Observation of Electromagnetically Induced Transparency in Collisionally Broadened Lead Vapor

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We report the observation of electromagnetically induced transparency on the collisionally broadened resonance line of Pb vapor. By applying a 1064-nm laser beam, the transmission at 283 nm is increased by at least a factor of exp(10), with nearly all of the Pb atoms remaining in the ground state.

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In a recent experiment, Boller, Imamoğlu, and Harris [1] have shown how, by applying an additional electromagnetic field, an optically thick transition whose upper level decays by autoionization can be rendered transparent to a probe frequency. Such transparency is a special case of the formalism and examples given by Lambropoulos [2,3], Eberly [4,5], Agarwal [6,7], Knight [8], and their co-workers. It is also of the same nature as that observed by Whitley and Stroud [9] and Alzetta *et al.* [10] in the Rayleigh scattering of radiatively broadened systems, and is also related to the Holtsmark profile of ion broadened lines [11]. The transparency, in essence, results from the combined ac-Stark splitting and quantum interference of the dressed states which are created by the additional electromagnetic field.

In this Letter, we extend the observation of electromagnetically induced transparency to the collisionally (pressure) broadened regime. We study the transmission of the 283-nm resonance transition of neutral lead vapor with, and without, a coupling laser at 1064 nm. The energy-level diagram of lead is shown in Fig. 1. In these experiments, the self-collisional broadening of the resonance transition exceeds its lifetime broadening by a factor of 40. We observe an increase in the transmission of optically thick Pb vapor from well below our dynamic range of $\exp(-14)$ to $\exp(-4)$.

Noting Fig. 1, in the absence of the 1064-nm coupling laser, a 283-nm probe will be absorbed by the strongly pressure broadened $|1\rangle$-$|3\rangle$ transition. With the 1064-nm coupling laser present, one expects both single-photon absorption on $|1\rangle$-$|3\rangle$, and also two-photon absorption on the $|1\rangle$-$|2\rangle$ transition. This expectation is correct when the Rabi frequency of the coupling laser is much less than the square root of the product of the Doppler width of the $|1\rangle$-$|2\rangle$ transition and the detuning of the probe laser from the $|1\rangle$-$|3\rangle$ transition (6 cm^{-1}, in this experiment). When the Rabi frequency of the coupling laser substantially exceeds this product the behavior of the system is very different [1,12]. The coupling laser dresses the system so as to create the equivalent energy-level diagram shown in the inset of Fig. 1. There is now a quantum interference between the $|1\rangle$-$|2d\rangle$ and $|1\rangle$-$|3d\rangle$ absorption paths. If a collision is fast (impact regime), it establishes a coherence between states $|2d\rangle$ and $|3d\rangle$. If a perturber interacts dominantly with bare state $|3\rangle$, this coherence results in a destructive interference and, in principle, zero absorption at line center. If a perturber interacts dominantly with bare state $|2\rangle$, then the interference is constructive and the probe laser sees the Lorentzian wings of the dressed states. Equivalently, when viewed in the bare-state basis, the superposition state of the atom has negligible amplitude in bare state $|3\rangle$ and is therefore nearly unaffected by $|1\rangle$-$|3\rangle$ collisions. For a system to show electromagnetically induced transparency in the collisionally broadened regime, it is therefore essential that a perturber interact dominantly with only bare state $|3\rangle$. In the present experiment, the principle perturbers are ground-state Pb atoms. State $|3\rangle$ lead atoms interact resonantly ($1/R^3$) with these atoms with a self-broadening coefficient of 0.018 cm^{-1} per 10^{17} atoms/cm^3. State $|2\rangle$ lead atoms interact nonresonantly ($1/R^6$) with these atoms, with a broadening of 0.0012 cm^{-1} per 10^{17} atoms/cm^3.

The characteristics of the coupling laser are critical to an experiment of this type. The present experiment was chosen because of the fortunate near coincidence of the $6s^2 6p7s\ ^3P_1^0$-$6s^2 6p7p\ ^3D_1$ Pb transition (calculated $gf=0.98$) [13], with the 1064-nm line of the Nd-doped yttrium-aluminum-garnet (Nd:YAlG) laser. This laser (Quanta-Ray DCR-2A) was injection seeded by a diode-

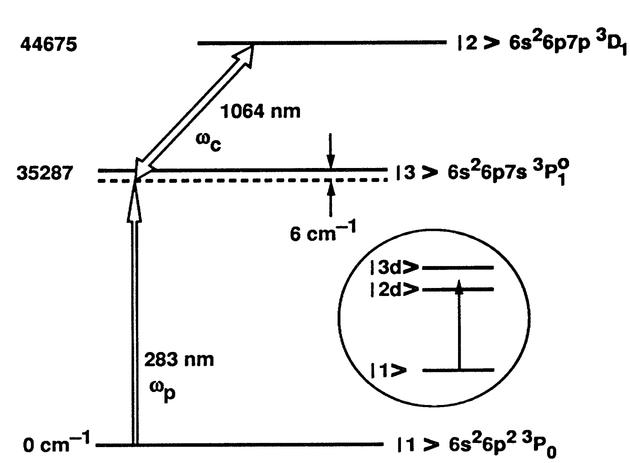

FIG. 1. Energy-level diagram of neutral Pb. Inset: Dressed-state picture.

pumped nonplanar ring resonator (Lightwave Electronics S-100). The 1064-nm laser was single mode and nearly transform limited with a pulse length of 8 ns. It has been shown that Wiener-Levy phase diffusion [14] of a laser will dephase state $|2\rangle$ in the same manner as do collisions with state $|2\rangle$. Therefore, to obtain the maximum transparency, the contribution to the linewidth of the coupling laser which results from phase diffusion must be small compared to the dephasing linewidth of state $|2\rangle$ ($0.0012\ \mathrm{cm}^{-1}$ in this experiment).

The characteristics of the probe laser (or even an incoherent probe source) are less important to an experiment of this type. If the coupling laser pulse is temporally smooth, then the only requirement on the probe laser is that its linewidth be small compared to the transparency width. The probe transition for this experiment was $6s^2 6p^2\,{}^3P_0$–$6s^2 6p7s\,{}^3P_1^0$ with a center frequency of $35\,287\ \mathrm{cm}^{-1}$ and a measured oscillator strength of $gf = 0.21$ [15]. The probe laser was a frequency-doubled dye laser which was in turn pumped by 532 nm light from the injection-seeded Quanta-Ray laser. The measured linewidth and pulse length of the undoubled dye laser were $1.3\ \mathrm{cm}^{-1}$ and 5 ns, respectively.

Figure 2 is a schematic of the experimental arrangement. The probe and coupling laser beams were orthogonally polarized and propagated collinearly through a lead vapor cell with a zone length of 9 cm. The diameter of the coupling laser beam was 3 mm, and to ensure good spatial overlap, the probe beam was apertured to a diameter of 0.5 mm. The energies of the pulses were typically 20 mJ and 10 nJ, respectively. The cell was operated in a heat pipe mode with the lead density determined by the He buffer gas. The cell was heated by molybdenum wire heaters which were enclosed in a surrounding vacuum chamber. Operating conditions were 15 torr at 1150 °C. The transmitted probe beam was detected by a Hamamatsu microchannel plate detector and averaged (10 Hz repetition rate) with a boxcar integrator. The ratio of maximum to minimum observable probe signal was about exp(14).

Figures 3 and 4 show measured transmission versus probe frequency at Pb pressures of 15 torr ($N = 1.0 \times 10^{17}$) and 18 torr, respectively. With the coupling laser off, Fig. 3(a), the line shape was non-Lorentzian with greater absorption on the low-frequency side [16] and a 50%-transmission linewidth of $61\ \mathrm{cm}^{-1}$. The long-wavelength skewing prevented a Lorentzian curve of growth estimate of the actual minimum transmission. Figure 3(b) shows transmission as a function of the probe frequency with the coupling laser on. Here, the maximum transmission is $\exp(-4)$, an increase of much greater than exp(10) compared to that at the same probe frequency with the coupling laser absent. By increasing the coupling laser intensity we were able to increase the maximum transmission to $\exp(-1)$.

The selection rules for the lead system require that the coupling and probe lasers be orthogonally polarized. The experiment was repeated with these lasers linearly polarized in the same direction. No transmitted probe signal was observed.

We have also observed a change in the group velocity of the probe pulse, depending on whether or not the coupling laser was present. For the conditions of Figs. 3 and 4, the relative delay of the probe pulse with respect to the coupling laser pulse was 2.5 to 3.0 nsec in the 9-cm zone length.

The solid curves in Figs. 3 and 4 show the calculated transmission as a function of probe frequency. These curves are obtained from Eq. (1) of Ref. [1], which was in turn obtained by solution of the density matrix equations with the introduction of macroscopic linewidths γ_{21}

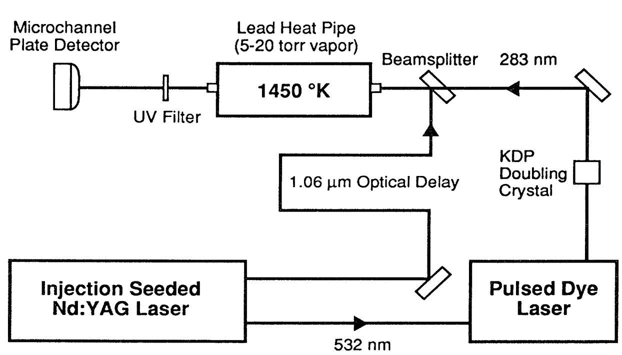

FIG. 2. Experimental apparatus.

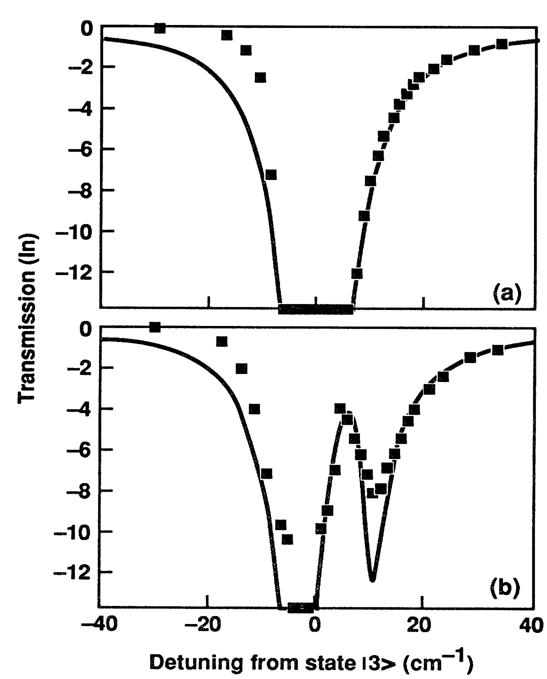

FIG. 3. Transmission vs probe laser detuning at 15 torr ($N = 1.0 \times 10^{17}$ atoms/cm^3) for (a) $\Omega_{23} = 0$ and (b) $\Omega_{23} = 8.5\ \mathrm{cm}^{-1}$. The solid curve is the fitted transmission spectrum.

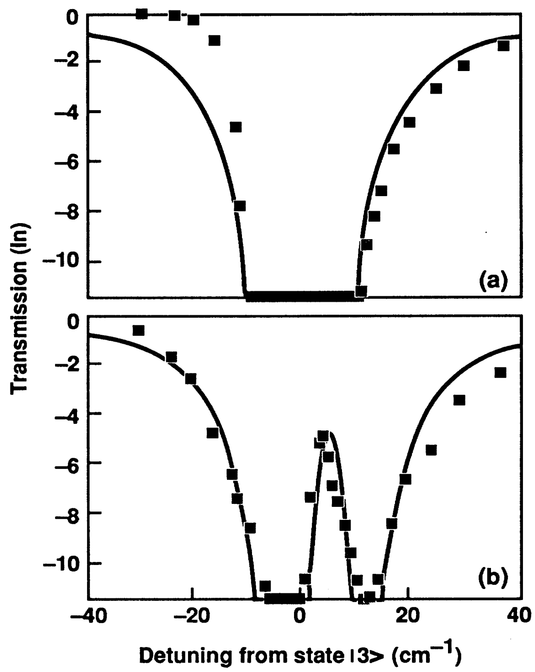

FIG. 4. Transmission vs probe laser detuning at 18 torr ($N=1.2\times10^{17}$ atoms/cm^3) for (a) $\Omega_{23}=0$ and (b) $\Omega_{23}=10.1$ cm^{-1}. The solid curve is the fitted transmission spectrum.

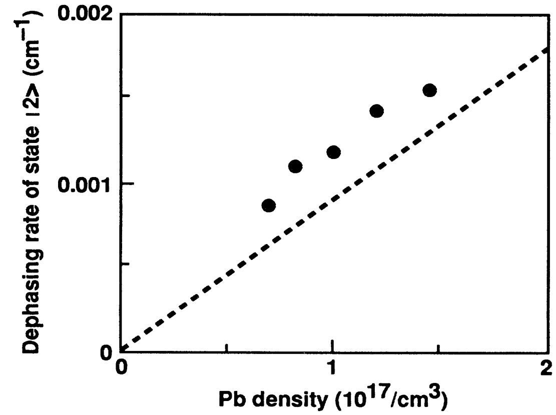

FIG. 5. State $|2\rangle$ dephasing rate γ_{21} vs Pb density. The data points are obtained by curve fitting to measured transmission profiles. The dashed curve is calculated.

and γ_{31} for the $|1\rangle$-$|2\rangle$ and $|1\rangle$-$|3\rangle$ transitions. For Fig. 4, the calculated peak absorption cross section is $\sigma_0=2.8\times10^{-12}$ cm^2, and the detuning of the 1064-nm coupling laser, corrected for ac-Stark shifts as caused by other levels, is $\Delta\omega_c=-4.5$ cm^{-1}. The linewidths γ_{21} and γ_{31} as well as the coupling laser Rabi frequency Ω_{32} are chosen to attain a best fit to the experimental data. For Fig. 3, we use $\gamma_{31}=0.0175$ cm^{-1}, $\gamma_{21}=0.0012$ cm^{-1}, $\Omega_{23}=8.5$ cm^{-1}; and for Fig. 4 we use $\gamma_{31}=0.0216$ cm^{-1}, $\gamma_{21}=0.00147$ cm^{-1}, $\Omega_{23}=10.1$ cm^{-1}. These parameters are each within a factor of 2 of the theoretically predicted values. The theoretical curves are also averaged over the 2-cm^{-1} linewidth of the probe laser assuming a Gaussian line shape. In Figs. 3 and 4, a larger absorption is predicted at the Autler-Townes sidebands than is actually observed. The spectral tails of the multimode probe laser are probably larger than that of a Gaussian; since the absorption peaks are very sharp, such an effect would explain the observed discrepancy.

In these experiments, the Rabi frequency of the coupling laser is very large compared to the linewidths γ_{21} and γ_{31}. In this case, Eq. (1) of Ref. [1] evaluated near the point of maximum transparency ($\Delta\omega_p=\Delta\omega_c$) is

$$\frac{\sigma}{\sigma_0}=4\frac{\gamma_{21}\gamma_{31}}{\Omega_{23}^2}+16\frac{(\Delta\omega_p-\Delta\omega_c)^2\gamma_{31}^2}{\Omega_{23}^4}\,,\qquad \sigma_0=\frac{1}{\hbar}\left(\frac{\mu_0}{\epsilon_0}\right)^{1/2}\frac{\omega_{31}|\mu_{31}|^2}{\gamma_{31}}\,, \tag{1}$$

where σ_0 is the peak absorption cross section of the $|1\rangle$-$|3\rangle$ transition in the absence of the coupling laser; $\Delta\omega_p=\omega_3-\omega_p-\omega_1$ and $\Delta\omega_c=\omega_3-\omega_c-\omega_2$ are the detunings of the probe and coupling lasers; $\omega_{31}=\omega_3-\omega_1$, and Ω_{23} is the Rabi frequency of the coupling laser.

The maximum transparency is thus determined by the linewidth γ_{21} of the $|1\rangle$-$|2\rangle$ transition and not by the (much broader) linewidth of the $|1\rangle$-$|3\rangle$ transition. To confirm this dependence, data were taken at each of several pressures, and as above, best fitted with the transmission profiles to obtain a value for γ_{21}. The experimental points in Fig. 5 are the result of this fitting procedure. The dashed line shows the calculated [17] value of γ_{21}.

In considering these results and in comparing them to those of Ref. [1], we first note that though the $|1\rangle$-$|3\rangle$ resonance transition is collision (instead of lifetime) broadened, the very large increase in transparency which is caused by the 1064-nm coupling laser is the result of an interference and not of simply the ac-Stark splitting of the resonance transition. If the transition were simply Stark split and (hypothetically) there were no interference between the two dressed states, then the calculated transmission at the transparency point $\Delta\omega_p=\Delta\omega_c$ for Fig. 3(b) would be $\exp(-36)$ and would not be observable.

There are problems with the use of the macroscopic density-matrix treatment. Resonance line collisions as caused by perturber Pb atoms have a characteristic impact time of about 20 ps, and this time is much greater than the reciprocal of Ω_{23}. In effect, the detuning of the probe frequency (when at the point of maximum transparency) is therefore too large for an impact-theory calculation. Because of the 6-cm^{-1} detuning of the 1064-nm laser from the $|2\rangle$-$|3\rangle$ transition frequency, this large Rabi frequency is required, and it is not possible to test Eq. (1) of Ref. [1] in the impact regime. The reasonable agreement of theory and experiment in Figs. 3, 4, and 5 is a result of the insensitivity to γ_{31}, or possibly, to the choice of fitting parameters. A complete treatment of this problem will have to include the effect of the coupling laser on the collision cross sections [18]. The issue is further complicated by the long-wavelength skewing of the Pb resonance transition. This skewing is probably also responsible for a discrepancy of a factor of 3 between the calculated and the measured group velocity.

In summary, we have reported the first observation of an electromagnetically induced transparency on a collisionally broadened resonance transition. Maximum transmission is shown to be determined by the smaller linewidth of a coupled nonresonantly broadened transition. As noted earlier [19], atoms which are excited to state $|2\rangle$ do not exhibit a destructive interference in their emission profile. Therefore, lasing may occur even though the sum of the population of states $|2\rangle$ and $|3\rangle$ is less than that of state $|1\rangle$. Because of easier pumping, pressure-broadened systems such as described here may be better candidates for inversion-free lasers than are lifetime-broadened systems. We also anticipate application to nonlinear optical processes [20,21]. In a recent experiment, Hakuta, Marmet, and Stoicheff, [22], working with atomic hydrogen, have demonstrated both reduced loss and a resonantly enhanced nonlinear susceptibility.

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