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Electromagnetically induced transparency in atoms with hyperfine structure

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We describe a method for obtaining population trapping and electromagnetically induced transparency in atoms with hyperfine structure and where, therefore, more than three states are coupled by two applied fields. [S1050-2947(97)50411-2]

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Electromagnetically induced transparency (EIT) is a technique for improving the transmission of laser beams through otherwise absorbing and refracting media. Most often, this is done in an atom that has three states that, at the density-length product that is to be made transparent, are resolvable. One applies two laser fields and uses either optical pumping (cw lasers) or adiabatic evolution (pulsed lasers) to produce a coherent superposition of the probability amplitudes of two states of the same parity, with a near-zero probability amplitude of a third state of opposite parity. Once in this superposition state, the atom does not interact with either of the applied fields [1–3]. Figure 1(a) shows an example of a prototype three-state system. Because the nuclear spin of ^{208}Pb is zero with linearly polarized light, the $J=0$ and $J=2$ fine-structure states of the $6p^2$ configuration, together with the $J=1$ state of the $6p7s$ configuration, form the necessary superposition state [4].

Figure 1(b) shows an energy schematic of ^{207}Pb. Here, the nuclear spin of $I=1/2$ causes the fine-structure states to split into their hyperfine components, with splittings of a few tenths of a cm^{-1}. Although at low atom density-length products (optically thin), the individual hyperfine components of the $|1\rangle\rightarrow|4\rangle$ and $|1\rangle\rightarrow|5\rangle$ transitions are resolvable, in a dense medium, they are not. Even though a particular three-state subsystem may be made transparent, other subsystems will produce a prohibitively large change in the complex refractive index, and the medium will be opaque. The question then arises: How should the lasers be tuned to render an optically thick, hyperfine-split system such as ^{207}Pb both transparent and nonrefracting?

In this paper we show that transparency and a refractive index near unity are attained when the two lasers are tuned to the respective center of gravity of the two transitions. No specific hyperfine state is in Raman resonance and the zero in the real part of the dipole moment results from the interference of the manifold of cooperating m states. At practical Rabi frequencies, the detuning from the individual Stark-shifted m states is sufficiently large that the nonzero imaginary part of the dipole moment is not of consequence. We also find, as is the case for the pure three-state system, that self-focusing and defocusing may be eliminated [5].

In related previous work, Ling *et al.* [6] and Milner *et al.* [7] have shown how degenerate m states may cooperate to form a population-trapped state. This work is similar to theirs in that we use a multistate interference. It differs from theirs in that the m_f states that we use are not degenerate [Fig. 1(b)]. General conditions for obtaining population trapping in multistate systems have also been given by Hioe and Carrol [8].

We begin the analysis with two assumptions that reduce the 18×18 Hamiltonian for ^{207}Pb to dimension 5×5. The first assumption is that the probe and coupling fields are linearly polarized in the $\overline{a_z}$ direction. The second is that there are no external magnetic fields so that the $m=\pm1/2$ spin states evolve similarly. For convenience, we assume, at $t=0$, that all of the ground-state atoms are in the $m=-1/2$ state. The five interacting m states are labeled in ascending numerical order [Fig. 1(b)].

It is the interrelation of the hyperfine splittings, and the matrix elements of ^{207}Pb, both determined by Racah algebra, which lead to the result of this work. We reference the frequencies of the hyperfine levels to the (nonsplit) fine-structure levels [Fig. 1(b)] of ^{208}Pb, or equivalently, to the center of gravity of the hyperfine levels of ^{207}Pb:

$$\delta\omega_i=\frac{A_J}{2}\left[F(F+1)-J(J+1)-I(I+1)\right]. \tag{1}$$

The quantity A_J is the (J dependent) hyperfine interaction strength [9]. For ^{207}Pb, $A_0=0$, $A_2=0.06$ cm^{-1}, and $A_1=0.2$ cm^{-1} [10].

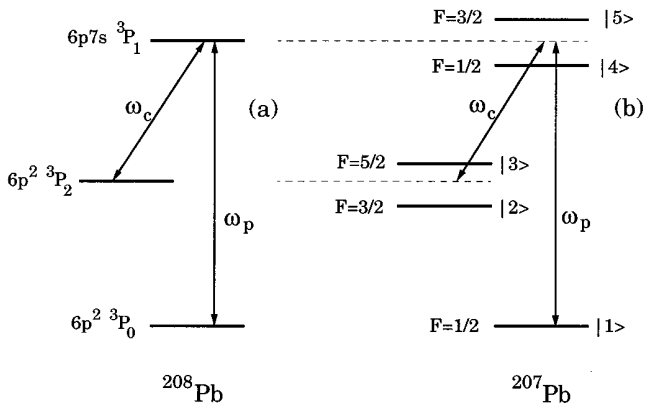

FIG. 1. Energy schematic for (a) ^{208}Pb and (b) ^{207}Pb. The nuclear spin of ^{208}Pb is zero and there is no hyperfine splitting. The center of gravity of the hyperfine split levels of ^{207}Pb coincides with the level frequencies of ^{208}Pb. As shown, the states of ^{207}Pb are numbered in ascending order.

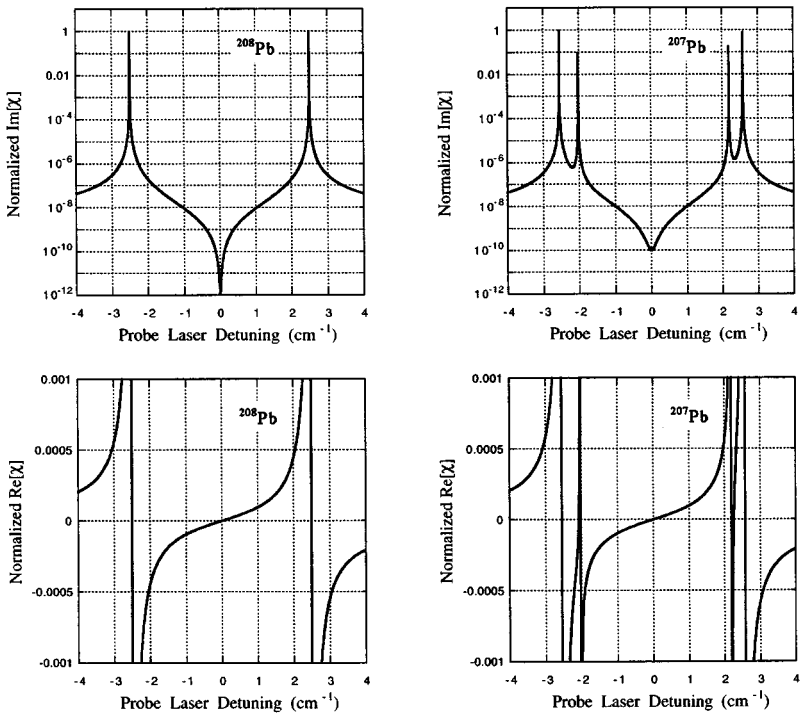

FIG. 2. Linear susceptibilities for a probe frequency ω_p in ^{208}Pb and ^{207}Pb. In ^{208}Pb the imaginary part of the susceptibility is zero at line center; in ^{207}Pb, because of the hyperfine splitting, it is small but finite. The real part of the susceptibilities, especially near line center, are very similar.

The Rabi frequencies of the probe and coupling lasers in ^{208}Pb are denoted by Ω_p and Ω_c, and we define the Rabi frequencies for ^{207}Pb in terms of these quantities. This is done so that, at large Rabi frequencies, the susceptibilities for ^{207}Pb will reduce explicitly to those of ^{208}Pb. Equivalently, only two independent matrix elements enter the problem. For ^{208}Pb these are defined as μ_p and μ_c. Using the appropriate line factors and $3-j$ symbols, the matrix elements for ^{207}Pb, with all atoms in the $m_F=-1/2$ state, are

$$\mu_{14}=\frac{1}{\sqrt{3}}\,\mu_p, \quad \mu_{15}=\sqrt{2/3}\mu_p, \quad \mu_{24}=\sqrt{5/6}\mu_c, \tag{2}$$

$$\mu_{25}=\frac{1}{2\sqrt{15}}\,\mu_c, \quad \mu_{34}=0, \quad \mu_{35}=\frac{3}{\sqrt{10}}\,\mu_c.$$

All other matrix elements are zero.

The detunings $\Delta\omega_p$ and $\Delta\omega_c$ of the probe and coupling lasers are also referenced to the center of gravity of the levels of ^{207}Pb. With the definitions of $\delta\omega_i$ of Eq. (1), these detunings are

$$\Delta\omega_p=(\omega_5-\delta\omega_5)-\omega_p, \qquad \Delta\omega_c=(\omega_3-\delta\omega_3)-(\omega_p-\omega_c). \tag{3}$$

The quantity $(\omega_5-\delta\omega_5)=(\omega_4-\delta\omega_4)$ is the frequency of the center of gravity of the $6p7s\ ^3P_1$ level. Similarly, $(\omega_3-\delta\omega_3)=(\omega_2-\delta\omega_2)$ is the center of gravity of the $6p^2\ ^3P_2$ level. We allow for the same Einstein A decay rate Γ of the hyperfine states $|4\rangle$ and $|5\rangle$ and also for a common decay or broadening rate γ from states $|2\rangle$ and $|3\rangle$, and define complex detunings as $\Delta\tilde{\omega}_p=\Delta\omega_p-j\Gamma/2$ and $\Delta\tilde{\omega}_c=\Delta\omega_c-j\gamma/2$.

The Hamiltonian is divided into a portion H_0, which is treated exactly, and a perturbation $H_{\rm pert}$. With the previous definitions and working in the interaction picture, H_0 and $H_{\rm pert}$ are

$$H_0=-\frac{1}{2}\begin{bmatrix} 0 & 0 & 0 & \frac{1}{\sqrt{3}}\,\Omega_p & \sqrt{\frac{2}{3}}\,\Omega_p \\ 0 & 0 & 0 & \sqrt{\frac{5}{6}}\,\Omega_c & \frac{1}{2\sqrt{15}}\,\Omega_c \\ 0 & 0 & 0 & 0 & \frac{3}{\sqrt{10}}\,\Omega_c \\ \frac{1}{\sqrt{3}}\,\Omega_p & \sqrt{\frac{5}{6}}\,\Omega_c & 0 & 0 & 0 \\ \sqrt{\frac{2}{3}}\,\Omega_p & \frac{1}{2\sqrt{15}}\,\Omega_c & \frac{3}{\sqrt{10}}\,\Omega_c & 0 & 0 \end{bmatrix}, \tag{4a}$$

$$H_{\text{pert}}=\begin{bmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & \Delta\tilde{\omega}_c-\frac{3A_2}{2} & 0 & 0 & 0 \\ 0 & 0 & \Delta\tilde{\omega}_c+A_2 & 0 & 0 \\ 0 & 0 & 0 & \Delta\tilde{\omega}_p-A_1 & 0 \\ 0 & 0 & 0 & 0 & \Delta\tilde{\omega}_p+\frac{A_1}{2} \end{bmatrix}. \tag{4b}$$

In this same basis, the dipole moment operator is

$$P=\begin{bmatrix} 0 & 0 & 0 & \frac{1}{\sqrt{3}}e^{-j\omega_p t}\mu_p & \sqrt{\frac{2}{3}}e^{-j\omega_p t}\mu_p \\ 0 & 0 & 0 & \sqrt{\frac{5}{6}}e^{-j\omega_c t}\mu_c & \frac{1}{2\sqrt{15}}e^{-j\omega_c t}\mu_c \\ 0 & 0 & 0 & 0 & \frac{3}{\sqrt{10}}e^{-j\omega_c t}\mu_c \\ \frac{1}{\sqrt{3}}e^{j\omega_p t}\mu_p & \sqrt{\frac{5}{6}}e^{j\omega_c t}\mu_c & 0 & 0 & 0 \\ \sqrt{\frac{2}{3}}e^{j\omega_p t}\mu_p & \frac{1}{2\sqrt{15}}e^{j\omega_c t}\mu_c & \frac{3}{\sqrt{10}}e^{j\omega_c t}\mu_c & 0 & 0 \end{bmatrix}. \tag{5}$$

The procedure of the following paragraphs is to find the eigenvector of the total Hamiltonian, which is analogous to the population-trapped eigenvector of the three-state system, and then to use the dipole moment operator of Eq. (5) to obtain the susceptibility at the probe frequency. We proceed first by perturbation and then numerically.

The eigenvalues and eigenvectors of H_0 are readily found algebraically. The population trapped eigenvector with zero eigenvalue is

$$|u_0^{(0)}\rangle=\frac{1}{\Omega_s}\left[\Omega_c,-\sqrt{\frac{2}{5}}\,\Omega_p,-\sqrt{\frac{3}{5}}\,\Omega_p,0,0\right], \tag{6}$$

where $\Omega_s=(\Omega_p^2+\Omega_c^2)^{1/2}$. In the same sense as in a three-state system the components of states $|1\rangle$, $|2\rangle$, and $|3\rangle$ cooperate to cause a zero probability amplitude of states $|4\rangle$ and $|5\rangle$. Also, as in the three-state system, when Ω_p is zero, this eigenvector coincides with the $(m_F=-1/2)$ ground state of the atom and may evolve adiabatically from this state. Since, here, the hyperfine structure is not yet included, the dipole moment that is obtained from this eigenvector is, as for ^{208}Pb, zero.

Including H_{pert}, to first order, the perturbed eigenvalue is

$$\lambda_0^{(1)}=\frac{\Omega_p^2}{\Omega_s^2}\,\Delta\tilde{\omega}_c. \tag{7}$$

When the Raman detuning is set to the center of gravity of the metastable transition, i.e., $\Delta\omega_c=0$, and with no dephasing of this transition, the quantity $\Delta\tilde{\omega}_c$ and the population-trapped eigenvalue are zero. Because of the balance of matrix elements and detunings, the hyperfine structure does not, in first order, cause a nonzero eigenvalue.

With $\Delta\tilde{\omega}_c=0$, the first-order correction to the population-trapped eigenvector is

$$|u_0^{(1)}\rangle=\frac{4A_2\Omega_p}{\sqrt{3}\Omega_c\Omega_s}\left[0,0,0,-1,\frac{1}{\sqrt{2}}\right]. \tag{8}$$

Unlike the three-state case, this eigenvector has nonzero probability amplitude in states $|4\rangle$ and $|5\rangle$. But, because of the symmetry, these probability amplitudes are phased so that the dipole moment at both ω_p and ω_c is zero. This is also the case in second order and, to obtain a nonzero dipole moment with $\Delta\tilde{\omega}_c=0$, we go to the third order of perturbation. With the dipole moment given by $\langle u_0|P|u_0\rangle$, we obtain the normalized (third-order) small probe susceptibility for ^{207}Pb. With the detunings and decay rates taken to be small as compared to the hyperfine constants A_1 and A_2, the real and imaginary parts of the susceptibility are

$$\begin{aligned}\text{Re}[\chi]&=-\frac{4\Delta\omega_c}{\Omega_c^2}+\frac{16A_1A_2^2}{\Omega_c^4}-\frac{32A_2}{\Omega_c^4}(\Delta\omega_pA_2+\Delta\omega_cA_1),\\ \text{Im}[\chi]&=-\frac{2\gamma}{\Omega_c^2}-\frac{16A_2}{\Omega_c^4}(\Gamma A_2+\gamma A_1).\end{aligned} \tag{9}$$

With N equal to the atom density, the actual susceptibility is obtained by multiplying these quantities by $(|\mu_p|^2N/\epsilon_0\hbar)$; the complex propagation constant is obtained by multiplying the susceptibility by $(-\pi/\lambda_p)$. In the limit of no hyperfine structure, i.e., $A_1=A_2=0$, the susceptibility of ^{207}Pb reduces to that of ^{208}Pb [11]. It is because of a cancellation that was introduced by tuning to the center of gravity that the effect of hyperfine splitting reduces as the fourth power of Ω_c; conversely, the effect of the noncompensated linear detuning $\Delta\omega_c$ reduces only quadratically.

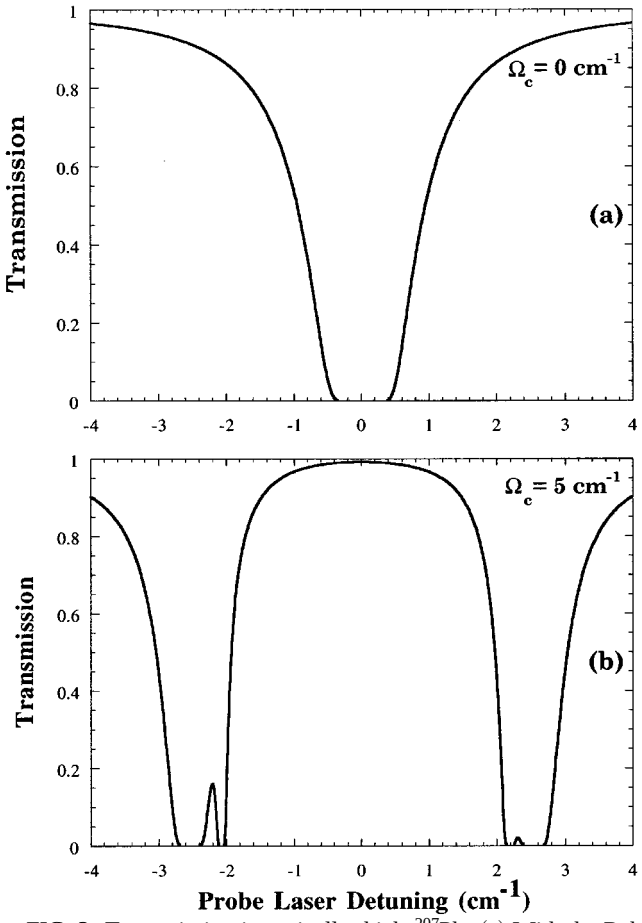

FIG. 3. Transmission in optically thick ^{207}Pb. (a) With the Rabi frequency of the coupling laser set to zero, the hyperfine components are masked and the power transmission at line center is $\exp(-10^5)$. (b) At $\Omega_c = 5\ \mathrm{cm}^{-1}$, transmission is restored. The parameters used here are an atom density of $2\times10^{15}\ \mathrm{cm}^{-3}$ and a cell length of 25 cm.

In Fig. 2 we compare the susceptibility of ^{207}Pb with ^{208}Pb. In either isotope, the total Einstein A decay rate of each m_F state of the $6s7p\ ^3P_1$ level is $1.88\times10^8\ \mathrm{s}^{-1}$. We assume no decay or dephasing of the metastable transition and take the Rabi frequency of the coupling laser as $\Omega_c = 5\ \mathrm{cm}^{-1}$. The coupling laser frequency is fixed, and probe frequency is tuned, with zero detuning at the center of gravity of the $6p^2\ ^3P_0 \rightarrow 6p7s\ ^3P_1$ transition. In ^{208}Pb, we note the Autler-Townes doublet with a spacing equal to the Rabi frequency of the coupling laser. In ^{207}Pb, the hyperfine structure results in four, instead of two, peaks. The imaginary part of the susceptibility (absorption) is zero in ^{208}Pb and small, but finite, in ^{207}Pb. The real part of the susceptibility (refractive index) versus frequency, in the vicinity of line center, is almost identical in both isotopes.

Figure 3 shows transmission versus probe frequency for ^{207}Pb with the same parameters as Fig. 2, but, here, with a atom density-length product that is comparable to that used in earlier experiments. The pressure broadening coefficients used here are $0.018\ \mathrm{cm}^{-1}$ at $10^{17}\ \mathrm{atoms/cm}^3$ for the resonantly self-broadened $6p^2\ ^3P_0 \rightarrow 6p7s\ ^3P_1$ transition [12] and $0.004\ \mathrm{cm}^{-1}$ at $10^{17}\ \mathrm{atoms/cm}^3$ for the $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_2$ metastable transition [5]. For these parameters, with $\Omega_c = 0$, the power transmission on the strongest of the hyperfine transitions is $\exp(-10^5)$. As expected from Eq. (9), the Rabi frequency of $\Omega_c = 5\ \mathrm{cm}^{-1}$ results in near perfect transmission at line center.

In summary, we have shown that hyperfine structure need not be an impediment for obtaining EIT. The principal idea is to use two lasers whose frequencies are equal to the center of gravity of the respective transitions and that are therefore not in Raman resonance with any particular hyperfine substate. The interplay of matrix elements and hyperfine splittings causes a cancellation such that, at sufficient coupling laser intensity, the transparency of the hyperfine split isotope will be approximately the same as that of the nonsplit isotope. This result is important for applications of EIT such as nonlinear optics [5], where, for example, the cost of a sufficient amount (20 g) of ^{208}Pb to load a 1-cm-diam, 20-cm-long heatpipe is about $60 000.

This idea will also apply to other types of splittings, but will require larger Rabi frequencies. In light atoms with small L-S coupling, fine-structure splitting may be overcome. It is likely that rotational splitting in molecules may also be overridden with this technique.

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