Light slowdown in the vicinity of cross-over resonances

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Pulse propagation is considered in an inhomogeneously broadened medium of three-level atoms in a V-configuration, dressed by a counter-propagating pump pulse. A significant signal slowdown is demonstrated in this of the three frequency windows of a reduced absorption and a steep normal dispersion, which is due to a cross-over resonance. Particular properties of the group index in the vicinity of such a resonance are demonstrated in the case of closely spaced upper levels.

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In recent years an interest has grown in studying light propagation in atomic media having very special controlled optical properties. The susceptibility of those media is steered by control or pump laser fields to create the conditions of a lowered absorption accompanied by a steep normal dispersion \[1\]. The majority of works in this field have been done for three-level atoms in the \(\Lambda\) configuration, for which it was possible to experimentally achieve \[2,3\] and theoretically describe \[4\] light slowdown or even light stopping in the conditions of the electromagnetically induced transparency \[5\]. Reviews of various aspects of "slow light" concerning both basic physical aspects as well as potential applications can be found, e.g., in Refs \[6,7\].

Another situation, in which such a behavior of the susceptibility is encountered, is typical of the Doppler-free saturated spectroscopy \(8\). As recently pointed out by Agarwal and Dey \(10\), in a region of the Lamb dip, created by a strong laser beam in a two-level system, conditions of a reduced absorption and steep dispersion are created for a counter-propagating signal pulse, being in resonance with the only transition in this system.

In this work we make a step further and study light propagation in an inhomogeneously broadened medium of three-level atoms in the \(V\) configuration. Although general optical properties of such media are well known, as concerns the possible enhancement of the index of refraction there exists a completely new element compared with the case of two-level systems. In addition to two resonances due to a resonant coupling between two states, there appears a cross-over resonance \(9\) and a region of a peculiar behavior of the susceptibility connected with it. In this region one observes a significant reduction of the absorption and a steep normal dispersion. We thus have to do with a kind of a transparency window, in which the group velocity is reduced but, in contrast to the case of the \(\Lambda\) configuration, the absorption is not completely suppressed. Note that cross-over resonances have recently been discussed in the context of double dark resonances appearing in generalized \(\Lambda\) systems with an additional coupling to a fourth level \(11\).

In this paper we will investigate the pulse propagation in the vicinity of a cross-over resonance. In particular we will evaluate the scale of the effect of the pulse slowdown and its dependence on the energy spacing of the upper levels.

Consider a three-level model atom with a single lower state \(b\) and two upper states \(a\) and \(d\). The lower state is coupled with the upper states by two counter-propagating fields: the weak signal field \(\frac{1}{2}\epsilon_1(z, t) \exp[i(k_1 z - \omega_1 t)] + \frac{1}{2}\epsilon_2(z, t) \exp[-i(k_1 z - \omega_1 t)]\) (the phase of the signal changes during the evolution) and a relatively strong control field \(\epsilon_2 \cos(k_2 z - \omega_2 t)\) \((k_2 < 0)\). The density matrix \(\rho\) fulfills the von Neumann equation completed with the phenomenological relaxation terms describing relaxation within the system. If we transform-off the terms rapidly oscillating in time and make the rotating wave approximation we obtain the following equations for the density matrix \(\sigma\) \((\sigma_{nn} = \rho_{nn} \exp[-i(k_2 z - \omega_2 t)], \sigma_{bd} = \rho_{bd} \exp[-i(k_2 z - \omega_2 t)], \sigma_{ij} = \rho_{ij}\) otherwise)

\[
\begin{align*}
    i\hbar \sigma_{aa} &= -d_{ab}\sigma_{ba} \left(\frac{1}{2} \epsilon_1 \exp[i(k_1 - k_2)z - i(\omega_1 - \omega_2)t] + \frac{1}{2} \epsilon_2\right) + \\
    &+ d_{ba}\sigma_{ab} \left(\frac{1}{2} \epsilon_1 \exp[i(k_2 - k_1)z - i(\omega_2 - \omega_1)t] + \frac{1}{2} \epsilon_2\right) - i\hbar \Gamma_{aa} \sigma_{aa}, \\
    i\hbar \sigma_{dd} &= -d_{db}\sigma_{bd} \left(\frac{1}{2} \epsilon_1 \exp[i(k_1 - k_2)z - i(\omega_1 - \omega_2)t] + \frac{1}{2} \epsilon_2\right) + \\
    &+ d_{bd}\sigma_{ad} \left(\frac{1}{2} \epsilon_1 \exp[i(k_2 - k_1)z - i(\omega_2 - \omega_1)t] + \frac{1}{2} \epsilon_2\right) - i\hbar \Gamma_{dd} \sigma_{dd}, \\
    i\hbar \sigma_{ba} &= (E_b + i\omega_2 - E_a)\sigma_{ba} + [d_{ba}(\sigma_{bb} - \sigma_{aa}) - d_{ba}\sigma_{da}] \left(\frac{1}{2} \epsilon_1 \exp[i(k_2 - k_1)z - i(\omega_2 - \omega_1)t] + \frac{1}{2} \epsilon_2\right) - i\hbar \Gamma_{ba} \sigma_{ba}, \\
    i\hbar \sigma_{bd} &= (E_b + i\omega_2 - E_a)\sigma_{bd} + [d_{bd}(\sigma_{bb} - \sigma_{dd}) - d_{bd}\sigma_{ad}] \left(\frac{1}{2} \epsilon_1 \exp[i(k_2 - k_1)z - i(\omega_2 - \omega_1)t] + \frac{1}{2} \epsilon_2\right) - i\hbar \Gamma_{bd} \sigma_{bd}, \\
    i\hbar \sigma_{ad} &= (E_a - E_d)\sigma_{ad} - d_{ab}\sigma_{bd} \left(\frac{1}{2} \epsilon_1 \exp[i(k_1 - k_2)z - i(\omega_1 - \omega_2)t] + \frac{1}{2} \epsilon_2\right) + \\
    &+ d_{bd}\sigma_{ab} \left(\frac{1}{2} \epsilon_1 \exp[i(k_2 - k_1)z - (\omega_2 - \omega_1)t] + \frac{1}{2} \epsilon_2\right) - i\hbar \Gamma_{ad} \sigma_{ad},
\end{align*}
\]

where \(d_{ij}\) are the dipole moment matrix elements and \(\Gamma's\) - the relaxation rates. Equations (1) are solved perturbatively with respect to the signal field \(\epsilon_1\), i.e. we write \(\sigma_{ij} = \sigma_{ij}^0 + \sigma_{ij}'\), where \(\sigma_{ij}^0\) are the solutions (stationary) of Eqs (1) without the signal field, while \(\sigma_{ij}'\) are linear in \(\epsilon_1\). In the Fourier picture the equations for \(\sigma_{ij}'(\omega)\) read

\[
\begin{align*}
    (\omega + i\Gamma_{aa})\sigma_{aa}' &= \frac{1}{2\hbar} d_{ab}\epsilon_2 \sigma_{ba}' - \frac{1}{2\hbar} d_{ba}\sigma_{ab}' = -\frac{1}{2\hbar} d_{ab}\sigma_{ba}^0 \exp[i(k_1 - k_2)z] \epsilon_1(\omega_1 - \omega_2) + \\
    &+ \frac{1}{2\hbar} d_{ba}\sigma_{ab}^0 \exp[i(k_2 - k_1)z] \epsilon_1^*(-\omega_1 - \omega_2), \\
    (\omega + i\Gamma_{dd})\sigma_{dd}' &= \frac{1}{2\hbar} d_{db}\epsilon_2 \sigma_{bd}' - \frac{1}{2\hbar} d_{bd}\sigma_{db}' = -\frac{1}{2\hbar} d_{bd}\sigma_{bd}^0 \exp[i(k_1 - k_2)z] \epsilon_1(\omega_1 - \omega_2) + \\
    &+ \frac{1}{2\hbar} d_{db}\sigma_{db}^0 \exp[i(k_2 - k_1)z] \epsilon_1^*(-\omega_1 - \omega_2). 
\end{align*}
\]
\[ (\omega + \omega_{ab} - \omega_2 + i\Gamma_{ba}) \sigma'_{ba} + \frac{1}{2\hbar} d_{ba} \epsilon_2 (\sigma'_{aa} - \sigma'_{bb}) = \frac{1}{2\hbar} d_{ba} \epsilon_2 \sigma'_{da} \]

\[ (\omega + \omega_{db} - \omega_2 + i\Gamma_{bd}) \sigma'_{bd} + \frac{1}{2\hbar} d_{bd} \epsilon_2 (\sigma'_{dd} - \sigma'_{bb}) = \frac{1}{2\hbar} d_{bd} \epsilon_2 \sigma'_{ad} \]

\[ [d_{ba} (\sigma'_{bb} - \sigma'_{aa}) - d_{bd} \sigma'_{da}] \]

\[ \frac{1}{\hbar} \exp[i(k_2 - k_1)z] \epsilon^*_1 (-\omega + \omega_1 - \omega_2), \]

where \( \omega_{ij} = (E_i - E_j)/h \). The equations for the other nondiagonal matrix elements follow from the relation \( \sigma_{ij} (\omega) = \sigma^*_{ji} (-\omega) \). Note that \( \sigma^*_{aa} + \sigma^*_{bb} + \sigma^*_{cc} = 1 \), \( \sigma^*_{aa} + \sigma^*_{bb} + \sigma^*_{cc} = 0 \).

The polarization of the medium is \( P(t) = P^+(t) + P^-(t) \) where

\[ P^+(t) = N [d_{ba} \sigma'_{ab}(t) + d_{bd} \sigma'_{bd}(t)] \exp[i(k_2 - k_1)z - i(\omega_2 - \omega_1)t], \]

\[ N \] being the medium density. Alternatively we may write

\[ P^+(\omega) = N [d_{ba} \sigma'_{ab}(\omega + \omega_1 - \omega_2) + d_{bd} \sigma'_{bd}(\omega + \omega_1 - \omega_2)] \exp[i(k_2 - k_1)z]. \]

This means that in the case of counter-propagating fields the contribution of the terms including \( \epsilon^*_1 \) in Eqs (2) is averaged to zero due to rapid spatial oscillations. The susceptibility is given by the formula \( \chi(\omega, \omega_1, \omega_2) = \sum_{i,j} \epsilon_{ij} \chi_{ij} \) \( \exp[i(k_1 - k_2)z] \), \( \epsilon_{0i} \) being the vacuum permittivity (the factor of 2 is due to the fact that we have included the factor of 1/2 in the definition of the electric fields). Because of the inhomogeneous broadening of the medium the calculated susceptibility is to be averaged over the atomic velocity distribution of the width \( D \)

\[ \chi_{av}(\omega) = \int_{-\infty}^{\infty} \chi(\omega, \omega_1, \omega_2) \frac{1}{\sqrt{\pi}D} \exp[-\frac{v^2}{D^2}] dv. \]

Due to the susceptibility being a rapidly varying function of \( \omega \) the group velocity \( v_g \) differs from \( c \) by the group index \( n_g = c/v_g \) of a large value

\[ n_g = 1 + \frac{\omega_1}{2} \frac{d}{d\omega} \text{Re} \chi_{av}(\omega). \]

The propagation inside the sample of a light pulse of a spectral shape \( g(\omega) \) is given by

\[ \epsilon_1(z, t) = \int_{-\infty}^{\infty} g(\omega) \exp[-i(\omega(t - z/c))] \exp[i(\omega_1z/2c) \chi_{av}(\omega)] dw. \]

For the sake of illustration we have performed numerical calculations of the susceptibility by finding a numerical solution of the stationary version of Eqs (1) with \( \epsilon_1=0 \) and then by solving Eqs (2). Because our discussion of the influence of cross-over resonances on laser pulse propagation has a rather general character we have adopted somewhat arbitrarily the input data being of order of those corresponding to rubidium and its hyperfine splitting. The obtained susceptibility was averaged with the Gaussian distribution with \( \kappa D = 4.58 \times 10^{-8} \) a.u. which corresponds to a Doppler FWHM of \( \Delta v = 502 \) MHz. The density of the medium was \( 3 \times 10^{-14} \) a.u. (2 \( \times \) \( 10^{11} \) cm\(^{-3} \)). The atomic parameters were taken \( \omega_{da} = 4.06 \times 10^{-8} \) a.u. (\( \nu = 267 \) MHz), \( \omega_{ab} = 5.845 \times 10^{-8} \) a.u. (\( \lambda = 789 \) nm), \( \Gamma_{aa} = \Gamma_{dd} = \Gamma = 8.63 \times 10^{-10} \) a.u. (\( \Delta \nu = 5.7 \) MHz), \( \Gamma_{ba} = \Gamma_{bd} = \Gamma_{ad}/2 = \Gamma/2 \), which corresponds to a spontaneous emission from both upper levels of a lifetime of 28 ns (a generalization for the case of different lifetimes of the two upper levels is straightforward). The matrix elements of the dipole moments were obtained from the values of the relaxation rates, assuming that the spontaneous emission occurs only to the lower state \( |b> \). The maximum pump field amplitude was \( 3.47 \times 10^{-10} \) a.u. (4.23 mW cm\(^{-2} \)). We have set \( \omega_1 = \omega_2 (k_2 = -k_1) \), \( h\omega_1 = (E_a + E_d)/2 - E_b \). Our scanning of the Fourier variable is equivalent to an independent scanning with the probe laser, as usual in the studies of electromagnetically induced transparency.
Fig. 1 presents the group index as a function of frequency. The left and right maxima correspond to those frequencies for which one observes a steep normal dispersion and a reduced absorption due to the excitation by the probe pulse of the lower and upper excited levels, respectively, in two groups of atoms of such velocities that their number in the ground state has been decreased because of transitions to the same level due to the pump. Each of the two maxima is an analogue of the single maximum observed in Ref. [10]. Our central maximum is a new phenomenon: we observe a reduction of the group velocity, connected with a steep dispersion and a reduced absorption in the frequency region which corresponds to a cross-over resonance, i.e. to an excitation of atoms from the holes of the velocity distribution to the upper states different from those which took part in the hole burning. The group index due to the cross-over resonance, similarly as that due to two-level resonances, can be as large as a few hundreds even for a small atomic density.

If the lasers are not tuned exactly in the middle between the upper levels the three peaks are shifted and made asymmetric, because the population of the atoms of such a velocity that they can be active in the transitions is changed.

Fig. 2 presents the shape of an initially Gaussian pulse inside the sample, propagating in the window of a reduced absorption around the cross-over resonance, obtained from Eq. (7). At the end of the 0.5 cm sample one observes a reduction of the height to the level of 3 % of the initial value due to the pulse absorption, which is a typical value in saturation absorption experiments, and a retardation by $4 \times 10^8$ a.u. (about 10 ns) due to the decrease of its group velocity.

Besides V systems composed of atomic hyperfine levels it is possible to realize such a system when the upper levels are chosen to be Zeeman sublevels of an excited level with properly chosen polarizations of the laser beams. The latter case allows for an additional control of the dispersive properties of the medium by changing the level spacings using an external magnetic field.

It is interesting to study the situation in which the energy spacing of the close upper levels is of order of the natural width. Fig. 3 shows the frequency dependence of the group index for different level spacings. Curve 1, corresponding to $\omega_{da} = 4\Gamma$ exhibits again three maxima, however with the central one lowered due a partial overlapping of the two holes burned out in the velocity distribution. Reducing the level spacing to $2\Gamma$ (curve 2) caused disappearing of the central peak, because the gap between the two holes has vanished. Merging the levels leads to a single peak (curve 3). This corresponds to a single dip in the absorption spectrum. It follows from the Bloch equations that such a situation is equivalent to the case of a two-level system, in which the upper state is a certain "bright" combination of $|a>$ and $|d>$ (the other orthogonal combination being decoupled from $|b>$), the transition dipole moment is $\sqrt{|d_{ab}|^2 + |d_{bd}|^2}$ and the dip is the Lamb dip. The widths of the holes depend on the intensity of the control field. Curve 1 in Fig. 4 is the same as curve 2 in Fig. 3. Decreasing the field intensity leads to reappearing of the central peak (curve 2), because the holes have been narrowed and thus separated again. Increasing $\epsilon_2$ causes such a broadening of the holes that they overlap, which yields the group index in the form of a single broad peak. The dependence of the heights of the peaks on the field intensity is connected with the fact that in weaker fields the holes are narrower, so are the transparency windows and the regions of a normal dispersion; as a consequence the dispersion curves are steeper and the group index is larger (Fig. 3).

We have demonstrated the possibility of a significant enhancement of the group index and a slowdown of a light pulse in an inhomogeneously broadened medium of three-level atoms in the V-configuration. The Doppler-averaged susceptibility exhibits three regions of a reduced absorption accompanied by a steep normal dispersion. We have examined laser pulse propagation in the central region corresponding to a cross-over resonance. The pulse slowdown is of the same order of magnitude as in the two remaining regions. In the case of closely spaced upper levels of a V system we have demonstrated how the frequency dependence of the group index changes depending on the level spacing and the pump field intensity.

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[1] M. O. Scully and M. S. Zubairy, *Quantum Optics*, (Cambridge University Press, 1997).
[2] C. Liu, Z. Dutton, C. H. Behroozi and L. V. Hau, Nature **409**, 490 (2001).
[3] D. F. Phillips, A. Fleischhauer, A. Mair, R. L. Walsworth and M. D. Lukin, Phys. Rev. Lett. **86**, 783 (2001).
[4] M. Fleischhauer and M. D. Lukin, Phys. Rev. Lett. **84**, 5094 (2000).
[5] O. Kocharovskaya, Y. Rostovtsev and M. O. Scully, Phys. Rev. Lett. **86**, 628 (2001).
[6] S. E. Harris, Phys. Today **507**, 36 (1997).
[7] A. B. Matsko, O. Kocharovskaya, Y. Rostovtsev, G. G. Welch, A. S. Zibrov and A. O. Scully, Adv. At. Mol. Phys. **46**, 191 (2001).
[8] Z. Dutton, N. S. Ginsberg, C. Slowe and L. V. Hau, Europhysics News **35**, 33 (2004).
[9] W. Demtröder, *Laser Spectroscopy*, (Springer, Berlin, 1996).
[10] G. S. Agarwal and Tarak Nath Dey, Phys. Rev. A **68**, 063816 (2003).
[11] G. Wąsik, W. Gawlik, J. Zachorowski and Z. Kowalczyk, Phys. Rev. A **64**, 051802(R) (2001).
FIG. 1: The group index for the data given in the text. $10^{-8}$ a.u. on the frequency axis corresponds to $12 \Gamma$.

FIG. 2: The shape of a signal pulse along the sample; left: from top to bottom at $z = 0$, $z = 2 \times 10^7$ a.u., $z = 6 \times 10^7$ a.u., $z = 10^8$ a.u. (0.53 cm) for the data as in the text; right: normalized pulse peak at the end of the sample (dashed line) compared with an analogous pulse traveling in vacuum (solid line). $10^{10}$ a.u. on the time axis is equal to $2.4 \times 10^{-7}$ s and corresponds to $8.63 \Gamma^{-1}$.

FIG. 3: The group index in the case of closely spaced upper levels for $\epsilon_2 = 3.47 \times 10^{-10}$ a.u.: $\omega_{da} = 4 \Gamma$ (line 1), $\omega_{da} = 2 \Gamma$ (line 2), $\omega_{da} = 0$ (line 3). $10^{-9}$ a.u. on the frequency axis corresponds to 1.2 $\Gamma$.

FIG. 4: The group index in the case of closely spaced upper levels for $\omega_{da} = 2 \Gamma$: $\epsilon_2 = 1.74 \times 10^{-10}$ (line 1), $\epsilon_0 = 3.47 \times 10^{-10}$ a.u. (line 2), $\epsilon_0 = 10.4 \times 10^{-10}$ a.u. (line 3). $10^{-9}$ a.u. on the frequency axis corresponds to 1.2 $\Gamma$. 
