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Electromagnetically induced absorption in a four-state system

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A simple theoretical model describing the positive sign of subnatural-width absorption resonances in the recent experiment of Akulshin and co-workers [Phys. Rev. A **57**, 2996 (1998)] is proposed. An analytical expression for the linear response to the weak probe field is found in the low-saturation limit with respect to the control field. It is shown that the positive sign of subnatural resonance is caused by the spontaneous transfer of the light-induced coherence from the excited level to the ground one.

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It is well known that nonlinear interference effects in the resonant atom-light interaction can lead to interesting and important phenomena, such as electromagnetically induced transparency (EIT) of atomic medium [1], coherent population trapping (CPT) [2], lasing without inversion [3], and others. Common to all these phenomena is the appearance of light-induced coherence between atomic levels, which are not coupled by dipole transitions. Recently Akulshin and coworkers have observed subnatural-width resonances in the absorption on the D_2 line of rubidium vapor under excitation by two copropagating optical waves with variable frequency offset [4]. In these experiments the control field was tuned to the center of the absorption line, while the frequency of the weak probe field was scanned around the control field frequency. Very narrow spectral structures with the widths about tens of kilohertz occurred at the zero-frequency offset. In the case of orthogonal polarizations of fields these resonances can be related to the ground-level Zeeman coherence, which is induced under simultaneous action of both fields. A prototype model for such resonances is a three-state Λ system, where two lower states belong to the ground level. In a Λ system, as is known [2], quantum coherence can lead to the destructive interference between two possible paths of excitation. As a result, if the control field is tuned in resonance, the probe absorption spectrum has a narrow dip at the two-photon resonance that can be interpreted as EIT caused by CPT. It is remarkable that, apart from expected EIT resonances with negative sign, the authors of Ref. [4] have detected positive resonances termed electromagnetically induced absorption (EIA). Later, basing on the experimental results and numerical calculations, they concluded [5] that EIA occurs in a degenerate two-level system whenever the following conditions are satisfied. First of all, the ground level must be degenerate in order to allow the long-lived Zeeman coherence. The next requirements specify what type of dipole transitions gives the positive-sign resonance. Namely, the total angular momentum of the excited level F_e must be larger than the total momentum of the ground level F_g, i.e., $F_e=F_g+1$, and, finally, the transition $F_g\rightarrow F_e$ should be a closed transition. However, the physical reasons for the sign change of the resonance, in our view, remained unexplained.

It should be noted that the positive subnatural-width spectral structures can appear in a Λ system in two cases. One possibility corresponds to an open Λ system when atoms are injected into the upper level. Under these conditions the probe field is amplified and the positive sign of resonance simply means a dip in the negative absorption [6]. Another case, arising when the control field is tuned far off resonance, is related to the second doublet of the resonance scattering (known as the Raman scattering) [7,8]. Obviously, both cases do not correspond to the experimental conditions of Refs. [4,5].

In comparison with three-state models, the four-state systems (in particular, double-Λ configuration) studied in a number of papers [9] open new possibilities in the quantum coherence manipulation for designing a desired atomic response. On the other hand, as was shown in Ref. [10], the spontaneous transfer of Zeeman coherence from the excited level to the ground level can change position, width, and amplitude (which is most important in the present context) of nonlinear resonances in the probe-field spectroscopy. It seems reasonable that this fundamental process discovered by Barrat and Cohen-Tannoudji [11] can be responsible for the positive sign of resonances in Refs. [4,5]. At the same time, simple theoretical models (Λ, V, double-Λ, and others) usually used for the description of quantum coherence and interference effects do not take into account the spontaneous coherence transfer. The exception is provided by the papers of Rautian on the spontaneous transfer of optical coherence [12].

In the present paper, motivated by the absence of EIA resonances in a closed three-state Λ system, we propose a simple theoretical model for EIA, which allows the spontaneous coherence transfer between levels, a four-state N-configuration atom. An analytical expression for the probe absorption as a function of frequency offset is found in the low-saturation limit with respect to the control field. It is shown that EIA resonance is caused by the spontaneous transfer of the light-induced low-frequency coherence from the excited level to the ground one.

Let us consider the resonant interaction of a bichromatic light field:

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}_1\exp[-i\omega_1 t+i(\mathbf{k}_1\cdot\mathbf{r})]+\mathbf{E}_2\exp[-i\omega_2 t+i(\mathbf{k}_2\cdot\mathbf{r})]+\text{c.c.} \quad (1)$$

with a four-state atom. This atomic system has four states $|i\rangle$, $i=1,\ldots,4$ (see Fig. 1). The odd-numbered states $|1\rangle$ and $|3\rangle$ belong to the nonrelaxing ground level with zero energy. The even-numbered states $|2\rangle$ and $|4\rangle$ form the ex-

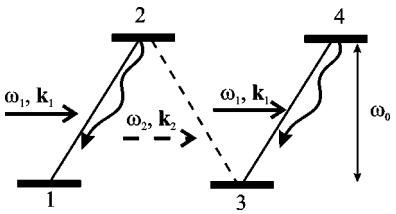

FIG. 1. N-configuration atom. The light-induced transitions are marked by solid (control field) and dashed (probe field) lines. Wavy lines show the spontaneous transfer of the excited-level coherence.

cited level with energy $\hbar\omega_0$ and spontaneous relaxation rate Γ. We assume that among the optical transitions between the ground and excited levels $|\text{odd}\rangle \rightarrow |\text{even}\rangle$ the transition $|1\rangle \rightarrow |4\rangle$ is forbidden due to some selection rule (for instance, with respect to the momentum projection). Let the first term in Eq. (1) (hereafter referred to as a control field) be sufficiently larger than the second one, which is a probe field. The control field with the vector amplitude $\mathbf{E}_1$ and frequency ω_1 drives two transitions simultaneously: $|1\rangle \rightarrow |2\rangle$ and $|3\rangle \rightarrow |4\rangle$. The weak probe filed $\mathbf{E}_2$ at frequency ω_2 excites the transition $|3\rangle \rightarrow |2\rangle$. In the rotating frame the Hamiltonian of a free atom reads

$$\hat{H}_0 = \hbar\delta_1 |1\rangle\langle 1| + \hbar\delta_2 |3\rangle\langle 3| + \hbar(\delta_2 - \delta_1)|4\rangle\langle 4|, \quad (2)$$

where $\delta_q = \omega_q - \omega_0 - (\mathbf{k}_q \cdot \mathbf{v})$ $(q = 1,2)$ are the detunings including the Doppler shifts. Using the rotating-wave and dipole approximations, we can write the atom-field interaction Hamiltonian in the form

$$\hat{H}_{AF} = \hbar\Omega_1 \hat{Q}_1 + \hbar\Omega_2 \hat{Q}_2 + \text{H.c.}. \quad (3)$$

Here Ω_q are the corresponding Rabi frequencies and the operators $\hat{Q}_q$ are given by

$$\hat{Q}_1 = A|2\rangle\langle 1| + |4\rangle\langle 3|, \quad \hat{Q}_2 = B|2\rangle\langle 3|, \quad A^2 + B^2 = 1, \quad (4)$$

where the real numbers A and B govern the relative transition amplitudes.

In the case of the pure radiative relaxation in a closed atomic system the optical Bloch equations for the atomic density matrix $\hat{\rho}$ have the form

$$\frac{d}{dt}\hat{\rho} = -\frac{i}{\hbar}[\hat{H}_0 + \hat{H}_{AF}, \hat{\rho}] - \frac{1}{2}\Gamma\{\hat{P}_e, \hat{\rho}\} + \Gamma \sum_{q=1,2} \hat{Q}_q^{\dagger}\hat{\rho}\hat{Q}_q, \quad (5)$$

where $\hat{P}_e = |2\rangle\langle 2| + |4\rangle\langle 4|$ is the projector onto the excited level. The second term on the right-hand side of Eq. (5) having the structure of the anticommutator describes the radiative damping of the excited-level populations and optical coherences. The last term on the right-hand side corresponds to the transfer of populations and low-frequency coherences from the excited level to the ground level.

It is well known that the probe field absorption is proportional to the real part of the product $i\Omega_2^* \rho_{23}$. The steady-state off-diagonal element ρ_{23} can be written as

$$\rho_{23} = [\Gamma/2 - i\delta_2]^{-1}\{-iB\Omega_2(\rho_{33} - \rho_{22}) - iA\Omega_1\rho_{13} + i\Omega_1\rho_{24}\}. \quad (6)$$

The last two terms (proportional to Ω_1) in curly brackets in Eq. (6) describe modifications occurring in absorption due to light-induced low-frequency coherence. In an ideal (with the lower coherence relaxation rate equal to zero) Λ system, the coherence ρ_{13} has such a phase and amplitude that the probe absorption vanishes at the exact two-photon resonance $\delta_2 = \delta_1$. This fact can be interpreted as a result of quantum destructive interference [2]. In the presence of dephasing collisions the absorption reduction is less pronounced. In the case under consideration an additional term in the equation for ρ_{13} arises from the spontaneous transfer of coherence $\rho_{24} \rightarrow \rho_{13}$ $(d\rho_{13}/dt = \cdots + A\Gamma\rho_{24})$. Let us consider the influence of this process on the probe absorption spectrum. For the sake of clarity, in the following analyses we use the first-order approximation in the probe amplitude Ω_2. Besides, we restrict ourselves to the low-saturation limit with respect to the control field, i.e., $\Omega_1 < \Gamma$. Then we replace Eq. (6) by

$$\rho_{23}^{(1)} = [\Gamma/2 - i\delta_2]^{-1}\{-iB\Omega_2\rho_{33}^{(0)} - iA\Omega_1\rho_{13}^{(1)}\}, \quad (7)$$

where the index in $\rho^{(n)}$ signifies the nth order of perturbation theory on Ω_2. Equation (7) is supplemented by the following equations for the first-order coherences:

$$\begin{aligned} &i(\delta_1 - \delta_2)\rho_{13}^{(1)} = iB\Omega_2\rho_{12}^{(0)} - iA\Omega_1^*\rho_{23}^{(1)} + i\Omega_1\rho_{14}^{(1)} + bA\Gamma\rho_{24}^{(1)}, \\ &[\Gamma + i(\delta_1 - \delta_2)]\rho_{24}^{(1)} = -iB\Omega_2\rho_{34}^{(0)} - iA\Omega_1\rho_{14}^{(1)} + i\Omega_1^*\rho_{23}^{(1)}, \\ &[\Gamma/2 + i(2\delta_1 - \delta_2)]\rho_{14}^{(1)} = i\Omega_1^*\rho_{13}^{(1)}. \end{aligned} \quad (8)$$

Here, in order to demonstrate the role of the spontaneous coherence transfer, we insert ''by hand'' the coefficient $0 \leq b \leq 1$, which governs the efficiency of the process. For example, if $b = 0$ the spontaneous coherence transfer is absent. The maximal efficiency $b = 1$ corresponds to the initial equations (5). This coefficient can be thought of as the branching ratio in a partially open system. However, for a self-consistent treatment of open systems we should introduce into Eq. (5) a source describing external pumping of levels and additional relaxation terms. As a result, the situation would be more complicated. Nevertheless, it can be shown that under certain conditions EIA resonances appear in an open four-state system as well.

From Eqs. (8) one can obtain the coupled equations for the low-frequency coherences:

$$\left[\frac{|A\Omega_1|^2}{\Gamma/2 - i\delta_2} + \frac{|\Omega_1|^2}{\Gamma/2 + i(2\delta_1 - \delta_2)} + i(\delta_1 - \delta_2)\right]\rho_{13}^{(1)} - bA\Gamma\rho_{24}^{(1)} = -\frac{AB\Omega_2\Omega_1^*}{\Gamma/2 - i\delta_2}\rho_{33}^{(0)} + iB\Omega_2\rho_{12}^{(0)}, \quad (9)$$

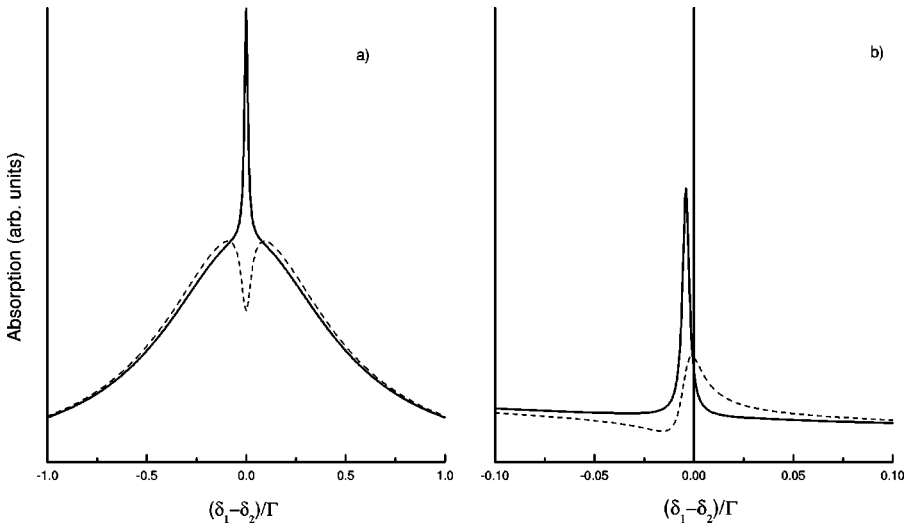

FIG. 2. Probe field absorption versus the frequency offset in the case of homogeneous broadening $\mathbf{v}=\mathbf{0}$. The control field detuning (a) $\delta_1=0$ and (b) $\delta_1=\Gamma$. Solid (dashed) curves correspond to the case of $b=1$ $(b=0)$. The other parameters are $A^2=B^2=1/2$, $\Omega_1=0.1\Gamma$.

$$[\Gamma+i(\delta_1-\delta_2)]\rho_{24}^{(1)}-\left\{\frac{A|\Omega_1|^2}{\Gamma/2-i\delta_2}+\frac{A|\Omega_1|^2}{\Gamma/2+i(2\delta_1-\delta_2)}\right\}\rho_{13}^{(1)}$$
$$=\frac{B\Omega_2\Omega_1^*}{\Gamma/2-i\delta_2}\rho_{33}^{(0)}-iB\Omega_2\rho_{34}^{(0)}.$$

The right-hand sides of Eqs. (9) are the interference terms describing the creation of $\rho_{13}^{(1)}$ and $\rho_{24}^{(1)}$. Equations (9) are not independent because of the second terms on the left-hand sides of both lines, which correspond to the spontaneous and induced coherence transfer between levels. In the low-saturation limit under consideration, the appearance of the excited-level coherence $\rho_{24}^{(1)}$ in the equation for the ground-level coherence $\rho_{13}^{(1)}$ is due to the spontaneous coherence transfer only. As one can see from Eqs. (9), this process changes the position, width, and amplitude of the nonlinear resonance caused by the low-frequency coherence. Since in the present paper we are interested in the subnatural-width resonance, the approximation $|\delta_1-\delta_2|\ll\Gamma$ is relevant. Using Eqs. (9) to eliminate the low-frequency coherence from Eq. (7), we arrive at the final result for the linear response:

$$\rho_{23}^{(1)}=\frac{-iB\Omega_2}{\Gamma/2-i\delta_2}\left\{\rho_{33}^{(0)}-\frac{(1-b)|A\Omega_1|^2\rho_{33}^{(0)}}{(\Gamma/2-i\delta_2)D}+\frac{iA\Omega_1(\rho_{12}^{(0)}-b\rho_{34}^{(0)})}{D}\right\},$$

$$D=\frac{|A\Omega_1|^2(1-b)}{\Gamma/2-i\delta_2}+\frac{|\Omega_1|^2(1-bA^2)}{\Gamma/2+i\delta_1}+i(\delta_1-\delta_2), \qquad (10)$$

where in the case of a closed system we should substitute $\rho_{11}^{(0)}=0$, $\rho_{12}^{(0)}=iA\Omega_1^*\rho_{11}^{(0)}/(\Gamma/2+i\delta_1)=0$, $\rho_{34}^{(0)}=i\Omega_1^*\rho_{33}^{(0)}/(\Gamma/2+i\delta_1)$. The positions and widths of nonlinear resonances are determined by the denominator $(\Gamma/2-i\delta_2)^2(\Gamma/2+i\delta_1)D$. In the general case, the frequencies of these resonances can be associated with transitions between two pairs of quasienergetic sublevels, which appear due to Rabi splitting of the singlets $|2\rangle$ and $|3\rangle$. In the low-saturation limit $\Omega_1<\Gamma$ all the resonances merge together and we can discuss the situation in terms of optical shifts and the field broadening of a single resonance.

For the sake of simplicity we will consider the case of $\mathbf{v}=\mathbf{0}$ (homogeneous broadening). Both nonlinear terms in Eq. (10) responsible for the subnatural-width resonance on the frequency offset $\delta_1-\delta_2$ are known in the probe-field spectroscopy [8]. These contributions have different origins. The first term proportional to $(1-b)$ can be related to the quantum interference between quasienergetic sublevels. It is similar to one for a closed Λ system, with changes caused by the light shift of the $|3\rangle$ state and by the spontaneous transfer of coherence. The last term in Eq. (10) can be interpreted as a consequence of mixing (or interference) of the states $|3\rangle$ and

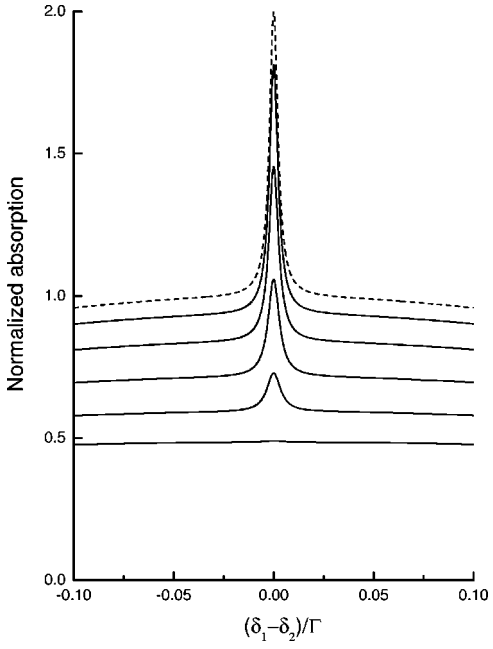

FIG. 3. Numerically generated probe absorption spectra. The probe field Rabi frequency is increased from top to bottom, $\Omega_2/\Gamma=0.01,0.02,0.03,0.04,0.05$. The rest of the parameters are $\delta_1=0$, $\Omega_1=0.05\Gamma$, $b=1$, and $A^2=B^2=1/2$. The dashed curve shows the analytical results of Eq. (10).

$|4\rangle$ as well as $|1\rangle$ and $|2\rangle$ (which should be populated for the nonvanishing effect) by the control field. Note that a similar term can appear in an open Λ system, when atoms are injected into the $|1\rangle$ state, but with the opposite sign. In the Λ case such a term allows the probe amplification [6]. Here it is possible to obtain a significant enhancement of the absorption. In the general case, there is a competition between these two terms that are controlled by the coefficient b. If the control field detuning $\delta_1=0$, the sign of the absorption resonance is determined by the sign of the factor $2b-1$. For example, in the absence of the spontaneous transfer of the low-frequency coherence ($b=0$) the resonance is negative, which corresponds to EIT. If $b>1/2$ the resonance is positive, i.e., we have EIA (see in Fig. 2). Compared to the linear response in the absence of the control field $\Omega_1=0$, the absorption is enhanced by the factor $1/[1-(2b-1)A^2]$, which exceeds 100% if $b=1$ and $A^2>1/2$.

In view of the many approximations made above in our derivations, it seems useful to compare the analytical results with exact numerical calculations. In the present case of a closed four-state system, such calculations can easily be done. Besides, using numerical methods, we can consider the probe absorption beyond our approximations. In particular, we can trace the spectrum modification with an increase of the probe field amplitude Ω_2. In Fig. 3 the probe absorption ($\mathrm{Re}\{i\Omega_2^*\rho_{23}\}$) normalized to the linear absorption at line center is shown as a function of the frequency offset. The curves correspond to different values of Ω_2, while the other parameters are fixed. It is seen that the contrast and amplitude of the resonance are significantly reduced with an increase of the probe amplitude. The decrease of amplitude can be understood as a result of the repumping of atoms from the state $|3\rangle$ to the state $|1\rangle$.

In conclusion, we note that the N-type interaction scheme can be easily organized in real atomic systems. For example, let us consider the closed transition $F_g=F\rightarrow F_e=F+1$ of the D_2 line of an alkali-metal atom that interacts with a circular polarized (σ_+) control field. In the absence of a probe field all atoms are completely pumped into the outermost Zeeman states $|F_g,m_g=F\rangle$ and $|F_e,m_e=F+1\rangle$. If the probe field has orthogonal circular polarization (σ_-), then to first order in the probe-field amplitude Ω_2 we have an N atom with the states $|1\rangle=|F_g,m_g=F-2\rangle$, $|2\rangle=|F_e,m_e=F-1\rangle$, $|3\rangle=|F_g,m_g=F\rangle$, and $|4\rangle=|F_e,m_e=F+1\rangle$. This scheme was implemented in Ref. [4] in experiments with circular polarized waves.

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