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Slowing and storing light processes without a trapping dark state in a double two-level system. Theoretical study

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We present a detailed theoretical study of a method for slowing light based on the oscillations of Zeeman coherences in a double two-level system [Hashmi, F.A.; Bouchene, M.A. *Phys. Rev. A* **2008**, *77*, 051803(R)]. This method does not require the presence of any trapping state. We focus on the properties and the limitations of such a method and compare it to previous ones. We also investigate the possibility of obtaining fast light and to store the light with such a method.

Keywords: slow light; wave mixing; light–atom interaction; polariton; stored light; fast light

1. Introduction

The interest in light propagation in resonant atomic media has been renewed these last years with the realization of spectacular experiments where slow and fast light can be produced [1]. These studies have opened up a new domain with an enormous potential for applications in optics. The key idea to slow light is to create a narrow transparency window in the absorption spectrum of a pulse as it propagates in a medium. Because of the Kramers–Krönig relations, an abrupt variation of the refraction index is in line with this transparency window leading to a strong reduction of the group velocity for a light pulse that propagates through the medium. The early demonstrations of slow light propagation [2,3] were achieved via electromagnetically induced transparency (EIT) [4]. The standard scheme for this effect is a three-level Λ system excited by a sequence of two fields – a weak probe and a much stronger control – each driving a separate transition of the system. When the two-photon resonance condition is realized, all the atoms can be coherently trapped in the dark state rendering the system transparent to both fields. This is coherent population trapping (CPT) that plays an important role in many phenomena in optics [5]. Slow light has been also investigated in double Λ systems that were intensively studied in the past in the context of resonant nonlinear optical phenomena [6]. These systems can exhibit EIT phenomena for equal strength pulses having special phase relation between them [7]. For these matching conditions, a dark state arises

in the system, leading to CPT and hence to slow [8] and stored light [9,10]. Another method to slow light makes use of coherent population oscillations (CPO) and can be implemented in a two-level system [11]. A pair of fields with different frequencies (pump and probe, respectively) induces a temporal grating in the population of the excited states. The self-diffraction of the pump beam by the grating reinforces the radiation at the probe frequency thus compensating for some absorption of the probe by the atomic medium. A spectral hole with a width fixed by the population relaxation rate is created inside the probe spectral absorption profile making slow light possible. A recent important development of slow light studies is the possibility to make optical quantum memories by stopping light [12]. Many alternative techniques have also been proposed to obtain optically controllable delays for telecommunication purposes [13] but light velocities achieved here are far from those obtained with EIT.

In a previous paper [14], we have presented the principle of an alternative method that can lead to very small group velocity based on Zeeman coherence oscillations (ZCO). It can be used for a linearly polarized probe pulse as it propagates through a double two-level system driven by an orthogonally polarized control field. The space/time modulation of the total polarization induces a grating in the Zeeman coherences which diffracts the control field into the probe field compensating for the absorption of the latter. In our situation, the control field is much

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stronger than the probe and an angle is introduced between the beams. The matching conditions are not satisfied and thus no dark state exists in the system, ruling out the possibility of transparency or slow light through CPT as was the case in previous studies in a four-level system [8–10]. Our method is in strong analogy with CPO in a non-collinear geometry [15] but in our case, it is the coherence that is oscillating and not the population. However, the transparency window exhibits characteristics similar to EIT method. This may suggest that EIT and CPO that are considered as fundamentally different methods are closer than expected. Indeed, all these methods (EIT, CPO and ZCO) can be described as different manifestations of the wave mixing induced by the two exciting fields. The transparency results from a balance between the absorption of the probe and diffraction of the control field off some grating. Indeed, the EIT method can also be described in this way for a degenerate Λ system and results from a scattering process in which the control beam is diffracted by the ground level Zeeman coherences [7,10]. This feature is generally overlooked in the literature because of the more powerful description based on the dark state, but it can be the key idea for implementation of slow light in more complex systems where no dark state exists.

In the present paper, we present a complete study of this phenomenon (ZCO) emphasizing the role of

limiting effects, and we also demonstrate the possibility to store light. This paper is organized as follows. In Section 2, we present the theoretical model giving the equations of evolution of the atomic quantities and the equation of propagation for the probe field. In Section 3, we determine the optical susceptibility for the probe emphasizing the ‘Zeeman coherence oscillation’ phenomenon. In Section 4, we discuss the process of slow and fast light using this effect and in Section 5 we discuss the limitation of this method emphasizing the role of the Doppler effect, the ground Zeeman coherence relaxations and non-linear effects. In Section 6, we demonstrate the propagation of a polariton and we show the possibility to store light. Finally, in Section 7, we conclude by summarizing the properties of this method and comparing it with other methods.

2. Theoretical model, equations of evolution

Consider on a single atom the $F=1/2 \rightarrow F=1/2$ transition with energy $\hbar\omega_0$ excited by two orthogonally polarized fields that propagate in different directions (Figure 1). The electric fields are $\mathbf{E}_\pi(\mathbf{r}, t) = e_\pi \varepsilon_\pi(\mathbf{r}) \exp[-i(\omega_\pi t - \mathbf{k}_\pi \mathbf{r})] + c.c.$ and $\mathbf{E}_\sigma(\mathbf{r}, t) = e_\sigma \varepsilon_\sigma(\mathbf{r}) \exp[-i(\omega_\sigma t - \mathbf{k}_\sigma \mathbf{r})] + c.c.$ We choose the axis of quantification such that \mathbf{E}_π (the control field) and \mathbf{E}_σ (the probe)

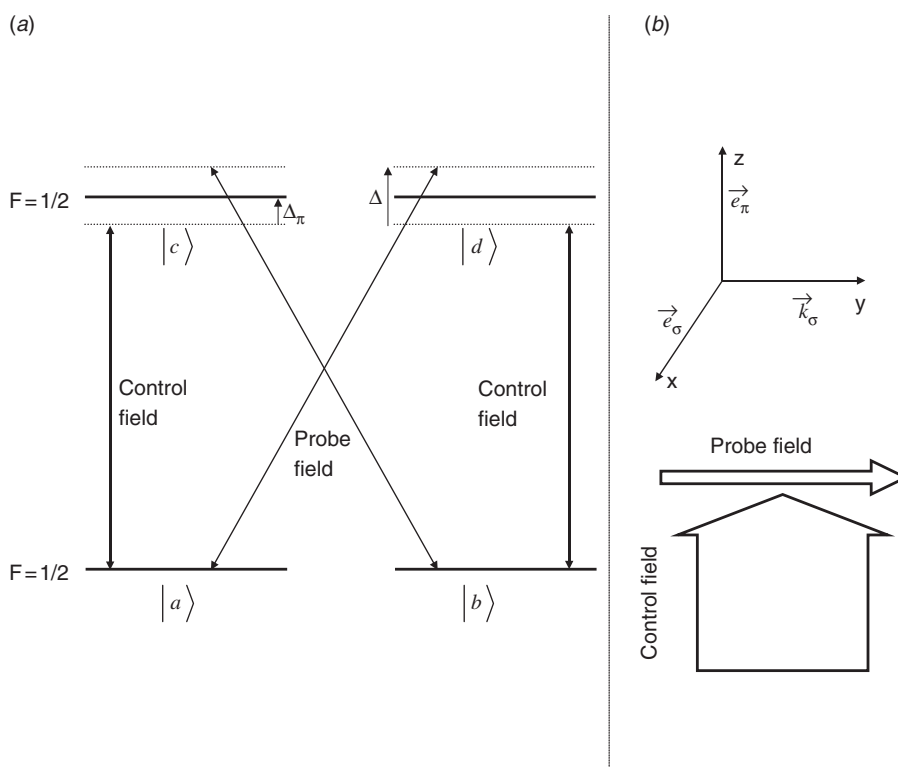


Figure 1. (a) The double two-level system and (b) the field configurations. See text for definition of parameters.

have, respectively, π and σ polarizations. The system is equivalent to a double two-level system in which the π polarized field excites the states with identical m_F and the σ polarized field couples cross-transitions. Throughout the paper, the control beam is considered to be much stronger than the probe.

The evolution of the density matrix ρ in the interaction picture (phase oscillation between ground and excited states in $\exp(-i\omega_\pi t)$) is given by:

$$i\hbar\partial_t\rho = [H, \rho] + \bar{\Gamma}_\rho. \quad (1)$$

With the Hamiltonian H given within the rotating wave approximation in the basis set $\{|a\rangle, |b\rangle, |c\rangle, |d\rangle\}$ by:

$$H = -\hbar \begin{pmatrix} 0 & 0 & \Omega_\pi^* & \Omega_\sigma^* \exp(i\Phi) \\ 0 & 0 & \Omega_\sigma^* \exp(i\Phi) & -\Omega_\pi^* \\ \Omega_\pi & \Omega_\sigma \exp(-i\Phi) & -\Delta_\pi & 0 \\ \Omega_\sigma \exp(-i\Phi) & -\Omega_\pi & 0 & -\Delta_\pi \end{pmatrix} \quad (2)$$

and the relaxation matrix $\bar{\Gamma}_\rho$ given by

$$\bar{\Gamma}_\rho = \begin{pmatrix} \Gamma_\pi\rho_{cc} + \Gamma_\sigma\rho_{dd} & -\Gamma_{zg}\rho_{ab} & -\Gamma_d\rho_{ac} & -\Gamma_d\rho_{ad} \\ -\Gamma_{zg}\rho_{ba} & \Gamma_\pi\rho_{dd} + \Gamma_\sigma\rho_{cc} & -\Gamma_d\rho_{bc} & -\Gamma_d\rho_{bd} \\ -\Gamma_d\rho_{ca} & -\Gamma_d\rho_{cb} & -\Gamma_\sigma\rho_{cc} & -\Gamma_{ze}\rho_{cd} \\ -\Gamma_d\rho_{da} & -\Gamma_d\rho_{db} & -\Gamma_{ze}\rho_{dc} & -\Gamma_\sigma\rho_{dd} \end{pmatrix}; \quad (3)$$

here $\Omega_\pi = d\varepsilon_\pi/\hbar$ and $\Omega_\sigma = d\varepsilon_\sigma/\hbar$ are the Rabi frequency associated with the π and σ polarized fields respectively, $\Delta_\pi = \omega_0 - \omega_\pi$ is the detuning of the control field and $\Phi = \Delta t - \delta\mathbf{k}\mathbf{r}$ is the dephasing between the two pulses ($\Delta = \omega_\sigma - \omega_\pi$ and $\delta\mathbf{k} = \mathbf{k}_\sigma - \mathbf{k}_\pi$). The excited state populations ρ_{cc} and ρ_{dd} relax with the rate $\pi = \Gamma/3$ into the ground state with identical m_F and $\Gamma_\sigma = 2\Gamma/3$ into the ground state with different m_F . The coherences ρ_{ac} , ρ_{ad} , ρ_{bc} and ρ_{bd} relax with the rate Γ_d , the ground and excited Zeeman coherences relax with the rate Γ_{ze} and Γ_{zg} , respectively. In the absence of non-radiative homogeneous dephasing processes, $(\Gamma_{zg}, \Gamma_{ze}, \Gamma_d)$ reduces to $(0, \Gamma, \Gamma/2)$.

The modification of the probe field is determined by the behavior of the coherence $\rho_\sigma = \rho_{cb} + \rho_{da}$ that radiates the σ polarized light. The evolution of ρ leads to the following equations where we have defined the ground and excited states populations $n_g = \rho_{aa} + \rho_{bb}$ and $n_e = \rho_{cc} + \rho_{dd} = 1 - n_g$, the coherence $\rho_\pi = \rho_{ca} - \rho_{db}$ responsible for the π polarized radiated field, and the imaginary parts of the ground and excited Zeeman coherences as $\rho_{zg} = 2i\text{Im}(\rho_{ab})$, $\rho_{ze} = 2i\text{Im}(\rho_{cd})$:

$$i\partial_t\rho_\sigma = -\Omega_\pi(\rho_{zg} + \rho_{ze}) + \Omega_\sigma \exp(-i\Phi)(\mathbf{r}, t)(n_e - n_g) + \bar{\Delta}_\pi^* \rho_\sigma, \quad (4)$$

$$i\partial_t\rho_\pi = \Omega_\pi(n_e - n_g) + \Omega_\sigma \exp[-i\Phi(\mathbf{r}, t)](\rho_{zg} + \rho_{ze}) + \bar{\Delta}_\pi^* \rho_\pi, \quad (5)$$

$$i\partial_t n_g = 2i \text{Im}(\Omega_\pi \rho_\pi^* + \Omega_\sigma \exp[-i\Phi(\mathbf{r}, t)]\rho_\sigma^*) + in_e\Gamma, \quad (6)$$

$$i\partial_t\rho_{zg} = 2 \text{Re}(\Omega_\sigma \exp[-i\Phi(\mathbf{r}, t)]\rho_\pi^* - \Omega_\pi \rho_\sigma^*) - i\Gamma_{zg}\rho_{zg}, \quad (7)$$

$$i\partial_t\rho_{ze} = i\partial_t\rho_{zg} - i(\Gamma_{ze}\rho_{ze} - \Gamma_{zg}\rho_{zg}), \quad (8)$$

with $\bar{\Delta}_\pi = \Delta_\pi + i\Gamma_d$. The relaxation rate for ground Zeeman coherence Γ_{zg} will be considered zero throughout this paper except in Section 5.2 when discussing the limitation of our method. In relations (4), only the imaginary parts of the Zeeman coherences ρ_{zg} and ρ_{ze} contribute to ρ_σ . This is a consequence of the symmetry of our system and can be understood in the following manner. The diffraction of the control field Ω_π from ρ_{ab} creates the coherence ρ_{cb} and (because of the opposite sign of dipole moment on $|b\rangle \leftrightarrow |d\rangle$ transition) the diffraction of $-\Omega_\pi$ from $(\rho_{ab})^*$ is involved in creation of ρ_{da} . This makes the contribution of the real part of ρ_{ab} to the total coherence $\rho_\sigma = \rho_{cb} + \rho_{da}$ go to zero. The same argument holds for the contribution of $\text{Re}(\rho_{cd})$ to ρ_σ and for the contribution of these Zeeman coherences to ρ_π . The probe field is modified during propagation according to the following equation with $y = \mathbf{k}_\sigma \mathbf{r} / \|\mathbf{k}_\sigma\|$:

$$\frac{\partial\Omega_\sigma}{\partial y} + \frac{1}{c} \frac{\partial\Omega_\sigma}{\partial t} = i\alpha_0\Gamma_d\tilde{\rho}_\sigma \quad (9)$$

with $\alpha_0 = Nd^2\omega_0/2c\hbar\varepsilon_0\Gamma_d$ the field absorption coefficient at the line-center, N is the atomic density and $\tilde{\rho}_\sigma$ is the part of the coherence ρ_σ that radiates in the direction of the σ field. The expression for $\tilde{\rho}_\sigma$ will be detailed below. We assume throughout the paper that the control field is such $|\Omega_\pi| \gg |\Omega_\sigma|$ and is not modified by the sigma pulse. During propagation, the control field experiences absorption and dispersion that are also assumed to be negligible. This imposes, however, a maximum propagation length. Next we determine the spectral response of the system with respect to the σ field. The spectral response of the system is characterized by the effective susceptibility $\chi = [(2\alpha_0/k)\Gamma_d]\tilde{\rho}_\sigma/\Omega_\sigma$ with $k = \omega_0/c$ and depends on the spatial configuration of the fields. When no control field is present, the resolution of (4) in the stationary regime, for a weak intensity of the probe ($|\Omega_\sigma| \ll (\Gamma\Gamma_d)^{1/2}$) gives $\chi = \chi_{\text{lin}}$ with $\chi_{\text{lin}} = (2\alpha_0\Gamma_d/k)/(-i\Gamma_d + \Delta_\sigma)$ the linear susceptibility ($\Delta_\sigma = \omega_0 - \omega_\sigma$).

3. Coherent Zeeman oscillations

When the beams propagate in different directions, the Hamiltonian and the polarization of the total field are modulated in space and time allowing the Floquet expansion of the density matrix as $\rho = \sum_{n=-\infty}^{n=+\infty} \rho^{(n)} \exp(-in\Phi)$. The part of the coherence that radiates in the direction of the σ probe field is thus $\tilde{\rho}_\sigma = \rho_\sigma^{(1)}$. An important remark has to be pointed out at this level. The part $\rho_\sigma^{(-1)}$ of the coherence radiates a field in the $2\mathbf{k}_\pi - \mathbf{k}_\sigma$ direction if phase matching is satisfied. Because the medium is optically thick, this radiation may modify substantially the behavior of $\rho_\sigma^{(1)}$. This requires that an angle $\theta > (\lambda/L)^{1/2}$ (L the length of the medium, λ the wavelength) be introduced between two pulses. When the probe is weak ($|\Omega_\sigma| \ll |\Omega_\pi|$, $(\Gamma\Gamma_d)^{1/2}$), the stationary solution for $\rho_\sigma^{(1)}$ can be derived from (4)–(9) as:

$$\rho_\sigma^{(1)} = (\bar{\Delta}_\pi^* - \Delta)^{-1} \left[\Omega_\sigma (n_g^{(0)} - n_e^{(0)}) + \Omega_\pi (\rho_{zg}^{(1)} + \rho_{ze}^{(1)}) \right]. \quad (10)$$

Here,

$$n_g^{(0)} - n_e^{(0)} = \frac{\Gamma_d^2 + \Delta_\pi^2}{4|\Omega_\pi|^2 \Gamma^{-1} \Gamma_d + \Gamma_d^2 + \Delta_\pi^2}$$

is the static part of the difference of population and the first term in Equation (10) shows the absorption of the probe by this population. This absorption is compensated by the second term which represents the scattering of the control field off the Zeeman spatial-temporal grating. This second term also accounts for the cross-Kerr effect [16]. In the stationary regime we have:

$$\rho_{zg}^{(1)} + \rho_{ze}^{(1)} = -\frac{\Omega_\sigma}{\Omega_\pi} (n_g^{(0)} - n_e^{(0)}) \left[1 - \Delta (\bar{\Delta}_\pi^* - \Delta) W(\Delta, \bar{\Delta}_\pi) \right] \quad (11)$$

with

$$W = \frac{[|\Omega_\pi|^2 M \bar{\Delta}_\pi^{-1} + (\Delta + \bar{\Delta}_\pi)]}{2|\Omega_\pi|^2 M (\Delta + i\Gamma_d) + \Delta (\bar{\Delta}_\pi^* - \Delta) (\Delta + \bar{\Delta}_\pi)} \quad (12)$$

and $M = (2\Delta + i\Gamma_{ze}) / (\Delta + i\Gamma_{ze})$. The modulation depth of the Zeeman grating is of course weak since $\rho_{zg}^{(1)} + \rho_{ze}^{(1)} \propto \Omega_\sigma / \Omega_\pi$ but the diffraction of the control field gives a contribution to the ρ_σ coherence that has the same order of magnitude as the absorption part. Perfect compensation between the two effects is obtained when the two fields have the same frequency ($\Delta = 0$). This creates a transparency window in the spectral absorption profile of the probe field. It is to be noted that this perfect transparency is realized without any dark state in the system. Indeed, we can identify two Λ systems $\{|a\rangle, |b\rangle, |c\rangle\}$ and $\{|a\rangle, |b\rangle, |d\rangle\}$ in our

double two-level system and define dark states $|D\rangle = -\Omega_\sigma \exp(-i\Phi)|a\rangle + \Omega_\pi|b\rangle$ and $|D'\rangle = \Omega_\pi|a\rangle + \Omega_\sigma \exp(-i\Phi)|b\rangle$, respectively. When the matching condition $[\Omega_\sigma \exp(-i\Phi)]^2 = -\Omega_\pi^2$ is satisfied, the two Λ systems share a common dark state making CPT possible. This situation has been identified and studied in four-level systems by many groups [8–10]. In our situation, the control field is much stronger than the probe $|\Omega_\pi| \gg |\Omega_\sigma|$, Φ is spatially dependent and so no dark state exists.

The effective susceptibility of the system for the probe field is $\chi = [(2\alpha_0/k)\Gamma_d]\rho_\sigma^{(1)}/\Omega_\sigma$. Using (6)–(8), we get:

$$\chi(\Delta, \Delta_\pi) = \Delta \frac{2\alpha_0\Gamma_d}{k} (n_g^{(0)} - n_e^{(0)}) W(\Delta, \bar{\Delta}_\pi). \quad (13)$$

Figure 2 shows the behaviour of the real and imaginary part of the susceptibility χ as a function of the detuning Δ between the fields for different strengths of the control field with $|\Omega_\pi| < \Gamma_d$. Two important features appear. First, the absorption profile $\text{Im}(\chi)$ exhibits a dip whose minimum is 0 for $\Delta = 0$, as has been discussed before. Secondly, the width of the dip decreases as the control intensity is decreased. Correspondingly, the dispersion profile $\text{Re}(\chi)$ exhibits a very abrupt variation around $\Delta = 0$. A simplified expression for the width of the dip can be derived in the limit $|\Omega_\pi|, \Delta_\pi, \Delta \ll \Gamma_d, \Gamma_{ze}$ as follows:

$$\text{Im}\chi(\Delta, \Delta_\pi) \simeq \frac{\alpha_0}{k} \frac{|\Omega_\pi|^{-4} \Gamma_d^2 \Delta^2 / 2}{1 + (|\Omega_\pi|^{-4} \Gamma_d^2 \Delta^2 / 4)}. \quad (14)$$

This is an inversed Lorentzian profile with a width $\sim 4|\Omega_\pi|^2 \Gamma_d^{-1}$. The relative width of the induced transparency window compared with the absorption profile

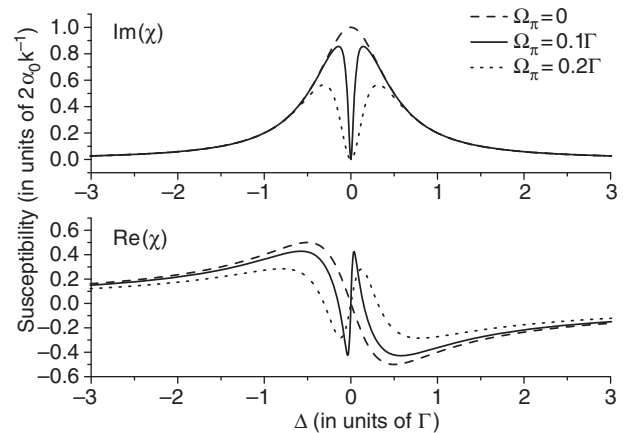


Figure 2. Profiles of the real and imaginary parts of the susceptibility for the probe beam for different values of the control Rabi frequency $|\Omega_\pi| < \Gamma$. The other parameters are $\Delta_\pi = 0$, $\Gamma_d = 0.5\Gamma$, $\Gamma_{ze} = \Gamma$ and $\Gamma_{zg} = 0$.

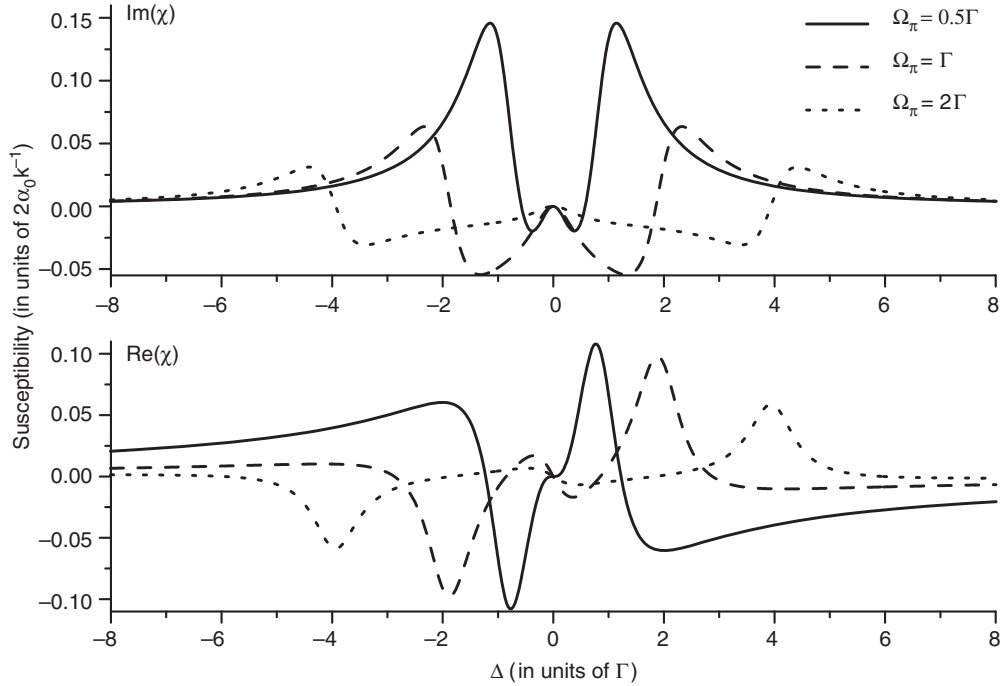


Figure 3. Profiles of the real and imaginary parts of the susceptibility for the probe beam when increasing the Rabi frequency of the control field ($|\Omega_\pi| \geq \Gamma$). The other parameters are $\Delta_\pi = 0$, $\Gamma_d = 0.5\Gamma$, $\Gamma_{ze} = \Gamma$ and $\Gamma_{zg} = 0$. Complex structures appear.

(linewidth $2\Gamma_d$) is then $2|\Omega_\pi/\Gamma_d|^2$ and can be reduced significantly by decreasing the control field intensity.

When $|\Omega_\pi| \geq \Gamma_d$, expression (14) for $\text{Im}(\chi)$ is no longer valid and the susceptibility exhibits different behaviour. These changes are shown in Figure 3. The absorption always vanishes for $\Delta = 0$, but the absorption profile breaks into several peaks with the possibility to obtain gain energy for the probe: the diffraction of the control field over compensates for the absorption of the probe. This is reminiscent of CPO behaviour in a two-level system when saturation occurs [17].

4. Slow and fast light

The opening of a narrow transparency window in the absorption profiles leads to the possibility of slow propagation of the probe provided its spectrum is contained within the window. For that, the pulse duration T of the probe must satisfy certain conditions. Indeed, $4|\Omega_\pi|^2\Gamma_d^{-1}$ is the spectral width over which $\text{Im}\chi$ is negligible but any small absorption of the probe outside this window is greatly enhanced during propagation. This absorption (for probe intensity) is given by $\exp(-kL\text{Im}\chi)$ and hence the transparency window is reduced to $4|\Omega_\pi|^2\Gamma_d^{-1}/(\alpha_0L)^{1/2}$. Thus, we need to have $T \gg (\alpha_0L)^{1/2}|\Omega_\pi|^{-2}\Gamma_d/4$ in order to

observe slow light effects without pulse distortion. The group velocity for the probe pulse is

$$v_g = c \left(1 + \frac{\text{Re}(\chi)}{2} + \frac{\omega_0}{2} \frac{\partial \text{Re}(\chi)}{\partial \Delta} \right)_{\Delta=0}^{-1}.$$

Using relation (13), we find that:

$$v_g = c \left(1 + \frac{c\alpha_0\Gamma_d}{2|\Omega_\pi|^2} g(\Omega_\pi, \Delta_\pi) \right)^{-1} \quad (15)$$

with

$$g(\Omega_\pi, \bar{\Delta}_\pi) = \frac{|\bar{\Delta}_\pi|^2 - |\Omega_\pi|^2}{4|\Omega_\pi|^2\Gamma_d\Gamma^{-1} + |\bar{\Delta}_\pi|^2}.$$

For weak intensities of the control field such as $|\Omega_\pi| \ll \Gamma_d$, we have $g \simeq 1$ and thus $v_g = c[1 + (c\alpha_0\Gamma_d/|\Omega_\pi|^2)]^{-1}$. Very small group velocities can be reached when decreasing the control field intensity and/or increasing the atomic density of the medium. Using $|\Omega_\pi| = (d/\hbar)(I_\pi/2c\epsilon_0)^{1/2}$, where I_π is the control field intensity, we obtain $v_g \simeq 2I_\pi/N\hbar\omega_0$. For $I_\pi = 1 \text{ mW cm}^{-2}$, $\lambda = 671 \text{ nm}$ and $N = 10^{12} \text{ at cm}^{-3}$, we get $v_g \simeq 60 \text{ m s}^{-1}$. The group delay $\tau = L(v_g^{-1} - c^{-1})$ can be approximated as $(\alpha_0L/2|\Omega_\pi|^2)\Gamma_d$. An important parameter for slow light methods is the figure of merit τ/T . Because $T \gg (\alpha_0L)^{1/2}|\Omega_\pi|^{-2}\Gamma_d/4$ ensures that the whole probe spectrum is located within the transparency window, the optical depth α_0L has to be large

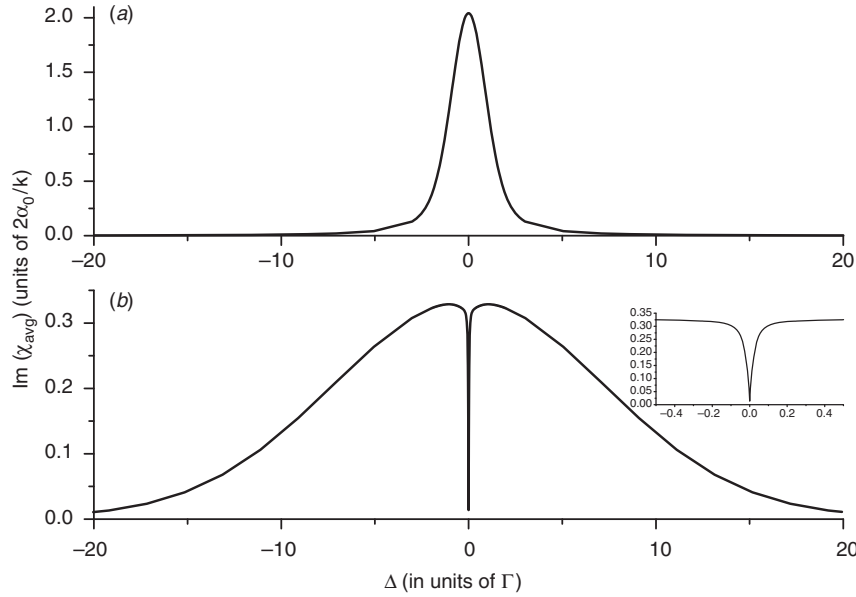


Figure 4. Influence of Doppler effect on the absorption profile ($\text{Im}(\chi_{\text{avg}})$) for two different situations (a) $\delta_d = \Gamma$ and $\theta = \pi/2$ (distributed pump intensity) (b) $\delta_d = 10\Gamma$ $\theta = 0.1$ mrad (nearly parallel beams). The other parameters are $\Omega_\pi = 0.1\Gamma$, $\Delta_\pi = 0$, $\Gamma_d = 0.5\Gamma$, $\Gamma_{ze} = \Gamma$ and $\Gamma_{zg} = 0$.

enough to obtain a good figure of merit. The maximum value for the optical depth is limited by the pump absorption and for a configuration where the pump and the probe beams have only a small angle between them, it can not exceed a few units. This difficulty can be overcome if a distributed configuration is used for which the control crosses the medium and the probe beam transversally [15]. The attenuation of the control field is then negligible and L is limited only by the transverse dimension of the pump beam.

For $|\Omega_\pi| \gg \Gamma_d$, $g \simeq -\Gamma_d^{-1}\Gamma/4$ and $v_g \simeq c[1 - (c\alpha_0\Gamma/8|\Omega_\pi|^2)]^{-1}$. The system turns into a fast light medium. The maximum value for v_g is obtained when $|\Omega_\pi| \simeq \Gamma_d = \Gamma/2$ and is $v_g \simeq c[1 - (c\alpha_0/2\Gamma)]^{-1}$. Even negative group index and backward propagation are possible if $c\alpha_0 > 2\Gamma$. For instance, if $\Gamma \simeq 37$ MHz (transition $^2S_{1/2}F=1/2 \rightarrow ^2P_{1/2}F=1/2$ of ^6Li) and $N = 10^{11}$ at cm^{-3} we get $v_g \simeq -2 \times 10^3 \text{ m s}^{-1}$.

5. Limiting effects

Many effects can modify the ideal behavior exposed in the previous paragraph. In addition to the absorption of the pump that can severely limit the propagation length if a distributed pump configuration is not used, three effects have to be studied: the Doppler effect that has to be taken into account for a vapour gas, the decoherence of the ground state Zeeman coherences due to for instance depolarizing collisions ($\Gamma_{zg} \neq 0$) and non-linear effects when the ratio $|\Omega_\sigma/\Omega_\pi|$

is not negligible. This latter situation is important with regard to storing light.

5.1. Doppler effect

In a vapour gas, the Doppler effect has to be taken into account. The averaged $\chi_{\text{avg}}(\Delta, \Delta_\pi)$ is obtained by summing the contributions of various velocities that follow a Maxwellian distribution $f(\mathbf{v}) = \prod_{i=x,y,z} f_i(v_i)$; $f_i(v_i) = (1/u\pi^{1/2}) \exp[-(v_i/u)^2]$, $u = (2k_B T/m)^{1/2}$ (k_B is the Boltzmann constant and T is the temperature):

$$\chi_{\text{avg}}(\Delta, \Delta_\pi) = \iiint \chi(\Delta + (\mathbf{k}_\sigma - \mathbf{k}_\pi)\mathbf{v}, \Delta_\pi - \mathbf{k}_\pi\mathbf{v}) f(\mathbf{v}) d^3\mathbf{v}. \quad (16)$$

The effect of Doppler averaging for large angle θ between the beams is dramatic and spoils the transparency. Indeed, the susceptibility is averaged over a frequency width of the order of $(2k \sin \theta/2)u$ around the detuning Δ . At room temperature, ku lies in the GHz region. In the distributed pump configuration $\theta = \pi/2$ and the susceptibility is thus averaged over a spectral domain that exceeds largely the transparency window width $4|\Omega_\pi|^2\Gamma_d^{-1}$ that lies in the MHz or less. Moreover, for $\theta = \pi/2$, the condition to obtain transparency is $ku \ll 4|\Omega_\pi|^2\Gamma_d^{-1}/2^{1/2}$ and from (15), we have $u \ll (\pi/2^{1/2})v_g(\lambda\alpha_0)$. In the distributed pump configuration, the maximum value of α_0^{-1} is given by the transverse dimension of the atomic beam. For $\lambda = 0.6 \mu\text{m}$, $\alpha_0^{-1} \simeq 3 \text{ mm}$ for the desired value of

group velocity $v_g = 300 \text{ m s}^{-1}$, we need $u \ll 0.1 \text{ m s}^{-1}$ corresponding to a temperature in the micro Kelvin range (for Li atom). Ultracold atoms have to be used.

The severity of the Doppler effect is exhibited in Figure 4(a) where the Doppler width $\delta_D = ku$ is equal only to Γ . The spectral hole in the imaginary part of the integrated susceptibility χ_{avg} is clearly washed out. We show now that the use of a smaller angle can reduce the Doppler effect. Indeed, for an angle θ between the beams such as $\theta \ll 4|\Omega_\pi|^2 \Gamma_d^{-1}/ku$, the susceptibility has to be averaged only around Δ_π and reduces to $\chi_{\text{avg}}(\Delta, \Delta_\pi) \simeq \int_{-\infty}^{+\infty} \chi(\Delta, \delta) f(\delta/k) d\delta/k$ (and $\Delta_\pi \ll \delta_D$). Figure 4(b) exhibits the sensitivity of the optical response to the Doppler effect in this case. The spectral hole induced by the presence of the control field still remains for $\delta_D = 10\Gamma$ indicating a robust behavior with respect to inhomogeneous effects. This result can be explained from the dependence of the susceptibility (13) with the detuning δ instead of Δ_π . As seen above, $\chi = 0$ for $\Delta = 0$ whatever δ is. The spectral hole is still present and only its width can be modified as a consequence. We can distinguish between atoms for which the detuning lies within the homogeneous absorption profile ($\delta < \Gamma_d$) and those outside this profile ($\delta > \Gamma_d$). The first kind of atoms will have identical contributions since $\bar{\delta} \simeq i\Gamma_d$ and the hole generated by these atoms exhibits an unchanged width $\sim 4|\Omega_\pi|^2 \Gamma_d^{-1}$. For atoms with detuning outside the homogeneous profile, we have $\bar{\delta} \simeq \delta$ and the susceptibility $\chi(\Delta, \delta)$ turns out to be an odd function of δ . The contribution of atoms outside the homogeneous profile is thus cancelled pair by pair. Only atoms with a resonance frequency located within the homogeneous profile contribute efficiently to the integrated susceptibility χ_{avg} . Note, however, that the situation of a small angle such as the Doppler width can be neglected correspond to a propagation length for the control field that is comparable to that for L of the probe. The maximum available optical depth $\alpha_0 L$ is only a few units making the figure of merit very small in this case.

5.2. Ground state Zeeman decoherence

When the relaxation of the ground Zeeman coherence is involved ($\Gamma_{zg} \neq 0$), the expression of susceptibility changes as:

$$\chi(\Delta, \Delta_\pi) = \frac{2\alpha_0 \Gamma_d}{k} \left(n_g^{(0)} - n_e^{(0)} \right) W'(\Delta, \bar{\Delta}_\pi) \quad (17)$$

with

$$W' = \frac{\left[\Delta |\Omega_\pi|^2 M' \bar{\Delta}_\pi^{-1} + (\Delta + i\Gamma_{zg})(\Delta + \bar{\Delta}_\pi) \right]}{2|\Omega_\pi|^2 M' (\Delta + i\Gamma_d) + (\Delta + i\Gamma_{zg})(\bar{\Delta}_\pi^* - \Delta)(\Delta + \bar{\Delta}_\pi)}$$

and

$$M' = \frac{2\Delta + i(\Gamma_{ze} + \Gamma_{zg})}{\Delta + i\Gamma_{ze}}.$$

It can be seen that susceptibility no longer vanishes at $\Delta = 0$. For $\Delta_\pi = 0$ and $|\Omega_\pi| \ll \Gamma_d$, the expression simplifies to:

$$\chi(0, 0) = \frac{2i\alpha_0}{k} \frac{1}{1 + 2|\Omega_\pi|^2 \Gamma_d^{-1} (\Gamma_{zg}^{-1} + \Gamma_{ze}^{-1})}. \quad (18)$$

Figure 5 shows the evolution of the optical response when the decoherence effects for the ground Zeeman coherence increases. For the chosen parameters, we have $2|\Omega_\pi|^2 \Gamma_d^{-1} = 10^{-4} \Gamma$. The contribution of Γ_{ze} is then negligible and the relevant parameter to characterize the deepness of the transparency windows is the ratio $\Gamma_{zg}/2|\Omega_\pi|^2 \Gamma_d^{-1}$.

For slow light and taking into account the propagation, the decoherence can be neglected only if $kL \text{Im}\chi \ll 1$. This is the case if $\alpha_0 L \Gamma_{zg} \ll 2|\Omega_\pi|^2 \Gamma_d^{-1}$: the ground state Zeeman coherence relaxation rate magnified by the optical depth should be smaller than the width of the spectral hole created in the susceptibility profile. This effect is illustrated in Figure 6 that shows the absorption of the transmitted probe pulse that result from the spoiling of the transparency when the ratio $\alpha_0 L \Gamma_{zg}/(2|\Omega_\pi|^2 \Gamma_d^{-1})$ is increased.

5.3. Non-linear effects

The result (9) is valid only at the first order with respect to the probe amplitude. Higher order terms may spoil the transparency for $\Delta \simeq 0$. The coherence ρ_σ can be calculated exactly for $\Delta = 0$ in the stationary regime. We found ($\Gamma_{zg} = 0$):

$$\rho_\sigma = \frac{[\bar{\Delta}_\pi (\Omega_\pi^2 + \Omega_\sigma^2 \exp(-2i\Phi))] \Omega_\sigma^* \exp(i\Phi)}{4\Gamma_d \Gamma^{-1} |\Omega_\pi^2 + \Omega_\sigma^2 \exp(-2i\Phi)|^2 + [|\bar{\Delta}_\pi|^2 (|\Omega_\pi|^2 + |\Omega_\sigma|^2)]}. \quad (19)$$

The σ coherence $\rho_\sigma^{(1)}$ that radiates in the direction of the probe is given by $\rho_\sigma^{(1)} = (1/2\pi) \int_{-\infty}^{+\infty} \rho_\sigma \times \exp(i\Phi) d\Phi$ and can be evaluated at the lowest non-vanishing order. From relations (1)–(3), we found:

$$\rho_\sigma^{(1)} \simeq \Omega_\sigma \left(\bar{\Delta}_\pi^* \right)^{-1} \frac{(\Omega_\sigma/\Omega_\pi)^2}{\left(1 + 4\Gamma_d \Gamma^{-1} |\bar{\Delta}_\pi|^{-2} \Omega_\pi^2 \right)^2}. \quad (20)$$

$\rho_\sigma^{(1)}$ is small if the control field intensity is much stronger than that of the probe ($|\Omega_\pi| \gg |\Omega_\sigma|$). However, this latter condition doesn't automatically ensure that the effects associated with this contribution can be

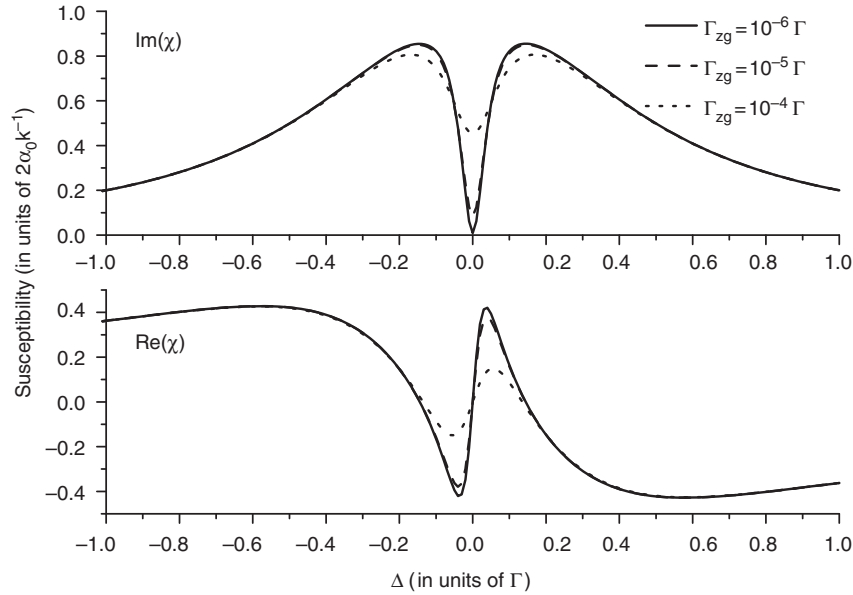


Figure 5. Influence of ground state Zeeman coherence on the (a) absorption and (b) dispersion profiles of the probe for the parameters $\Omega_\pi=0.1\Gamma$, $\Delta_\pi=0$, $\Gamma_d=0.5\Gamma$, $\Gamma_{ze}=\Gamma$ and different values of Γ_{zg} . We have $2|\Omega_\pi|^2\Gamma_d^{-1}=10^{-4}\Gamma$ and the ratio $\Gamma_{zg}/2|\Omega_\pi|^2\Gamma_d^{-1}$ is 0.01, 0.1 and 1 for $\Gamma_{zg}=10^{-6}\Gamma$, $10^{-5}\Gamma$ and $10^{-4}\Gamma$, respectively.

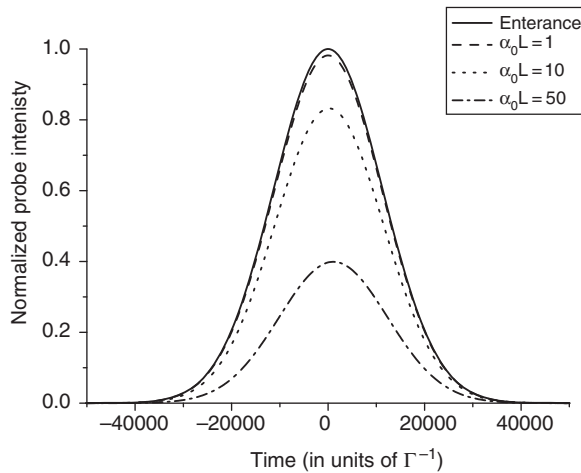


Figure 6. Influence of ground state Zeeman coherence on the profile of the transmitted probe pulse. The parameters are $\Omega_\pi=0.1\Gamma$, $\Delta_\pi=0$, $\Gamma_d=0.5\Gamma$, $\Gamma_{ze}=\Gamma$ and thus $\Gamma_{zg}/(2|\Omega_\pi|^2\Gamma_d^{-1})=0.01$ (situation of curve in solid line in Figure 5). The values of the optical depth are $\alpha_0L=1$, 10 and 50 corresponding to a value of 0.01, 0.1 and 0.5 for the relevant parameter $\alpha_0L\Gamma_{zg}/(2|\Omega_\pi|^2\Gamma_d^{-1})$, respectively. The absorption of the probe is clearly enhanced during propagation.

neglected. Indeed, the probe field amplitude obeys during propagation Equation (9). Any small contribution can be amplified during propagation and can spoil transparency. This requires an additional condition $X=\alpha_0L(\Omega_\sigma/\Omega_\pi)^2\ll 1$ (for $|\Delta_\pi|<\Gamma_d$).

6. Polariton and stored light

An important application of slow light is the possibility to store the light by switching off the control field making the group velocity of the probe go to zero [12]. The optical probe pulse is coherently absorbed and its properties are then transferred to an atomic coherence where they survive as long as decoherence can be neglected. By turning on the control field again, the atomic coherence properties can be transferred again to light and the probe pulse can be retrieved from the system. We show next that such a process can also be realized in our system and discuss the limitations.

6.1. Polariton

An elegant formalism to describe these features is to introduce a dark state polariton that couples the propagating beam and the atomic coherence [12]. Choosing in our method a distributed pump configuration, the control field remains unaffected by the propagation and we can define the dark polariton as (for simplicity $\Delta_\pi=0$):

$$\psi(y, t) = \cos \theta \Omega_\sigma(y, t) - \sin \theta \kappa^{1/2} \rho_{zg}^{(1)}(y, t) \quad (21)$$

with $\kappa=(c\alpha_0/2)\Gamma_d$ and $\tan \theta=\kappa^{1/2}/\Omega_\pi$. If we consider the slow light situation ($|\Omega_\pi|\ll\Gamma_d$), neglect the ground Zeeman decoherence ($\Gamma_{zg}\simeq 0$), the non-linear effects ($X\ll 1$) and if transparency is ensured for the entire probe spectrum, relation (7) still holds whereas

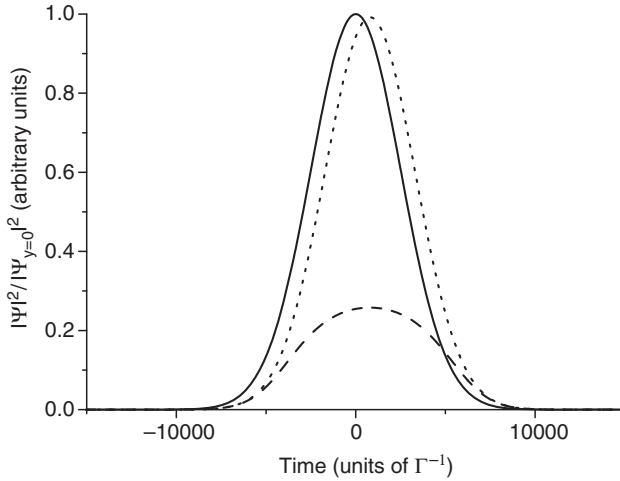


Figure 7. Non-linear effects and polariton dynamics. Temporal profile of the polariton intensity for different values of X . We have $\Omega_\sigma(t, y=0) = \Omega_{\sigma 0} \exp[-(t/T)^2]$, $\Omega_\pi = 0.2\Gamma$ (constant in time), $\Delta_\pi = 0$, $\Gamma_d = 0.5\Gamma$, $\Gamma_{ze} = \Gamma$, $\Gamma_{zg} = 0$, $T = 5000\Gamma^{-1}$ and $\alpha_0 L = 200$. Solid line: polariton at the entrance ($y=0$). Dotted line: $\Omega_{\sigma 0} = 0.001\Gamma$ and thus $X \simeq 0.006$, transparency is ensured and the polariton is delayed in time. Dashed line: $\Omega_{\sigma 0} = 0.02\Gamma$ and thus $X \simeq 2.5$. The polariton is damped and distorted.

relations (4)–(5) give $\rho_{ze}^{(1)}(y, t) \simeq 0$. The instantaneous Zeeman coherence involved in (17) can be approximated as $\rho_{zg}^{(1)}(y, t) \simeq -(\Omega_\sigma(y, t)/\Omega_\pi)$. Using $\rho_\sigma^{(1)}(t) = \int \rho_\sigma^{(1)}(\Delta) \exp(-i\Delta t) dt$ and from Equations (6)–(7) we get $\rho_\sigma^{(1)}(t) \simeq (i/4\Omega_\pi^2)\dot{\Omega}_\sigma(t)$. The polariton propagates within the material along with the probe according to the shape preserved equation:

$$\partial\psi/\partial t + c \cos^2 \theta \partial\psi/\partial y = 0. \quad (22)$$

The important difference with dark polaritons in Λ system [12] is that here only a part of the (ground) Zeeman coherence ($\rho_{zg}^{(1)}$) constitutes the bound part of the polariton. When damping effects or higher order terms given by relation (16) become important, the polariton is modified during propagation, experiencing both damping and shape distortion. Figure 7 shows the non-linear effects on the polariton dynamics. This may happen if we decrease the intensity of the control field or if the probe pulse is not enough weak. For the chosen parameters, the dominant contribution for the polariton is the bound material part. When X is small, perfect transparency is obtained for the probe and the polariton is only temporally delayed, whereas for large X , the polariton is damped.

6.2. Stored light

We discuss in this subsection, the evolution of the photonic and bound part of the polariton when the

control field is switched off and switched on after a time delay. Stored light is associated with the conversion of the photonic part into the bound part that no longer propagates inside the medium (stopped). The process can be described as follows. When the probe pulse enters the medium, the field amplitude is preserved due to continuity at the interface and the incoming travelling wave of the form $\Omega_\sigma(t - y/c)$ in free space turns into the form $\Omega_\sigma(t - y/v_g)$ as it propagates through the medium. Both the spatial profile and the carried energy $\int |\Omega_\sigma(t - y/v_g)|^2 dy$ undergo an important compression of the order of v_g/c . The pulse duration and the spectral width of the pulse remain unaffected by the slowing process only if the probe spectrum is contained within the transparency window ($T \gg (\alpha_0 L)^{1/2} \Gamma_d / |\Omega_\pi|^2$) to avoid absorption and distortion of the pulse profile. Most of the energy has been extracted from the probe pulse and the residual energy is negligible. This energy is transferred to the control field whereas the bound part of the polariton does not carry energy and propagates along with the probe pulse.

Once the probe pulse is entirely contained within the medium $\Delta l = v_g T \ll L$, the storing process can be set off by switching off the control field. When the control field is abruptly switched off, the bound part of the polariton is stopped whereas the residual (negligible) energy is absorbed [18]. If the control field is adiabatically reduced, the spectral narrowing of both the transparency window and the pulse spectrum when decreasing the control field intensity allows the complete transformation of the light pulse into the Zeeman coherence [12]. The information content of the probe is stored into the bound part that survives as long as permitted by decoherence effects. When the control field is turned on again, the light pulse reemerges in the medium. This effect is shown in Figure 8 where we represent the profile of the probe intensity for a different distance of propagation. The light pulse disappears when the control field is reduced and reappears when it is applied again. Because our medium is a slow light medium only if $|\Omega_\pi| < \Gamma_d$, the non-linear effects and the condition for the probe spectrum to be contained inside the transparency window impose some restrictions on the choice of the probe pulse. The first condition ($X \ll 1$) gives a maximal value for the Rabi frequency of the probe pulse of the order of $|\Omega_\sigma| \sim \Gamma_d (\alpha_0 L)^{1/2}$. The latter condition imposes a minimum value for the duration of the probe pulse to be stored of the order of $T \sim (\alpha_0 L)^{1/2} / \Gamma_d$. The small damping observed in Figure 8 where $X = 0.005$ is due to residual absorption whereas in Figure 9, the non-linear effects ($X = 0.5$ at $\alpha_0 v = 5000$) are dominant for the

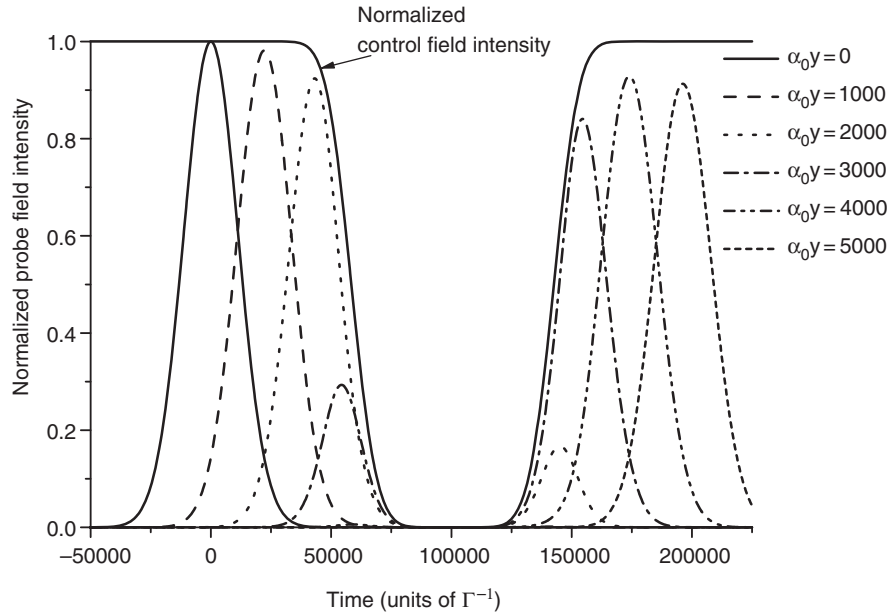


Figure 8. Stored light. Deceleration and acceleration $X \ll 1$. Here, $\Omega_{\sigma}(t, y=0) = \Omega_{\sigma 0} \exp[-(t/T)^2]$ and $\Omega_{\pi} = \Omega_{\pi 0}(1 - \exp\{-(t - 10^5 \Gamma^{-1})/4 \times 10^4 \Gamma^{-1}\}^2)$. The time here is the reduced time ($\equiv (t - y/c)\Gamma$). The value of the parameters are $\Gamma_d = 0.5\Gamma$, $T = 22500\Gamma^{-1}$, $\Omega_{\sigma 0} = 0.0001\Gamma$, $\Omega_{\pi 0} = 0.1\Gamma$ and thus $X = 0.005$ for $\alpha_0 y = 5000$.

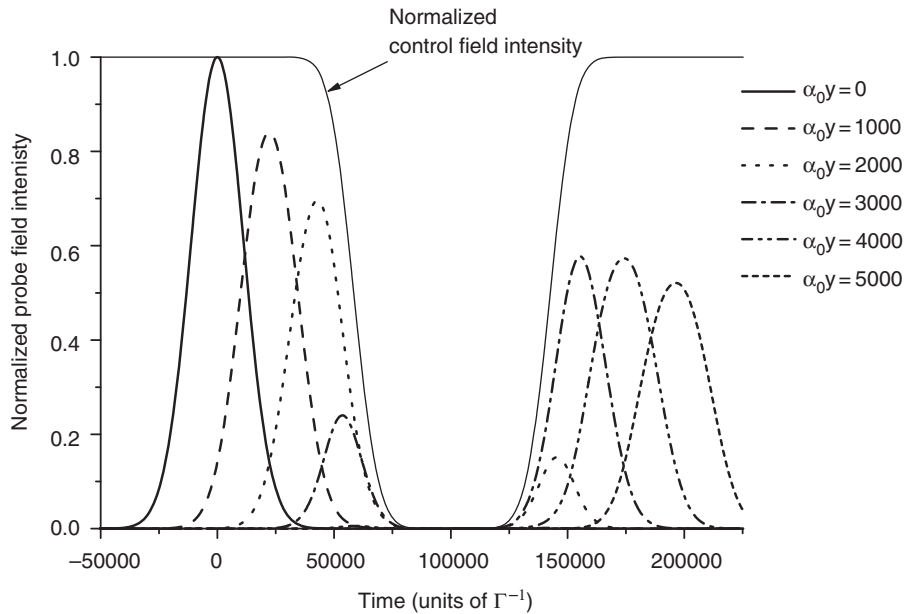


Figure 9. The same than Figure 8 but $\Omega_{\sigma 0} = 0.001\Gamma$ and thus $X = 0.5$ for $\alpha_0 y = 5000$.

damping of the field. However, the storing process still occurs.

7. Summary, experimental implementation and comparison with others methods

We have presented in this paper a detailed theoretical study of slowing and storing light in a double two-level

system. No trapping dark state exists and the underlying phenomenon for the cancellation of the absorption is a non-linear effect: the diffraction of the control beam from the Zeeman coherence grating induced by the polarization grating of the total field. We summarize also in Table 1, the main characteristics (a) and the main limitations (b) of our method based on CZO. To store the light, we also need $v_g \ll c(|\Omega_{\pi}|^2 \ll c\alpha_0\Gamma_d)$ and $\alpha_0 L(\Omega_{\sigma}/\Omega_{\pi})^2 \ll 1$ for a probe pulse with a time duration

Table 1. Summary of (a) the main characteristics and (b) the main limitations of our method based on coherent Zeeman oscillations (CZO). In part (b), we give the conditions such as the cited effects can be neglected. See text for the signification of the parameters.

Part (a)			
Slow/fast light	Group velocity (slow light), $ \Delta_\pi < \Gamma_d$	Transparency window (slow light)	Figure of merit (slow light)
$ \Omega_\pi < \tilde{A}_\pi / \Omega_\pi > \tilde{A}_\pi $	$v_g = c(1 + c\alpha_0\Gamma_d \Omega_\pi ^2)^{-1}$	$4 \Omega_\pi ^2\Gamma_d^{-1}/(\alpha_0L)^{1/2}$	$\alpha_0L\Gamma_d/2 \Omega_\pi ^2T$
Part (b)			
Conjugate wave contribution	Doppler effect $ \Delta_\pi < ku$	Ground Zeeman coherence $ \Delta_\pi < \Gamma_d$	Non-linear effects $ \Delta_\pi < \Gamma_d$
$\theta \gg (\lambda/L)^{1/2}$	$\theta(ku) \ll 4 \Omega_\pi ^2\Gamma_d^{-1}$	$\Gamma_{zg} \ll 2 \Omega_\pi ^2\Gamma_d^{-1}/\alpha_0L$	$\alpha_0L(\Omega_\sigma/\Omega_\pi)^2 \ll 1$

$T \gg (\alpha_0L)^{1/2}/4|\Omega_\pi|^2\Gamma_d^{-1}$ and $T < L/v_g$. The first condition ensures almost total conversion between the photonic and the bound part of the polariton, whereas the second and third conditions ensure small damping of the polariton due to the non-linear effects and absorption respectively. Finally, the last condition ensures that the entire polariton can be contained inside the medium.

Experimental demonstration of these phenomena can be obtained in principle in any $F=1/2 \rightarrow F=1/2$ transition excited by a sequence of two polarized fields. The laser fields can be obtained by sending a single beam into an acousto-optics to generate the control and the probe fields. The detuning between the fields can thus be controlled. The characteristics of performances one can obtain depend strongly on the nature of the sample. The severity of the Doppler effect and the necessity to have a non-vanishing angle to remove the conjugate wave makes the method inefficient in hot gases. If a gas of ultracold atoms is used, the Doppler effect can be neglected and very small group velocities can be obtained as discussed previously in the text. For instance, a possible candidate for the demonstration of this effect is the transition $^2S_{1/2}F=1/2 \rightarrow ^2P_{1/2}F=1/2$ of ^6Li at 671 nm. Samples with densities as high as 10^{12}cm^{-3} can be obtained with temperatures around the μK [19]. Very small group velocities are thus possible in such systems as suggested by the numerical example in Section 4. To avoid the conjugate wave contribution, it is necessary to choose a geometrical configuration with an angle $\theta > (\lambda/L)^{1/2} \simeq 45\text{ mrad}$ ($L \simeq 300\mu\text{m}$). The small dimension of the sample allows one also to neglect the non-linear effects ($X \ll 1$) for a probe pulse less intense than the control.

We give now a small comparison of our method with already existing methods of slowing light. Qualitatively, our method resembles CPO in which the control field is diffracted off an oscillating

population but in our case it is the coherence that is oscillating. However, quantitatively this method is closer to the EIT schemes that realize the dark state in the system and make the system transparent. Indeed, the transparency window width in both cases is proportional to the control field intensity and can be arbitrary reduced [2,3]. In contrast, in the CPO technique, the width of the spectral hole is at a minimum given by the population relaxation rate and the dip can not be decreased further [11,17]. Note that EIT and ZCO share a common limitation: the ground Zeeman coherence relaxation limits the depth of the spectral hole.

On the other hand, by increasing the control intensity, the absorption profile in EIT schemes have a broader transparency window and Stark shift effects dominate for $|\Omega_\pi| > \Gamma_d$. The corresponding shape is always related to a normal dispersion profile and fast light can not be obtained. In ZCO, when the intensity of the control pulse is increased, the absorption dip splits into multiple fragments making superluminal and background propagation possible.

It was discussed that for good figure of merit the optical depth α_0L has to be important. In EIT, the optical depth can be increased theoretically as much as required since transparency is achieved for both the control and the probe. But in CPO and ZCO the control field is always subjected to absorption that limits optical depths and distributed pump configurations have to be used to overcome this hurdle.

In a distributed pump configuration, Doppler broadening is a serious limitation for the ZCO scheme. Cold atoms have to be used here. An alternative is to make a small angle between the control and the probe beams in order that the optical response is immune to the Doppler effect. But, in turn this enhances the pump absorption limiting the efficiency of the slowing process. The EIT schemes are shown to be robust against the Doppler effect [3], whereas

in CPO the Doppler effect can be overcome only by counter-propagating fields [20].

Finally, ZCO and EIT can be used as storing light process whereas this is not the case in the CPO method (with transfer from light to population and not the coherence). The population damping represents then a serious limitation. In ZCO, the higher non-linear effects that spoil the transparency limits the validity of our method to probe pulses with intensities such as $X = \alpha_0 L (\Omega_\sigma / \Omega_\pi)^2 \ll 1$. This limitation does not exist in EIT since the coherence rigorously vanishes for the two-photon resonance condition.

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