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Subnatural linewidth for probe absorption in an electromagnetically-induced-transparency medium due to Doppler averaging

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We obtain subnatural linewidth (i.e., $<\Gamma$) for probe absorption in room-temperature Rb vapor using electromagnetically induced transparency (EIT) in a Λ system. For stationary atoms, the EIT dip for a resonant control laser is roughly one-half as wide as the control Rabi frequency Ω_c. But in thermal vapor, the moving atoms fill the transparency band so that the final EIT dip remains subnatural even when $\Omega_c \gg \Gamma$. We observe a linewidth of $\Gamma/4$ for $\Omega_c = 8\Gamma$ in the D_2 line of Rb.

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Laser spectroscopy in a room-temperature gas is often limited by Doppler broadening due to the thermal velocity of gas particles. While techniques such as saturated-absorption spectroscopy can be used to eliminate the first-order Doppler effect and get linewidths close to the natural linewidth, the natural linewidth itself appears as a fundamental limit to the resolution that can be achieved in precision spectroscopy. In addition, when lasers are locked to atomic transitions (for use as frequency standards), the natural linewidth determines the tightness of the lock. It is therefore desirable to develop techniques for getting below the natural linewidth.

In this work, we demonstrate a technique to obtain subnatural linewidth in a Doppler broadened medium. The technique has been adapted from recent developments in the use of control lasers in three-level systems as a means of modifying the absorption properties of a probe beam [1], in what is generally called coherent-control spectroscopy. More specifically, we use the phenomenon of electromagnetically induced transparency (EIT) in a Λ-type system, in which an initially absorbing medium is rendered transparent to a weak probe when a strong (independent) control laser is applied to a second transition [2,3]. It is well known that the EIT dip on resonance for stationary atoms can be subnatural ($<\Gamma$) if the Rabi frequency of the control laser (Ω_c) is sufficiently small, i.e., $\Omega_c < 2\Gamma$ [4]. However, in thermal vapor, the effect of the large Doppler width was thought to have a detrimental effect on observing any subnatural features. Indeed, theoretical work in such Doppler-broadened media predicted that one can achieve at best sub-Doppler resolution by detuning the control laser [5]. It is only recently that theoretical analysis in hot vapor has shown that the EIT linewidth actually gets narrowed due to thermal averaging [6,7].

In earlier work [8], we have shown that we can obtain subnatural linewidth in hot vapor either by detuning the control by an amount that is larger than the Doppler width, or by using a slightly detuned control along with a counterpropagating pump beam that allows the probe to address only zero-velocity atoms. Here, we show that one can observe subnatural linewidth for the EIT dip even when the control is on resonance. While there have been some previous observations of subnatural linewidth for the EIT dip in room-temperature vapor [4,9,10], the fact that the linewidth actually gets reduced by thermal averaging has not been well appreciated. This is why all of the previous experiments were done with values of the control Rabi frequency less than 2Γ, where the dip is subnatural even for zero-velocity atoms. By contrast, we will see that our linewidth is $\Gamma/4$ even when the Rabi frequency is as large as 8Γ.

We must also contrast this with the related phenomenon of coherent-population trapping (CPT) in Λ systems [11,12], where the linewidth can be extremely small compared to the linewidth of the excited state because it is limited only by the decoherence rate between the two ground levels. In contrast to EIT experiments, CPT experiments require (i) the use of phase coherent control and probe beams, (ii) roughly equal powers in the two beams, (iii) detuning of the two beams (equally) from resonance to decrease the decohering effect of the excited state, and (iv) buffer gas filled vapor cells to increase the ground coherence time. Under these conditions, the control and probe beams pump the atoms into a dark nonabsorbing state and probe transmission shows a narrow peak, with a linewidth of 50 Hz being observed in room-temperature Cs vapor [13]. Applications of CPT include atomic clocks [11] and storage of light [14]. By contrast, EIT occurs because of the ac Stark shift of the excited state by the strong control laser. On resonance, this creates two dressed states [15] that are shifted equally from the unperturbed level, and the probe absorption again shows a minimum at line center. Since CPT is a ground-state coherence phenomenon, it is effectively decoupled from the excited state and is used for spectroscopy on the ground hyperfine interval, which in the case of Cs is used in the SI definition of the second. Indeed, the linewidth of 50 Hz is not really subnatural because the natural linewidth of the upper ground hyperfine level is <1 Hz. On the other hand, in earlier work [16] we have shown that the subnatural linewidth of the EIT resonance can be used for high-resolution hyperfine spectroscopy on the excited state.

We first consider the theoretical analysis of the three-level Λ system shown in Fig. 1. For specificity, we show the relevant hyperfine levels in the D_2 line of ^{87}Rb. The strong control laser drives the $|1\rangle \leftrightarrow |2\rangle$ transition with Rabi frequency Ω_c and detuning Δ_c, while the weak probe is scanned across the $|1\rangle \leftrightarrow |3\rangle$ transition. The spontaneous decay rate

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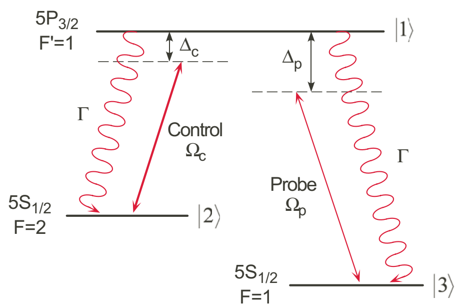

FIG. 1. (Color online) Three-level Λ system in the D_2 line of ^{87}Rb.

from the upper state to either of the ground states is Γ, which is $2\pi\times 6$ MHz in Rb. The absorption of the weak probe is well known from a density matrix analysis of the system [5–7,17]. It is given by $-\mathrm{Im}(\rho_{13}\Gamma/\Omega_p)$, where ρ_{13} is the induced polarization on the $|1\rangle\leftrightarrow|3\rangle$ transition. In the weak probe limit, all of the atoms will get optically pumped into the $|3\rangle$ state, so that $\rho_{33}\approx 1$ and $\rho_{22}\approx 0$. From the steady state solution of the density matrix equations, we obtain (to first order in Ω_p)

$$\rho_{13} = -\frac{i\Omega_p/2}{\Gamma - i\Delta_p + i\frac{|\Omega_c/2|^2}{\Delta_p-\Delta_c}}. \tag{1}$$

The pole structure of this equation shows that there will be a zero in the absorption when $\Delta_p=\Delta_c$, and this minimum will occur exactly on resonance if the control is on resonance. This is the phenomenon of EIT.

The above analysis is correct for a stationary atom. For an atom moving along the direction of the beams with a velocity v, the detuning of the two beams will change by $\pm kv$, where k $(=2\pi/\lambda)$ is the photon wave vector and the sign depends on whether the atom is moving away from or toward the beams. Thus, to obtain the complete probe absorption in a gas of moving atoms, the above expression must be corrected for the velocity of the atom and then averaged over the one-dimensional Maxwell-Boltzmann distribution of velocities.

The results of such a calculation for room-temperature Rb atoms with $\Delta_c=0$ and different values of Ω_c are shown in Fig. 2. First let us look at the curves for zero-velocity atoms shown on the left-hand side. As is well known, the absorption splits into an Autler-Townes doublet and shows a classic EIT dip in the center. The doublet peaks are the two symmetric dressed states created by the control laser [15], and their separation is exactly equal to the value of Ω_c. Thus, the linewidth of the EIT dip (defined as the full width at half-maximum) depends on the value of Ω_c. It is subnatural (i.e., $<\Gamma$) only when Ω_c is less than 2Γ, and arises because the absorption on resonance goes to zero due to quantum interference between the dressed states [4]. But the linewidth increases quickly when Ω_c is increased, so that the width is 3Γ when $\Omega_c=4\Gamma$.

Now consider the probe response after thermal averaging shown on the right-hand side. The scale of absorption has decreased by a factor of 30 as the absorption spreads over the

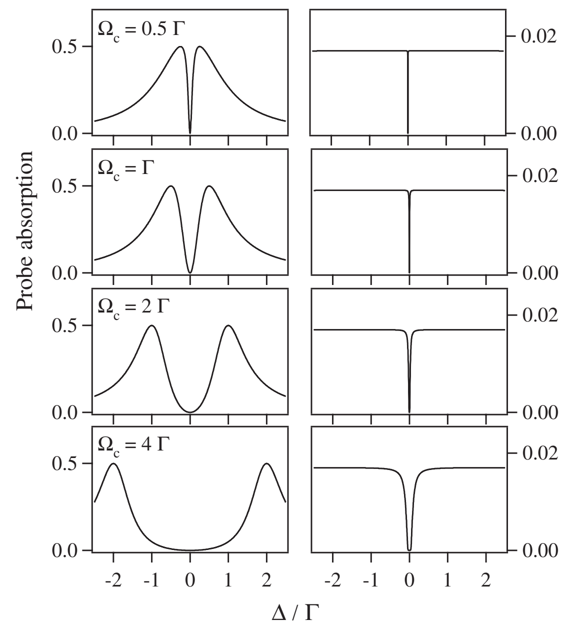

FIG. 2. Calculated probe absorption for four values of Ω_c. The curves on the left-hand side are for zero-velocity atoms, while the curves on the right-hand side are after thermal averaging in room-temperature vapor. Note the decreased scale on the right-hand side.

different velocity groups, but the linewidth of the EIT dip remains extremely small. Indeed it only increases to $\Gamma/6$ when $\Omega_c=4\Gamma$, and remains subnatural for much higher values of Ω_c.

The prediction that the linewidth of the EIT dip in a Λ system becomes narrower after thermal averaging is both surprising and counterintuitive, and has been appreciated theoretically only recently [6,7]. This can be understood better by considering the effect of velocity on the EIT line shape, as shown in Fig. 3 for $\Omega_c=4\Gamma$. The solid curve is for stationary atoms, while the dashed (dotted) curve is for atoms moving with $v=+10$ (-10) m/s. The Autler-Townes doublet for the moving atoms is shifted to the right or left of center so that they fill in the transparency region for stationary atoms. The overall transparency window shrinks and the effective EIT linewidth decreases. Note that Fig. 2 shows

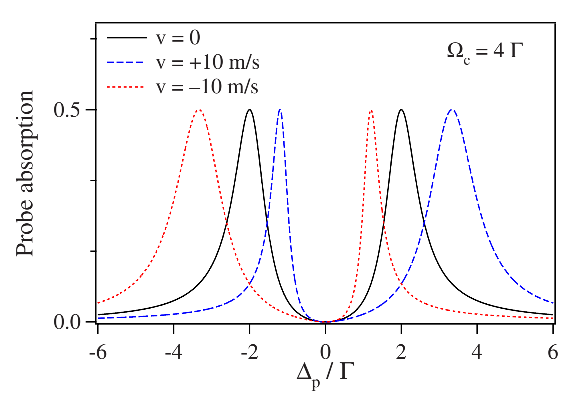

FIG. 3. (Color online) Effect of velocity on probe absorption. The curves are for zero velocity and for atoms moving with 10 m/s toward the right and left.

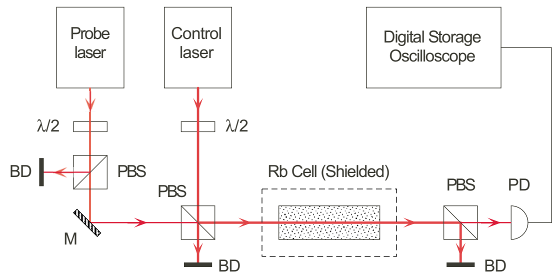

FIG. 4. (Color online) Schematic of the experiment. Figure key: $-\lambda/2$, half-wave plate; PBS, polarizing beam splitter; BD, beam dump; M, mirror; PD, photodiode.

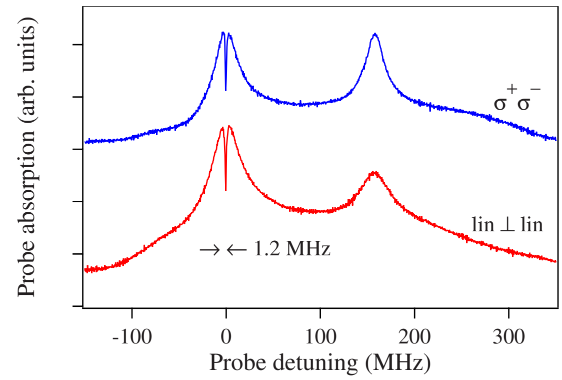

FIG. 5. (Color online) Probe absorption showing a narrow EIT dip for orthogonal circular ($\sigma^+\sigma^-$) and linear (lin $\perp$ lin) polarizations. Probe detuning is measured from the $F'=1$ level.

that this linewidth reduction is accompanied by a change in the EIT line shape as well [7].

Such a surprising reduction in linewidth after thermal averaging also happens for EIT in a ladder-type system. This has been predicted and observed by us in earlier work with room-temperature Rb atoms [18]. As in the present case, thermal averaging results in two additional features: (i) The scale of the transparency is reduced; and (ii) the line shape is modified from that for zero-velocity atoms. This prediction of a modified line shape showing enhanced absorption near resonance has recently been observed for EIT with Rydberg atoms [19]. However, the observed linewidth in ladder systems is usually large; for example, the EIT linewidth obtained by us using the same D_2 line of Rb for the lower transition was 50 MHz [18]. Narrow linewidths can instead be obtained using resonant two-photon excitation to access the upper state [20].

We now turn to the experimental demonstration of these results. The experimental schematic is shown in Fig. 4. The probe and control beams are derived from independent home-built frequency-stabilized diode laser systems tuned to the 780 nm D_2 line of Rb [21]. The linewidth of the lasers after stabilization is about 1 MHz. The two beams are about 2 mm in diameter each. The beams copropagate through a room-temperature vapor cell with orthogonal polarizations. The cell has a magnetic shield around it so that the residual field (measured with a three-axis fluxgate magnetometer) is <5 mG.

In Fig. 5, we show a typical probe absorption spectrum taken with a control power of 0.21 mW, corresponding to a Rabi frequency of about 9 MHz (1.5Γ), and a probe power of 0.07 mW. The most striking feature of the curve is the 1.2 MHz ($\Gamma/5$) wide EIT dip at the line center, which would be $\Gamma/1.33$ wide if we were dealing only with stationary atoms and not with hot vapor. The signal-to-noise ratio of the dip is more than 20, and it appears exactly at line center. Note that the control laser is locked to the $F=2 \rightarrow F'=1$ transition using standard saturated-absorption spectroscopy, where the observed transition linewidths are about 15 MHz. Therefore, one expects residual frequency jitter of the lock point on the order of a few hundred kHz. Furthermore, the linewidth of the probe laser is of the same order as the calculated EIT linewidth. The observed linewidth can also become broader if there is a small misalignment angle between the control and probe beams [10]. Despite these broadening effects, the observed dip is only 1.2 MHz wide. The narrow resonance is quite robust and appears whether the two beams have orthogonal linear or circular polarizations, as seen from the figure.

This kind of narrow resonance can be used for tight locking of the probe laser, i.e., locking the laser with reduced statistical uncertainty. Of course, this does not guarantee that there will be no systematic errors in the lock point. Since the EIT resonance condition corresponds to the difference between the control and probe laser frequencies, any systematic shift of the control laser from resonance would cause an equal systematic shift of the probe laser. Such systematic errors can arise in any locking scheme, but the advantage of using the EIT resonance for locking is that the statistical errors will be more than an order of magnitude smaller. This method of locking the probe laser to an atomic transition is to be distinguished from schemes where only the difference (rf) frequency between the lasers is important, which are important in atomic clocks [11] and locking of two lasers with a fixed frequency offset [9]. Such difference-frequency resonances (which depend only on the ground hyperfine interval) can achieve much narrower linewidth by using the related CPT phenomenon and buffer gas filled cells, as mentioned earlier.

Another interesting feature of the spectrum shown in Fig. 5 is that the narrow EIT dip appears in the middle of a 40 MHz wide peak. A similar peak also appears at a frequency 157 MHz higher, which is precisely the hyperfine interval between the $F'=1$ and $F'=2$ levels of the excited state. We thus conclude that this peak is due to additional velocity-dependent optical pumping by the control laser. Note that the earlier density matrix analysis assumes complete optical pumping by the strong control laser [17]. However, such population transfer will be true mainly for zero-velocity atoms for which the control laser is on resonance, but will not be very effective for moving atoms for which the control is detuned, particularly when we consider that the probe has finite power and is no longer in the weak probe limit. Thus, the optical pumping will be velocity dependent, and the population as a function of velocity will deviate from the Maxwell-Boltzmann distribution with a peak near zero velocity.

In Fig. 6, we compare the calculations of probe absorption in room-temperature vapor with and without such optical

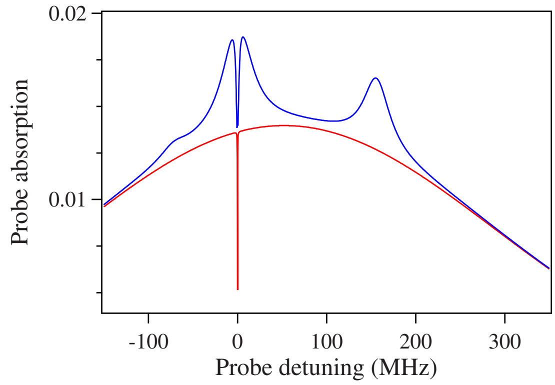

FIG. 6. (Color online) Calculated probe absorption spectrum in room-temperature vapor. The lower curve is for a Maxwell-Boltzmann velocity distribution, while the upper curve is obtained after taking into account additional optical pumping by the control laser for zero velocity atoms.

pumping. The velocity-dependent optical pumping is incorporated phenomenologically by assuming a population transfer that is proportional to the scattering rate for control photons. The peak near zero in the velocity distribution will show up as two additional peaks in the probe absorption spectrum, one when these atoms come into resonance with the $F=1 \rightarrow F'=2$ transition (at $\Delta_p=+157$ MHz) and other for the $F=1 \rightarrow F'=0$ transition (at $\Delta_p=-72$ MHz). A close examination of the observed spectrum in Fig. 5 does show a small third peak at $\Delta_p=-72$ MHz. As expected, the size of the optical pumping effect and the relative height of the three broad peaks are different for the two polarizations, but the line shape is close to the calculated one.

Further consideration of velocity-dependent optical pumping shows that there will be a second velocity class that will get optically pumped into the $F=1$ level. This happens for atoms moving toward the control beam with a velocity near 122 m/s (corresponding to a Doppler shift of 157 MHz), for which the control appears resonant with the $F=2 \rightarrow F'=2$ transition. This will cause two additional peaks in the probe absorption, at $\Delta_p=-157$ and -229 MHz. Since such optical pumping is a competition between probe power and control detuning, we see these additional peaks in our measured spectra only when the probe power is reduced considerably.

We finally consider the effect of control power on the EIT linewidth. From the theoretical analysis presented earlier, the EIT dip should remain subnatural for quite high values of Rabi frequency. This is confirmed from the three spectra shown in Fig. 7(a), taken with control powers of 0.02 mW ($\Omega_c \sim 0.5\Gamma$), 2 mW (5Γ), and 6.5 mW (8Γ), respectively. As the power is increased, the overall size of the broad optical pumping peak increases. But the linewidth of the EIT dip only increases from 0.88 MHz ($\Gamma/7$) to 1.5 MHz ($\Gamma/4$). The slow increase of the linewidth with Ω_c is seen more clearly

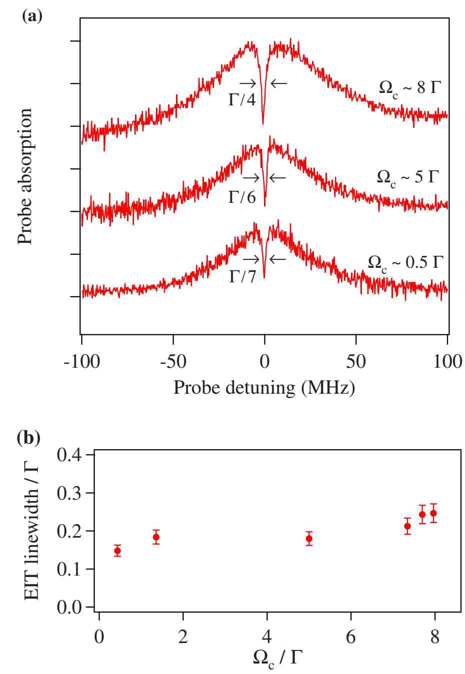

FIG. 7. (Color online) Dependence of EIT linewidth on control power. In (a), we show three absorption curves with increasing values of control Rabi frequency (Ω_c), as indicated. The linewidth increases only slightly with Ω_c, as shown in (b).

in Fig. 7(b), where we plot the width of the EIT dip for six values of Ω_c. This clearly demonstrates that the EIT linewidth in thermal vapor is well below the control Rabi frequency, or equivalently the separation of the dressed states.

In conclusion, we have demonstrated subnatural width for the EIT dip in thermal Rb vapor at large values of control power. While the transparency band for stationary atoms is about one-half the Rabi frequency of the control laser, in thermal vapor the moving atoms fill up this band in such a manner that the residual EIT dip remains extremely narrow even for large values of Rabi frequency. The observed line shape is described well by a density-matrix treatment of the three-level system with thermal averaging [6,7]. The most important advantage of the narrow feature over our previous work with detuned control lasers [8] is that the dip appears exactly at line center. Thus, the feature can be used for high resolution spectroscopy and tight laser locking. The narrow feature is robust in terms of laser polarization and detuning. This could be important for applications of EIT such as nonlinear optics, gain without inversion, slowing of light, and quantum-information processing.

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