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MOLECULAR SPECTROSCOPY

Fluorescence and Absorption Spectra of the Λ-System in the Case of Raman Resonance

I. V. Bargatin and B. A. Grishanin

International Laser Center, Faculty of Physics, Moscow State University, Moscow, 119899 Russia

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Abstract—Analytic formulas are obtained that describe the fluorescence and absorption spectra of the Λ-system in the case of Raman resonance for unequal intensities of the fields and nonzero one-photon detuning in the rotating wave approximation in the limit of the saturating field. It is shown that, along with a group of Lorentzian lines, the spectra contain an alternating non-Lorentzian part caused by the quantum specificity of the dynamics of the Λ-system, which is of fundamental importance for an adequate explanation of important physical features, such as the decay of spectral line wings, the position of maxima of spectral lines, and the behavior of the absorption coefficient of a probe field near laser frequencies. The relation between theoretical models describing the stationary and nonstationary dynamics of formation of the response of individual atoms is considered, and the possibility of the universal use of the phenomenological stationary model is substantiated.

INTRODUCTION

One of the most interesting and extensively studied phenomena observed in three-level systems is the coherent population trapping (CPT) (see reviews [1, 2]). This effect manifests itself most distinctly in the Λ-configuration of the three-level systems, where dipole transitions between the upper and each of the two lower energy levels are allowed (Fig. 1). Upon excitation of such a system by two lasers at frequencies ω_L and ω'_L in the case of the Raman resonance $\delta_R = \omega'_L - \omega_L - \omega_{12} = 0$ [where δ_R is the Raman (two-photon detuning) and ω_{12} is the frequency of the transition between two lower levels], virtually the total population proves to be concentrated in the coherent superposition of the two lower states (the dark state), which does not interact with the laser field. Upon detuning the laser frequency near the Raman resonance, the CPT is manifested, in particular, as a narrow hole in the absorption and fluorescence bands.

The resonance fluorescence and absorption spectra of the probe field can be studied using three different experimental setups, which require different theoretical descriptions. In the first case, an atomic gas of the active substance is studied in a cell with a buffer gas [3, 4], the elastic dephasing of the active atoms being determined by collisions with atoms of the buffer gas. Because the $|1\rangle$ and $|2\rangle$ levels in the Λ-system have the same parity and transitions between them are forbidden in the dipole approximation, the corresponding radiative relaxation parameters are negligible and relaxation is determined by collisions with atoms of the buffer gas and cell walls. Therefore, if the effect of collisions on the population decay of the two lower levels is neglected, the only relaxation parameter, which should be taken into account for the construction of the adequate theory, is the rate of elastic dephasing Γ_{12}. Under such conditions, as shown below, the fluorescence intensity is proportional to Γ_{12}, because an emitting atom is found in a stationary state, which differs from the dark state only when $\Gamma_{12} \neq 0$.

In the second case, a buffer gas is absent [5], and the characteristic time of the elastic dephasing of the $|1\rangle \longleftrightarrow |2\rangle$ transition caused by collisions in pure vapors of active atoms is much longer than the time of

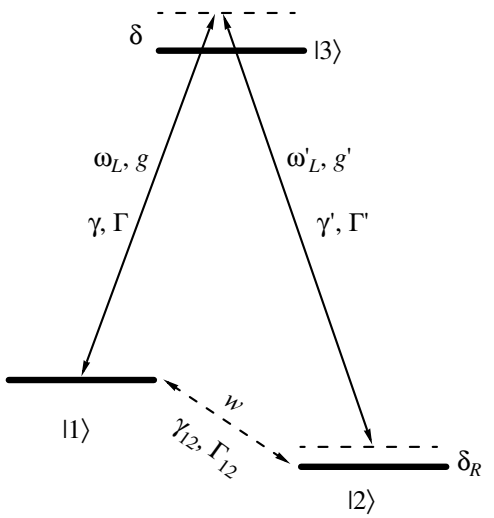

Fig. 1. Configuration and parameters of the Λ-system excited by two monochromatic laser fields at frequencies ω_L and ω'_L; γ, γ', and γ_{12} are relaxation rates of the upper levels; Γ_{13}, Γ_{23}, Γ_{12} are dephasing rates; and w is the rate of incoherent pumping to the level 1.

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flight through a laser beam. In this case, the time dependence of the Rabi frequencies entering the Liouvillian plays a decisive role, and the stationary density matrix can no longer describe the state of an individual atom.

In the third case, a beam of active atoms is produced, which interacts with a laser beam in the region of their intersection [6]. The difference of this variant from the preceding one consists in a rigid fixation of atomic velocities and, thereby, detunings of laser frequencies. This results in an absence of the Doppler broadening, which simplifies the theoretical description of experiments.

In this paper, we performed analytic calculations of fluorescence and absorption spectra of the Λ-system in the case of the Raman resonance under the continuous action of laser fields. In the first two cases discussed above, the calculation of experimental spectra should be carried out taking into account the Doppler shift $\delta \longrightarrow \delta + \mathbf{kv}$, where $\mathbf{k}$ is the wave vector and $\mathbf{v}$ is the velocity of an atom. In addition, the residual two-photon Doppler shift $\delta_R \longrightarrow \delta_R + (\mathbf{k}_L - \mathbf{k}'_L)\mathbf{v}$ can result in the violation of conditions of the Raman resonance for a fraction of atoms, which makes the theoretical description presented below invalid. However, the width of the Raman resonance substantially increases in sufficiently intense fields [7] and it can be sufficiently large for all the atoms interacting with the laser field to be under conditions of the Raman resonance. Our calculations describe the resonance fluorescence spectra for *individual* atoms only in strong fields, when the generalized Rabi frequency greatly exceeds the one-photon detuning. If such a condition is satisfied for all the atoms in a cell, the observed spectrum can be calculated by averaging the spectra of individual atoms over the Doppler distribution of atomic velocities in a cell. In the third case, the detuning δ is the same for all the atoms in well-collimated atomic beams, and the observed spectrum coincides with the spectrum of an individual atom.

We obtained analytic formulas of the stationary theory corresponding to the first experimental setup. The formulas describe the fluorescence power spectrum and the dependence of the absorption coefficient on the probe field frequency in the rotating wave approximation (RWA) in the limit of the saturating field. The cases of unequal intensities of the fields and nonzero one-photon detuning are considered, which generalize the results [8]. A qualitatively new feature of the calculation is a consideration of the non-Lorentzian contribution, which allowed us to find important features of wings of the fluorescence band and of the behavior of the absorption coefficient near laser frequencies.

In the case of the second and third experimental setups, we obtained expressions for the fluorescence spectrum based on the dynamic theory considering atoms flying through a laser beam with a rectangular intensity profile, and compared the results of the stationary and dynamic theories.

LIOUVILLIAN OF THE Λ-SYSTEM IN THE ROTATING WAVE APPROXIMATION

The Hamiltonian of a system excited by two laser fields can be represented in the form

$$\begin{aligned}\hat{\mathcal{H}} &= -\hbar\omega_{12}|2\rangle\langle 2| + \hbar\omega_{13}|3\rangle\langle 3| \\ &\quad + \hbar g\cos(\omega_L t + \varphi)|1\rangle\langle 3| \\ &+ \hbar g'\cos(\omega'_L t + \varphi')|2\rangle\langle 3| + \text{H.c.},\end{aligned} \tag{1}$$

where the energy of the level $|1\rangle$ is set at zero, so that the projection operator $|1\rangle\langle 1|$ is absent in the Hamiltonian. The interaction constants g and g', i.e., the Rabi frequencies, depend on amplitudes A_{ω_L} and $A_{\omega'_L}$ of an external field and dipole matrix elements d_{13} and d_{23}: $g = \frac{1}{\hbar} d_{13} A_{\omega_L}$, $g' = \frac{1}{\hbar} d_{23} A_{\omega'_L}$. The only case of interest is of one-photon resonances, when ω_L and ω'_L are close to ω_{13} and ω_{23}, respectively. Then, by passing to the interaction representation, in which the unperturbed dynamics is described by the unitary transformation $\mathcal{U}_0(t) = \exp[-(i/\hbar)(\hbar\omega_L|3\rangle\langle 3| - \hbar\Delta|2\rangle\langle 2|)t]$, where $\Delta = \omega'_L - \omega_L \approx \omega_{12}$ is the biharmonic frequency detuning, we neglect in the RWA [9], the rapidly oscillating terms in $\hat{\mathcal{H}}_\Lambda = \mathcal{U}_0^{-1}(t)\hat{\mathcal{H}}\mathcal{U}_0(t)$, so that the Hamiltonian (1) takes the form

$$\begin{aligned}\hat{\mathcal{H}}_\Lambda &= \hbar\Big[-\delta|3\rangle\langle 3| + \delta_R|2\rangle\langle 2| \\ &+ \left(\frac{g}{2}|1\rangle\langle 3| + \frac{g'}{2}|2\rangle\langle 3| + \text{H.c.}\right)\Big],\end{aligned} \tag{2}$$

where $\delta = \omega_L - \omega_{13}$ and $\delta_R = \omega'_L - \omega_L - \omega_{12}$ describe the one-photon detuning for the $|1\rangle \longrightarrow |3\rangle$ transition and two-photon Raman detuning, respectively.

The dynamic part of the Liouvillian corresponding to the RWA Hamiltonian (2) has the form $\mathcal{L}_\Lambda = (i/\hbar)[\hat{\mathcal{H}}_\Lambda, \odot]$, and the total Liouvillian $\mathcal{L}_{\text{RWA}}$ includes the relaxation operator. In the case of the symmetrical Λ-system ($\gamma' = \gamma$ and $\Gamma' = \Gamma$) and the Raman resonance $\delta_R = 0$, we obtain in the Hermitian basis $\{\hat{e}_k\} = \{|3\rangle\langle 3|, |1\rangle\langle 1|, |2\rangle\langle 2|, (|1\rangle\langle 2| + |2\rangle\langle 1|)/\sqrt{2}, -i(|1\rangle\langle 2| - |2\rangle\langle 1|)/\sqrt{2}, (|1\rangle\langle 3| + |3\rangle\langle 1|)/\sqrt{2}, -i(|1\rangle\langle 3| - |3\rangle\langle 1|)/\sqrt{2}, (|2\rangle\langle 3| + |3\rangle\langle 2|)/\sqrt{2}, -i(|2\rangle\langle 3| - |3\rangle\langle 2|)/\sqrt{2}\}$.

$$\mathcal{L}_{\mathrm{RWA}} = \begin{pmatrix} -2\gamma & \gamma & \gamma & 0 & 0 & 0 & g/\sqrt{2} & 0 & g'/\sqrt{2} \\ 0 & -\gamma_{12} & \gamma_{12} & 0 & 0 & 0 & -g/\sqrt{2} & 0 & 0 \\ 0 & w & -w & 0 & 0 & 0 & 0 & 0 & -g'/\sqrt{2} \\ 0 & 0 & 0 & -\Gamma_{12} & 0 & 0 & -g'/2 & 0 & -g/2 \\ 0 & 0 & 0 & 0 & -\Gamma_{12} & g'/2 & 0 & -g/2 & 0 \\ 0 & 0 & 0 & 0 & -g'/2 & -\Gamma & \delta & 0 & 0 \\ -g/\sqrt{2} & g/\sqrt{2} & 0 & g'/2 & 0 & -\delta & -\Gamma & 0 & 0 \\ 0 & 0 & 0 & 0 & g/2 & 0 & 0 & -\Gamma & \delta \\ -g'/\sqrt{2} & 0 & g'/\sqrt{2} & g/2 & 0 & 0 & 0 & -\delta & -\Gamma \end{pmatrix}. \quad (3)$$

Here, γ is the rate of the population decay of the level $|3\rangle$ to levels $|1\rangle$ and $|2\rangle$; Γ is the dephasing rate for the $|1\rangle \longleftrightarrow |3\rangle$ and $|2\rangle \longleftrightarrow |3\rangle$ transitions; and Γ_{12}, γ_{12}, and w are the rates of dephasing, decay, and incoherent pumping in the system of lower levels $|1\rangle$ and $|2\rangle$. The quantity Γ_{12} can describe collisional dephasing and other equivalent mechanisms, for example, the phase instability of laser pumping.

CALCULATION OF FLUORESCENCE SPECTRA

The spectral density of radiation of an excited atom is determined by the normally ordered, two-time correlation function of radiation emitted by the atom [10, 11]. By assuming the Markov behavior of atomic fluctuations, i.e., the independence of noise at moments t and $t + \tau$, the correlation function can be written in the form [12]

$$\mathcal{H}(\tau) = \langle \hat{\rho}_0 S(0, t) | \hat{\sigma}^-(t)[S(t, t+\tau)\hat{\sigma}^+(t+\tau)] \rangle, \quad (4)$$

where $\hat{\sigma}^{\pm}(t)$ are positive- and negative-frequency Heisenberg operators for the $|k\rangle \longleftrightarrow |3\rangle$ atomic transitions ($k = 1, 2$). Superoperators $S(t_1, t_2) = T\exp[\int_{t_1}^{t_2} \mathcal{L}(\tau) d\tau]$, where T is the symbol of the time ordering, describe the relaxation and interaction of the atom with exciting laser fields during time intervals $(0, t)$ and $(t, t + \tau)$, respectively, and $\hat{\rho}_0 S(0, t)$ represents the density matrix $\hat{\rho}(t)$ at the moment t. The emission spectrum can be calculated as the Fourier transform of the correlation function (4) (Fig. 2a).

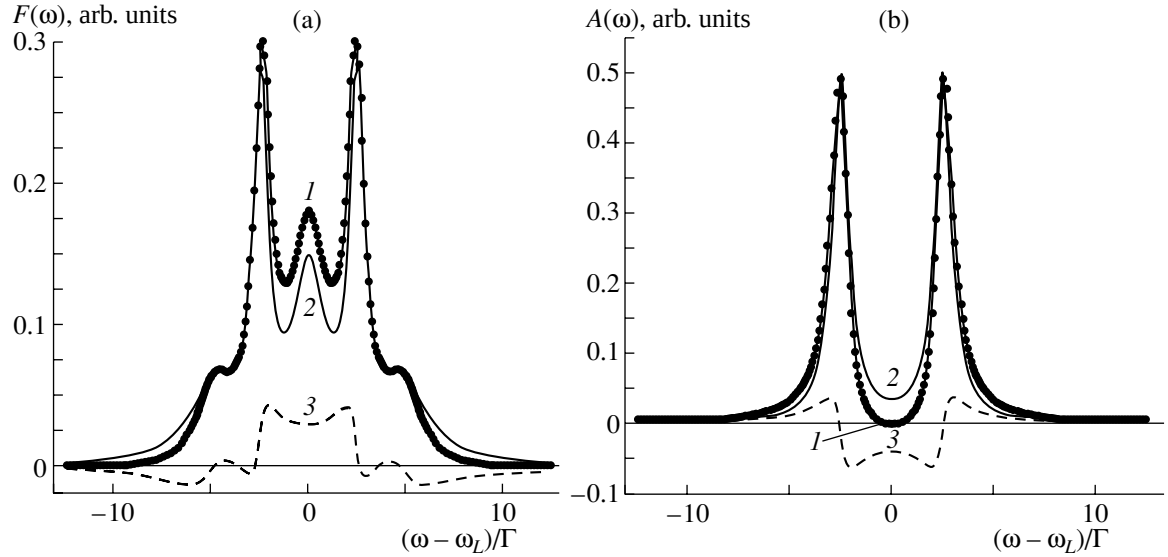

Fig. 2. (a) Fluorescence and (b) absorption spectra. (*1*) The total spectrum; (*2*) and (*3*) are Lorentzian and non-Lorentzian parts of the spectrum, respectively. Parameters of the system: $g_\Lambda = 5\Gamma$, $\delta = 0$, $\gamma = \Gamma$, $\Gamma_{12} = 0.001\Gamma$, $\varphi = \pi/2$.

Stationary Theory of Fluorescence Spectra

In the stationary theory, the density matrix in the vector representation is a zero vector $\langle 0|$ of matrix (3) of the RWA Liouvillian $\mathcal{L}_{\text{RWA}}$. By describing the dynamics of the Λ-system with the help of the Liouvillian $\mathcal{L}_{\text{RWA}}$ and expanding it in eigenprojectors, we obtain for the correlation function (4) the expression

$$\mathcal{H}(\tau) = \sum_{k=0}^{8}\left[C_k^{13}e^{(\lambda_k - i\omega_L)\tau} + C_k^{23}e^{\left(\lambda_k - i\omega_L'\right)\tau}\right]. \quad (5)$$

Here, C_k^{l3}, (l = 1, 2) are the intensity coefficients defined by expressions

$$\begin{aligned} C_k^{13} &= \langle 0|\hat{\sigma}_{13}^- \cdot |k\rangle\rangle\langle\langle k|\hat{\sigma}_{13}^+\rangle, \\ C_k^{23} &= \langle 0|\hat{\sigma}_{23}^- \cdot |k\rangle\rangle\langle\langle k|\hat{\sigma}_{23}^+\rangle, \end{aligned} \quad (6)$$

where a symbol "·" means the multiplication of operands according to the multiplication rules for the 3×3 matrices describing atomic operators and the representation of the result as a ket vector; and λ_k, $|k\rangle\rangle$, and $\langle\langle k|$ are eigenvalues and eigenvectors of the $\mathcal{L}_{\text{RWA}}$ matrix. The power spectrum can be written in the form

$$\mathcal{F}(\omega) = 2\text{Re}\left[\sum_{k=0}^{8}\frac{C_k^{13}}{i(\omega-\omega_L)+\lambda_k} + \frac{C_k^{23}}{i(\omega-\omega_L')+\lambda_k}\right]. \quad (7)$$

Nine eigenvalues determine the maximum number of lines in the fluorescence spectrum of the three-level atom. However, as follows from (15), under the condition of the Raman resonance, the spectrum exhibits, in the general case, two groups of five lines each, because only five coefficients C_k^{l3} are nonzero for each of the transitions under study (l = 1, 2). In the case of the nonzero Raman detuning, the spectrum exhibits two groups consisting of seven lines each [13].

Dynamic Theory of Fluorescence in the Case of Instantaneous Variation of the Liouvillian

The model assumes that atoms are irradiated by an electromagnetic pulse of duration T_p corresponding to the time of flight of the atom through a laser beam with the rectangular intensity profile. At the leading edge of the pulse, the transient process proceeds for the time of the order of $1/\gamma$, during which the density matrix transfers from the initial state $\hat{\rho}_0$ to the state $\hat{\rho}_{\text{st}}$ determined by the condition $\langle\langle\hat{\rho}_{\text{st}}|\mathcal{L}_{\text{RWA}} = 0$. During the time between successive entries of the atom to the region of the laser beam, the atom experiences numerous collisions with other atoms and cell walls. This suggests that during the successive entry to the region of the laser beam, the atom is in the thermodynamic equilibrium with the environment, and the corresponding density matrix is described by nonzero matrix elements $\rho_{11} = n$, $\rho_{22} = 1 - n$, with $n = \exp(-\hbar\omega_{12}/k_BT_{\text{eff}})[1 + \exp(-\hbar\omega_{12}/k_BT_{\text{eff}})]^{-1}$ corresponding to the effective temperature T_{eff}. During the transient process, the atom emits a light pulse with the spectrum to be calculated.

In accordance with the finite time T_p of generation of radiation, the spectrum is defined as radiation of an emitter modulated by a rectangular pulse of the unit amplitude in the interval $(0, T_p)$

$$\mathcal{F}(\omega) = \left\langle \mathcal{N}\left|\int_0^{T_p}\hat{\sigma}^+(t)e^{i\omega t}dt\right|^2\right\rangle, \quad (8)$$

where $\hat{\sigma}^+(t)$ describes the stochastic positive-frequency amplitude of the quantum vibrational mode, $\mathcal{N}$ is the normal ordering, and $\langle\cdot\rangle$ means averaging over fluctuations of the bath. By uncovering the ordered square of the modulus and integrating, we obtain in the most interesting case $T_p \gg 1/\gamma$

$$\mathcal{F}_{\text{ns}}(\omega) = 2\text{Re}\sum_{k=0}^{8}\sum_{\lambda_j\neq 0}\frac{1}{\lambda_j} \times\left[\frac{C_{jk}^{13}}{i(\omega-\omega_L)+\lambda_k} + \frac{C_{jk}^{23}}{i(\omega-\omega_L')+\lambda_k}\right], \quad (9)$$

where coefficients C_{jk}^{l3} are expressed in terms of the eigenvectors of the Liouvillian using the modified expressions (6) $C_{jk}^{l3} = \langle\hat{\rho}_0|j\rangle\rangle\langle\langle j|\hat{\sigma}_{l3}^- \cdot |k\rangle\rangle\langle\langle k|\hat{\sigma}_{l3}^+\rangle$, l = 1, 2.

By neglecting the free decay of polarization of the lower system of levels, i.e., for $\Gamma_{12} = 0$, the atom does not emit radiation in the stationary state, and the fluorescence detected represents the incoherent superposition of radiation pulses from individual atoms. If we assume that the pulses come to a photodetector with the average period T, then the spectral intensity averaged in time is described by the expression

$$\mathcal{F}_{\text{avr}}(\omega) = \Gamma_T\mathcal{F}_{\text{ns}}(\omega), \quad (10)$$

where $\Gamma_T = 1/T$ plays the role of the new effective relaxation parameter.

CALCULATION OF ABSORPTION SPECTRA

The probability density of absorption of a probe-field photon of frequency ω near the absorption resonance is represented by the Fourier transform of the asymmetric part of the atomic correlation function—

the average commutator [8, 14]

$$P(\omega) = g_{\rm pr}^2 \mathcal{A}(\omega), \quad \mathcal{A}(\omega) = \int_{-\infty}^{\infty} \mathcal{C}(\tau) e^{i\omega\tau} d\tau. \quad (11)$$

Here, $g_{\rm pr}$ is the Rabi frequency corresponding to the probe field, and

$$\mathcal{C}(\tau) = \langle [\sigma^+(t), \sigma^-(t+\tau)] \rangle = \langle \sigma^+(t)\sigma^-(t+\tau) \rangle - \langle \sigma^-(t+\tau)\sigma^+(t) \rangle.$$

In the stationary case, results are similar to (7):

$$\mathcal{A}(\omega) = 2g_{\rm pr}^2 {\rm Re}\left[\sum_{k=0}^{8} \frac{D_k^{13}}{i(\omega - \omega_L) + \lambda_k} + \frac{D_k^{23}}{i(\omega - \omega_L') + \lambda_k} \right], \quad (12)$$

where coefficients D_k^{l3}, $l = 1, 2$ are defined by expressions

$$D_k^{l3} = \langle 0|\hat{\sigma}_{l3}^- \cdot |k\rangle\langle k|\hat{\sigma}_{l3}^+\rangle - \langle 0|\hat{\sigma}_{l3}^+ \cdot |k\rangle\langle k|\hat{\sigma}_{l3}^-\rangle. \quad (13)$$

RESULTS OF ANALYTIC CALCULATIONS

To perform analytic calculations using the general relations presented above, it is necessary to find the eigenvalues and eigenvectors of the Liouvillian (3). In the general case, this problem is very complicated, however, it can be solved in the limit of the saturating field $g_\Lambda = \sqrt{g^2 + g'^2} \gg \gamma, \Gamma, \delta$ with the help of computer analytic calculations using the perturbation theory. A part of the $\mathcal{L}_{\rm RWA}$ matrix, containing only asymptotically large terms $\sim g, g'$ corresponding to the interaction with the laser field, is treated as the unperturbed Liouvillian, while the rest of the total Liouvillian $\mathcal{L}_{\rm RWA}|_{g = g' = 0}$ is considered as a small perturbation. In [8, 12], only the leading zero-order asymptotics was considered. Here, we also calculated the first-order contributions over the parameter $(\gamma + \delta)/g_\Lambda$.

By neglecting the population decay of the lower levels ($\gamma_{12} \longrightarrow 0$, $w \longrightarrow 0$) and assuming that $\Gamma_{12} \ll \Gamma$, we find the eigenvalues of the Liouvillian (3):

$$\lambda_0 = 0, \ \lambda_1 = -\frac{\gamma}{2}, \ \lambda_2 = -\Gamma,$$

$$\lambda_3 = ig_\Lambda - \frac{3\gamma}{4} - \frac{\Gamma}{2},$$

$$\lambda_4 = -ig_\Lambda - \frac{3\gamma}{4} - \frac{\Gamma}{2}, \ \lambda_5 = i\frac{g_\Lambda + \delta}{2} - \frac{\Gamma}{2}, \quad (14)$$

$$\lambda_6 = i\frac{g_\Lambda - \delta}{2} - \frac{\Gamma}{2}, \ \lambda_7 = i\frac{-g_\Lambda + \delta}{2} - \frac{\Gamma}{2},$$

$$\lambda_8 = i\frac{-g_\Lambda - \delta}{2} - \frac{\Gamma}{2}.$$

Fluorescence in the Stationary Case

In the approximations used, we obtain from (6)

$$C_0^{13} = C_0^{23} = C_1^{13} = C_1^{23} = 0,$$

$$C_2^{13} = \frac{\Gamma_{12}}{\gamma}\cos^4\varphi\sin^2\varphi,$$

$$C_2^{23} = \frac{\Gamma_{12}}{\gamma}\cos^2\varphi\sin^4\varphi,$$

$$C_3^{13} = \frac{\Gamma_{12}}{2\gamma}\cos^4\varphi\sin^2\varphi\left(1 + \frac{i(2\Gamma - 11\gamma)}{64g_\Lambda}\right),$$

$$C_4^{13} = (C_3^{13})^*,$$

$$C_3^{23} = \frac{\Gamma_{12}}{2\gamma}\cos^2\varphi\sin^4\varphi\left(1 + \frac{i(2\Gamma - 11\gamma)}{64g_\Lambda}\right), \quad (15)$$

$$C_4^{23} = (C_3^{23})^*,$$

$$C_5^{13} = C_5^{23} = C_7^{13} = C_7^{23} = 0,$$

$$C_6^{13} = \frac{\Gamma_{12}}{\gamma}\cos^4\varphi\sin^2\varphi\left(1 - \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right),$$

$$C_6^{23} = \frac{\Gamma_{12}}{\gamma}\cos^2\varphi\sin^4\varphi\left(1 - \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right),$$

$$C_8^{13} = \frac{\Gamma_{12}}{\gamma}\cos^4\varphi\sin^2\varphi\left(1 + \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right),$$

$$C_8^{23} = \frac{\Gamma_{12}}{\gamma}\cos^2\varphi\sin^4\varphi\left(1 + \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right).$$

Here, $\cos\varphi = g/g_\Lambda$ and $\sin\varphi = g'/g_\Lambda$. Spectrum (7), as coefficients (15), is proportional to the dephasing rate Γ_{12} of the lower system of levels.

This is reflected by the CPT effect under conditions being studied, because the ratio Γ_{12}/γ under usual experimental conditions is several orders of magnitude less than unity (10^{-2}–10^{-5}). Consider separately each of the factors affecting the shape of the spectrum.

Inaccuracy of the resonance ($\delta \neq 0$). The effect of the nonzero one-photon detuning is manifested, first, in the additional shift of the inner lines, shifted by $\pm g_\Lambda/2$, to the atomic lines. Second, the symmetry in the line intensities is violated, which are proportional, according to (7), to real parts of coefficients C_k. The intensity of the lines shifted to the laser frequency increases, and *vice versa.* (Fig. 3). It is interesting that in the two-level atom, corrections to the coefficients C_k and shifts ${\rm Im}\lambda_k$ related to the resonance inaccuracy are only of the second order in δ/g_Λ. Thus, the Λ-system proves to be more sensitive to the one-photon detuning than the two-level atom. The additional shift of the inner lines results in their Doppler broadening already in a linear approximation (for the rest of the lines, these corrections are

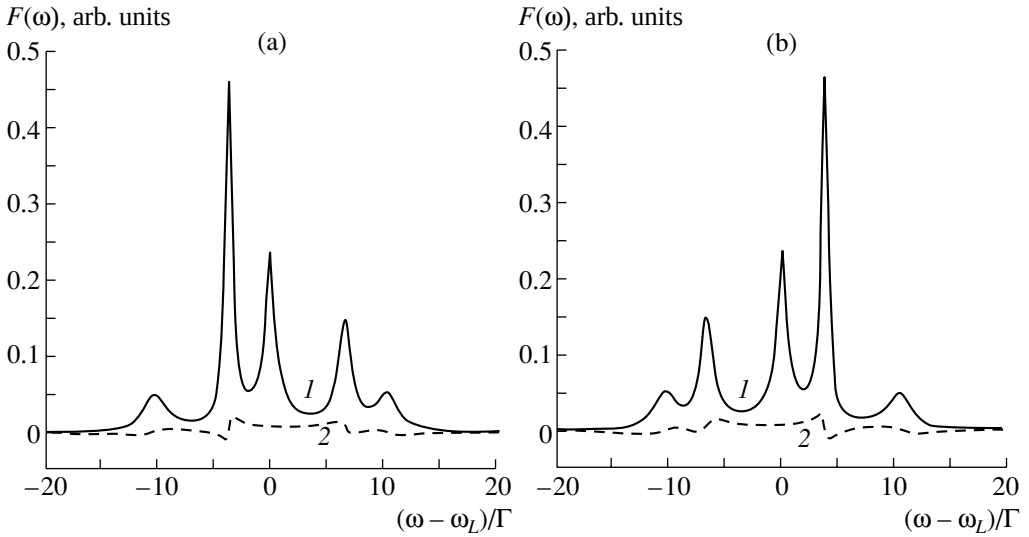

Fig. 3. Fluorescence spectrum at both transitions of the Λ-system in the case of the inexact resonance. (*1*) Total spectrum; (*2*) non-Lorentzian part. Parameters of the system: $g_\Lambda = 10\Gamma$, $\varphi = \pi/4$, $\gamma = \Gamma$, $\Gamma_{12} = 0.001\Gamma$; (a) $\delta = -3\Gamma$; (b) $\delta = 3\Gamma$.

quadratic as minimum), so that in sufficiently strong fields $g_\Lambda \gg |\mathbf{k}|\sqrt{k_B T/m}$, only the inner lines will show the appreciable Doppler broadening.

Dependence on the ratio of laser-field intensities. One can see from (15) that the line intensities depend only on the ratio of the Rabi frequencies characterizing laser-field intensities, which is reflected in the transition saturation by the laser field. The lines shifted by $\pm g_\Lambda$ and the unshifted line corresponding to the response of the two-level atom to the saturating laser field depend on the intensity ratio differently than the lines shifted by $\pm g_\Lambda/2$. In particular, when one of the Rabi frequencies is much higher than another one (let for definiteness $g \gg g'$), the $|1\rangle \longleftrightarrow |3\rangle$ transition spectrum exhibits only three components corresponding to the response of the two-level atom (Fig. 4). At the same time, the $|2\rangle \longleftrightarrow |3\rangle$ transition spectrum exhibits only two lines shifted by $\pm g_\Lambda/2$, i.e., the Autler–Towns structure [15]. Thus, the total spectrum observed, for example, in the case of equal fields and consisting of five lines, is distributed over the transition frequencies. At the transition where the laser field is mainly concentrated, three lines remain which correspond to the response of the two-level atom, while at another transition, two lines shifted by $\pm g_\Lambda/2$ remain.

Non-Lorentzian distribution. One can see from expression (7) that the total spectrum of the fluorescence power at each of the transitions contains, along with Lorentzian lines whose intensity is determined by the real parts of coefficients C_k, a Lorentzian part whose intensity is proportional to the imaginary parts of these coefficients, and the frequency dependence is similar to that for dispersion. Unlike the Lorentzian part, which is positive for $\mathrm{Re}\, C_k > 0$, this spectral contribution has no definite sign, although the total spectrum is, of course, always positive. It follows from the form of coefficients (15) that in the saturating field approximation, the Lorentzian part represents a small correction of the order of γ/g_Λ; however, it is this correction that determines a fall of the spectral density at spectral line wings. Although the non-Lorentzian contribution corresponding to one coefficient falls at the wings as $1/\Delta\omega$, the account for the total contribution from all the coefficients results in the fall of the total non-Lorentzian part proportional to $\propto 1/\Delta\omega^2$, as for the Lorentzian part. Moreover, for $\Gamma = \gamma$, i.e., in the absence of elastic dephasing at dipole transitions, the non-Lorentzian and Lorentzian parts totally compensate each other in the wings in the order $1/\Delta\omega^2$, so that the total spectrum falls much steeper, as $\mathcal{F}(\Delta\omega) \propto 1/\Delta\omega^4$. In this case, the ratio of the total spectrum to its Lorentzian part falls as $1/\Delta\omega^2$. Using coefficients (15), the asymptotics of the spectrum for large $\Delta\omega$ has the form

$$\mathcal{F}(\Delta\omega) = \frac{2\Gamma_e\Gamma_{12}\sin^2\varphi\cos^2\varphi}{\gamma}\frac{1}{\Delta\omega^2} + O\left(\frac{1}{\Delta\omega^4}\right). \quad (16)$$

Here, $\Gamma_e = \Gamma - \gamma$ is the rate of elastic dephasing of dipole transitions.

It is interesting that for two-level atoms, for which exact formulas for fluorescence spectra have long been known [14, 16], the fall of spectral line wings is also determined by the non-Lorentzian part [17] and, therefore, by the presence or absence of elastic dephasing. In this case, the asymptotic analysis of spectral wings yields the dependence

$$\mathcal{F}(\Delta\omega) = \frac{g^2\Gamma_e}{g^2 + \gamma\Gamma}\frac{1}{\Delta\omega^2} + O\left(\frac{1}{\Delta\omega^4}\right). \quad (17)$$

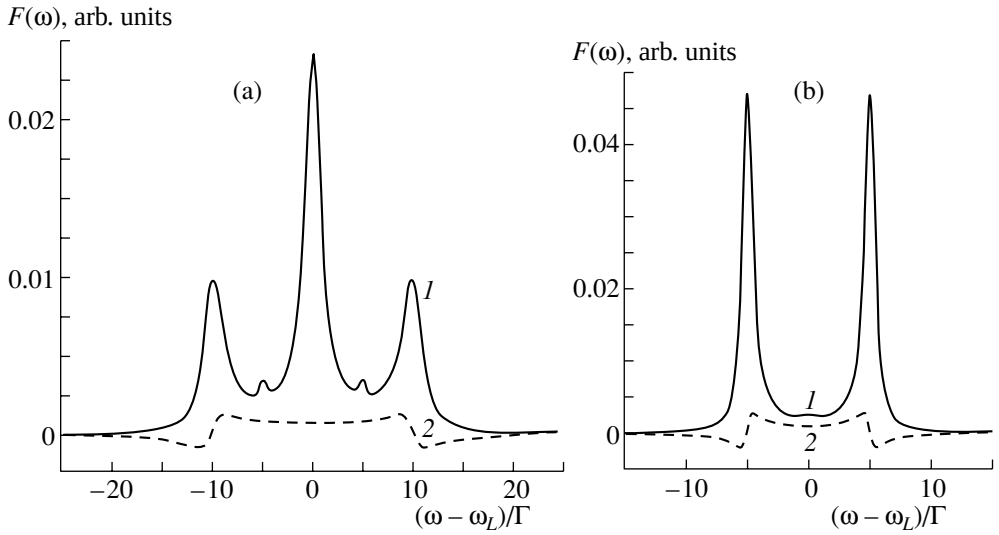

Fig. 4. Fluorescence spectrum of the Λ-system at the $|1\rangle \longleftrightarrow |3\rangle$ transition frequency for unequal intensities of laser fields. (*1*) Total spectrum; (*2*) non-Lorentzian part. Parameters of the system: $g_\Lambda = 10\Gamma$, $\delta = 0$, $\gamma = \Gamma$, $\Gamma_{12} = 0.001\Gamma$; (a) $\varphi = \pi/20$; (b) $\varphi = 9\pi/20$.

The elastic dephasing is defined for the two-level atom as the difference $\Gamma_e = \Gamma - \gamma/2$.

Fluorescence in the Case of an Abrupt Change in the Liouvillian

Using expressions (9) and (10) for the averaged intensity coefficients $C_k^{i3} = \Gamma_T \sum_{\lambda_j \neq 0} C_{jk}^{i3}/\lambda_j$, which are analogous to coefficients (15), we obtain

$$C_0^{13} = C_0^{23} = C_1^{13} = C_1^{23} = 0,$$

$$C_2^{13} = \frac{\Gamma_T}{\gamma}\cos^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi),$$

$$C_2^{23} = \frac{\Gamma_T}{\gamma}\sin^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi),$$

$$C_3^{13} = \frac{\Gamma_T}{\gamma}\cos^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi) \times\left(1 + \frac{i(2\Gamma - 11\gamma)}{64 g_\Lambda}\right),$$

$$C_4^{13} = C_3^{13*},$$

$$C_3^{23} = \frac{\Gamma_T}{2\gamma}\sin^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi) \times\left(1 + \frac{i(2\Gamma - 11\gamma)}{64 g_\Lambda}\right),$$

$$C_4^{23} = C_3^{23*},$$

$$C_5^{13} = C_5^{23} = C_7^{13} = C_7^{23} = 0, \qquad (18)$$

$$C_6^{13} = \frac{\Gamma_T}{\gamma}\cos^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi) \times\left(1 - \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right),$$

$$C_6^{23} = \frac{\Gamma_T}{\gamma}\sin^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi) \times\left(1 - \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right),$$

$$C_8^{13} = \frac{\Gamma_T}{\gamma}\cos^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi) \times\left(1 + \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right),$$

$$C_8^{23} = \frac{\Gamma_T}{\gamma}\sin^2\varphi(n\cos^2\varphi + (1-n)\sin^2\varphi) \times\left(1 - \frac{i(2\Gamma - \gamma) - \delta}{g_\Lambda}\right).$$

One can see from expressions (15) and (18) that the effect of the one-photon detuning and non-Lorentzian part on the shape of the spectrum in the dynamic theory is completely analogous to that in the stationary theory. Only the dependence on the angle φ changes, which describes the relative distribution of the intensity over laser-field frequencies, because a new parameter n appears which characterizes excitation of the lower system of levels in the $\hat{\rho}_0$ state.

To construct a correct phenomenological theory, it is necessary to know for which values of specified phenomenological relaxation parameters the results of cal-

culations coincide with the results of the dynamic theory. It can be shown that if, instead of the approximation $\gamma \longrightarrow 0$, $w \longrightarrow 0$ used in the stationary theory, we specify the relaxation parameters as $\Gamma_{12} \longrightarrow \Gamma_T$, $\gamma \longrightarrow (1 - n)\Gamma_T$, $w \longrightarrow n\Gamma_T$ and calculate coefficients (6) within the framework of the stationary theory, we obtain instead of (15) the expressions that completely coincide with formulas (18) of the nonstationary theory. Thus, the phenomenological description of relaxation of the lower levels allows one to take into account a nonstationary response of the system within the framework of a stationary phenomenological theory.

Absorption of the Probe Field

Using expressions (13) for coefficients determining absorption of the probe field, we obtain nonzero coefficients

$$\begin{aligned} D_5^{13} &= \frac{1}{2}\sin^2\varphi\left(1 + \frac{i\Gamma + \delta}{g_\Lambda}\right), \\ D_5^{23} &= \frac{1}{2}\cos^2\varphi\left(1 + \frac{i\Gamma + \delta}{g_\Lambda}\right), \\ D_7^{13} &= \frac{1}{2}\sin^2\varphi\left(1 - \frac{i\Gamma + \delta}{g_\Lambda}\right), \\ D_7^{23} &= \frac{1}{2}\cos^2\varphi\left(1 - \frac{i\Gamma + \delta}{g_\Lambda}\right). \end{aligned} \tag{19}$$

One can see from these expressions that absorption of light at one transition is proportional to the ratio of the light intensity at another transition to the total intensity of the laser field. By considering for definiteness the $|1\rangle \longleftrightarrow |3\rangle$ transition, we see from (13) and (19) that for this transition $\mathcal{A}^{13} \propto \sin^2\varphi = g'^2/g_\Lambda^2$, which at first glance may seem unexpected for $g = 0$. When the laser field is mainly concentrated at the $|2\rangle \longleftrightarrow |3\rangle$ transition, absorption at this transition is absent because of the self-induced transparency of the matter—one of the manifestations of CPT. In the case under study, all the population proves to be concentrated in the $|1\rangle$ state, so that photons of the probe field cannot be absorbed at the $|2\rangle \longleftrightarrow |3\rangle$ transition. At the same time, absorption at the $|1\rangle \longleftrightarrow |3\rangle$ transition achieves its maximum value upon the resonance.

In the case of the inexact resonance, two absorption lines shifted by $\pm g_\Lambda/2$ relative to the laser-field frequency acquire the additional shift $\delta/2$. Unlike the shift of fluorescence lines, this shift is directed away from atomic frequencies. A change in the line intensity also occurs in the opposite way: the intensity of lines shifted to the laser frequency decreases, and *vice versa*.

The non-Lorentzian and Lorentzian parts of the spectrum compensate each other for small $\Delta\omega = \omega - \omega_L$, resulting in almost zero total absorption coefficient for $\Delta\omega = 0$ (Fig. 2b), which corresponds to the self-induced transparency. This should take place for $\Delta\omega = 0$, when the probe field does not differ from the pump field and its absorption should be weak upon CPT.

CONCLUSIONS

Thus, the account for the first-order corrections in the saturating field approximation allows one to improve the accuracy of previously known formulas resulting in good agreement with numerical calculations already for comparatively low Rabi frequencies $g_\Lambda \sim 5\gamma$ and also take into account more detailed features of the generation of the response of Λ-systems. In particular, for small one-photon detunings δ, corrections to the fluorescence and absorption spectra of the Λ-system are linear in δ, whereas these corrections for the two-level atom are quadratic. When one of the Rabi frequencies is much greater than another one, fluorescence at one transition represents a standard triplet, which is typical for the two-level atom, and fluorescence at another transition is additive to the five-component spectrum of the Λ-system. Although the non-Lorentzian part is small in the approximation considered, it determines the fall of fluorescence line wings and the consistent frequency dependence of the absorption coefficient in the vicinity of laser frequencies. In the absence of elastic dephasing, the usual Lorentzian fall $\mathcal{F}(\Delta\omega) \propto 1/\Delta\omega^2$ is replaced by the steeper fall $\mathcal{F}(\Delta\omega) \propto 1/\Delta\omega^4$, and the absorption coefficient virtually vanishes at laser pump frequencies, as should be upon CPT.

Based on the dynamic model of generation of the response at low decay rate Γ_{12} of the polarization in the ground state, we showed that the results of spectral calculations for the abrupt change in the field in the case of the rectangular profile of a beam can be reduced to the results of the stationary theory by introducing additional relaxation parameters that describe the population redistribution in the ground state. This allows one to replace dynamic models by their simpler stationary phenomenological analogs.

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