PHYSICAL REVIEW A VOLUME 59, NUMBER 5 MAY 1999

Electromagnetically induced transparency with spectator momenta

Hui Xia, A. J. Merriam, S. J. Sharpe, G. Y. Yin, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 18 May 1998)

We describe a method and present experimental results for obtaining electromagnetically induced transparency by adjusting the frequencies of two lasers so that they coincide with the centers of gravity of two hyperfine split transitions. No hyperfine states are in Raman resonance and the zero of the dipole moment results from the interference of the manifold of cooperating *m* states. [S1050-2947(99)50805-6]

PACS number(s): 42.50.Gy, 32.80.Qk, 33.15.Pw, 42.65.An,

Electromagnetically induced transparency (EIT) enables transparent and distortion-free propagation of laser beams through an otherwise absorbing and refracting medium. The prototype situation for EIT occurs in an ideal three-state atom. Here, two lower states, $|1\rangle$ and $|2\rangle$, are coupled by laser frequencies ω_p and ω_c to a common state $|3\rangle$ [Fig. 1(a)]. The three-state system is then optically pumped (cw lasers) or adiabatically evolved (pulsed lasers) into a superposition state that has zero probability amplitude in state $|3\rangle$. In the ideal case of stationary atoms and no collisions, a pair of matched pulses will then propagate as in vacuum [1–3].

But, most often, atoms have additional angular momenta that enlarge the manifold of coupled states, but that are largely unaffected by the application of optical-frequency laser fields. Such additional momenta are termed ''spectators,'' and their interaction with other angular momenta causes additional energy-level splittings. The example of this work is ^{207}Pb, where a nuclear spin of $I=1/2$ splits the fine-structure levels of nuclear spin-free ^{208}Pb into their hyperfine components (Fig. 1). For sufficiently small opacity ($N\sigma L \ll 1$), one can often use light of appropriate polarization and tune to a three-state subset of the hyperfine states to obtain transparency. At higher opacities, however, even though there may be near complete energy transmission, other m_F states of the system can significantly affect the phase, resulting in severe wave-front distortion. Such phase distortion will greatly reduce the effectiveness of EIT for both nonlinear optics and measurement.

This Rapid Communication reports experimental results demonstrating EIT and phase protection in such a hyperfine system at high opacities, using the technique suggested by Xia *et al.* [4]. In the presence of spectator angular momenta, rather than tune the lasers to a three-state Raman-like subset of the multistate system, one should instead tune the lasers to the center of gravity of the splittings. This center of gravity is determined by both the transition matrix elements and frequency splittings and always coincides with the positions of the nonsplit transitions [5].

This work is, in part, based on the establishment of coherence among more than three cooperating nondegenerate *m* states. Prior to this work, Ling *et al.* [6] and Milner and Prior [7] have shown how degenerate *m* states may cooperate to form a population-trapped state. The general conditions for obtaining populating trapping in multistate systems have also been given by Hioe and Carroll [8].

In the ^{207}Pb ground level, atoms are in either of the degenerate states $m_F=\pm 1/2$ (Fig. 2). We take the probe frequency ω_p to have polarization σ_+ and the coupling laser ω_c to have polarization σ_-. As in [4], we denote Ω_p and Ω_c as the Rabi frequencies of the probe and coupling lasers of these same polarizations in ^{208}Pb. Using the *m*-state selection rules, one sees that atoms in the $m_F=+1/2$ state evolve as a three-component eigenvector, while those in the $m_F=-1/2$ state evolve (independently) as a five-component eigenvector. If the probe and coupling lasers are tuned to the center of gravity of both transitions (see Fig. 1), then, in the interaction picture, to first order in perturbation theory, and with $\Omega_c \gg A_2$, these eigenvectors are

$$|+\rangle=\frac{1}{\Omega_s}\left(-\Omega_c|1\rangle+\Omega_p|2\rangle+\frac{2A_2\Omega_c\Omega_p}{\Omega_s^2}|3\rangle\right),$$

$$|-\rangle=\frac{1}{\Omega_s}\left(-\Omega_c|1\rangle+\frac{2}{\sqrt{5}}\Omega_p|2\rangle+\frac{1}{\sqrt{5}}\Omega_p|3\rangle - \sqrt{\frac{8}{3}}\frac{A_2\Omega_p}{\Omega_c}\frac{\Omega_p^2+2\Omega_c^2}{\Omega_s^2}|4\rangle + \frac{2}{\sqrt{3}}\frac{A_2\Omega_p}{\Omega_c}\frac{2\Omega_p^2+\Omega_c^2}{\Omega_s^2}|5\rangle\right). \quad (1)$$

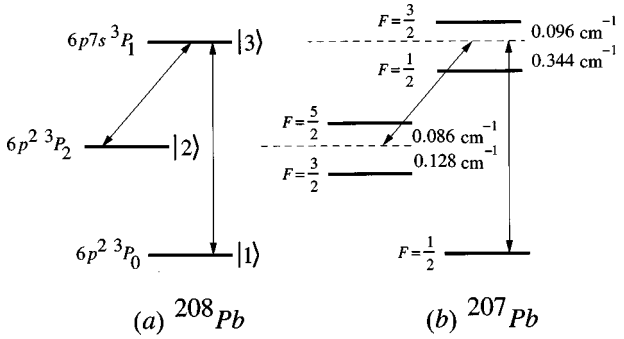

FIG. 1. Energy schematic for (a) ^{208}Pb and (b) ^{207}Pb. The nuclear spin of ^{208}Pb is zero and there is no hyperfine splitting. Neglecting isotopic shifts, the center of gravity of the hyperfine split levels of ^{207}Pb coincides with the level frequencies of ^{208}Pb.

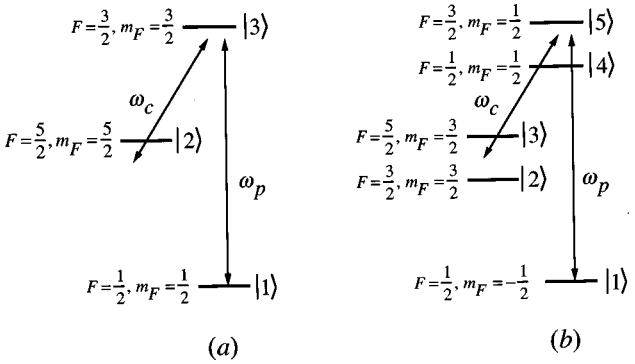

FIG. 2. In ^{207}Pb, when the probe laser ω_p has polarization σ_+ and the coupling laser ω_c has polarization σ_-, (a) the $m_F=1/2$ state of ground level is coupled with two other states, and (b) the $m_F=-1/2$ state is coupled with four other states. The coupled states are numbered in ascending order.

Here, $|+\rangle$ denotes that eigenvector that has evolved from the $m_F=+1/2$ ground state, and $|-\rangle$ denotes the eigenvector that has evolved from the $m_F=-1/2$ eigenstate. The quantity A_2 is the (J-dependent) hyperfine interaction strength of level $6p^2\ ^3P_2$ [5], and $\Omega_s=(\Omega_p^2+\Omega_c^2)^{1/2}$.

The dipole moment operators of those atoms that have evolved from the $m_F=\pm 1/2$ states are

$$P_+=\mu_p e^{-j\omega_p t}|1\rangle\langle 3|+\mu_c e^{-j\omega_c t}|2\rangle\langle 3|+\text{H.c.},$$

$$P_-=\mu_p e^{-j\omega_p t}\left(\sqrt{\frac{2}{3}}|1\rangle\langle 4|+\frac{1}{\sqrt{3}}|1\rangle\langle 5|\right)$$

$$+\mu_c e^{-j\omega_c t}\left(\sqrt{\frac{5}{6}}|2\rangle\langle 4|+\frac{1}{\sqrt{15}}|2\rangle\langle 5|+\sqrt{\frac{3}{5}}|3\rangle\langle 5|\right)$$

$$+\text{H.c.} \qquad (2)$$

The numerical factors that appear in Eq. (2) are obtained from Racah algebra, with the quantities μ_p and μ_c defined as the dipole matrix elements for light of the same circular polarization acting on the respective (unsplit) transitions of ^{208}Pb.

The expected value of the dipole moment operator for each class of atoms is

$$\langle +|P_+|+\rangle=\frac{2A_2\Omega_p\Omega_c}{\Omega_s^3}\left(-\frac{\Omega_c}{\Omega_s}\mu_p e^{-j\omega_p t}+\frac{\Omega_p}{\Omega_s}\mu_c e^{-j\omega_c t}\right)$$

$$+\text{c.c.}, \qquad (3)$$

$$\langle -|P_-|-\rangle=-\langle +|P_+|+\rangle.$$

Therefore, both classes of atoms have a first-order cancellation of the imaginary part of their dipole moments (absorption), but have finite real parts. If the initial populations are equal, then the sum of the dipole moments results in a (first-order) cancellation of the real parts, and therefore a near-unity refractive index. (In Ref. [4], with linear polarization both the real and imaginary cancellations occur atom by atom. Here the cancellation is macroscopic.)

In our experiments, we use Pb with an isotopic mixture of 92% ^{207}Pb, 6% ^{208}Pb, and 2% ^{206}Pb (Oak Ridge National Laboratory). The cell is a 10-cm-long sealed fused-silica sidearm cell, which, depending on its temperature, operates at an atom density varying between 10^{12} atoms/cm^3 and 5×10^{14} atoms/cm^3. The sidearm cell is not magnetically shielded. As in earlier work [9], the 406- and 283-nm laser beams are obtained from independent frequency-doubled and -tripled single longitudinal mode Ti:sapphire laser systems operating near 812 and 850 nm, respectively. Because the center of gravity of the hyperfine split levels of ^{207}Pb coincides with the nonsplit level frequencies of ^{208}Pb, we use another sidearm cell filled with 99.9% isotopically pure ^{208}Pb as a frequency reference. By tuning the probe laser to the center of the transition $6p^2\ ^3P_0\rightarrow 6p7s\ ^3P_1$ of the pure ^{208}Pb and tuning the coupling laser to maximize the EIT effect in the reference cell, we are able to tune both lasers to the respective centers of gravity of the two transitions of ^{207}Pb. (This technique ignores the isotopic shifts in ^{207}Pb, which change the position of the center of gravity by 4×10^{-4} cm^{-1} for the $6p^2\ ^3P_2\rightarrow 6p7s\ ^3P_1$ transition and 0.05 cm^{-1} for the $6p^2\ ^3P_0\rightarrow 6p7s\ ^3P_1$ transition.)

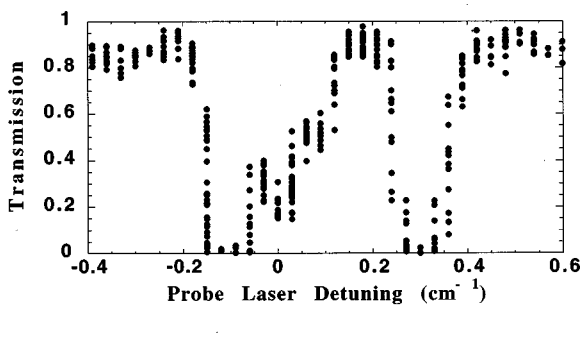

FIG. 3. Absorption profile of a weak probe laser (0.3 W/cm^2) of 283 nm tuned near the $6p^2\ ^3P_0\rightarrow 6p7s\ ^3P_1$ transition of ^{207}Pb. The cell contains 92% ^{207}Pb, 6% ^{208}Pb, and 2% ^{206}Pb, at a density of 1×10^{12} atoms/cm^3. At this low density, the hyperfine transitions $F=1/2\rightarrow1/2$ and $F=1/2\rightarrow3/2$ are distinct.

The probe and coupling lasers have pulse durations of about 19 and 32 ns, respectively. The beams are focused into the center of the cell and have beam diameters of 0.24 and 2.14 mm. The probe beam is centered on the central portion of the larger coupling laser beam. The timing of the lasers is set so that the coupling laser enters the cell before the probe and is present until after the probe has exited the cell. All data points in this work are from individual pulses and are not averaged.

Figure 3 shows the probe transmission as a function of the probe frequency with no coupling laser present. The spontaneous decay time of each m_F state of the $6p7s\ ^3P_1$ level is 5.2 ns. The probe has a Rabi frequency of 6×10^{-4} cm^{-1} (0.018 GHz) and the cell is operated at a density of 1×10^{12} atoms/cm^3. At this low density, the absorption at the probe frequency of the individual peaks ($F=1/2\rightarrow1/2$ and $F=1/2\rightarrow3/2$) of the hyperfine split levels [Fig. 1(b)] is observable. The absorption dip at the zero detuning is due to the presence of 6% ^{208}Pb and 2% ^{206}Pb in the cell.

In Fig. 4(a) the density is increased to 5×10^{14} atoms/cm^3. Here, the absorption of the hyperfine components is merged and is not observable. At this density with a cell temperature of 800 °C (Doppler width=0.06 cm^{-1} for the $6p^2\ ^3P_0\rightarrow 6p7s\ ^3P_1$ transition), the transmission at the

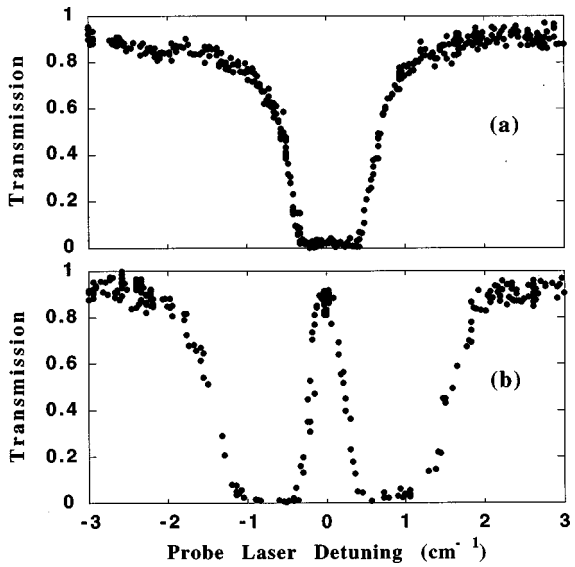

FIG. 4. Transmission vs probe laser frequency in ^{207}Pb vapor. The atom density is 5×10^{14} atoms/cm^3. (a) Probe alone, no coupling laser present. At this density, the transitions of Fig. 3 are merged. (b) A coupling laser with Rabi frequency 1.7 cm^{-1} is applied and tuned to the center of gravity of the $6p^2\ ^3P_2\rightarrow 6p7s\ ^3P_1$ transition, and the Pb cell becomes nearly transparent at line center.

resonance of the $F=1/2\rightarrow 1/2$ transition is calculated to be $\exp(-4100)$ and that of the $F=1/2\rightarrow 3/2$ transition to be $\exp(-8200)$.

Figure 4(b) shows the transmission of the probe laser beam as a function of the detuning from the center of gravity of the $6p^2\ ^3P_0\rightarrow 6p7s\ ^3P_1$ transition when a coupling laser is applied with a Rabi frequency of 1.7 cm^{-1} (51 GHz) and tuned to the center of gravity of the transition $6p^2\ ^3P_2 \rightarrow 6p7s\ ^3P_1$. The observed transmission of the probe laser, when tuned directly to the center of gravity, increases from near zero to about 90%.

In Fig. 5 we compare the transmission of ^{207}Pb ($I=1/2$) to that of ^{208}Pb ($I=0$) as a function of the coupling laser Rabi frequency. In this figure, both lasers are tuned to the respective centers of gravity of each transition and the intensity of

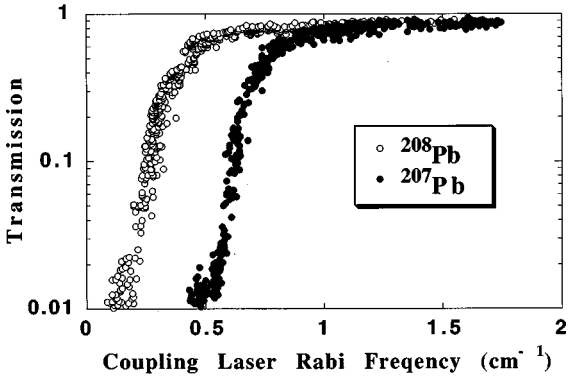

FIG. 5. Transmission of the probe laser as a function of the coupling laser Rabi frequency for ^{208}Pb and ^{207}Pb. The probe and coupling lasers are tuned to the center of gravity of the respective transitions. The Pb vapor atom density is $N=5\times10^{14}$ atoms/cm^3 in both cases.

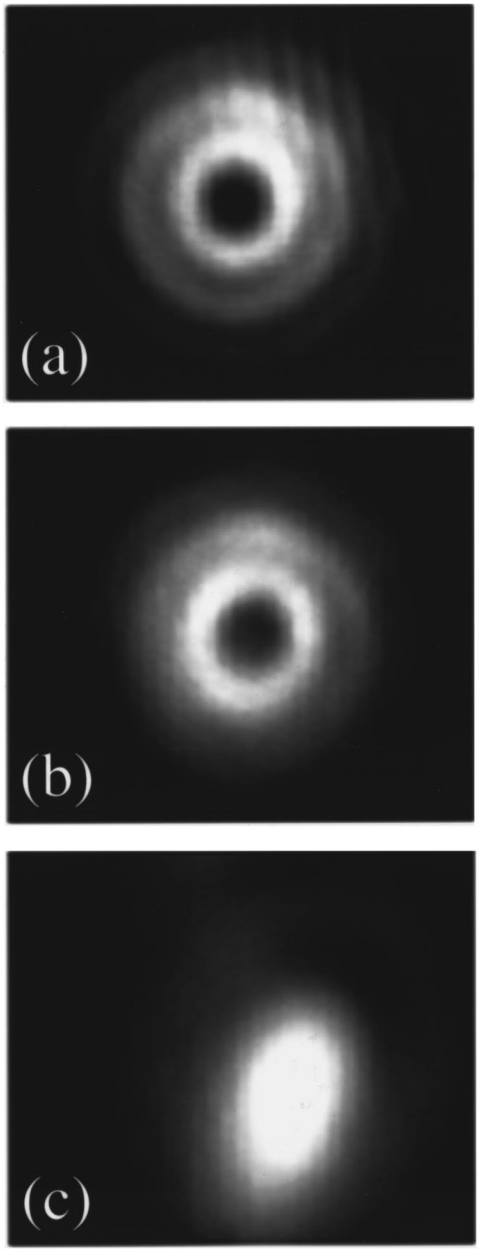

FIG. 6. Images of the diffraction patterns of the probe beam after passage through the ^{207}Pb cell. (a) Probe alone in a cold cell. (b) ^{207}Pb density$=5\times10^{14}$ atoms/cm^3 with a coupling Rabi frequency of 1.7 cm^{-1}, and both lasers tuned to respective centers of gravity of the transitions. (c) The same condition as in (b), except that the two lasers are tuned to the Raman resonance of a three-state subsystem (coupling laser resonant with the transition $6p^2\ ^3P_2$, $F=5/2\rightarrow 6p7s\ ^3P_1$, $F=3/2$; probe laser resonant with the $6p^2\ ^3P_0$, $F=1/2\rightarrow 6p7s\ ^3P_1$, $F=3/2$ transition). [In (a), (b), and (c), the Rabi frequency of the probe laser is 0.1 cm^{-1}.]

the coupling laser is varied. We find, in agreement with numerical calculations, that the hyperfine split ^{207}Pb requires a somewhat higher Rabi frequency to obtain transparency. In particular, in ^{207}Pb, under these experimental conditions, transparency is not observed until the coupling laser Rabi frequency is several times the value of the hyperfine interaction strength A_2, which is on the order of 0.1 cm^{-1} for ^{207}Pb.

Figure 6 shows how EIT may result in near-diffraction-

limited propagation in otherwise opaque ^{207}Pb. The geometry, with imaging of the focal plane onto a charge-coupled device (CCD) camera, is similar to that of the experiments of Kasapi *et al.* [9]. The probe beam passes through a 2-mm-diam aperture, travels 34 cm to a 1-m lens, and is focused at the cell center. The position of the CCD camera is chosen such that the central portion of the (Airy disk) diffraction pattern for free-space propagation is dark [Fig. 6(a)]. The coupling laser, at beam center, has a Rabi frequency of 1.7 cm^{-1} and, as a result of its spatially varying intensity, acts as a spatial filter for the probe beam. We observe that, with the lasers tuned to the respective centers of gravity of each transition, the diffraction pattern [Fig. 6(b) is nearly the same as that for free-space propagation. If the lasers are tuned to the Raman resonance of a three-state subsystem [Fig. 6(c)], though there is still reasonable energy transmission, there is substantial absolute and differential phase accumulation on the probe beam and the diffraction pattern is badly distorted.

In summary, we have presented experimental results demonstrating transparency and high-quality beam transmission in atoms with hyperfine structure. This is done by tuning the frequencies of two lasers to coincide with the centers of gravity of the hyperfine split transitions. Due to the high cost of obtaining nuclear spin-free isotopes ($5000/g for ^{208}Pb), this result is important for applications of EIT that require heat-pipe technology. One example is high-efficiency frequency conversion [10]. In this work we have used two circularly polarized beams. But we have shown analytically that, irrespective of optical polarization, the center-of-gravity method described in the previous paragraphs remains valid in more general cases with any value of nuclear spin, subject to the conditions that the ground level of the atom has $J=0$ and that all of the m_F hyperfine states are equally populated. This method of tuning to the centroid rather than to discrete sublevels should also apply, but at a higher coupling laser intensity, to *LS* coupling in light atoms and, perhaps, to special situations in molecules.

The authors gratefully acknowledge helpful discussions with Alexei Sokolov. This work was supported by the U.S. Office of Naval Research, the U.S. Air Force Office of Scientific Research, and the U.S. Army Research Office.

[1] S. E. Harris, Phys. Today **50** (7), 36 (1997).

[2] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, London, 1997).

[3] E. Arimondo, in *Progress in Optics*, edited by E. Wolf (Elsevier Science, Amsterdam, 1996), pp. 257–354.

[4] H. Xia, S. J. Sharpe, A. J. Merriam, and S. E. Harris, Phys. Rev. A **56**, R3362 (1997).

[5] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California, Berkeley, 1981).

[6] H. Y. Ling, Y. Q. Li, and M. Xiao, Phys. Rev. A **53**, 1014 (1996).

[7] V. Milner and Y. Prior, Phys. Rev. Lett. **80**, 940 (1998).

[8] F. T. Hioe and C. E. Carroll, Phys. Rev. A **37**, 3000 (1988).

[9] A. Kasapi, M. Jain, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **74**, 2447 (1995); A. Kasapi *ibid.* **77**, 1035 (1996).

[10] M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, Phys. Rev. Lett. **77**, 4326 (1996).