Sub-Doppler resolution in inhomogeneously broadened media using intense control fields

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We propose a scheme for obtaining sub-Doppler resolution for one transition of an inhomogeneously broadened, three-level atomic system, by using an intense control field at the other transition. Analytical and numerical calculations are presented to delineate the mechanism responsible for this sub-Doppler resolution, and quantify the extent to which Doppler broadening can be reduced.

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Manipulation of atom response with intense control fields has attracted tremendous attention in recent years [1–8]. Generation of large nonlinear optical cross sections [1], electromagnetically induced transparency (EIT) [2,3,7], enhancement of refractive index in atomic media [4], lasing without population inversion (LWI) [5,6], and efficient population transfer to highly excited metastable states [8], are all examples of phenomena where the control field at one transition of a three-level atom is used to modify the properties of the other transition. Typically, the control field induces coherence between the levels to which it is coupled, and the phenomena listed above exploit this atomic coherence. Thus in EIT, for example, a field that is resonant with an atomic transition can experience diminished absorption, provided a suitable control field acts on the other transition.

Experiments in nonlinear optics and spectroscopy are often performed in atomic vapors, where the linewidths of the transitions of interest are dominated by Doppler broadening. In fact, all experiments on LWI [6], and several related to EIT [3], have been done in atomic vapors. In this paper, we demonstrate the possibility of obtaining sub-Doppler resolution for one transition of a three-level Doppler-broadened atomic system by utilizing an intense control field at the other transition. This work represents an interesting class of phenomena in nonlinear optics, where the atomic coherence induced by a control field is used to influence inhomogeneous broadening. Variations in Doppler widths, based on whether pump and probe fields are copropagating or counterpropagating, have been discussed in the context of gas laser amplifiers [9]. Previously, Reynaud *et al.* [10] reported alteration of Doppler broadening through velocity-dependent shifts of atomic energy levels. Analogous effects have been observed in other fields; for example, double-resonance methods have been used to mitigate spatial inhomogeneity effects on the nuclear-magnetic-resonance linewidths in liquids [11].

For specificity, we focus on EIT, and present results for three-level lambda (Λ) and ladder systems, two atomic level schemes that are easily available in atomic rubidium, and can be accessed by diode lasers [3]. First consider the Λ scheme in the inset of Fig. 1(a), where the intense control field of Rabi frequency G and frequency ω_2 couples the $|1\rangle\leftrightarrow|2\rangle$ transition, and a weak probe, with Rabi frequency g and frequency ω_1, is scanned across the $|1|\leftrightarrow|3\rangle$ transition. $2\gamma_1$ ($2\gamma_2$) is the radiative width of the $|1\rangle\leftrightarrow|3\rangle$ ($|1\rangle\leftrightarrow|2\rangle$) transition. It is straightforward to derive equations for the time evolution of relevant density-matrix elements, which we have explicitly written in Ref. [12].

The absorption of the weak probe A (in units of weak field resonant absorption in the absence of the control field) is given by

$$A=-\operatorname{Im}\left(\frac{\rho_{13}\gamma_1}{g}\right), \tag{1a}$$

where ρ_{13} is the induced polarization on the $|1\rangle\leftrightarrow|3\rangle$ transition. The real part of ρ_{13} is related to the refractive index η through

$$\eta=\operatorname{Re}\left(\frac{\rho_{13}\gamma_1}{g}\right). \tag{1b}$$

From the time-dependent density-matrix equations, a steady-state solution for ρ_{13} can be obtained analytically, to first order in g, as

$$\rho_{13}=\frac{g(\Delta_1-\Delta_2)}{|G|^2-i(\gamma_1+\gamma_2-i\Delta_1)(\Delta_1-\Delta_2)}, \tag{2}$$

where $\Delta_1=\omega_{13}-\omega_1$ and $\Delta_2=\omega_{12}-\omega_2$.

To obtain the probe response in a Doppler-broadened medium, ρ_{13} should be averaged over the velocity distribution of the moving atoms. For a single atom, moving with a velocity v along the z axis, the probe frequency $\omega_1(v)$ and the control field frequency $\omega_2(v)$, as seen by the atom, are given by

$$\omega_1(v)=\omega_1\left(1\pm\frac{v}{c}\right), \quad \omega_2(v)=\omega_2\left(1\pm\frac{v}{c}\right), \tag{3}$$

where the lower (upper) sign corresponds to a copropagating (counterpropagating) atom and probe. We denote by $\delta_1(v)$ and $\delta_2(v)$ the detunings of the probe and the control field from their respective transitions in the atom frame; i.e., $\delta_1(v)=\omega_{13}-\omega_1(v)$ and $\delta_2(v)=\omega_{12}-\omega_2(v)$, $\delta_2(v)$ can be written in terms of $\delta_1(v)$ and the stationary atom parameters as

$$\delta_2(v)=\Delta_2\pm[\delta_1(v)-\Delta_1], \quad \delta_1(v)=\Delta_1\mp\omega_1\frac{v}{c}, \tag{4}$$

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where we have set $\omega_1=\omega_2$ for simplicity. By assuming a Maxwell-Boltzmann distribution for the atomic velocities, we obtain a probability distribution function for $\delta_1(v)$ [13],

$$\rho(\delta_1)=\frac{1}{\sqrt{2\pi D^2}}\,e^{-(\delta_1-\Delta_1)^2/2D^2} \tag{5}$$

where D is the width of the Gaussian. Probe response is then obtained by replacing Δ_1 and Δ_2 in Eq. (2) by $\delta_1(v)$ and $\delta_2(v)$, and performing the average over the probability distribution given by Eq. (5).

For the Λ system, we present results for copropagating probe and control fields, since this geometry is known to yield optimal results in two-photon spectroscopy, EIT and LWI [13]. Figure 1 shows the results of our numerical calculations. Difficulties associated with numerical algorithms at $D=0$ were circumvented by taking the results for $D=0.01$ as representative of a stationary atom (this assertion has been carefully checked previously [13]). Figure 1(a), the absorption spectrum, has the usual Autler-Townes doublet. The asymmetry in the widths arises because the control field detuning is nonzero. The two peaks, located at $\Delta_1=(\Delta_2/2)\pm\frac{1}{2}\sqrt{\Delta_2^2+4|G|^2}$, result from the dressing of the $|1\rangle\leftrightarrow|2\rangle$ transition by the control field. *We have determined the linewidths of the two dressed-state transitions in Fig. 1(a), and find that one has a linewidth greater than D, while the other has a linewidth less than D*. We also find that the decrease in the width of one line is *precisely equal* to the increase in the width of the other line. Thus, by choosing the sign of Δ_2 appropriately, one can selectively narrow one of the lines to a width significantly less than D. The consequences are immediately apparent—if in the absence of the control field the $|1\rangle\leftrightarrow|3\rangle$ transition is Doppler broadened, by employing a control field at the $|1\rangle\leftrightarrow|2\rangle$ transition, sub-Doppler resolution can be realized in the vicinity of the $|1\rangle\leftrightarrow|3\rangle$ transition.

We now provide an analytical argument to elucidate the underlying mechanism responsible for this sub-Doppler resolution. This analysis will enable us to determine the extent of line narrowing that can be achieved for a given set of atom and field parameters, and address the question of why the control field *must* be detuned from resonance. For a stationary atom, Δ_1 and Δ_2 have no velocity dependence, and the pole structure of Eq. (2) gives

$$\Delta_1=\frac{\Delta_2-i(\gamma_1+\gamma_2)}{2}\pm\tfrac{1}{2}\sqrt{\Delta_2^2+4|G|^2-(\gamma_1+\gamma_2)^2+2i(\gamma_1+\gamma_2)\Delta_2}. \tag{6}$$

By expanding the term under the radical, the corresponding linewidths (β) are given by

$$\beta=\frac{\gamma_1+\gamma_2}{2}\left(1\mp\frac{\Delta_2}{\sqrt{\Delta_2^2+4|G|^2}}\right). \tag{7}$$

Equation (7) is the usual asymmetry in the widths of the two Autler-Townes lines when Δ_2 is nonzero.

Next we examine the consequences of a moving atom by replacing, in Eq. (2), Δ_1 by Δ_1+x, and Δ_2 by Δ_2+x, where $x=kv$, and k is the wave vector of the probe. For copropagating fields in a Doppler-broadened Λ system, terms involving $(\Delta_1-\Delta_2)$ in Eq. (2) will have no velocity dependence, and so the only term that is modified is the Δ_1 term. The poles of ρ_{13} are now given by

$$\Delta_1=\frac{\Delta_2-i(\gamma_1+\gamma_2)-x}{2}\pm\tfrac{1}{2}\sqrt{\Delta_2^2+4|G|^2-(\gamma_1+\gamma_2-ix)^2+2i\Delta_2(\gamma_1+\gamma_2-ix)}, \tag{8}$$

which, on expanding the radical, can be written as

$$\Delta_1=\frac{\Delta_2-i(\gamma_1+\gamma_2)-x}{2}\pm\frac{\sqrt{\Delta_2^2+4|G|^2}}{2}\left\{1+\frac{i\Delta_2(\gamma_1+\gamma_2-ix)}{\Delta_2^2+4|G|^2}\right\}. \tag{9}$$

Note that $\langle x\rangle=0$ and $\langle x^2\rangle=\langle k^2v^2\rangle=D^2$, where the angle brackets denote averaging over the distribution in Eq. (5). Since the uncertainty in x will determine the uncertainty in the position of the peak, the fluctuations in x give rise to the widths of the peaks. By combining the contribution from the radiative damping terms in Eq. (7), the net widths for the lines located at $(\Delta_2/2)\pm\frac{1}{2}\sqrt{\Delta_2^2+4|G^2}$ are given by

$$\beta=\frac{\gamma_1+\gamma_2+D}{2}\left(1\mp\frac{\Delta_2}{\sqrt{\Delta_2^2+4|G|^2}}\right). \tag{10}$$

It is clear from Eq. (10) that, for $\Delta_2=0$, both lines would have identical widths, equal to $(\gamma_1+\gamma_2+D)/2$, and so for $D\gg\gamma_1,\ \gamma_2$, the linewidth would be dominated by D. For $\Delta_2\neq0$, and with the parameters of Fig. 1(a), the term in the parentheses of Eq. (10) is approximately 1.7 or 0.3. Hence one transition has a width greater than D, but the other is narrowed to a width much less than D. The widths of the two lines, determined from our numerical calculations, are in exact agreement with those given by Eq. (10). Furthermore, this analysis suggests that the width of one of the lines [at $\Delta_1=(\Delta_2/2)+\frac{1}{2}\sqrt{\Delta_2^2+4|G|^2}$] will be reduced, due to the control field, by a factor of $[1-\Delta_2/(\sqrt{\Delta_2^2+4|G|^2})]$. Thus, by choosing appropriate values of the control field intensity (G) and frequency (Δ_2), one can manipulate the linewidth of the $|1\rangle\leftrightarrow|3\rangle$ transition, and obtain sub-Doppler resolution. Equation (10) clearly shows the necessity of detuning the control field from resonance, since only a nonzero Δ_2 will lead to the differential broadening effect on the two lines that is described here. It is also obvious from Eq. (10) that for a given G and Δ_2, the decrease in the width of one line will be precisely compensated for by an increase in the width of the other line. We note from Fig. 1(a) that the position of the absorption maxima shift slightly with an increase in the Doppler width D. These shifts, which are small, can be estimated by examining the coefficient of x^2 in Eq. (8).

Equation (2) indicates that for a stationary atom, when $\Delta_1=\Delta_2$, one obtains zero absorption (100% transparency). We find from Fig. 1(a) that this condition persists even for a Doppler-broadened medium. Finally, in Fig. 1(b) we show the behavior of the real part of the induced polarization on the $|1\rangle\leftrightarrow|3\rangle$ transition, which also shows the differential line-broadening effect. Profiles such as these are experimentally measurable [3].

We next look at the ladder system in the inset of Fig. 2(a)

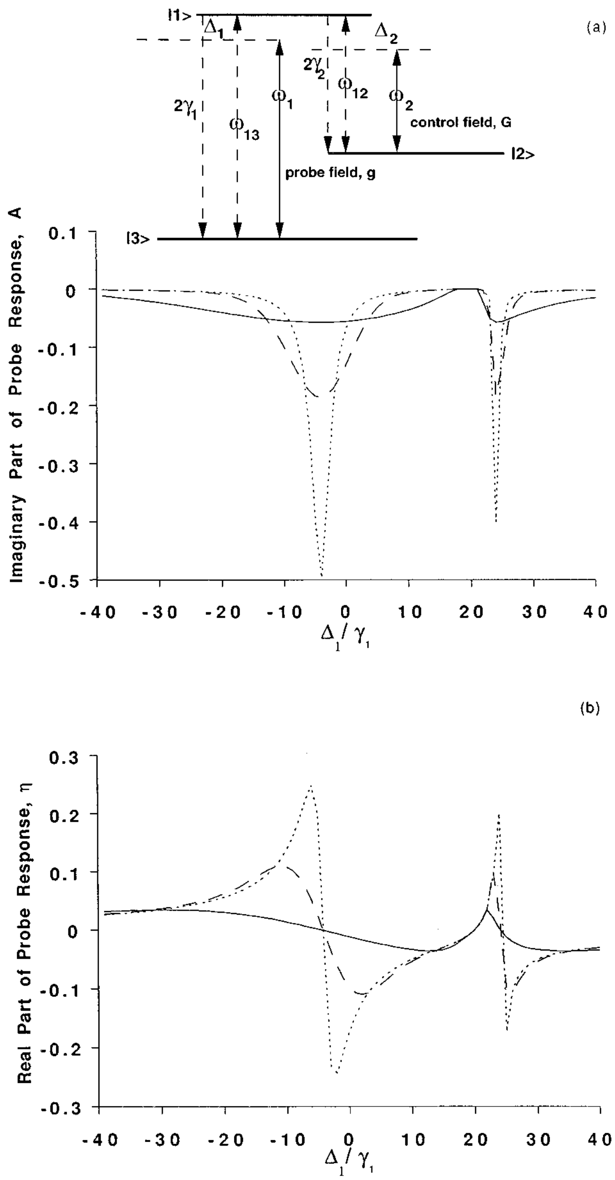

FIG. 1. (a) Probe absorption spectrum in a Λ system for Doppler widths D of 0.01 (dot), 5 (dash), and 20 (solid). Other parameters are $G=10\gamma_1$, $\Delta_2=20\gamma_1$, and $\gamma_2=\gamma_1$. Inset: Schematic representation of a three-level Λ system. The spontaneous decay rates from $|1\rangle$ to $|3\rangle$ and $|1\rangle$ to $|2\rangle$ are $2\gamma_1$ and $2\gamma_2$, respectively. ω_{12} and ω_{13} are the resonance frequencies of the two allowed transitions. (b) Real part of probe response in Λ system for parameters identical to (a).

[12], where the control field, at frequency ω_2 and Rabi frequency G, drives the $|2\rangle \leftrightarrow |3\rangle$ transition, and a weak probe field, at frequency ω_1 and Rabi frequency g, is tuned across the $|1\rangle \leftrightarrow |2\rangle$ transition. From the equations describing time evolution of density-matrix elements [12], the steady-state value of ρ_{12} (related to probe response) can be derived analytically, to first order in g, as

$$\rho_{12}=ig\,\frac{(\gamma_1+i\Delta_1+i\Delta_2)\rho_{22}^0-iG^*\rho_{23}^0}{(\gamma_1+i\Delta_1+i\Delta_2)(\gamma_1+\gamma_2+i\Delta_1)+|G|^2},\qquad(11)$$

where ρ_{22}^0 and ρ_{23}^0 are the zeroth-order contributions, given by

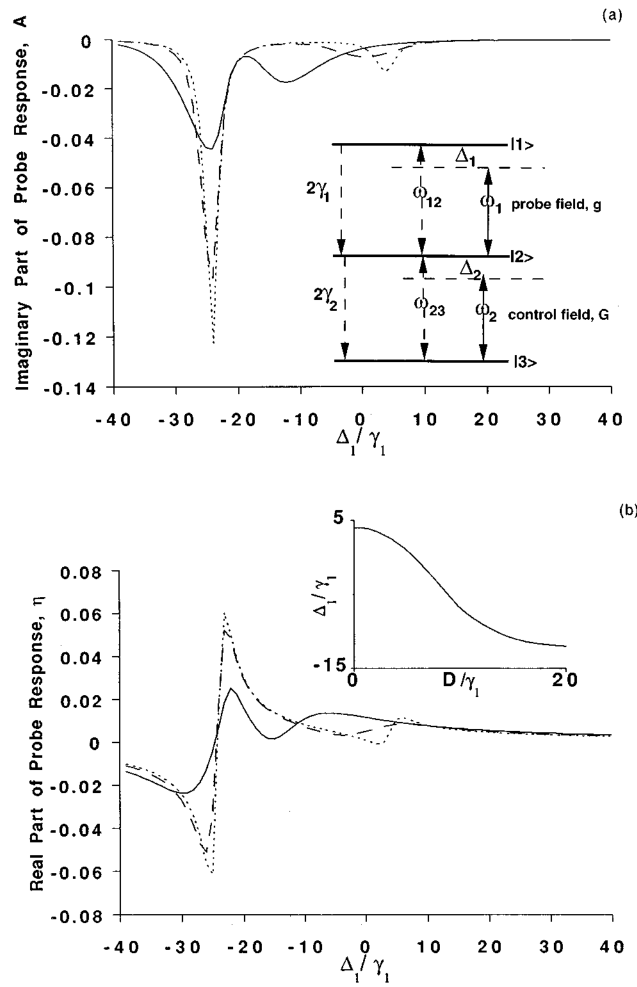

FIG. 2. (a) Probe absorption spectrum in a ladder system for Doppler widths D of 0.01 (dot), 5 (dash), and 20 (solid). Other parameters are $G=10\gamma_1$, $\Delta_2=20\gamma_1$, and $\gamma_2=\gamma_1$. Inset: Schematic representation of a three-level ladder system. The spontaneous decay rates from $|1\rangle$ to $|2\rangle$ and $|2\rangle$ to $|3\rangle$ are $2\gamma_1$ and $2\gamma_2$, respectively. ω_{12} and ω_{23} are the resonance frequencies of the upper and lower transitions, respectively. (b) Real part of the probe response in ladder system for parameters identical to (a). Inset: Value of Δ_1 at which maximum absorption occurs for the line at $\Delta_1=(-\Delta_2/2)+\frac{1}{2}\sqrt{\Delta_2^2+4|G|^2}$, as a function of D.

$$\rho_{22}^0=\frac{|G|^2}{\gamma_2^2+\Delta_2^2+2|G|^2}\qquad(12a)$$

and

$$\rho_{23}^0=\frac{iG(\gamma_2-i\Delta_2)}{\gamma_2^2+\Delta_2^2+2|G|^2}.\qquad(12b)$$

The probe absorption for this case is

$$A=-\mathrm{Im}\left(\frac{\rho_{12}\gamma_1}{g}\right),\qquad(13a)$$

and the refractive index is

$$\eta=\mathrm{Re}\left(\frac{\rho_{12}\gamma_1}{g}\right).\qquad(13b)$$

The prescription outlined in Eqs. (3)–(5) can be directly applied here to average the probe response over the Doppler distribution. The numerical results of probe absorption for counterpropagating probe and control fields (this is the optimal geometry for ladder systems) are shown in Fig. 2(a). Once again, we find that one transition [at $\Delta_1=(-\Delta_2/2)-\frac{1}{2}\sqrt{\Delta_2^2+4|G|^2}$] is narrowed to a width less than D, while the other is broadened to more than D. Just as we did for the Λ system, we can determine the linewidths here by examining the pole structure of the expression for ρ_{12} in Eq. (11), and these widths, in the presence of Doppler broadening, are

$$\beta=\gamma_1+\left(\frac{\gamma_2+D}{2}\right)\left(1\pm\frac{\Delta_2}{\sqrt{\Delta_2^2+4|G|^2}}\right). \qquad (14)$$

For the parameters of Fig. 2(a), $[1-(\Delta_2/\sqrt{\Delta_2^2+4|G|^2})]$ is approximately 1.7 or 0.3, indicating that we can obtain sub-Doppler resolution. The reduction in linewidth due to the control field is by the same factor as for the Λ system, i.e., $(1-(\Delta_2/\sqrt{\Delta_2^2+4|G|^2}))$. While the linewidths determined from our analytical formalism are identical to the numerically calculated widths for a Λ system, there is a small discrepancy ($\sim$5%) between the numerical and analytical results for a ladder system due to our not having accounted for the velocity dependence of ρ_{22} and ρ_{23}. Figure 2(a) also indicates that, unlike the Λ system, the ladder system does not show a point of zero absorption. A striking feature seen in Fig. 2(a) is that the shifts in the positions of the absorption maxima, as a function of D, are now quite substantial, and in fact should be observable in experiments (these shifts can be estimated in the same way as for the Λ system). The inset to Fig. 2(b) shows the calculated position of the absorption maxima [for the line at $\Delta_1=(-\Delta/2)+\frac{1}{2}\sqrt{\Delta_2^2+4|G|^2}$], versus D. Lastly, the refractive index profiles in Fig. 2(b), for parameters identical to Fig. 2(a), also exhibit differential line-broadening effects.

In summary, we have presented a scheme for obtaining sub-Doppler resolution on an inhomogeneously broadened atomic transition, by the application of a control field at a different transition. This scheme utilizes the ideas of atomic coherence and interference and can be easily implemented, provided the following requirements are met: (i) the control field must act on a transition which shares an energy level with the transition for which sub-Doppler resolution is desired, and (ii) the control field must be detuned from the transition it is coupled to. The first condition is usually satisfied in most LWI and EIT schemes. The second condition is, in general, not crucial to demonstrate either EIT or LWI, but is *necessary* here to provide differential broadening in the Autler-Townes doublet. For the parameters chosen in our calculations, the linewidth is reduced by a factor greater than 3. Note that the line-narrowing effect proposed here can be observed for much larger and realistic Doppler widths (say, $D=100$), provided G and Δ are more than D (e.g., $G=\Delta=500$). The amount of linewidth reduction can be controlled by optimizing the control field intensity and frequency. Finally, we have demonstrated that the decrease in the width of one dressed-state transition is exactly matched by the increase in the width of the other dressed-state transition. The results for counterpropagating (copropagating) fields in Λ (ladder) systems are not presented, since typical experiments utilize the configuration chosen in this work. However, the formulation presented here may be readily extended to other geometries, as well as to systems with four or more energy levels.

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