Electromagnetically induced absorption

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A large increase in atomic absorption due to coherent interaction with resonant radiation is predicted for a closed transition between two degenerate atomic levels verifying $0<F_g<F_e$ (F_g and F_e are the total angular momentum of the ground and the excited levels, respectively). In good agreement with the theoretical prediction, a total absorption enhancement by a factor 1.7 was obtained on the D_2 line of ^{85}Rb in a vapor cell experiment. [S1050-2947(99)04106-2]

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The possibility of rendering transparent an atomic medium around an absorbing optical transition has attracted considerable attention in recent years. This effect, named electromagnetic induced transparency (EIT), results from the coherent interaction of the medium with a resonant driving field and has found a large variety of applications [1]. EIT is intimately linked to the coherent population trapping (CPT) [2] of the system in a superposition of states (dark state) not coupled to the radiation. Most studies on EIT are concerned with three-level systems (mainly Λ systems) [1–3]. Degenerate and multilevel systems have been less explored [4–8]. Nevertheless, the existence of dark states and thus the possibility of CPT in degenerate two-level atoms has been theoretically analyzed [9,10].

While large reductions of the atomic absorption via EIT have been observed and understood, the opposite effect, an enhancement of the absorption resulting from atomic coherence induced by optical radiation, has remained so far unexplored. In this paper we show that a significant enhancement of the absorption of a probe field can occur in a degenerate two-level system under the action of a driving (pump) field provided that three requirements are satisfied: (i) $F_e=F_g+1$ (F_g and F_e are the total angular momentum of the ground and excited levels, respectively); (ii) transition $g\rightarrow e$ must be closed; (iii) the ground state must be degenerate ($F_g>0$). Notice that the first condition corresponds to the requirement for the nonexistence of a dark state in the ground level under excitation by an elliptically polarized wave [10]. Under these conditions, the increase in the absorption relative to the absorption without pump can exceed 100%. We have termed this effect electromagnetically induced absorption (EIA) [7].

Consider an atom at rest with a ground level g and an excited level e with angular momentum F_g and F_e, respectively, and energy separation $\hbar\omega_0$ (see inset in Fig. 1). We suppose that level e decays by spontaneous emission into g at a rate $b\Gamma$ where b is a branching ratio constant ($0\leqslant b\leqslant 1$). $b=1$ corresponds to a close transition, $1-b$ denotes the probability for an excited atom to decay to a level other than g. No specific relaxation mechanism is considered for the ground state. However, to account in a simplified way for the finite interaction time of the atoms with the light, we assume an overall relaxation of the density matrix towards equilibrium (i.e., all atoms isotropically distributed in the ground level) at a rate γ. We suppose $\Gamma\gg\gamma$. The atomic system is submitted to the action of a magnetic field B and two classical monochromatic electromagnetic fields: $\vec{E}_1(t)=E_1\hat{e}_1\exp(i\omega_1 t)$, $\vec{E}_2(t)=E_2\hat{e}_2\exp(-i\omega_2 t)$, $\hat{e}_{1,2}$ are complex unit polarization vectors. Neglecting the rapidly evolving antiresonant terms in the coupling of the atom with the optical fields (usual rotating wave approximation), the total Hamiltonian of the system is $H(t)=H_0+V_1+V_2$ with $V_j=E_j\hat{e}_j\cdot\vec{D}_{ge}\exp(i\omega_j t)+E_j^*\hat{e}_j^*\cdot\vec{D}_{ge}^{\dagger}\exp(-i\omega_j t)$ ($j=1,2$). H_0 is the atom Hamiltonian including Zeeman effect. $\vec{D}_{ge}=P_g\vec{D}P_e$ is the lowering part of the atomic dipole operator (P_g and P_e are projectors on the ground and excited subspaces, respectively).

The time evolution of the density matrix ρ is governed by the master equation [11]:

$$\frac{\partial\rho}{\partial t}=-\frac{i}{\hbar}[H,\rho]-\frac{(\Gamma+\gamma)}{2}\{P_e,\rho\}+b\Gamma\sum_{q=-1,0,1}Q_{ge}^q\rho Q_{eg}^q-\frac{\gamma}{2}\{P_g,\rho-\rho_0\}. \quad (1)$$

ρ_0 is the equilibrium density matrix (in the absence of the optical fields) taken as $\rho_0=Pg/(2F_g+1)$. $Q_{ge}^q=Q_{eg}^{q\dagger}$ ($q=-1,0,1$) are the standard components of the vectorial operator defined by $\vec{Q}_{ge}=\sqrt{2F_e+1}\vec{D}_{ge}\langle g\|\vec{D}\|e\rangle^{-1}$, where $\langle g\|\vec{D}\|e\rangle$ is the reduced matrix element of the dipole operator between g and e [11].

To find the steady-state solution of Eq. (1) to all orders in E_1 (pump field) with $E_2=0$ we consider the slowly varying matrix $\sigma_0=P_g\rho P_g+P_e\rho P_e+P_g\rho P_e\exp(-i\omega_1 t)+P_e\rho P_g\exp(i\omega_1 t)$. Substituting into Eq. (1) (with $V_2=0$) one gets for the steady-state value of σ_0 the equation

$$-\gamma\rho_0=-\frac{i}{\hbar}[H_0+\overline{V}_1-\hbar\omega_1 P_e,\sigma_0]-\frac{(\Gamma+\gamma)}{2}\{P_e,\sigma_0\}+b\Gamma\sum_{q=-1,0,1}Q_{ge}^q\sigma_0 Q_{eg}^q-\frac{\gamma}{2}\{P_g,\sigma_0\}, \quad (2)$$

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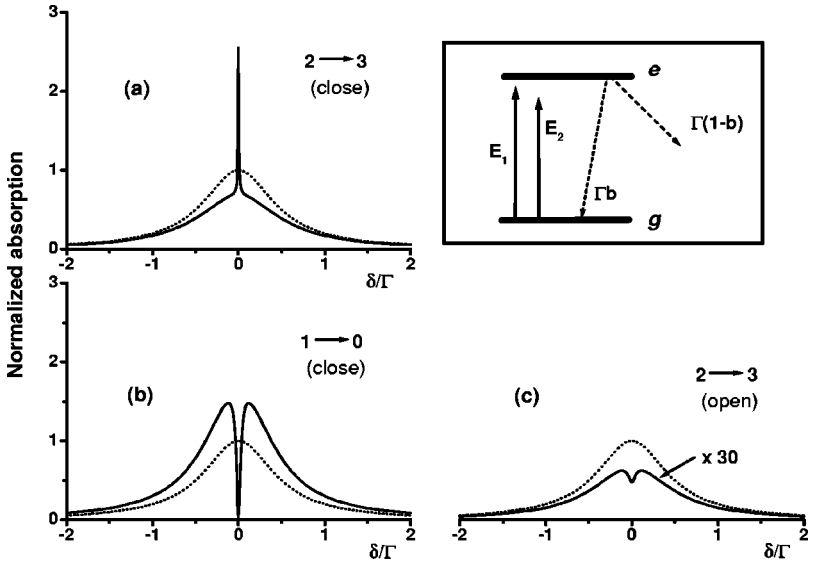

FIG. 1. Calculated probe absorption as a function of the frequency offset δ between pump and probe in absence (dotted) and in presence (solid) of the pump field for orthogonal and linear pump and probe polarizations [$\omega_1=\omega_0$, $\Gamma/\gamma=1000$, $\Omega_1=(2E_1/\hbar)\langle g\|\vec{D}\|e\rangle=0.4\Gamma$, $B=0$, $b=1$ (a), (b), $b=0.44$ (c)] for different transitions. The absorption is normalized to the maximum linear absorption of the probe field.

with $\overline{V}_1=E_1\hat{e}_1\cdot\vec{D}_{ge}+E_1^*\hat{e}_1^*\cdot\vec{D}_{ge}^{\dagger}$. We have solved Eq. (2) numerically operating in a Liouville space. The presence in Eq. (2) of the source term $-\gamma\rho_0$ and the possible escape out of the degenerate two-level system (if $b<1$) modify the total population [$\mathrm{Tr}(\sigma_0)$]. To account for this, the numerical solution of Eq. (2) is normalized by a factor $N=n_g+n_e[1+(\Gamma/\gamma)(1-b)]$ where n_g and n_e are the stationary populations obtained from Eq. (2) for the ground and excited levels, respectively.

We consider now the probe absorption by the atom plus pump field system. Neglecting higher harmonics of the fields, the relevant quantity is the term of $\rho_{ge}(t)=P_g\rho P_e$ evolving at frequency ω_2. Let $\sigma_{ge}\exp(i\omega_2 t)$ be this term. Considering only contributions to first order in E_2, we obtain: $\sigma_{ge}=P_g\sigma P_e$ where σ satisfies

$$i\left[\frac{\Omega_{ge}}{2},\sigma_0\right]=-\frac{i}{\hbar}[H_0+\overline{V}_1-\hbar\omega_1 P_e,\sigma]-i\delta\sigma-\frac{(\Gamma+\gamma)}{2}\{P_e,\sigma\}-\frac{\gamma}{2}\{P_g,\sigma\}+b\Gamma\sum_{q=-1,0,1}Q_{ge}^q\sigma Q_{eg}^q. \quad (3)$$

Here $\delta=\omega_2-\omega_1$ and $\Omega_{ge}=(2E_2/\hbar)(\hat{e}_2\cdot\vec{D}_{ge})$. The numerical solution of Eq. (3) is computed by the same methods used to solve Eq. (2). The probe absorption coefficient is given by $\alpha(\omega_2)\propto\mathrm{Im}[\hat{e}_2\cdot\mathrm{Tr}(\sigma_{ge}\vec{D}_{eg})]$. A large variety of absorption spectra can be generated depending on the angular momentum of the chosen atomic levels, the polarization and detunings of the pump and probe fields, as well as the magnitude of the magnetic field. We plan to present a detailed survey elsewhere.

Consider the specific case of a closed $F_g\rightarrow F_e=F_g+1$ transition. Figure 1(a) shows (in solid line) the absorption spectra calculated for an $F_g=2\rightarrow F_e=3$ transition with zero external magnetic field, and linear and orthogonal pump and probe polarizations. The dotted lines in Fig. 1 correspond to the probe absorption spectra in the absence of a pump field whose width is determined by Γ. The large amplitude sub-natural-width resonance occurring in the presence of the pump is a clear manifestation of EIA. The enhancement factor of the probe absorption due to EIA, relative to the absorption without pump, can be rather large (2.6 in this case). It depends on the Γ/γ ratio and is an increasing function of F_g. At low pump intensities, the width of the EIA resonance is 2γ [half-width at half maximum (HWHM)]. The spectra corresponding to a different type of closed transition $F_g=1\rightarrow F_e=0$ are shown in Fig. 1(b). Here the narrow resonance corresponds to EIT. Finally, Fig. 1(c) illustrates the case of an open $F_g=2\rightarrow F_e=3$ transition. Here again the coherent resonance corresponds to EIT in spite of the fact that no dark state exists in the ground level [10].

Figure 2 shows the probe absorption at $\delta=0$ for two choices of the optical polarizations and different integer values of the ground-state angular momentum F_g ($F_e=F_g+1$, $b=1$), as a function of the pump saturation parameter (proportional to pump intensity). Also shown in Fig. 2 is the incoherent optical pumping contribution to the probe absorption obtained taking $\overline{V}_1=0$ in Eq. (3). In the case of linear and orthogonal optical polarizations [Figs. 2(a)–2(c)], the absorption enhancement is essentially due to the coherent interaction between the driven atom and the probe beam and not to the pump-beam-induced optical pumping among ground- state Zeeman sublevels. For circular and equal polarizations [Fig. 2(d)—2(f)], the incoherent optical pumping contribution is larger, as expected [12]. Finally, notice that for $F_g=0$ the absorption at $\delta=0$ is always smaller than in the absence of the pump. In particular, this is the case in Fig. 2(d) that corresponds to a pure two-level system [13]. Notice that the largest resonances are predicted for pump intensity below saturation.

When the two optical waves propagate in the same direction, the resonance condition for a Raman transition between

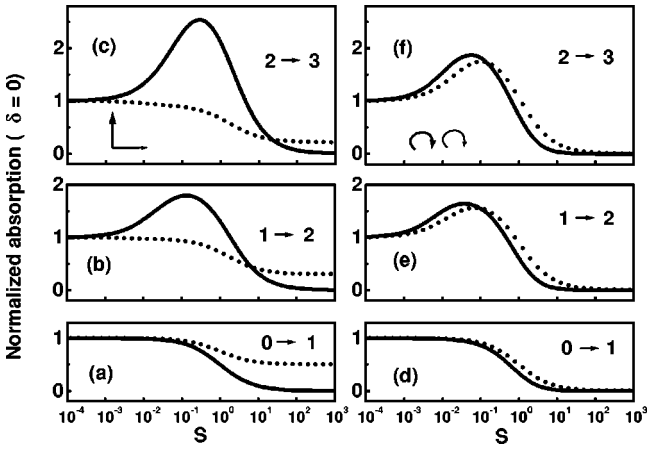

FIG. 2. Calculated absorption for zero pump-probe frequency offset ($\delta=0$) for different $F\rightarrow F+1$ transitions as a function of the pump saturation parameter $S=2\Omega_1^2/\Gamma^2=2(2E_1/\hbar\Gamma)^2\langle g\|\vec{D}\|e\rangle^2$ for $\omega_1=\omega_0$, $\Gamma/\gamma=1000$, $B=0$, and $b=1$. Solid: total absorption. Dotted: incoherent contribution (see text). The pump and probe polarizations are linear and orthogonal (a)–(c) or circular and equal (d)–(f).The absorption is normalized to the maximum linear absorption of the probe field.

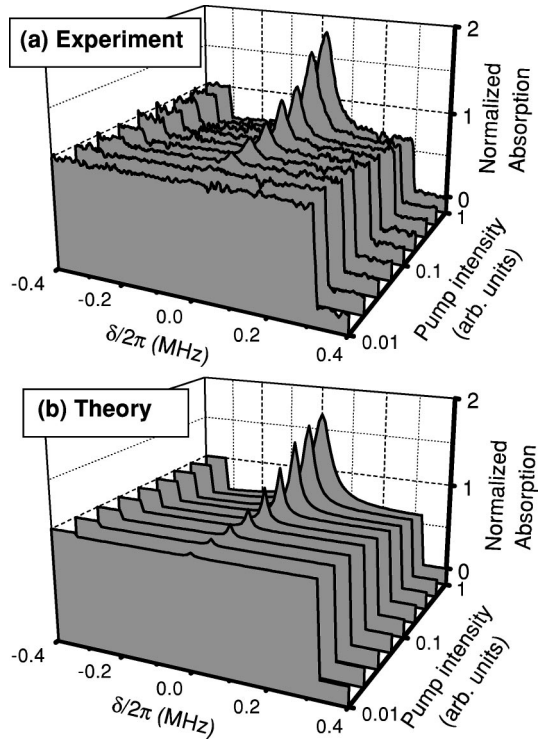

FIG. 3. (a) Experimental weak probe absorption spectra as a function of the frequency offset δ between pump and probe for different pump intensities. The leftmost parts of the spectra were recorded in the absence of the pump. The pump frequency is kept fixed at the ^{85}Rb $5S_{1/2}(F=3)\rightarrow 5P_{3/2}(F=4)$ transition except for the rightmost part of the spectra where both fields were shifted outside the absorption line. (b) Numerical simulation corresponding to the experimental conditions. The absorption is normalized to the maximum linear absorption of the probe field.

Zeeman sublevels of the ground state is negligibly affected by the atomic motion. However, the amplitude of the coherent resonances depends on the detunings of the fields relative to the optical transition which is a function of the atomic velocity through the Doppler effect. In consequence, the velocity distribution of the atoms must be taken into account by a numerical integration.

The experiment was realized on rubidium vapor using a setup similar to the one described in [7]. We used an extended cavity diode laser (sub-MHz linewidth). The laser frequency was locked to the $5S_{1/2}(F=3)\rightarrow 5P_{3/2}(F=4)$ transition of ^{85}Rb with the help of a servoloop based on the saturation absorption signal from an auxiliary Rb cell (frequency fluctuations within 1 MHz). The probe beam was frequency shifted with respect to the pump by two consecutive acousto-optic modulators, one driven by a variable rf source. In this way two mutually coherent pump and probe waves were generated with tunable frequency offset δ. The polarizations of the two fields were independently controlled. Perfect overlap between the two waves was achieved by propagating them simultaneously along a 50 cm single-mode optical fiber that did not significantly modify the polarizations. After the fiber, the light was sent through a 2 cm long cylindrical cell containing rubidium vapor at room temperature. Maximum power of the pump and probe at the cell were 0.8 mW and 50 μW, respectively. When necessary, the pump intensity was attenuated with neutral density filters. The atomic cell was placed within Helmholtz coils for magnetic field control. After the cell, the pump beam was blocked by a linear polarizer (extinction ratio larger than 200) while the probe intensity was monitored with a photodiode. A fraction of the probe beam, collected before the atomic cell, was simultaneously monitored on a reference photodiode. The output of the signal photodiode was divided by the output of the reference photodiode to compensate probe intensity fluctuations.

In order to maximize the absorption enhancement, the optical field intensity and the relaxation rate γ (determined by time of flight) were varied using the full available power and modifying the beam diameter at the cell. The largest absorption enhancement was obtained for a 1 cm beam diameter ($2\gamma\simeq 12$ kHz [7], $I_{\text{pump}}\simeq 0.8$ mW/cm^2). Careful cancellation of the magnetic field was necessary for maximum amplitude of the coherent resonance.

Figure 3(a) shows a series of experimental probe absorption spectra obtained with the pump frequency tuned to the $5S_{1/2}(F=3)\rightarrow 5P_{3/2}(F=4)$ transition of ^{85}Rb. Notice that the scanning range of δ is small compared to Γ ($\Gamma/2\pi=6$ MHz). The polarizations of the optical fields are linear and orthogonal. The ambient magnetic field has been compensated ($B=0\pm 5$ mG). On the leftmost part of the spectra the absorption was monitored in the absence of the pump beam (sample absorption 14%) while the rightmost part was recorded with ω_1 and ω_2 shifted outside the Doppler broadened absorption line. Visible in Fig. 3(a) is the increase of the transparency of the sample over most of the frequency range due to saturation by the pump field. However, in agreement with our theoretical prediction, a sharp increase of the absorption develops around $\delta=0$. The magnitude of the EIA peak overcomes the reduction in the absorption due to saturation and reaches at high pump intensity a 1.7 increase relative to the absorption without pump.

Since a linear polarizer in front of the detector was used to block the pump beam (to prevent detector saturation), pos-

sible contributions to the absorption spectra due to induced birefringence [5] have been investigated although no birefringence is expected in our case due to symmetry. This was checked by changing the polarizer angle a few degrees around the position for minimum pump transmission. No change was visible in the spectra. In fact, EIA resonances are clearly visible even when the detector polarizer is removed.

EIA resonances correspond to a significant change in the atomic fluorescence as was confirmed by monitoring the fluorescence from the atomic cell. This clearly indicates that the effect of EIA is associated to a modification of the atomic dissipation.

To compare the spectra presented in Fig. 3(a) with theory it is necessary to take into account that, due to the Doppler effect, three different atomic transitions, one closed ($F_g=3 \rightarrow F_e=4$) and two open ($F_g=3 \rightarrow F_e=2,3$), contribute to the signal. While the closed transition gives rise to EIA, both open ones, which are not totally bleached by optical pumping [8], give rise to EIT. The contributions to the absorption of the three transitions were independently computed and added. According to the experimental conditions, the pump laser was assumed to be on resonance with the $F_g=3 \rightarrow F_e=4$ transition. For each of the transitions involved ($F_e=2,3,4$), the reduced matrix element of the dipole moment was scaled (relative values: 0.35, 0.66, and 1, respectively) and the corresponding branching ratio b used ($b=0.22$, 0.56 and 1 respectively) [14]. Finally, the contribution of each transition was weighted by the relative contribution of the corresponding velocity class in the Maxwell-Boltzmann distribution at room temperature. No free parameter was varied in the calculation. Based on the experimental measurement of the resonance width at low intensity we used $\Gamma/\gamma = 1000$. The pump intensity was chosen to correspond to the maximum EIA effect. The results of the calculations are presented in Fig. 3(b). A good agreement with the observations is obtained. The maximum absorption enhancement observed and calculated is 1.7. According to the theoretical model, this result corresponds to an EIA enhancement factor of 3.3 for the closed transition alone.

In conclusion, we have demonstrated the possibility of a large resonant increase in atomic absorption due to coherent interaction with optical radiation. The phenomenon of EIA is in many aspects complementary to EIT. It may extend the range of the suggested applications of coherently prepared atomic systems and open the way to new possibilities such as the realization of steep anomalous dispersion. Finally, the connection between the increased dissipation associated to EIA and the mechanical effects of radiation on atoms [15] deserves to be examined.

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