frequency dependence of the atomic susceptibility is absolutely similar to the one that is typical for EIT.

III. PULSE PROPAGATION THROUGH A DOMAIN BLEACHED BY A TRANSVERSE PUMP

We consider the propagation of a probe pulse in a thin slab of an optically dense absorptive medium shown in Fig. 2. This slab has a length l and a thickness $d(d \ll l)$. The probe pulse propagates along the slab (this path is assumed to be optically thick). Before the probe pulse is applied, the slab is prepared by the pump field. The pump is applied perpendicular to the slab (this path is assumed to be optically thin). The pump makes a "trench" of optically bleached atoms shown by the area confined between the dashed lines. Since $d \ll l$, the slab is thin for the pump, but thick for the probe field. Such a geometry of the pump-probe excitation of an optically dense medium was experimentally developed in Refs. [25,26].

We do not consider the transverse structure of the probe field and describe its propagation inside the slab by the onedimensional wave equation for the slowly varying amplitude of the probe, $E_{\text{prob}}(z, t)$,

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right) E_{\text{prob}}(z,t) = i\frac{\alpha_0\hbar}{d_{eg}}\langle\sigma(z,t)\rangle, \qquad (14)$$

where the right-hand side is the response function of the atoms located in a plane with coordinate z (for a derivation of this equation see, for example, Ref. [27]). This function $\langle \sigma(z,t) \rangle$ is the inverse Fourier transform of $\langle \sigma(z,\nu) \rangle$. The parameter $\alpha_0 = 2 \pi N_0 |d_{eg}|^2 \omega / \hbar c$ is the resonant absorption coefficient of the sample with concentration N_0 of the two-level atoms. The probe pulse is considered as a plane wave with wave vector \mathbf{k}_{prob} parallel to the z axis directed along the slab. The variation of the probe field amplitude in the x-y plane perpendicular to z is neglected and it is assumed that E_{probe} is confined in the domain between the dashed lines shown in Fig. 2.

The wave equation (14) for the Fourier transform of the probe field envelope, $E_{\text{prob}}(z, \nu)$, can be rewritten as

$$\left(\frac{\partial}{\partial z} - \frac{i}{c}\nu + A(\nu)\right) E_{\text{prob}}(z,\nu) = 0, \qquad (15)$$

$$A(\nu) = \frac{2\pi\omega}{c} [\chi''(\nu) - i\chi'(\nu)].$$
 (16)

This equation is integrated as $E_{\text{prob}}(z, \nu) = E_{\text{prob}}(0, \nu) \exp[(i\nu z/c) - A(\nu)z]$. If one takes the inverse Fourier transform, the resulting expression for the probe pulse envelope is

$$E_{\text{prob}}(z,t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} E_{\text{prob}}(0,\nu) \exp\left[-i\nu\left(t-\frac{z}{c}\right) - A(\nu)z\right] d\nu.$$
(17)

This is the general solution for the propagation of a small probe pulse, $E_{\text{prob}}(z, t)$, in a sample with arbitrary thickness containing two-level atoms prepared by the pump field $E_{\text{pump}}(t)$.

We consider the case when the spectral half width of the incoming probe pulse, Δ_{in} , is smaller than the half width of the hole, Δ_{hp} . Then one can expand the function $A(\nu)$ in a power series near $\nu=0$

$$A(\nu) = \frac{\alpha}{2} \sum_{k=0}^{\infty} (-i\nu)^k a_k,$$
 (18)

where $\alpha = 2\alpha_0/\Gamma_0$ is the Beer's law attenuation coefficient for the probe propagating in the medium not excited by the pump field. According to Eqs. (10) and (11), this expansion converges if $\nu < \Delta_{hp}$. Below we show that, if $\Delta_{in} < \Delta_{hp}$, three terms of expansion (18) are sufficient to describe the probe pulse propagation in the resonant medium saturated by the pump. These terms have the following coefficients:

$$a_0 = \frac{\Gamma_0 \Gamma}{(\Gamma_{\rm inh} + \Delta_h) \Delta_h},\tag{19}$$

$$a_1 = \frac{1}{\Delta_h} \left[1 - \frac{2\Gamma}{\Delta_{hp}} - \frac{\Delta_h^2 + \Gamma^2}{(\Gamma_{\rm inh} + \Delta_h)\Delta_{hp}} \right],\tag{20}$$

$$a_2 = \frac{1}{(\Gamma_{\text{inh}} + \Delta_h)\Delta_{hp}} \left[\frac{2\Gamma}{\Gamma_0} - \frac{(\Delta_h - \Gamma)(\Gamma_0 + \Delta_h)}{\Delta_{hp}\Delta_h} \right].$$
(21)

For the input probe pulse at z=0 we take a pulse with a Gaussian envelope $E_{\text{prob}}(0,t)=E_0\exp[-(\Delta_{\text{in}}t/2)^2]$. The Fourier transform of this pulse is $E_{\text{prob}}(0,\nu) = (E_0 2\sqrt{\pi}/\Delta_{\text{in}})\exp(-\nu^2/\Delta_{\text{in}}^2)$ and the half width of its spectrum is Δ_{in} . If we take into account only three terms of the expansion of $A(\nu)$, i.e., $A(\nu) \approx \alpha(a_0 - ia_1\nu - a_2\nu^2)/2$, the integral (17) for the Gaussian pulse can be easily calculated,

$$E_{pA}(z,t) = \frac{\Delta_{\text{out}}}{\Delta_{\text{in}}} E_0 \exp\left[-a_0 \frac{\alpha z}{2} - \frac{1}{4} \Delta_{\text{out}}^2 (t - t_d)^2\right], \quad (22)$$

where *t* is actually the local time t-z/c and the subscript *A* designates that this is an approximate solution. Here $\Delta_{out} = \Delta_p / \sqrt{1 - a_2 \Delta_{in}^2 (\alpha z/2)}$ is the half width of the spectrum of the output probe pulse (a_2 is assumed to be negative in our case), $t_d = a_1 \alpha z/2$ is the delay time of the probe pulse.

The first term of the $A(\nu)$ expansion, with the coefficient a_0 , describes the absorption of a zero frequency Fourier com-

where

ponent of the probe pulse envelope (ν =0), i.e., the absorption exactly at the center of the hole. This Fourier component is the pulse area, which is the time integral of the pulse, i.e.,

$$\theta_A(z) = \frac{2d_{eg}}{\hbar} \int_{-\infty}^{+\infty} E_{\text{prob}}(z,t) dt = \theta(0) e^{-a_0 \alpha z/2}.$$
 (23)

Here $\theta(0) = 4\sqrt{\pi}d_{eg}E_0/\Delta_{in}\hbar$ is the pulse area at the input (z = 0).

We consider the case of strong inhomogeneous broadening of the absorption line: $\Gamma_{inh} \gg \Gamma$. We also assume that $\Gamma \gg \gamma$ or, in terms of the relaxation times, that $T_2 \ll T_1$. The pump satisfies the condition of strong saturation: $\Omega_{pump}^2 T_1 T_2 \gg 1$, which leads to the approximation $\Delta_{hp} \approx \Delta_h \approx p = \Omega_{pump} \sqrt{T_1/T_2}$. In this case the coefficient a_0 is approximated as $a_0 \approx 1/(\Omega_{pump} \sqrt{T_1T_2})$. In Eq. (22) the first term in the square brackets of the exponent, $-a_0 \alpha z/2$, reflects the absorption at the center of the hole, which follows Beer's law if $a_0 = 1$. In case of a deep hole, the absorption coefficient is reduced by a factor $\Omega_{pump} \sqrt{T_1T_2}$.

The second term of the $A(\nu)$ expansion, with coefficient a_1 , describes the pulse delay t_d . This delay originates from the steep dispersion at the center of the hole, which results in slow group velocity of the pulse,

$$V_g = c/[1 + ca_1(\alpha/2)].$$
(24)

If the pump is not applied (p=0), the slope of the dispersion at the line center is negative $\sim -1/\Gamma_0$. If the pump burns a deep hole in the absorption spectrum, the dispersion slope at the hole center is positive and steep $\sim 1/\Omega_{\text{pump}}\sqrt{T_1/T_2}$ [see Fig. 1(b)]. We assume that $\Gamma_0 \ge \Omega_{\text{pump}}\sqrt{T_1/T_2} \ge \Gamma$. In this case the delay time of the pulse is approximated as $t_d \approx \alpha z/2\Delta_h$. The group velocity of the pulse reduces as V_g/c $\simeq 2\Delta_h/c\alpha$, i.e., as the ratio of the hole width $(2\Delta_h)$ and the product of the speed of light in vacuum (c) and Beer's constant (α) .

The third term of the expansion $A(\nu)$ with coefficient a_2 produces probe pulse broadening in time or its spectrum narrowing with distance. The spectral half width of the output probe pulse can be expressed as follows

$$\Delta_{\rm out} = \frac{\Delta_{\rm in}}{\sqrt{1 + (\Delta_{\rm in}/\Delta_{\rm eff})^2}},\tag{25}$$

where

$$\Delta_{\rm eff} = \sqrt{\frac{2(\Gamma_{\rm inh} + \Delta_h)\Delta_{hp}}{\alpha z \left[\frac{(\Delta_h - \Gamma)(\Gamma_0 + \Delta_h)}{\Delta_{hp}\Delta_h} - \frac{2\Gamma}{\Gamma_0}\right]}}$$
(26)

is the effective half width of the hole at distance z for a thick sample. If the saturation is strong $(\Omega_{pump}^2 T_1 T_2 \ge 1)$, the effective half width is $\Delta_{eff} \approx \Delta_h \sqrt{2/\alpha_z}$, where Δ_h can be considered as the hole half width for a thin sample. Thus for a thick sample the effective half width of the hole, Δ_{eff} , narrows as $\sim 1/\sqrt{z}$. It follows from Eqs. (25) and (26) that the spectral width of the output pulse, Δ_{out} , tends to Δ_{eff} with the increase of z.

The energy of the output pulse or its time integrated intensity decreases with distance as



FIG. 3. (a) Comparison of the analytical solution $E_{pA}(l,t)$, for the output probe pulse, Eq. (22), shown by the thick line, with the field of the probe that would travel the same distance in free space, shown by the thin solid line. Here for $E_{\text{prob}}(0,t)$ we take z=0 and the distance l, which the probe would travel in free space, is taken only in the local time t, i.e., t-l/c. (b) Comparison of the analytical approximation $E_{pA}(l,t)$ (solid line) with the numerical calculation of the envelope of the output probe pulse, $E_{\text{prob}}(l,t)$ (dots), given by Eq. (17) where $A(\nu)$ is not approximated but taken from Eq. (16). All plots are normalized to the amplitude E_0 . Time is in units $\Delta_{\text{in}}t$, where t is the local time t-l/c. The parameters are $\Gamma_{\text{inh}}=9.5$ GHz, $\Gamma=353.7$ kHz, $\gamma=38$ Hz, $\Omega_{\text{pump}}=500$ kHz, $\Delta_{\text{in}}=30$ MHz, and $\alpha l/2=15$.

$$U_{A}(z) = \int_{-\infty}^{+\infty} |E_{pA}(z,t)|^{2} dt = U(0) \frac{\Delta_{\text{out}}}{\Delta_{\text{in}}} e^{-a_{0}\alpha z}, \qquad (27)$$

where $U(0) = \sqrt{2\pi |E_0|^2 / \Delta_{\text{in}}}$ is the input pulse energy. Since $\Delta_{\text{out}}/\Delta_{\text{in}} \sim 1/\sqrt{1 + \alpha z \Delta_{\text{in}}^2 / 2\Delta_h^2}$, the attenuation of the probe pulse violates Beer's law in the case of hole burning.

Figure 3(a) shows a comparison of the output probe pulse, Eq. (22) (thick line), with a pulse that would propagate the same distance in free space (thin line). The delay of the pulse is obvious. The pulse amplitude decreases because of the residual absorption at the center of the hole and due to the pulse broadening in time. Figure 3(b) shows a comparison of our analytical solution Eq. (22) (solid line) with the numerical calculation of the integral (17) where $A(\nu)$ is not approximated but given by Eq. (16). The parameters are Γ_{inh} =9.5 GHz, T_2 =0.45 μ sec, T_1 =4.2 msec, and Ω_{pump} =500 kHz. They are typical, for example, for the R_1 line in ruby (Czochralski-grown crystal of concentration 0.05 wt. % Cr_2O_3), which is excited by a laser beam of moderate power at liquid helium temperature and 3-kG magnetic field; see, for example, Ref. [28]. The half width of the hole is Δ_h =48.3 MHz. The spectral half width of the input pulse is taken Δ_{in} =30 MHz. The delay time of the pulse is t_d =48.5 ns for the sample with α =12 cm⁻¹ and l=2.5 cm. During a time interval of t_d =48.5 ns, the pulse would propagate 14.5 m in free space. For ruby crystal with 0.05% atomic weight doping by Cr^{3+} , we estimate the group velocity of 4.75×10^5 m/sec , which is $\sim 10^3$ times smaller than speed of light in vacuum.

Figure 3(b) shows a small deviation of the analytical approximation, Eq. (22), from the integral (17). The fit can be improved if one takes into account the fourth term of the expansion (18) with the coefficient

$$a_{3} = -\frac{1}{\Gamma_{0}^{3}} + \frac{p^{2}}{\Delta_{h}\Delta_{hp}^{4}} + \frac{p^{2}\left[\Gamma\Gamma_{0}^{3} + \Delta_{h}\Delta_{hp}(\Gamma_{0}^{2} + \Gamma_{0}\Delta_{hp} + \Delta_{hp}^{2})\right]}{\Delta_{h}\Delta_{hp}^{4}\Gamma_{0}^{3}(\Delta_{h} + \Gamma_{inh})}.$$
(28)

Then $A(\nu) \approx \alpha (a_0 - ia_1\nu - a_2\nu^2 + ia_3\nu^3)/2$ and the output pulse is presented as a convolution of Eq. (22) and the Airy function, Ai(x), i.e.,

$$E_{pA1}(z,t) = \frac{\Delta_{\text{out}}}{\Delta_{\text{in}}} E_0 e^{-a_0 \alpha z/2} \int_{-\infty}^{+\infty} \Delta_{\text{dist}} \operatorname{Ai}(\Delta_{\text{dist}} \tau)$$
$$e^{-(\Delta_{\text{out}}^2/4)(t - t_{dA} - \tau)^2} d\tau, \qquad (29)$$

where $\Delta_{dist} = (3\alpha a_3 z/2)^{-1/3}$ is a pulse distortion parameter. This expression originates from the integral representation of the Airy function [29],

$$|\Delta_{\text{dist}}|\text{Ai}(\pm|\Delta_{\text{dist}}|\tau) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \exp\left[-i\left(\nu\tau \pm \frac{\nu^3}{3|\Delta_{\text{dist}}^3|}\right)\right] d\nu,$$
(30)

and the convolution theorem

$$\frac{1}{2\pi} \int_{-\infty}^{+\infty} F_1(\nu) F_2(\nu) e^{-i\nu t} d\nu = \int_{-\infty}^{+\infty} f_1(t-\tau) f_2(\tau) d\tau, \quad (31)$$

where $F_k(\nu)$ is the Fourier transform of $f_k(t)$, k=1, 2.

If $\Gamma_0 \gg \Omega_{\text{pump}} \sqrt{T_1/T_2} \gg \Gamma$, the pulse distortion parameter is $\Delta_{\text{dist}} \approx \Delta_h / \sqrt[3]{3\alpha z/2}$. It can be shown [30] that the pulse distortion is small if $\Delta_{\text{out}} < \Delta_{\text{dist}}$. This condition is well satisfied for all *z*'s if the spectral width of the input pulse is smaller than the spectral width of the transparency window ($\Delta_{\text{in}} < \Delta_h$). Thus for any optically thick absorber a Gaussian pulse of spectral width smaller than Δ_h keeps its Gaussian shape with distance.

IV. SIMILARITY BETWEEN THE PERSISTENT HOLE BURNING AND EIT

One can show that there is a strong similarity between persistent hole burning and population trapping in the dark state. In persistent hole burning, an ion or molecule is trapped in the state t with the help of the pump inducing a transition of the ground-state particle to the excited state eand then via a fast nonradiative decay or via a molecule transformation the particle ends up in the long-lived state t[see Fig. 4(a)]. In this way a medium becomes transparent for the probe field.

EIT can be explained if one introduces a new basis of dark d and bright b states. Figure 4(b) shows the excitation



FIG. 4. (a) Excitation scheme for the persistent hole burning. g, e, and t are the ground, excited, and trapping states. The pump field is shown by the bold arrow. Γ_{fast} is a fast nonradiative process shown by the bold arrow with the white strip. For a dye molecule in a polymer, this can be a photoinduced transformation of the molecule. γ is the decay rate of the trapping state. The decay process is shown by the dashed arrow. The lifetime T_1 of the trapping state tends to infinity for the molecule. For Cr³⁺ this is a radiative lifetime, which is long because of the small dipole matrix element for this transition. The open circle shows a "hole" in the population of the ground state. The filled circle in state t shows population trapping. (b) Excitation scheme for EIT. g, e, and m are the ground, excited, and metastable states. The thin arrow shows the excitation by the probe. The bold arrow shows the interaction with the coupling field. The filled circle shows the population of the ground state. (c) The same excitation scheme as in (b) but in the basis of dark d and bright b states. The continuous arrow shows the effective coupling of the bright and excited states. The strength of the effective coupling is defined as a particular combination of the Rabi frequencies of the probe and coupling fields. The dashed arrow shows the radiative decay from e to d. The open circle shows a hole in the population of the bright state. The filled circle shows population trapping in the dark state.

scheme of a three-level atom interacting with a probe and a coupling field. An excited state e is a short-lived state, decaying to the ground state g. A metastable state m has a long lifetime. Therefore the coherence g-m, induced by a twoquantum excitation of the ground-state atom by the probe and coupling fields, has also long lifetime. One can find an eigenstate of the Hamiltonian of the three-level atom interacting with the probe and coupling fields. This is a dark state (or uncoupled state), which is an asymmetric superposition of ground g and metastable m states [9]. One can also introduce another superposition of the same states, e and m, but orthogonal to the dark state. This is a bright state (or coupled state) [9]. The dark state is uncoupled with the excited state because of destructive interference of the transition paths, while the bright state is coupled with the excited state because of constructive interference. Initially the atom is in the ground state g and hence both dark and bright states are populated. Continuous excitation by a cw probe and coupling field depopulates the bright state; see Fig. 4(c). With the help of radiative decay of the state e, this population is collected and eventually trapped in the dark state. Therefore the medium becomes transparent for the probe field. Some recent discussion of the atom evolution in the basis of dark and bright states can be found in Ref. [31].

For EIT and hole burning we have a dip in the absorption line. In case of EIT this dip appears for a single atom since a single atom (or each atom of an atomic ensemble) is trapped in the dark state. This trapping is very sensitive to the Raman resonance condition, which is $\omega_{pr} - \omega_{coup} = \omega_{mg}$. Here ω_{pr} is the frequency of the probe field, ω_{coup} is the frequency of the coupling field, and ω_{mg} is the resonant frequency for the transition m-g. Actually the latter is represented by the Raman induced coherence ρ_{mg} . Its decay rate, γ_{mg} , specifies the depth of the hole for a given value of the Rabi frequency for the coupling field, Ω_{coup} , i.e., the absorption is reduced by a factor of $\Omega_{coup}^2/(\Gamma \gamma_{mg})$, where Γ is the decay rate of the coherence g-e induced by the probe. For $\Omega_{coup} < \Gamma$, the halfwidth of the EIT hole is Ω_{coup}^2/Γ and the dispersion slope is also close to Ω_{coup}^2/Γ .

For hole burning each particle is saturated differently depending on its detuning from the frequency of the pump field. The absorption of the resonant particles is reduced by a factor $\Omega_{\text{pump}}^2/(\Gamma \gamma)$. For particles slightly detuned from resonance, the absorption decreases less. The response of an ensemble of inhomogeneously broadened particles exhibits a hole in the absorption spectrum for the probe. The half width of this hole is $\Omega_{\text{pump}}\sqrt{\Gamma/\gamma}$ and the slope of the dispersion is defined by this half width. Comparing EIT and hole burning, we find that in both cases the depth of the hole is strongly dependent on the decay rate of the trapping (or dark) state. However, in case of EIT the hole width is narrower than for HB if $\Omega_{coup} < \Gamma$ and we take the same Rabi frequencies for the coupling field and the pump ($\Omega_{coup} = \Omega_{pump}$), the same decay rate for the coherence induced by the probe (Γ) and the same values for the decay rate of the trapping states $(\gamma = \gamma_{mg}).$

V. CONCLUSION

In this paper, we have shown how persistent hole burning can be used to produce slow light. A transverse pump field is applied in advance to saturate resonant ions or molecules in a solid. If the relaxation time of the population difference is long, the probe field can be applied after some delay time. This method has several aspects in common with electromagnetically induced transparency. The pump field creates a transparency window for the probe. The dispersion of the resonant particles has a steep slope and its derivative has a sign opposite to the sign of the usual resonant dispersion at the line center. Therefore the group velocity of the pulse appreciably slows down. In the EIT scheme, one can make a transparency window as large as the Rabi frequency for a strong-coupling field Ω_c . Usually, the width of the EIT window is much smaller and roughly equal to Ω_c^2/Γ if $\Omega_c \ll \Gamma$, where Γ is the decay rate of the polarization induced by the probe field. Persistent hole burning due to the saturation broadening phenomenon allows the creation of a much broader transparency window $\approx \Omega_{pump} \sqrt{\Gamma/\gamma} \gg \Omega_{pump}$, where γ is the decay rate of the population difference $(\gamma \ll \Gamma)$. Therefore hole burning can produce slow light for much shorter pulses whose spectral width must, however, be limited by the hole width. One further advantage of persistent spectral hole burning over "regular" EIT is that the shape of the spectral hole can be arbitrary, as long as the features in the spectrum are not narrower than the homogeneous linewidth. For example, one can make a spectral hole which has steep edges, but a very low flat absorption in the middle. The group delay produced by such a box-shaped hole will still be large, but the residual absorption will be less than for a regular shape of the hole.

ACKNOWLEDGMENTS

This work was supported by the Fonds voor Wetenschappelijk Onderzoek Vlaanderen, FNRS and the IAP program of the Belgian government. R.N.S. also acknowledges support from CRDF CGP (Grant No. RP1-2560-KA-03) and ISTC (Grant No. 2121).

- L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Nature (London) **397**, 594 (1999).
- [2] M. M. Kash, V. A. Sautenkov, A. S. Zibrov, L. Hollberg, G. R. Welch, M. D. Lukin, Y. Rostovtsev, E. S. Fry, and M. O. Scully, Phys. Rev. Lett. 82, 5229 (1999).
- [3] D. Budker, D. F. Kimball, S. M. Rochester, and V. V. Yashchuk, Phys. Rev. Lett. 83, 1767 (1999).
- [4] C. Liu, Z. Dutton, C. H. Behroozi, and L. V. Hau, Nature (London) 409, 490 (2001).
- [5] D. F. Phillips, A. Fleischhauer, A. Mair, R. L. Walsworth, and M. D. Lukin, Phys. Rev. Lett. 86, 783 (2001).
- [6] O. Kocharovskaya and Ya. I. Khanin, Zh. Eksp. Teor. Fiz. 90, 1610 (1986) [Sov. Phys. JETP 63, 945 (1986)].
- [7] K.-J. Boller, A. Imamoğlu, and S. E. Harris, Phys. Rev. Lett. 66, 2593 (1991).
- [8] J. E. Field, K. H. Hahn, and S. E. Harris, Phys. Rev. Lett. 67, 3062 (1991).
- [9] E. Arimondo, Coherent Population Trapping in Laser Spectroscopy, edited by E. Wolf, Progress in Optics No. 35

(Elsevier Science, Amsterdam, 1996), pp. 257–354, and references therein.

- [10] S. E. Harris, Phys. Today 50 (7), 36 (1997).
- [11] E. Arimondo and G. Orriols, Lett. Nuovo Cimento Soc. Ital.
 Fis. 17, 333 (1976); G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento Soc. Ital. Fis., B 36B, 5 (1976); G. Alzetta, L. Moi, and G. Orriols, *ibid.* 52B, 209 (1979); R. M. Whitley and C. R. Stroud, Jr., Phys. Rev. A 14, 1498 (1976).
- [12] G. Orriols, Nuovo Cimento Soc. Ital. Fis., B 53B, 1 (1979).
- [13] D. A. Braje, V. Balić, S. Goda, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. 93, 183601 (2004).
- [14] M. S. Bigelow, N. N. Lepeshkin, and R. W. Boyd, Phys. Rev. Lett. **90**, 113903 (2003); Science **301**, 200 (2003).
- [15] G. S. Agarwal and T. N. Dey, Phys. Rev. A 68, 063816 (2003).
- [16] S. E. Schwartz and T. Y. Tan, Appl. Phys. Lett. 10, 4 (1967).
- [17] M. Sargent III, Phys. Rep. 43, 223 (1978).
- [18] L. W. Hillman, R. W. Boyd, J. Krasinsky, and C. R. Stroud, Jr., Opt. Commun. 45, 416 (1983).
- [19] L. Allen and J. H. Eberly, Optical Resonance and Two-Level

Atoms (Wiley, New York, 1975).

- [20] A. Renn, U. P. Wild, and A. Rebane, J. Phys. Chem. A 106, 3045 (2002).
- [21] H. Schwoerer, D. Erni, and A. Rebane, J. Opt. Soc. Am. B 12, 1083 (1995).
- [22] R. Jaaniso and H. Bill, Europhys. Lett. 16, 569 (1991).
- [23] R. Bauer, A. Osvet, I. Sildos, and U. Bogner, J. Lumin. 56, 57 (1993).
- [24] A. Javan, O. Kocharovskaya, H. Lee, and M. O. Scully, Phys. Rev. A 66, 013805 (2002).
- [25] M. Tschanz, A. Rebane, and U. P. Wild, Opt. Eng. (Bellingham) 34, 1936 (1995).

- [26] M. Tschanz, A. Rebane, D. Reiss, and U. P. Wild, Mol. Cryst. Liq. Cryst. Sci. Technol., Sect. A 283, 43 (1996).
- [27] N. Bloembergen, in *Quantum Optics*, p. 355 (Academic Press, London and New York, 1970).
- [28] A. Szabo and J. Heber, Phys. Rev. A 29, 3452 (1984).
- [29] *Handbook of Mathematical Functions*, edited by M. Abramovitz and I. A. Stegun (Dover, New York, 1965).
- [30] R. N. Shakhmuratov and J. Odeurs, Phys. Rev. A 71, 013819 (2005).
- [31] R. N. Shakhmuratov, J. Odeurs, R. Coussement, and A. Szabo, Laser Phys. 14, 39 (2004).