Narrowing of electromagnetically induced transparency resonance in a Doppler-broadened medium

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We derive an analytic expression for the linewidth of electromagnetically induced transparency (EIT) resonance in a Doppler-broadened system. It is shown here that for relatively low intensity of the driving field the EIT linewidth is proportional to the square root of intensity and is independent of the Doppler width, similar to the laser-induced line narrowing effect described by Feld and Javan. In the limit of high intensity we recover the usual power-broadening case where the EIT linewidth is proportional to the intensity and inversely proportional to the Doppler width.

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Because of the Doppler effect the atoms in a gas experience a radiation field with shifted frequency. Hence the macroscopic polarization representing the medium's response to the radiation needs to be averaged over the frequency distribution determined by the velocity distribution of the atoms. By and large, all sorts of phenomena in gas lasers are related to Doppler broadening [1] and it is also the origin of the famous hole burning [2] and Lamb dip [3,4]. It was more than 30 years ago that the laser-induced line narrowing effect in a three-level Doppler broadened system was discovered by Feld and Javan [5]. Notably, Feld and Javan found the spectral width of the narrow line to be linearly proportional to the driving field Rabi frequency. Various aspects of this effect have been investigated [6–8].

The interest in the narrow nonabsorption resonances imposed on the Doppler profile has resumed recently in connection with electromagnetically induced transparency (EIT) experiments, which have produced ultraslow light propagation [9–11] with spatial compression (group velocity less than tens of m/s) and have made it possible to enhance nonlinear optical processes by orders of magnitude [12–15].

The steepness of the dispersion function with respect to frequency plays the key role for the small group velocity of light, and is directly related to the transmission width [16–18]. Hence the behavior of the transmission linewidth in terms of experimental parameters is of a great deal of interest. In high-resolution spectroscopy and high-precision magnetometry based on a narrow EIT line [19–24] the experiments are usually carried out with atomic cell configurations so that the effect of Doppler broadening on EIT is also an important concern for the performance of the devices.

Doppler-broadening effects in EIT and lasing without inversion have been studied by a number of authors [25–29]. Sub-Doppler resolution spectroscopy using the EIT sideband has also been proposed [30]. Most of this work focused on the possibilities of absorption cancellation and preferable field configurations (copropagation of probe and drive lasers in folded schemes, counterpropagation in cascade schemes). In the limit of vanishing probe field and under the assumption that all atoms were trapped in the dark state it was found that power broadening of the EIT line takes place: $\Gamma_{EIT}=\Omega^2/W_D$ (where Ω is the Rabi frequency of the driving field and W_D is the Doppler linewidth), which is similar to the well-known result for a homogeneously broadened system: $\Gamma_{EIT}=\Omega^2/\gamma$ (where γ is the homogeneous linewidth). This dependence was experimentally verified in [10]. In the limit of relatively low probe field intensity, $\alpha \ll (\gamma/W_D)\Omega$, and under the same assumption of full coherent trapping (i.e., neglecting the two-photon coherence decay) it leads to the following result for the EIT linewidth: $\Gamma_{EIT}=\alpha\Omega/\gamma$, where α is the Rabi frequency of the probe field [31].

In this paper, we find an explicit expression for the linewidth of EIT resonance in a Doppler-broadened three-level system in the linear approximation with respect to the probe field taking into account the finite decay time of the low-frequency coherence. In the limit of very large intensity it is reduced to the power-broadening case. However, for the intermediate range of intensities the coherent population trapping is velocity selective, i.e., it occurs only for those atoms whose frequencies are close to resonance with the driving field. In this case we find that the width of EIT resonance is proportional to the Rabi frequency of the driving field (similar to the result of Feld and Javan [5]) and to the square root of the ratio of the relaxation times of the coherence at the two-photon (low-frequency) and of the population difference at the one-photon (optical) transitions:

$$\Gamma_{EIT} \Rightarrow \sqrt{\frac{2\gamma_{bc}}{\gamma}}\Omega. \tag{1}$$

This regime corresponds to the narrowest possible EIT linewidth and therefore it is very favorable for realization of efficient EIT-based nonlinear transformations and light storage.

Let us consider the closed atomic model scheme depicted in Fig. 1. In this three-level Λ scheme one of the two lower levels is coupled to the upper level ($a\rightarrow c$) by a coherent

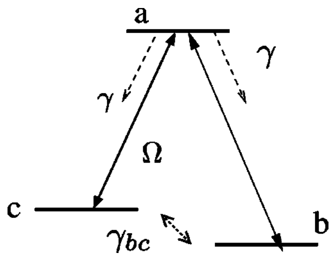

FIG. 1. Three-level model scheme. The upper level *a* decays to *b* and *c* with decay rate γ. The relaxation rate between levels *b* and *c* is denoted as γ_{bc}, which is assumed to be small compared to γ.

drive laser and the transition $a \to b$ is probed by a weak coherent field. The atomic decays are confined among the given levels. Note that such a model gives a description almost equivalent to the one for an open system in which atoms decay (out of the interaction region) with the rate γ_{bc}, and atoms are coming into the interaction region with equally populated lower levels. A detailed comparison of our model with the open system will be published elsewhere.

If the system is Doppler broadened, the susceptibility should be averaged over the entire velocity distribution such that [1]

$$\chi = \int d(kv) f(kv)\, \eta \left\{ \frac{\rho_{ab}(kv)}{\alpha} \right\}, \tag{2}$$

where k is the wave number of the probe field, $f(kv)$ is the velocity distribution function, $\rho_{ab}(kv)$ is the coherence between states a and b induced by the radiation fields, $\eta \equiv (3/8\pi) N \gamma \lambda^3$, N is the atomic density, and λ is the wavelength. For a stationary atom, in the first order of the probe field, ρ_{ab} can be written as

$$\rho_{ab} = \frac{-i\alpha}{\Gamma_{ab}\Gamma_{cb} + \Omega^2} \times \left[\Gamma_{cb}(\rho_{aa}^{(0)} - \rho_{bb}^{(0)}) + \frac{\Omega^2}{\Gamma_{ca}}(\rho_{cc}^{(0)} - \rho_{aa}^{(0)}) \right], \tag{3}$$

where the $\rho_{ii}^{(0)}$'s are the zeroth-order populations (in the probe field) and $\Gamma_{ij} \equiv \gamma_{ij} + i\Delta_{ij}$ with the off-diagonal decay rates γ_{ij} given by $\gamma_{ab} = \gamma_{ac} = (\gamma + \gamma' + \gamma_{bc})/2$, $\gamma_{cb} = \gamma_{bc}$. The Δ_{ij}'s are defined as $\Delta_{ab} = \omega_{ab} - \nu \equiv \Delta$, $\Delta_{ac} = \omega_{ac} - \nu_0$, and $\Delta_{cb} = \Delta_{ab} - \Delta_{ac}$, where ν and ν_0 are the frequencies of the probe and drive fields, respectively.

In the present analysis we use the following assumptions. (1) The decay rates in the transitions $a \to b(\gamma)$ and $a \to c(\gamma')$ are assumed to be the same (γ) and defined by spontaneous emission, which is typically the case for dilute gases. (2) The decay rates of the population difference and coherence at the low-frequency transition $b \leftrightarrow c$ are the same (γ_{bc}), which is typically the case when this decay is determined by the time of flight through the interaction region. (3) The probe field is weak such that a first-order analysis is valid. (4) The driving field is on resonance for a stationary atom: $\omega_{ac} = \nu_0$. (5) The probe field and driving field propagate in the same direction, and the frequency difference between the transitions $a \to b$ and $a \to c$ is small enough such that the residual Doppler shift $(k - k')v$ can be ignored. (6) The EIT condition for the homogeneously broadened system ($\Omega^2 \gg \gamma\gamma_{bc}$) is valid. (7) The inhomogeneous linewidth (W_D) is large enough that $W_D \gg \gamma, \Omega$.

Under these assumptions the atomic populations ρ_{ii}^0 can be written as

$$\rho_{aa}^{(0)} = \frac{2\gamma_{bc}\Omega^2}{2D}, \quad \rho_{cc}^{(0)} = \frac{4\gamma X \gamma_{bc} + 2\gamma_{bc}\Omega^2}{2D},$$

$$\rho_{bb}^{(0)} = \frac{4\gamma X \gamma_{bc} + 2\gamma_{bc}\Omega^2 + 2\Omega^2\gamma}{2D}, \tag{4}$$

where $X = [\gamma^2 + (kv)^2]/2\gamma$ and $D = 4\gamma X \gamma_{bc} + 3\gamma_{bc}\Omega^2 + \Omega^2\gamma$. Then, for an atom with its velocity v, the off-diagonal element of the density matrix $\rho_{ab}(kv)$ is found as

$$\rho_{ab} = \frac{i\alpha}{Y}\frac{1}{2D}\left[\Gamma_{cb}(4\gamma X\gamma_{bc} + 2\Omega^2\gamma) - \frac{\Omega^2 4\gamma X \gamma_{bc}}{\gamma + \gamma_{bc}/2 + ikv} \right], \tag{5}$$

where $Y = (\gamma + \gamma_{bc}/2 + i\Delta + ikv)(\gamma_{bc} + i\Delta) + \Omega^2$.

Doppler broadening is usually modeled by convolution of a given function over a Maxwell-Boltzmann velocity distribution. Due to the complexity in the integration with a Gaussian distribution, however, explanations of the results obtained usually rely on numerical analysis [26–29]. In order to obtain a simple expression of the linewidth, we approximate the usual Gaussian distribution with a Lorentzian function; this leads to a rather simple form of the inhomogeneously broadened susceptibility with which detailed analysis is possible.

If we use a Lorentzian profile as the velocity distribution function $f(kv)$ with full width half maximum (FWHM) $2W_D$ such that $f(kv) = (1/\pi) W_D / [W_D^2 + (kv)^2]$, Eq. (2) can be evaluated by contour integration in the complex plane, which contains two poles in the lower half plane, viz., $kv = -iW_D$ and $kv = -i\sqrt{\Omega^2\gamma/2\gamma_{bc}}$. After straightforward calculation of the contributions from the two poles, one can find the complex susceptibility. In particular, the minimum absorption at the line center is obtained as

$$\chi''(\Delta = 0) = \frac{\eta\gamma_{bc}}{\gamma_{bc}W_D + \Omega^2}\left[\frac{\sqrt{x}}{1 + \sqrt{x}} \right], \tag{6}$$

where $x = \Omega^2\gamma/2\gamma_{bc}W_D^2$. We note that, as long as $\Omega^2 \gg \gamma\gamma_{bc}$, the expression is vanishingly small as $\eta\sqrt{x}/W_D$ when $x \ll 1$, and also as $\eta\gamma/W_D^2$ when $x \gg 1$, so that the EIT (i.e., strong suppression of absorption in the presence of a driving field at $\Delta = 0$) is preserved. The maximum of χ'', on the other hand, can be found as $\chi''_{\max} \approx \eta/W_D$ at $\Delta \approx \pm\Omega$.

Since the absorption at the line center is negligibly small, given by Eq. (6), we evaluate Δ, which defines Γ_{EIT} as $\chi''(\Delta = \Gamma_{EIT}) = \eta/2W_D$. The FWHM of the EIT resonance (Γ_{EIT}) is then obtained as

$$\Gamma_{EIT}^2 = \frac{\gamma_{bc}}{\gamma}\Omega^2(1+x)\left[1+\left\{1+\frac{4x}{(1+x)^2}\right\}^{1/2}\right]. \tag{7}$$

This is the main result of the present article. Here we can see the two extreme cases, namely,

$$\Gamma_{EIT} \Rightarrow \begin{cases} \sqrt{\frac{2\gamma_{bc}}{\gamma}}\Omega & (x \ll 1), \quad \text{(8a)} \\ \frac{\Omega^2}{W_D} & (x \gg 1). \quad \text{(8b)} \end{cases}$$

Note that the range of x is $(\gamma/W_D)^2 \ll x \ll \gamma/\gamma_{bc}$. In the expression (8a) corresponding to the limit $x \ll 1$, the linewidth of EIT is linearly proportional to Ω, the Rabi frequency of the driving field (i.e., to the square root of the driving field intensity) and it is independent of the Doppler width W_D.

A similar linear dependence of the linewidth on the Rabi frequency was previously obtained in Ref. [5]. The earlier work [5] dealt with a laser gain system where the weak transitions between the lasing levels were used. The decays out of the lasing levels were the main relaxation mechanisms while the spontaneous decays between levels were not taken into account. These so-called open systems have relaxation of the low-frequency coherence (γ_{bc}) of the same order of magnitude as relaxation of the population difference at the optical transitions (γ), i.e., $\gamma_{bc} \approx \gamma$. In this case we have $x \approx \Omega^2/2W_D^2$; Eq. (8a) takes the form $\Gamma_{EIT} \approx \Omega$. Since $\Omega \ll W_D$, the linewidth, in turn, is much smaller than W_D. This limit fully corresponds to the experimental conditions of Ref. [5].

In the limit $x \gg 1$ (corresponding to small γ_{bc} or a strong driving field) Γ_{EIT} is proportional to the intensity Ω^2, and inversely proportional to W_D. Many recent EIT experiments were performed in alkali-metal vapors where the two-photon coherence (ρ_{bc}) was built among the hyperfine levels of the ground state. In these systems the low-frequency coherence relaxation time is determined by the time of flight of the atom through the interaction region, and it is large compared to the lifetime of the excited optical state.

In Fig. 2, we plot the EIT linewidth as a function of the Rabi frequency of the driving field. Due to the fact that the difference between Lorentzian and Gaussian velocity distributions lies mainly at the tail and concerns only far off-resonant atoms whose contribution is negligible, the plots (a) and (b) coincide with high accuracy. We note that $\Gamma_{EIT} \geq \Omega\sqrt{2\gamma_{bc}/\gamma}$ for any value of Ω. Apparently, a smaller ratio γ_{bc}/γ leads to a smaller EIT width at $x \ll 1$, and to a smaller value of Ω at which the linear dependence of Γ_{EIT} on Ω ($\Gamma_{EIT} \propto \Omega$) changes to a quadratic dependence ($\Gamma_{EIT} \propto \Omega^2$). In both the $x \ll 1$ and $x \gg 1$ limits, for a given value of intensity, the width of the EIT resonance in the inhomogeneously broadened medium is smaller than in a homogeneously broadened medium with the same homogeneous linewidth at resonant driving. In the limit $x \gg 1$ this fact was outlined earlier in [31].

This line narrowing effect has a simple physical explanation; namely, it is due to the reduced power broadening for the off-resonant atoms. At the same time it is worth noting that the width of EIT resonance in a Doppler-broadened system can never be reduced beyond the ultimate limit defined by the low-frequency coherence decay time: $\Gamma_{EIT} \geq \gamma_{bc}$. It reaches this limit when EIT sets in with $\Omega^2 \geq \gamma\gamma_{bc}$ independently if the optical line is homogeneously or inhomogeneously broadened. In the case $x \gg 1$ the EIT linewidth exceeds this minimum value at least by the factor W_D/γ.

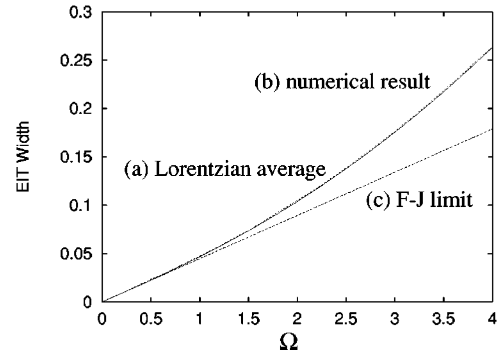

FIG. 2. EIT linewidth (in units of γ) as a function of Ω (also in units of γ), with Doppler width $2W_D = 100\gamma$ and $\gamma_{bc} = 10^{-3}\gamma$. The plot (a) of Eq. (7) by averaging over the Lorentzian distribution function (solid line) is almost identical to that (b) of the numerical result from Eq. (2) using the Maxwell-Boltzmann distribution (dotted line). (c) Feld-Javan (F-J) limit given by Ref. [5] denotes the value of $\Omega\sqrt{2\gamma_{bc}/\gamma}$.

The physical meaning of the parameter x can be understood in the following way. First, let us suppose the system is homogeneously broadened. The optical pumping rate from the level c to b is Ω^2/γ for the resonant driving field. In order to have complete coherent optical pumping in the case of resonant driving this rate should be much bigger than the pumping rate from b to c: $\Omega^2/\gamma \gg \gamma_{bc}$. This means that the driving field should be sufficiently strong: $\Omega^2 \gg \Omega_{hom}^2 \equiv \gamma_{bc}\gamma$. For atoms with velocity v, then, the optical pumping rate is $\Omega^2\gamma/[\gamma^2 + (kv)^2]$. Then, in order to have a complete coherent optical pumping in a Doppler-broadened system we need to require $\Omega^2\gamma/(\gamma^2 + W_D^2) \gg \gamma_{bc}$, which corresponds to $x \gg 1$, i.e., $\Omega^2 \gg \Omega_{inhom}^2 \equiv 2\gamma_{bc}W_D^2/\gamma$. Hence, the parameter x represents the degree of optical pumping from the level c to b within the inhomogeneous linewidth ($x = \Omega^2/\Omega_{inhom}^2$).

With the notion of an effective width δ_{eff}, the width of the EIT resonance can always be regarded as

$$\Gamma_{EIT} \sim \frac{\Omega^2}{\delta_{\text{eff}}}, \tag{9}$$

which is equivalent to the EIT linewidth for the homogeneously broadened medium (where $\Gamma_{EIT} = \Omega^2/\gamma$). The effective width δ_{eff} is defined as the magnitude of the maximum detuning for which atoms are optically pumped into the level b (and hence can interact with a probe field) for a fixed value of Ω.

For $\Omega_{hom} \ll \Omega \ll \Omega_{inhom}$, δ_{eff} can be estimated as $\Omega^2(\gamma/\delta_{\text{eff}}^2) \sim \gamma_{bc}$, yielding $\delta_{\text{eff}} \sim \sqrt{\Omega^2\gamma/\gamma_{bc}}$. Therefore, an increase of intensity of the driving field makes the number of trapped atoms increase, which results, according to Eq. (9), in the linear dependence of t he EIT resonance width: $\Gamma_{EIT} \sim \Omega \sqrt{\gamma_{bc}/\gamma}$ [see Eq. (8a)]. When $\Omega \gg \Omega_{inhom}$ the number of optically pumped atoms is not increased further (since all of them are already optically pumped into the level b), so that $\delta_{\rm eff} \sim W_D$ yielding $\Gamma_{EIT} = \Omega^2/W_D$.

It is worth noting that the results obtained can be used for description of EIT experiments not only in gaseous media with Doppler broadening but also in solids with long lived spin coherence, for example, in rare-earth ion-doped crystals at low temperature [32] when inhomogeneous line broadening of optical transitions plays a major role while inohomogeneous broadening of the spin transitions is negligible. On the other hand, they are not directly applicable for EIT experiments involving a buffer gas in a cell or paraffin coating since collisions of the operating atoms with the buffer gas or wells can essentially disturb the Doppler velocity distribution.

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