Sub-Doppler Absorption Resonances of Three-Level Quantum Systems in Thin Gas Cells

A. Namdar*, H. Tajalli*, M. Kalafi*, and A. Ch. Izmailov**

* Center for Applied Physics Research, University of Tabriz, Tabriz, Iran
** Institute of Physics, Azeri Academy of Sciences, Baku 370143 Azerbaijan

e-mail: physic@lan.ah.az
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Abstract—Interaction of probe and pump monochromatic waves is analyzed with the Λ-system of three quantum levels of atoms (molecules) in the thin gas cell, whose length is much less than the diameter of light beams. Nontrivial peculiarities were discovered of sub-Doppler absorption resonances, caused by the light-induced coherence of lower long-lived quantum states of the Λ-system. Given resonances may be used for the stabilization of both frequencies of waves and the difference of these frequencies.

With optical pumping the laser beam diameter is usually much smaller than the length of the gas cells [1]. Therefore, the relaxation of particles (atoms, molecules) resulting from the finite time of their flight along a cell is, as a rule, ignored. However, such relaxation may lead to new nontrivial results if the cell length a cell is, as a rule, ignored. However, such relaxation may lead to new nontrivial results if the cell length is much smaller than the beam diameter D of the pumping radiation [2–5]. In reality, we consider quantum transitions from the ground (metastable) atomic (or molecular) state. Then displays of nonlinear optical effects are determined by the transit time τ = l/|v| between walls of the cell for atoms (with velocity projection v on the wave vector), which effectively interact with the pumping radiation. Thus, the time τ of the transit relaxation depends on frequencies of pumping waves with nonhomogeneous broadening of spectral lines of resonance transitions. In consequence nontrivial sub-Doppler resonances arise in the absorption of given waves. Earlier such narrow resonances were investigated in the case of the monochromatic wave interacting with the two-level quantum system [2–5].

At the same time, it is interesting to analyze the interaction of bichromatic radiation with the three-level atomic (molecular) Λ-system (Fig. 1) between the excited quantum state |3⟩ and long-lived states |1⟩ and |2⟩. Then, unlike the case of the two-level system, the coherence is essential between lower levels |1⟩ and |2⟩ [6, 7]. Displays of such coherence for a usual cell (where l >> D) were investigated earlier [6, 7]. However, in the case of the thin cell (where D ≫ l) with a rarefied gas, the relaxation of the coherence between levels |1⟩ and |2⟩ (Fig. 1) will be determined by the transit time τ = l/D|v| of atoms (molecules), which effectively interact with the bichromatic radiation. Therefore, as we show in the given paper, nontrivial sub-Doppler resonances arise in the absorption of the probe wave. These resonances may be used for the stabilization of both frequencies of waves and the difference of these frequencies.

We consider the field ε of two plane monochromatic light waves, propagating through the thin gas cell along the axis z:

\[ ε = \sum_{j=1}^{2} I_j^{1/2} e_j \exp \{i(\omega_j t - k_j z)\} + c. c., \] (1)

where \(\omega_j\) is the frequency, \(e_j\) is the unit polarization vector, \(I_j\) is the intensity, \(k_j\) is the projection of the wave vector (on the axis z) for the wave \(j (j = 1, 2)\). Wave frequencies \(\omega_1\) and \(\omega_2\) are close to centers \(\Omega_{31}\) and \(\Omega_{32}\) of optical transitions |1⟩–|3⟩ and |2⟩–|3⟩ of the Λ-system of levels (Fig. 1), respectively.

It is assumed that the gas in the cell is so rarefied that we can neglect an interaction between atoms (molecules). We consider sufficiently low wave intensities (1), when the following conditions take place:

\[ g_j^2 \ll \gamma^2, \] (2)

where \(g_j = \frac{1}{2} I_j^{1/2} |e_j d_j| \hbar^{-1}\) is the Rabi frequency for the resonance transition |3⟩–|2⟩ with the matrix element \(d_j\) (\(j = 1, 2\)) of the dipole moment and homogeneous half-width \(\gamma\) of the spectral line. Population of the excited level |3⟩ is negligible in comparison with the population of long-lived levels |1⟩ and |2⟩ at conditions (2) [6]. It is convenient also to describe wave intensities (1) by the following dimensionless parameters:

\[ p_j = \frac{g_j^2}{\gamma} \sqrt{k_j l}, \quad (j = 1, 2). \] (3)

We consider the case, when wavelengths of the radiation (1) are much less than the cell length \(l\), that is, \(k_j l \gg 1\) and \(k_j j \gg 1\). Essential nonlinear phenomena take place at the condition \(p_1 + p_2 \gg 1\), which may be realized side by side with restrictions (2).
It is not difficult to receive in our case the following
equations for stationary populations $\rho_{11}$, $\rho_{22}$ and the
cohere $\tilde{\rho}_{12} = \tilde{\rho}_{12} \exp[i(\delta_1 - \delta_2)t - (k_1 - k_2)z]$ of states
the density matrix of the $A$-system (Fig. 1) in the
resonance bichromatic radiation (1):

$$\nu \frac{\partial \rho_{11}}{\partial z} = A_1 \rho_{11} + \gamma_1 B_2 \rho_{22} + L_{12} \tilde{\rho}_{12} + L^*_{12} \tilde{\rho}_{21},$$

$$\nu \frac{\partial \rho_{22}}{\partial z} = A_2 \rho_{22} + \gamma_2 B_1 \rho_{11} + L_{21} \tilde{\rho}_{21} + L^*_{21} \tilde{\rho}_{12},$$

$$\nu \frac{\partial \tilde{\rho}_{12}}{\partial z} = -(M_1 \rho_{11} + M_2 \rho_{22} + M_3 \tilde{\rho}_{12}),$$

where

$$\tilde{\rho}_{21} = \tilde{\rho}_{12}^*, \quad A_m = \frac{2g^2_m(\gamma_m - \gamma)}{\gamma + \xi_m}, \quad B_m = \frac{2g^2_m}{\gamma + \xi_m},$$

$$L_{mn} = \frac{g^2_m g_n}{\gamma} \left( \frac{\gamma_m - \gamma}{\gamma + i\xi_m} + \frac{\gamma_m}{\gamma - i\xi_m} \right) (m, n = 1, 2; m \neq n),$$

$$M_1 = \frac{g_1 g_2}{\gamma + i\xi_1}, \quad M_2 = \frac{g_1 g_2}{\gamma + i\xi_2},$$

$$M_3 = i(\xi_1 - \xi_2) + \frac{g_1^2}{\gamma + i\xi_1} + \frac{g_2^2}{\gamma + i\xi_2},$$

$\xi_j = \delta_j - k_j \nu$, $\delta_j = (\omega_j - \Omega_3)$ is the frequency detuning
from the resonance for the wave $j$ ($j = 1, 2$). Values $2\gamma_1$ and $2\gamma_2$ in (4) are partial rates of the radiative decay
from the level [3] to states [1] and [2] (Fig. 1), respectivey, $(\gamma_1 + \gamma_2) \leq \gamma$. It is necessary to supplement (4)
with boundary conditions for values $\rho_{11}$, $\rho_{22}$, and $\tilde{\rho}_{12}$.

We can assume that equilibrium distributions for both
atomic velocities and populations of quantum levels are
established due to collisions of atoms (molecules) with
walls of the thin cell [2–5]. Then the following boundary
conditions take place for populations $\rho_{11}$, $\rho_{22}$ and the
cohere $\tilde{\rho}_{12}$ of lower levels [1] and [2] at coordinates $z = 0$ and $z = l$ of walls:

$$\rho_{11}(z = 0, \nu > 0) = \rho_{11}(z = l, \nu < 0) = \rho_{11}^{(0)} F(\nu),$$

$$\rho_{22}(z = 0, \nu > 0) = \rho_{22}(z = l, \nu < 0) = \rho_{22}^{(0)} F(\nu),$$

$$\tilde{\rho}_{12}(z = 0, \nu > 0) = \tilde{\rho}_{12}(z = l, \nu < 0) = 0,$$

where $F(\nu)$ is the one-dimensional Maxwellian distri-
bution of atoms on the velocity projection $\nu$ and $\rho_{11}^{(0)}$
and $\rho_{22}^{(0)}$ are the equilibrium populations of levels [1]
and [2], respectivey (in the absence of radiation). It is
not difficult to receive the analytical solution of (4), (5)
in the case of the weak probe wave 1 at the intensity
parameter $p_1 \ll 1$ (3). Then equations (4) have the form

$$\nu \frac{\partial \rho_{11}}{\partial z} = \frac{2\gamma_1 g^2_2}{(\gamma + \xi_1)} \rho_{22},$$

$$\nu \frac{\partial \rho_{22}}{\partial z} = \frac{2(\gamma_2 - \gamma) g^2_2}{(\gamma + \xi_2)} \rho_{22},$$

$$\nu \frac{\partial \tilde{\rho}_{12}}{\partial z} = - \frac{g_1 g_2}{(\gamma + i\xi_1)} \rho_{11}.$$

We receive the following solution of (6) at boundary
conditions (5) (in the region $0 \leq z \leq l$):

$$\rho_{11} = \{\theta(\rho_{11}^{(0)} - 1)[\eta(\nu) + \eta(-\nu)e^{i\beta}]e^{-\beta z}$$

$$+ (1 - \theta)\rho_{11}^{(0)} + \theta \} F(\nu),$$

$$\rho_{22} = \rho_{22}^{(0)}[\eta(\nu) + \eta(-\nu)e^{i\beta}]e^{-\beta z} F(\nu),$$

$$\tilde{\rho}_{12} = \left[ \frac{R_3}{R_3 + (R_3 - \beta)} \right] F(\nu),$$

$$+ S_1[\eta(\nu) + \eta(-\nu)e^{R_1\beta}]e^{-R_1\beta} \right] F(\nu),$$

where $\eta(\nu) = \frac{1}{\cosh^2(\nu)}$. The
above solution is obtained by calculating the solution
of the system of differential equations (4) by the method
of separation of variables.
where $\eta(\nu)$ is the stepped function $[\eta(\nu) = 1$ if $\nu \geq 0$ and $\eta(\nu) = 0$ when $\nu < 0$],

$$\beta = \frac{2g_2^2(\gamma - \gamma_2)}{(\gamma + \xi_2^2)^2}, \quad \theta = \frac{\gamma_1}{(\gamma - \gamma_2)},$$

$$R_3 = \left[i(\xi_1 - \xi_2) + \frac{g_2^2}{\gamma + i\xi_1}\right]\frac{1}{\nu},$$

$$R_5 = -[(1 - \theta)\rho_1^{(0)} + \theta]R_1,$$

$$R_4 = \rho_2^{(0)}[\eta(\nu) + \eta(-\nu)e^{i\theta}][R_1, \theta - R_2],$$

$$R_1 = \frac{g_1g_2}{(\gamma + i\xi_1)^2}, \quad R_2 = \frac{g_1g_2}{(\gamma - i\xi_1)^2},$$

$$S_1 = -\frac{R_5}{R_3} - \frac{(R_1, \theta - R_2)\rho_2^{(0)}}{(R_3 - \beta)}.$$

Further we will analyze the absorption coefficient $\alpha_1$ of the probe wave $1$ (1), which, according to known relationships [6], in our case has the form:

$$\alpha_1 = 4\pi\omega_1|P_{11}|^2 \frac{\hbar c}{I},$$

$$\times \left\{ \gamma P_{11} + g_2 g_1^{-1}[\gamma \text{Re}(P_{12}) + \xi_1 \text{Im}(P_{12})] \right\}$$

$$\times (\gamma^2 + \xi_1^2)^{-1}d\nu,$$

where

$$P_{ij} = \frac{i}{\pi} \int \rho_{ij}dz, \quad (j = 1, 2), \quad P_{12} = \frac{i}{\pi} \int \delta_{12}dz.$$

At the numerical calculation we will plot the value $\alpha_1$ (8) in units of the following linear absorption coefficient $\alpha_0$ of the probe wave $1$ in the thin cell at $\delta_1 = 0$ and $I_2 = 0$:

$$\alpha_0 = \frac{4\pi\omega_1|P_{11}|^2}{\hbar c} \rho_1^{(0)} \int_{-\infty}^{+\infty} \frac{\gamma F(\nu)}{(\gamma^2 + k_2^2\nu^2)}d\nu.$$

According to the Doppler effect, pump and probe waves (1) may induce the essential coherence between long-lived levels $|1\rangle$ and $|2\rangle$ (Fig. 1) only for atoms (molecules) with the definite velocity projections $\nu$.

$$|\delta_1 - \delta_2 - (k_1 - k_2)| \leq \frac{1}{\tau(\nu)} + \frac{\sigma g_2^2}{\gamma} = \Delta,$$

where in our case $\tau(\nu) = l/|\nu|$ is the time of the transit relaxation of given atoms between walls of the thin cell, numerical factor $\sigma = 1$. We note that in (10) the value $\Delta \approx \gamma$ for velocity projections $|\nu| \ll \gamma$ and at intensities of the radiation under consideration (2). Further we will analyze the case of copropagating waves (1) with close frequencies, when $|k_1 - k_2| \ll |k_1|$. Then the coherence condition (10) may be realized for atoms (molecules) with various velocities.

Let us at first consider the closed $\Lambda$-system (Fig. 1), where the excited level $|3\rangle$ may radiatively decay only to the state $|1\rangle$ or $|2\rangle$, that is, $\gamma = (\gamma_1 + \gamma_2)$ in (4), (6). For example, such a situation is typical for atoms of alkali metals, whose ground term consists of two sublevels of the hyperfine structure [8]. Figure 2 shows the dependence of the absorption coefficient $\alpha_1(\delta_1)$ (8) of the probe wave 1 on the frequency detuning $\delta_1 = (\omega_1 - \Omega_{11})$ at the fixed frequency $\omega_2$ of the pump wave 2. In Fig. 2 the Doppler broadening of the spectral line $ku = 50\gamma$, where $k_1 = k_2 = k$. Dependence $\alpha_1(\delta_1)$ at the value $\delta_1 = 0$ has the symmetric sub-Doppler structure near the point $\delta_1 = 0$. This structure consists of the peak with the characteristic width $G \approx \gamma$, and the much more narrow dip (curve I, Fig. 2). The given peak is caused by the growth of the population of the level $|1\rangle$ because of the induced transition $|1\rangle \rightarrow |3\rangle$ (by the pump wave 2) and the following radiative decay on the channel $|3\rangle \rightarrow |1\rangle$ [9].

Such a process is realized in atoms with definite velocity projections $\nu$ [1, 9]:

$$|k_\nu - \delta_1| \leq G \ll ku,$$

where the value $G \geq \gamma$ increases with the growth of the intensity $I_2$. Value $G = \gamma$ at the sufficiently small intensity $I_2$, when the parameter $p_2 \ll 1$ (3). Resonances of the optical pumping arise in the absorption of the probe wave 1, when $|\delta_1 - \delta_2| \approx G$. A much more narrow dip at
the small difference $|\delta_1 - \delta_2|$ (10) is caused by the coherence of states $|1\rangle$ and $|2\rangle$. Taking into account the averaging in velocity projections of atoms, interacting with the resonance radiation (11), we can write on the basis of inequality (10) the following relationship for the region of the coherence dip in the dependence $\alpha_1(\delta_1)$ (8):

$$|\delta_1 - \delta_2| \leq \tilde{\Delta},$$

(12)

where the width $\tilde{\Delta}$ depends on the frequency detuning $\delta_2$. Thus, according to relationships (10) and (11), the width $\tilde{\Delta} = |[\delta_2](kl)^{-1} + \sigma_1^\gamma^{-1}|$ at the detuning $|\delta_2| > G$.

A sharp decrease is the absorption of the wave 1 at the condition (12) is the result of the direct two-quantum transition $|2\rangle \rightarrow |1\rangle$ (without a population of the excited level $|3\rangle$) [7]. Curves 2 and 3 in Fig. 2 correspond to the detuning $|\delta_3| > \gamma$. Then the dependence $\alpha_1(\delta_1)$ has the peak and dip in the vicinity of the value $\delta_1 = \delta_2$, as in the case of curve 1 in Fig. 2. Moreover, curves 2 and 3 in Fig. 2 have the peak near the value $\delta_1 = 0$. The given peak is caused by the more intensive optical repumping of the population on the channel $|2\rangle \rightarrow |3\rangle \rightarrow |1\rangle$ for atoms with the comparatively large time of transit relaxation, which effectively interact exactly with the wave 1 at the sufficiently small detuning $|\delta_1| \leq \gamma$. The growth of the intensity $I_1$ of the pump wave 2 leads to the increase of both amplitudes and widths of all sub-Doppler resonances in Fig. 2 (curves 2 and 3) because of the intensification of nonlinear optical processes. It is important to note that nonlinear resonances in the dependence $\alpha_1(\delta_1)$ decrease with the growth of the detuning $|\delta_2|$ at the fixed intensity $I_2$ (curves 1 and 2 in Fig. 2). Really, the smaller the value $|\delta_3|$ the stronger the displays of nonlinear optical effects because of the growth of the time $\tau = I_1/|\nu|$ of the transit relaxation for atoms (11), which effectively interact with the pump wave 2.

Figure 3 presents the dependence of the absorption coefficient $\alpha_1(\delta_1)$ on the frequency detuning $\delta_1$ at the fixed frequency difference $(\omega_2 - \omega_0)$ (1). We see the rise of the sub-Doppler structure at the small detuning $|\delta_3| \leq G$ (11). Given resonances are caused by the intensification of nonlinear optical processes for atoms with the comparatively large time of transit relaxation, which interact with the radiation at the decrease of the value $|\delta_3|$. Dependence $\alpha_1(\delta_1)$ peaks in the region $|\delta_3| \leq G$ (curves 2 in Fig. 3) at the breach of the coherence condition (12). The given peak is connected with the amplification of the population, repumping from the state $|2\rangle$ to the state $|1\rangle$ (Fig. 1) because of the excitation on the transition $|2\rangle \rightarrow |3\rangle$ and the following radiative decay on the channel $|3\rangle \rightarrow |1\rangle$. At the same time, dependence $\alpha_1(\delta_1)$ has the sub-Doppler dip in the region $|\delta_3| \leq G$ at equal detunings $\delta_1 = \delta_3$ (curves 1 and 5 in Fig. 3). This dip is caused by the decrease of the absorption of the wave 2 because of the coherence of states $|1\rangle$ and $|2\rangle$ at $\delta_1 = \delta_3$ (12). Thus, the sub-Doppler structure of the dependence $\alpha_1(\delta_1)$ may radically change at variations of the difference $|\delta_1 - \delta_2|$ in the narrow interval from 0 to the value $\tilde{\Delta}$ (12). We note that at the condition $0 < |\delta_2 - \delta_1| < \tilde{\Delta}$ the dependence $0 < |\delta_2 - \delta_1| < \tilde{\Delta}$ has both the peak and the dip, which are shifted from the point $\delta_1 = 0$ (curves 3 and 4 in Fig. 3). Discovered peculiarities of sub-Doppler resonances in thin gas cells (Figs. 2 and 3) may be used for the stabilization of both frequencies of waves (1) and their difference $(\omega_2 - \omega_0)$. Growth of the pump intensity leads to the increase of given sub-Doppler resonances (curves 1 and 5 in Fig. 3).

Now let us analyze also the case of an open $\Lambda$-system (Fig. 1), where probabilities of the radiative decay of the excited state $|3\rangle$ on initial long-lived levels $|1\rangle$ and $|2\rangle$ are negligible, that is $\gamma \ll \gamma_1 + \gamma_2$. For example, such a situation is typical for molecules, whose ground term consists of many sublevels [8]. We will consider the case when the state $|1\rangle$ has a little more energy than the state $|2\rangle$ (Fig. 1). Therefore, the relation $\rho_2^{(0)} \gg \rho_1^{(0)}$ takes place for equilibrium populations of given levels. Then, unlike the case of the closed $\Lambda$-system (Fig. 2), the dependence $\alpha_1(\delta_1)$ (Fig. 4) has no peak caused by the optical repumping of the population from the level $|2\rangle$ to the level $|1\rangle$ by means of the radiative decay of the excited state $|3\rangle$ (Fig. 1). At the same time, the dependence $\alpha_1(\delta_1)$ for the open $\Lambda$-system (Fig. 4) has a narrow dip in the region $|\delta_1 - \delta_2| \leq \tilde{\Delta}$ (12), where the coherence of states $|1\rangle$ and $|2\rangle$ is realized. Dependences of the amplitude and width of given coherence reso-
Fig. 4. Absorption coefficient $\alpha_1(\delta_1)$ of the probe wave versus its frequency detuning $\delta_1$ at the fixed frequency $\omega_0$ of the pump wave. Case of the open $\Lambda$-system of levels, when $(\gamma_1 + \gamma_2) \ll \gamma$, $k_2 = 1.01k_1$, $k_1u = 50\gamma$, $k_1l = 100$, $p_2^{(0)} = 19p_1^{(0)}$, $p_1 = 0.03$, $(I$ and $2)$ $p_2 = 0.5$, $(3)$ $1$, $(I)$ $\delta_2/\gamma = 0$, $(2)$ $3$, $(3) -3$.

Fig. 5. Absorption coefficient $\alpha_1(\delta_1)$ of the probe wave versus its frequency detuning $\delta_1$ at the fixed frequency difference $(\omega_2 - \omega_1)$. A case of the open $\Lambda$-system of levels, when $(\gamma_1 + \gamma_2) \ll \gamma$, $k_2 = 1.01k_1$, $k_1u = 50\gamma$, $k_1l = 100$, $p_2^{(0)} = 19p_1^{(0)}$, $p_1 = 0.03$, $(I, 3–5)$ $p_2 = 0.5$, $(2)$ $0.25$, $(I$ and $2)$ $(\delta_2 - \delta_1)/\gamma = 0$, $(3) 0.025$, $(4) -0.025$, $(5) 0.25$.

Absorption coefficient $\alpha_1(\delta_1)$ of the probe wave. Case of the open $\Lambda$-system (Fig. 2). However, we note that the absorption coefficient $\alpha_1(\delta_1)$ may be negative for the open $\Lambda$-system in the region of the coherence dip (curves 1 and 3 in Fig. 4). Really, the direct two-quantum transition $|2 \rightarrow 1|$ may lead to the amplification of the wave 1 at the population difference $p_2^{(0)} \gg p_1^{(0)}$.

Figure 5 presents the dependence of the absorption coefficient $\alpha_1(\delta_1)$ on the frequency $\delta_1$ at the fixed frequency difference $(\omega_2 - \omega_1)$. Dependence $\alpha_1(\delta_1)$ has the sub-Doppler dip in the region $|\delta_1| \ll G$ (11) at equal detunings $\delta_1 = \delta_2$ (curves 1 and 2 in Fig. 5), which is caused by the coherence of states $|1\rangle$ and $|2\rangle$. The given dip disappears at a breach of the coherence condition (12) (curve 5 in Fig. 5). We see that the sub-Doppler structure of the dependence $\alpha_1(\delta_1)$ radically changes at small variations of the difference $|\delta_2 - \delta_1|$ from 0 to $\Delta$ (Fig. 5). It is important to note that the absorption coefficient $\alpha_1(\delta_1)$ may be negative in the region of sufficiently small frequency detuning $|\delta_1| < G$. Thus, the amplification of the probe radiation is possible in the definite frequency interval $|\delta_1| < G$ because of the coherence of long-lived states $|1\rangle$ and $|2\rangle$. The given interval increases at the growth of the intensity $I_2$ of the pump wave 2 (curves 1 and 2 in Fig. 5).

The mathematical model used in the given work does not take into account the fact that transverse dimensions of light beams are finite. However, according to previous investigations of optical effects in thin cells [2, 3], results obtained here are valid if the diameter $D$ of light beams satisfies the condition $D \geq l|k_1| + |k_2|u/\gamma$. The last condition signifies that the time required for a particle to travel across light beams is large enough for the optical pumping of long-lived states. We note that ultrathin gas cells with length $l \sim 10 \mu m$ were manufactured and used in experiments [4, 5]. Therefore the results of this paper may be used at usual diameters of laser beams $D \sim 1$ mm even at the ratio $\gamma/((|k_1| + |k_2|)u) \sim 10^{-2}$.

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