Four-state $N$-atom as a simple theoretical model for electromagnetically induced absorption

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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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I. INTRODUCTION

As is well-known the nonlinear interference effects in the resonant atom-light interaction can lead to the electromagnetically induced transparency (EIT) of atomic medium [1] as well as to other interesting phenomena [2]. The key point of all these phenomena is the light-induced coherence between atomic levels, which are not coupled by dipole transitions. Recently Akulshin and co-workers 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 [3]. Surprisingly, that apart from EIT-resonances with negative sign, they have detected positive resonances termed in ref. [3] as electromagnetically induced absorption (EIA). Basing on the experimental results and numerical calculations, authors of ref. [3] have deduced [4] that EIA occurs in a degenerate two-level system when the three conditions are satisfied: i) The excited-state total angular momentum $F_e = F_g + 1$. ii) Transition $F_g → F_e$ is closed. iii) The ground state is degenerate $F_g > 0$.

In the present paper, motivated by the absence of EIA-resonances in three-state $\Lambda$ and $V$-systems, we propose a simple theoretical model for EIA – four-state $N$-atom. Namely, we consider an atom with four states $|i\rangle, \; i = 1 \ldots 4$. The odd states $|1\rangle$ and $|3\rangle$ are degenerate and belong to the ground level, while the even states $|2\rangle$ and $|4\rangle$ (also degenerate) form the excited level. All optical transitions $|odd\rangle → |even\rangle$ are permitted except for $|1\rangle → |4\rangle$, that is forbidden. The control field with frequency $\omega_1$ drives the transitions $|1\rangle → |2\rangle$ and $|3\rangle → |4\rangle$. The weak probe at $\omega_2$ is applied to the $|3\rangle → |2\rangle$ transition. An analytical expression for the probe absorption as a function of frequency offset $\omega_1 - \omega_2$ is found. It is shown that EIA occurs due to the spontaneous transfer of the light-induced low-frequency coherence from the excited level to the ground one. Obviously, both $\Lambda$ and $V$-systems do not describe such a process. The sign of the subnatural resonance depends on the branching ratio constant $0 < b \leq 1$ and becomes positive for closed transition $b = 1$. Velocity averaging in the case of Doppler broadening is briefly discussed.

It is worth noting, effects of the spontaneous coherence transfer on nonlinear resonances in the probe field spectroscopy were first considered from the general point of view by S. G. Rautian [5].

II. FORMULATION OF THE PROBLEM

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

$$E(r, t) = E_1 \exp[-i\omega_1 t + i(k_1 r)] + E_2 \exp[-i\omega_2 t + i(k_2 r)] + c.c.$$  \hspace{1cm} (1)

with a four-state atom. This atomic system has four states $|i\rangle, \; i = 1 \ldots 4$ (see in fig.1). The two odd states $|1\rangle$ and $|3\rangle$ are degenerate and belong to the ground level with zero energy and zero relaxation rate. The even states are also degenerate and form the excited level with the energy $\bar{E}$ and relaxation rate $\Gamma$. We assume that among optical transitions between the ground and excited levels $|odd\rangle → |even\rangle$ the transition $|1\rangle → |4\rangle$ is forbidden due to some selection rule (for instance, with respect to the momentum projection). Let the first term in eq.(1) (will be referred as a control field) is sufficiently larger than the second one, which is a probe field. The control field with the vector amplitude $E_1$ and frequency $\omega_1$ drives simultaneously two transitions $|1\rangle → |2\rangle$ and $|3\rangle → |4\rangle$. The weak probe field $E_2$ at the frequency $\omega_2$ induces the $|3\rangle → |2\rangle$ transition. In the rotating frame the Hamiltonian for the free atom reads...
\[ \hat{H}_0 = \hbar \delta_1 |1\rangle \langle 1| + \hbar \delta_2 |3\rangle \langle 3| + \hbar (\delta_2 - \delta_1) |4\rangle \langle 4|, \] (2)

where \( \delta_q = \omega_q - \omega_0 - (k_q v) \) \((q = 1, 2)\) are the detunings including the Doppler shifts. Using the rotating wave approximation, we write the atom-field interaction Hamiltonian in the form

\[ \hat{H}_{AF} = \hbar \Omega_1 \hat{Q}_1 + \hbar \Omega_2 \hat{Q}_2 + h.c. . \] (3)

Here \( \Omega_q \) are the corresponding Rabi frequencies and the operators \( \hat{Q}_q \) are given by

\[ \begin{align*}
\hat{Q}_1 &= A |2\rangle \langle 1| + |4\rangle \langle 3| \\
\hat{Q}_2 &= B |2\rangle \langle 3| , \quad A^2 + B^2 = 1 ,
\end{align*} \] (4)

where the real numbers \( A \) and \( B \) govern the relative amplitudes of transitions in the model under consideration.

In the case of the pure radiative relaxation the optical Bloch equations for the atomic density matrix \( \hat{\rho} \) read

\[ \frac{d}{dt} \hat{\rho} + \frac{i}{\hbar} \left[ \hat{H}_0 + \hat{H}_{AF}, \hat{\rho} \right] + \frac{1}{2} \Gamma \left\{ \sum_{q=1,2} \hat{Q}_q \hat{Q}_q^\dagger, \hat{\rho} \right\} - b \Gamma \sum_{q=1,2} \hat{Q}_q^\dagger \hat{Q}_q \hat{\rho} = \hat{\bar{R}} , \] (5)

where the third term on the l.h.s. has a structure of anticommutator and describes the radiative damping of the excited-level populations and optical coherences. The last term on the l.h.s. corresponds to the transfer of the populations and low-frequency coherences from the excited level to the ground one under the spontaneous emission.

The low-saturation limit for the control field, i.e. \( \Omega \) has such phases that the absorption of the probe field is reduced at the two-photon resonance \( \delta_2 = \delta_1 \). We note that these two terms in eq. \ref{eq:4} have opposite signs. In the case under consideration an additional term in equation for \( \rho_{13} \) arises from the spontaneous transfer \( \rho_{24} \rightarrow \rho_{13} \) \((d \rho_{13}/dt = \ldots + bA \rho_{24})\). Hence, the phase of \( \rho_{24} \) giving the transparency through the term \( i\Omega_1 \rho_{24} \) can give the increase of absorption through the term \( -iA \Omega_1 \rho_{13} \). For the sake of clarity, in the following analyses we use two approximations: i) The first order in the probe amplitude \( \Omega_2 \); and ii) The low-saturation limit for the control field, i.e. \( \Omega_1 < \Gamma \). In this case instead of eq. \ref{eq:4} we write

\[ \rho_{23}^{(1)} = [\Gamma/2 - i \delta_2]^{-1} \{-iB \Omega_2 (\rho_{23}^{(0)} - \rho_{22}) - iA \Omega_1 \rho_{14} + iA \Omega_1 \rho_{24} \} , \] (6)

The last two terms in the curly brackets in eq. \ref{eq:5} (proportional to \( \Omega_1 \)) describe modifications of the absorption due to the light-induced low-frequency coherences. It is well-known that in both \( \Lambda \) and \( V \) systems the coherences \( \rho_{13} \) and \( \rho_{24} \) have such phases that the absorption of the probe field is reduced at the two-photon resonance \( \delta_2 = \delta_1 \). We note that these two terms in eq. \ref{eq:6} have opposite signs. In the case under consideration an additional term in equation for \( \rho_{13} \) arises from the spontaneous transfer \( \rho_{24} \rightarrow \rho_{13} \) \((d \rho_{13}/dt = \ldots + bA \rho_{24})\). Hence, the phase of \( \rho_{24} \) giving the transparency through the term \( i\Omega_1 \rho_{24} \) can give the increase of absorption through the term \( -iA \Omega_1 \rho_{13} \). For the sake of clarity, in the following analyses we use two approximations: i) The first order in the probe amplitude \( \Omega_2 \); and ii) The low-saturation limit for the control field, i.e. \( \Omega_1 < \Gamma \). In this case instead of eq. \ref{eq:4} we write

\[ \rho_{23}^{(1)} = \frac{i(\delta_1 - \delta_2) \rho_{13}^{(0)}}{\Gamma/2 + i \delta_1} ; \quad \rho_{34}^{(0)} = \frac{i\Omega_1 \rho_{33}^{(0)}}{\Gamma/2 + i \delta_1} , \] (7)

where the index over \( \rho^{(n)} \) means that this element is taken in the \( n \)-th order on \( \Omega_2 \). The equation \ref{eq:4} should be completed by the following equations for the first-order coherences:

\[ \begin{align*}
[i(\delta_1 - \delta_2) \rho_{13}^{(1)}] &= iB \Omega_2 \rho_{12}^{(0)} - iA \Omega_1 \rho_{14}^{(1)} + bA \rho_{24}^{(1)} \\
[\Gamma + i(\delta_1 - \delta_2)] \rho_{24}^{(1)} &= -iB \Omega_2 \rho_{24}^{(0)} - iA \Omega_1 \rho_{14}^{(1)} + i\Omega_1 \rho_{23}^{(1)} \\
[\Gamma/2 + i(2 \delta_1 - \delta_2)] \rho_{14}^{(1)} &= i\Omega_1 \rho_{13}^{(1)} \\
\rho_{12}^{(0)} &= \frac{iA \Omega_1 \rho_{11}^{(0)}}{\Gamma/2 + i \delta_1} ; \quad \rho_{34}^{(0)} = \frac{i\Omega_1 \rho_{33}^{(0)}}{\Gamma/2 + i \delta_1} .
\end{align*} \] (8)
Here we assume that the term $\hat{R}$ in eq. (3) is diagonal and, consequently, gives contributions into eqs. (8) implicitly through the zero-order populations $\rho_{11}^{(0)}$ only. From eqs. (8) one can get the coupled equations for the low-frequency coherences:

$$
\begin{align*}
\left[ \frac{|A\Omega_1|^2}{\Gamma/2 - i\delta_2} + \frac{|\Omega_1|^2}{\Gamma/2 + i(2\delta_1 - \delta_2)} \right] \rho_{13}^{(1)} - bA\Gamma\rho_{24}^{(1)} = & \frac{AB\Omega_2\Omega_1^*}{\Gamma/2 - i\delta_2} \rho_{33}^{(0)} - \frac{AB\Omega_2\Omega_1^*}{\Gamma/2 + i\delta_1} \rho_{11}^{(0)} \\
\left[ \Gamma + i(\delta_1 - \delta_2) \right] \rho_{24}^{(1)} = & \left\{ \frac{A\Omega_1^2}{\Gamma/2 - i\delta_2} + \frac{A\Omega_1^2}{\Gamma/2 + i(2\delta_1 - \delta_2)} \right\} \rho_{13}^{(1)} = \left\{ \frac{B\Omega_2\Omega_1^*}{\Gamma/2 - i\delta_2} + \frac{B\Omega_2\Omega_1^*}{\Gamma/2 + i\delta_1} \right\} \rho_{33}^{(0)}.
\end{align*}
$$

(9)

The right-hand sides of eqs. (8) are the field interference terms describing the creation of $\rho_{13}^{(1)}$ and $\rho_{24}^{(1)}$. In the square bracket of the first line the field broadening and optical shifts of the ground-level states are present. Equations (8) are not independent due to the second terms on l.h.s. of both lines, which correspond to the spontaneous and induced coherence transfer between levels. In the low-saturation limit the excited-level coherence $\rho_{24}^{(1)}$ enters into the equation for the ground-level coherence $\rho_{13}^{(1)}$ through the term describing the spontaneous coherence transfer. As it can be seen, this process leads to changes in the position, width and amplitude of nonlinear resonances connected with the low-frequency coherence. Since in the present paper we are interested in the subnatural-width resonance, the ansatz $|\delta_1 - \delta_2| \ll \Gamma$ is relevant. Using eqs. (8), we can eliminate the low-frequency coherence in eq. (7) and arrive at the final result for the linear response:

$$
\rho_{23}^{(1)} = \frac{-iB\Omega_2}{\Gamma/2 - i\delta_2} \left\{ \rho_{33}^{(0)} + \frac{(b - 1)\rho_{33}^{(0)}|A\Omega_1|^2}{|A\Omega_1|^2(1 - b) + |\Omega_1|^2(1 - bA^2)\frac{\Gamma/2 - i\delta_2}{\Gamma/2 + i\delta_1} + i(\delta_1 - \delta_2)(\Gamma/2 - i\delta_2)} \right. \\
+ \left. \frac{(b\rho_{33}^{(0)} - \rho_{11}^{(0)})|A\Omega_1|^2}{|A\Omega_1|^2(1 - b)\frac{\Gamma/2 - i\delta_2}{\Gamma/2 + i\delta_1} + |\Omega_1|^2(1 - bA^2) + i(\delta_1 - \delta_2)(\Gamma/2 + i\delta_1)} \right\}.
$$

(10)

A. Homogeneous broadening

Consider first the case of $v = 0$. The steady-state zero-order populations $\rho_{11}^{(0)}$ and $\rho_{33}^{(0)}$ are governed by the equilibrium between the excitation and relaxation processes in the absence of the probe field. Obviously, these values can not contain structures with the width less than $\Gamma$. Then, only the last two terms on the r.h.s. of (10) are responsible for the subnatural-width resonance on the frequency offset $\delta_1 - \delta_2$. If $\delta_1 = 0$, the sign of the absorption resonance is determined by the sign of the expression $(2b - 1)\rho_{33}^{(0)} - \rho_{11}^{(0)}$ and, consequently, depends on both the branching ratio constant and zero-order populations. For example, in the absence of the spontaneous transfer of the low-frequency coherence ($b = 0$) the resonance is always negative, that corresponds to EIT. In the opposite case of the closed transition ($b = 1$) the resonance is positive if $\rho_{33}^{(0)} > \rho_{11}^{(0)}$, i.e. we have EIA (see in fig. 2). The position and the width of the EIA-resonance is determined by the real and imaginary parts of the linear combination of the complex ground-level optical shifts:

$$(1 - b)\Delta \varepsilon_1 - (1 - bA^2)\Delta \varepsilon_2 = (1 - b)\frac{|A\Omega_1|^2}{\delta_1 - i\Gamma/2} - (1 - bA^2)\frac{|\Omega_1|^2}{\delta_1 + i\Gamma/2}.$$

It is remarkable that the coefficients of this combination depend on the branching ratio $b$.

B. Doppler broadening

In the case of atomic gas the EIA-resonance is a sum of structures with different amplitudes, positions, and width. Here we consider the result of such velocity averaging in one specific case. Let the copropagating control and probe fields have the approximately equal Doppler shifts $(k_1 v) \approx (k_2 v) \approx k v_z$. Besides, we assume that the transition is closed $b = 1$, and the zero-order populations $\rho_{11}^{(0)} = 0$ and $\rho_{33}^{(0)} = f_M(v)$ is the Maxwell distribution. The averaged optical coherence is expressed through the error function:

3
\[
\langle \rho_{23} \rangle_v = -i B \Omega_2 \left\{ V(\delta_2) + \frac{|A \Omega_1|^2}{|A \Omega_1|^2 + i \Gamma (\delta_1 - \delta_2) - (\delta_1 - \delta_2)^2} \left[ V(\delta_2) + V(-\delta_1 + |B \Omega_1|^2/\delta_1 - \delta_2) \right] \right\},
\]

where \( \delta_q = \omega_q - \omega_0, \quad \tau = \sqrt{2k_B T/M}, \) and \( V(x) \) is the well-known Voight contour. In the case of large Doppler broadening \( k\tau \gg \Gamma \) the probe field absorption \( \sim \Re \{ i \Omega_2^* \langle \rho_{23} \rangle_v \} \) as a function of the frequency offset \( \delta_1 - \delta_2 \) contains two structures situated at \( \delta_1 - \delta_2 = 0 \) with different width and signs. One of them described by the Lorentzian \( |A \Omega_1|^2/(|A \Omega_1|^2 + i \Gamma (\delta_1 - \delta_2)) \) has the width \( |A \Omega_1|^2/\Gamma \) and gives the rising of absorption. The last Voight function in eq.(11) \( V(-\delta_1 + |B \Omega_1|^2/(\delta_1 - \delta_2)) \) describes a very narrow dip in the absorption spectrum (see in fig.3.a). This structure with the width \( |B \Omega_1|^2/(k\tau) \) is the result of averaging. For an atom with given velocity \( v \) the effective detuning is \( \delta_1 - kv \) due to the Doppler shift. As is seen from eq.(11) and fig.2.b the subnatural resonance is optically shifted with respect to the point \( \delta_1 - \delta_2 = 0 \). The sum of such shifted resonances with amplitudes and width dependent \( v_z \) gives a dip. If \( \delta_1 \neq 0 \), the absorption as a function of \( \delta_1 - \delta_2 \) becomes asymmetric (see in fig.3.b).

IV. CONCLUSION

To conclude we note the four-state N-type interaction scheme can be easily organized in real atomic systems. For example, let us consider a closed \( F_g = F \rightarrow F_e = F + 1 \) transition of the \( D_2 \) line of an alkali atom interacting with the \( \sigma_+ \) polarized control field. In the steady state all atoms are completely pumped into the stretched states \( |F_g, m_g = F \rangle \) and \( |F_e, m_e = F + 1 \rangle \). If the probe field has \( \sigma_- \) polarization, then in the first order on the probe field amplitude \( \Omega_2 \) we have \( N \)-atom with the states \( |1\rangle = |F_g, m_g = F - 2 \rangle, \quad |2\rangle = |F_e, m_e = F - 1 \rangle, \quad |3\rangle = |F_g, m_g = F \rangle, \) and \( |4\rangle = |F_e, m_e = F + 1 \rangle \).

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

FIG. 2. The probe field absorption versus the frequency offset in the case of homogeneous broadening \( v = 0 \). The control field detuning \( \delta_1 = 0 \) (a) and \( \delta_1 = 1 \) (b). Solid (dashed) curves correspond to the case of \( b = 1 \) (b = 0). Other parameters are \( A^2 = B^2 = 1/2, \quad \Omega_1 = 0.1 \Gamma, \quad \rho^{(0)}_{11} = 0 \) and \( \rho^{(0)}_{13} = 1 \).

FIG. 3. The probe field absorption versus the frequency offset in the case of Doppler broadening \( k\tau = 10\Gamma \). The control field parameters: detuning \( \delta_1 = 0 \) (a) and \( \delta_1 = 10 \Gamma \) (b), the Rabi frequency \( \Omega_1 = 0.1 \Gamma \); and \( A^2 = B^2 = 1/2 \).
Fig. 1
Fig. 2
Fig. 3

Absorption (arb. units)

\[ \frac{(\delta_1 - \delta_2)}{\Gamma} \]

\[ \frac{(\delta_1 - \delta_2)}{\Gamma} \]