

# Optical-collisional nonlinearity and the coherent population trapping effect

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The interaction between intense two-frequency laser radiation and three-level atoms in the vapor phase is considered. The effect of coherent population trapping (CPT) is studied, and the intensities of the components of the fluorescence and absorption spectra of a probe signal are calculated for  $\Lambda$  and  $V$  configurations of the levels. It is shown that the change in the atomic collision dynamics in a strong field gives rise to an exponential decrease in the collisional relaxation properties as a function of field strength, and consequently to extinction of the lines of the fluorescence and absorption spectra. The width of the CPT dip in the profile (the “black line”) is calculated.

## INTRODUCTION

The interaction between atoms with a three-level excitation scheme and two monochromatic fields when the coherent population trapping (CPT) effect arises has been actively investigated in recent years both theoretically<sup>1-6</sup> and experimentally.<sup>7-10</sup> The considerable attention devoted to this problem stems both from the fundamental nature of the phenomenon and from physical applications in high-resolution spectroscopy,<sup>11</sup> frequency stabilization,<sup>10,12,13</sup> optically bistable systems,<sup>14,15</sup> for atomic cooling,<sup>16-18</sup> etc.

In this paper we consider the CPT effect and calculate the components of the fluorescence and absorption spectra of a probe signal when intense two-frequency laser radiation acts on a gaseous medium. Coherent population trapping arises when two aligned laser fields with frequencies  $\omega_1$  and  $\omega_2$  interact with a three-level system having levels in a  $\Lambda$  configuration, provided the two-photon resonance condition  $\omega_1 - \omega_2 = \omega_{ac}$  holds, where  $\omega_{ac}$  is the difference in the energies of the two lower atomic levels (see Fig. 1a; here  $\hbar = 1$ ). When this happens the atom is transformed by the field into a coherent superposition state made up of the two lower levels, while the upper level  $b$  is not excited. Under these conditions the incident radiation is not absorbed. This is responsible for the narrow sharp dip in the fluorescence signal, which is known as the “black line”. The medium becomes transparent with respect to the incident radiation.<sup>6</sup>

But the CPT effect occurs only if the coherence of the lower levels relaxes substantially more slowly than the radiative relaxation rate of the uppermost level. In gaseous media the coherence relaxation rate is determined primarily by elastic collisions between the active atoms and atoms of the buffer gas. Here we assume that the lower levels  $a$  and  $c$  are not components of a hyperfine structure of the same electronic state. In previous studies, collisions were taken into account by introducing appropriate phenomenological constants. But if the laser radiation is sufficiently intense, it affects the dynamics of the atomic collisions (the optical-collisional nonlinearity).<sup>19-23</sup> This makes the collisional relaxation characteristics depend strongly on the field strength and the departure from resonance. As the field strength increases, in particular, the collisional relaxation rate of the coherence must decrease, tending to zero in the limit  $\Omega \gg \Omega_W$ , where  $\Omega$  is the Rabi frequency and  $\Omega_W$  is the Weisskopf frequency. Consequently, if the CPT effect is absent at moderate field strengths due to collisions, it must

reappear in sufficiently strong fields  $\Omega \gtrsim \Omega_W$ , corresponding to incident radiation with intensity  $I \gtrsim 10^7 \text{ W cm}^{-2}$ . It follows that, as noted by Vdovin and Travençolo,<sup>5</sup> the CPT effect can be a useful tool for experimentally verifying theoretical predictions regarding changes in the dynamics of atomic collisions in strong fields. Reference 24 discusses some closely related ideas. As will be shown below, the study of the corresponding coherent effect for  $V$ -shaped configurations of the levels also opens up interesting possibilities.

The theoretical aspects of the CPT effect in strong laser fields are of interest in their own right, since they offer a way to calculate the width of the dip, the strengths and widths of the fluorescence spectrum, the absorption lineshape of the probe radiation, and the properties of the “window of transparency” that occurs when strong two-frequency laser light propagates in such a medium under these conditions.

## 1. ATOMIC BASIS AND THE “DRESSED-STATE” BASIS

We will assume that the structure of the atomic levels has a  $\Lambda$  or  $V$  shape (Fig. 1). The field at frequency  $\omega_1$  with amplitude  $E_1$  induces transitions between the  $a$  and  $b$  atomic states, and the field at frequency  $\omega_2$  with amplitude  $E_2$  induces transitions between the  $b$  and  $c$  states. The deviations from resonance are written as  $\Delta_1$  and  $\Delta_2$ , where

$$\Delta_1 = |\omega_{ba} - \omega_1|, \quad \Delta_2 = \omega_2 - |\omega_{bc}|, \quad \delta = \Delta_1 + \Delta_2. \quad (1.1)$$

In what follows we will omit the index from  $\Delta_1$  and write  $\Delta_1 \equiv \Delta$ .

Since two strong resonant fields are acting on the three-level system, it is convenient to go over to the dressed-state basis.<sup>25</sup> The wave functions of the dressed states,  $|\mu, n_1, n_2\rangle$ ,  $\mu = \alpha, \beta, \gamma$ , are the eigenfunctions of the Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{V}_{AL}, \quad (1.2)$$

Here we have written  $\hat{H}_0 = \hat{H}_A + \hat{H}_L$  where  $\hat{H}_A$  and  $\hat{H}_L$  are the Hamiltonians of the free atom and the field, and  $\hat{V}_{AL}$  is the operator of the interaction between the atom and the field:

$$|\mu, n_1, n_2\rangle = \sum_{i=0}^2 T_{i\mu} |i\rangle, \quad (1.3)$$

where  $|i\rangle$ ,  $i = 0, 1, 2$ , are the eigenfunctions of the operator  $\hat{H}_0$ , and  $n_1$  and  $n_2$  are the numbers of photons of the laser

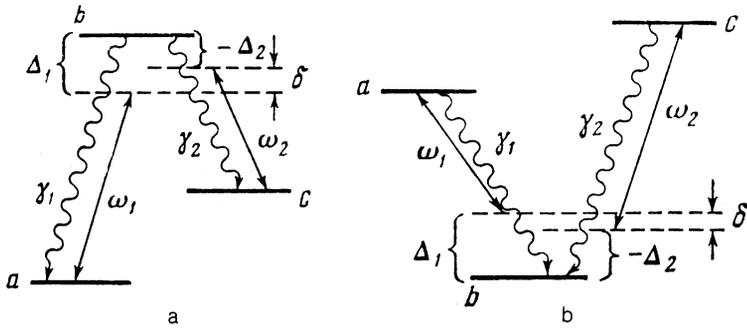


FIG. 1. Level diagram in the atomic basis: a)  $\Lambda$  configuration b)  $V$  configuration.

field at the frequencies  $\omega_1$  and  $\omega_2$ . For the  $\Lambda$  configuration (Fig. 1a) we have

$$|0\rangle = |a, n_1, n_2\rangle, \quad |1\rangle = |b, n_1-1, n_2\rangle, \quad (1.4)$$

$$|2\rangle = |c, n_1-1, n_2+1\rangle.$$

For the  $V$  configuration (Fig. 1b) we have

$$|0\rangle = |a, n_1-1, n_2\rangle, \quad |1\rangle = |b, n_1, n_2\rangle, \quad |2\rangle = |c, n_1, n_2-1\rangle. \quad (1.5)$$

The dressed-state energy levels are determined by the equation for the eigenvalues<sup>2,3</sup>

$$\omega_\mu^3 - (\Delta + \delta)\omega_\mu^2 - (G^2 - \Delta\delta)\omega_\mu + G_1^2\delta = 0, \quad (1.6)$$

where

$$T^+ = \begin{vmatrix} \alpha & 0 & 1 \\ \beta & -(1-x)^{1/2} & 0 \\ \gamma & [x(1-x)/2]^{1/2} & [(1+x)/2]^{1/2} \end{vmatrix}, \quad (1.9)$$

with  $x = \Delta/\Omega$  and  $\kappa = G_1^2/G^2$ . If (1.7) does not hold but we have  $\delta \ll G$ , then as will be seen, it suffices to apply a correction to just one element of the matrix  $T$ :

$$T_{1\alpha} = (T^+)_{\alpha 1} = -[x(1-x)]^{1/2}\delta/G. \quad (1.10)$$

Note, however, that when we go over the dressed-state basis it makes sense to consider the structure of the states when they are well separated, i.e., for  $\Omega \gg \Gamma$ , where  $\Gamma$  is the spectral width of the corresponding line. Moreover, we assume that the frequencies  $\omega_1$  and  $\omega_2$  are sufficiently separated:

$$|\omega_1 - \omega_2| \gg \Omega. \quad (1.11)$$

The probabilities of the radiative transitions  $\nu \rightarrow \mu$  in the dressed basis are

$$\begin{aligned} \gamma_{\mu\nu}^{(1)} &\propto |\langle \mu, n_1-1, n_2 | \hat{d} | \nu, n_1, n_2 \rangle|^2, \\ \gamma_{\mu\nu}^{(2)} &\propto |\langle \mu, n_1, n_2-1 | \hat{d} | \nu, n_1, n_2 \rangle|^2, \\ \gamma_{\mu\nu} &= \gamma_{\mu\nu}^{(1)} + \gamma_{\mu\nu}^{(2)}, \end{aligned} \quad (1.12)$$

where  $\hat{d}$  is the operator of the atomic dipole moment. Taking into account that in the atomic basis only the matrix ele-

$$G_1 = |d_{ab}E_1|/2, \quad G_2 = |d_{bc}E_2|/2, \quad G^2 = G_1^2 + G_2^2.$$

When the two-photon resonance condition

$$\delta = 0, \quad (1.7)$$

is satisfied, i.e., when  $\omega_1 - \omega_2 = \pm \omega_{ca}$  holds, where the upper and lower signs refer to the  $\Lambda$  and  $V$  configurations, respectively, we have

$$\omega_\alpha = 0, \quad \omega_\beta = (\Delta + \Omega)/2, \quad \omega_\gamma = (\Delta - \Omega)/2, \quad (1.8)$$

where  $\Omega = (\Delta^2 + 4G^2)^{1/2}$  is the Rabi frequency. For the  $V$  configuration it is necessary to make the replacement  $\Delta \rightarrow -\Delta$  in (1.8).

When the condition (1.7) holds, the transition matrix takes the form

ments  $d_{ab}$  and  $d_{bc}$  are nonzero (the  $a-c$  transition is forbidden) and making use of the expansion (1.3), we find for the  $\Lambda$  configuration

$$\gamma_{\mu\nu}^{(1)} = T_{0\mu}^2 T_{1\nu}^2 \gamma_1, \quad \gamma_{\mu\nu}^{(2)} = T_{2\mu}^2 T_{1\nu}^2 \gamma_2, \quad (1.13)$$

and for the  $V$  configuration

$$\gamma_{\mu\nu}^{(1)} = T_{0\nu}^2 T_{1\mu}^2 \gamma_1, \quad \gamma_{\mu\nu}^{(2)} = T_{2\nu}^2 T_{1\mu}^2 \gamma_2. \quad (1.14)$$

where  $\gamma_1$  and  $\gamma_2$  are the radiative transition probabilities corresponding to the  $a-b$  and  $b-c$  transitions. The positions of the levels of the dressed atom and the transitions between them are shown in Fig. 2. When the condition (1.17) for two-photon resonance in the  $\Lambda$ -system of levels is fulfilled, the  $|\alpha, n_1, n_2\rangle$  state does not relax radiatively, i.e.,  $\gamma_{\mu\alpha} = 0$ ,  $\mu = \alpha, \beta, \gamma$ . This is true because for  $\delta = 0$  it is a superposition of states corresponding to the lower states  $a$  and  $c$  alone. For the  $V$ -configuration of levels, in contrast, for  $\delta = 0$  there are no transitions to the states  $|\alpha, n_1, n_2\rangle$ , and we have  $\gamma_{\alpha\mu} = 0$ .

## 2. COLLISIONS: THE OPTICAL-COLLISIONAL TRANSITION MATRIX

We consider the collisions between active three-level atoms and structureless buffer gas atoms. In order to ac-

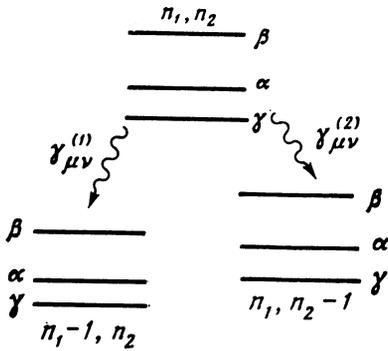


FIG. 2. Level diagram in the dressed basis.

count for collisional relaxation in a strong field, Bakaev *et al.*<sup>20</sup> showed that the dressed states of the atom are the most appropriate choice of basis. Now elastic collisions between the atoms and those of the inert gas give rise to inelastic transitions between states of the dressed atom. In the dressed-state basis the collision integral in the equation for the density matrix takes the form

$$\begin{aligned} \langle \mu, n_1, n_2 | \left( \frac{\partial \hat{\rho}}{\partial t} \right)_{\text{col}} | \mu', n_1', n_2' \rangle \\ = - \sum_{\nu, \nu'} \Gamma_{\mu\mu'}^{\nu\nu'} \langle \nu, n_1, n_2 | \hat{\rho} | \nu', n_1', n_2' \rangle, \end{aligned} \quad (2.1)$$

where  $\hat{\Gamma}$  is the optical-collisional transition matrix:

$$\Gamma_{\mu\mu'}^{\nu\nu'} = N \left\langle \nu \int_0^\infty 2\pi b db (\delta_{\mu\nu} \delta_{\mu'\nu'} - S_{\mu\nu} S_{\mu'\nu}') \right\rangle_\nu, \quad (2.2)$$

$N$  is the number density of the buffer gas,  $b$  is the impact parameter,  $\langle \dots \rangle_\nu$  denotes an average over the relative velocity  $\nu$ , and  $S_{\mu\nu}$  are the elements of the scattering matrix in the dressed-state basis, obtained by solving the equations

$$iS_{\mu\nu}(t) = \sum_{\mu'} V_{\mu\mu'} S_{\mu'\nu}(t) \exp(i\omega_{\mu\mu'} t), \quad (2.3)$$

$$S_{\mu\nu}(-\infty) = \delta_{\mu\nu}, \quad S_{\mu\nu}(+\infty) = S_{\mu\nu}, \quad \omega_{\mu\mu'} = \omega_\mu - \omega_{\mu'}.$$

Here  $V$  is the operator that describes the interaction between a dressed atom and an atom of the buffer gas, with

$$V_{\mu\mu'} = \sum_{i=0}^n T_{i\mu} T_{i\mu'} V_{ii}, \quad (2.4)$$

where the  $T_{i\mu}$  are the elements of the transition matrix (1.9),

$$V_{ii} = C_i / R^n, \quad (2.5)$$

where the  $C_i$  are constants specific to the interactions with atoms in the states  $i = 0, 1, 2$ , and  $n$  is an integer. The value  $n = 6$  corresponds to the Van der Waals interaction.

In weak fields, for which the approximation  $\Omega \ll \Omega_W \sim \tau$  holds, where  $\tau$  is the collision time, the system of dynamical equations (2.3) can be solved immediately. Furthermore, the elements of the matrix  $\Gamma$  can easily be obtained from the corresponding elements in the atomic basis using the transformation (1.3) (cf., e.g., Berman and Salomaa<sup>26</sup>):

$$\Gamma_{\mu\mu'}^{\nu\nu'} = \sum_{ijj'j'} T_{i\mu} T_{j\nu} T_{i'\mu'} T_{j'\nu'} \Gamma_{ij}^{j'j'}. \quad (2.6)$$

When (1.7) is satisfied, we have in particular using (1.9)

$$\Gamma_{\alpha\alpha}^{\beta\beta} = \Gamma_{\beta\beta}^{\alpha\alpha} = -\kappa(1-\kappa)(1-x) \text{Re } \Gamma_{\alpha\alpha}^{\alpha\alpha}, \quad (2.7)$$

where  $\text{Re } \Gamma_{\alpha\alpha}^{\alpha\alpha}$  is the rate at which the coherence of the lower levels relaxes collisionally:

$$\text{Re } \Gamma_{\alpha\alpha}^{\alpha\alpha} = Nu(C/u)^{2/(n-1)}, \quad (2.8)$$

where  $C = C_2 - C_0$ ,  $u = (kT/M)^{1/2}$  is the thermal speed,  $M$  is the reduced mass of the colliding atoms, and  $\Gamma_{\gamma\gamma}^{\alpha\alpha}$  differs from  $\Gamma_{\beta\beta}^{\alpha\alpha}$  through the replacement  $x \rightarrow -x$ .

In the strong-field limit

$$\Omega \gg \Omega_W \sim \tau^{-1}, \quad (2.9)$$

the collision dynamics is affected by the field and in the atomic basis the collision integral assumes a complicated form. Going over to the dressed basis simplifies matters. Now it is necessary to solve Eqs. (2.3) and use (2.2) to express the elements of  $\Gamma$ .

Equations (2.3) can be solved analytically only if we make some further simplifying assumptions. We are interested in the strong-field limit, when the energy levels of the dressed states spread apart to a considerable degree and transitions between them become unlikely. Then we can neglect transitions between the  $\beta$  and  $\gamma$  levels, the ones which are farthest apart, leaving only transitions between the neighboring levels,  $\beta$  and  $\alpha$  and  $\alpha$  and  $\gamma$ . Now Eqs. (2.3) go over to two systems of the same form, each of which contains only two equations. This type of system has been solved approximately for the case of a power-law interaction potential.<sup>27</sup> Using these results we obtain value of  $\Gamma_{\mu\mu}^{\nu\nu}$  for  $\nu = \mu$ :

$$\Gamma_{\alpha\alpha}^{\mu\mu} = \Gamma_{\mu\mu}^{\alpha\alpha} = \text{Re } \Gamma_{\alpha\alpha}^{\alpha\alpha} a_n f_\mu \beta_\mu^{-(n+1)n} \exp(-2a_n f_\mu \beta_\mu^{(n-1)/n}), \quad (2.10)$$

where

$$\beta_\mu = |\omega_{\mu\alpha}| / \Omega_W, \quad \Omega_W = (u^n / C)^{1/(n-1)},$$

$$a_n = 2^{1/n} \sin(\pi/2n), \quad f_{\beta,\gamma} = [\kappa(1-\kappa)(1 \mp x)/2]^{1/2n}.$$

Expression (2.10) is valid in the strong-field limit, when the distance between the levels of the dressed atom satisfies  $\omega_{\mu\alpha} \sim \Omega \gg \Omega_W$ . In this case we have  $\beta_\mu \gg 1$  and the characteristics  $\Gamma_{\mu\mu}^{\alpha\alpha}$ , as seen from (2.10), are exponentially small in comparison with (2.7). From the unitarity of the scattering matrix  $S$  it follows that

$$\sum_\nu \Gamma_{\mu\mu}^{\nu\nu} = 0, \quad \sum_\mu \Gamma_{\mu\mu}^{\nu\nu} = 0. \quad (2.11)$$

Since we need no matrix elements other than those of the form (2.7) and (2.10), we do not exhibit them here.

### 3. LEVEL POPULATIONS

The kinetic equation for the dressed atom can be written down in the form

$$\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}] + \left( \frac{\partial \hat{\rho}}{\partial t} \right)_{\text{rad}} + \left( \frac{\partial \hat{\rho}}{\partial t} \right)_{\text{col}}, \quad (3.1)$$

where the last two terms describe relaxation due to radiative transitions and to elastic collisions with buffer-gas atoms

[see Eq. (2.1)]. We assume that the secularity condition holds:

$$\Omega \gg \gamma, |\Gamma_{\mu\mu'}^{vv'}|. \quad (3.2)$$

In this limit we can neglect the off-diagonal elements of the density matrix, which are of order  $|\Gamma|/\Omega$ , and pass to a system of equations for the level populations only (balance equations):

$$\dot{\rho}_{\mu\mu} = -\gamma_{\mu}\rho_{\mu\mu} + \sum_{\nu} (\gamma_{\mu\nu} - \Gamma_{\mu\nu}^{v\nu})\rho_{\nu\nu}, \quad (3.3)$$

where

$$\rho_{\mu\mu} = \sum_{n_1, n_2} \langle \mu, n_1, n_2 | \hat{\rho} | \mu, n_1, n_2 \rangle, \text{ and } \gamma_{\mu} = \sum_{\nu} \gamma_{\nu\mu}$$

is the radiative width of the level. The condition that the system be closed takes the form

$$\sum_{\mu} \rho_{\mu\mu} = 1. \quad (3.4)$$

We look for a stationary solution of Eqs. (3.3). The effects we are interested in appear for  $|\Gamma| \lesssim \gamma$ . Using expressions (1.13) and (1.14) for  $\gamma_{\mu\nu}$  and assuming  $\delta \ll G$ ,  $|\Gamma| \ll \gamma_1, \gamma_2$ , and  $G_1 \sim G_2$ , we find for the  $\Lambda$ -configuration of levels

$$\rho_{\beta\beta} = -\Gamma_{\beta\alpha}\gamma^{-1}[(1+x)^{-1} + \xi y/2] - \Gamma_{\gamma\alpha}\gamma^{-1}\xi y/2 + \xi\kappa(1-\kappa)y\delta^2, \quad (3.5)$$

where

$$\Gamma_{\mu\nu} = \Gamma_{\mu\nu}^{v\nu}, \quad \gamma = (\gamma_1 + \gamma_2)/2, \quad \delta = \delta/G,$$

$$\xi = (\gamma_1 G_1^2 + \gamma_2 G_2^2) / (\gamma_1 G_2^2 + \gamma_2 G_1^2),$$

$$y = \frac{1-x}{1+x},$$

$\rho_{\gamma\gamma}$  differs from  $\rho_{\beta\beta}$  through the replacements  $\beta \rightarrow \gamma$ ,  $\gamma \rightarrow \beta$ ,  $x \rightarrow -x$ , and  $y \rightarrow 1/y$ , and

$$\rho_{\alpha\alpha} = 1 - \rho_{\beta\beta} - \rho_{\gamma\gamma}.$$

If an elastic transition takes place between the atomic levels  $a$  and  $c$  ( $a \rightleftharpoons c$ ) with probability  $\mathcal{T}$  ( $\mathcal{T} \ll \gamma$ ), then changes  $\delta\rho_{\beta\beta}$  and  $\delta\rho_{\gamma\gamma}$  occur in the populations of the  $\beta$ - and  $\gamma$ -levels:

$$\delta\rho_{\beta\beta} = (2\kappa - 1)^2 (\xi + 1) y \mathcal{T} / 2\gamma, \quad (3.6)$$

where  $\delta\rho_{\beta\beta}$  and  $\delta\rho_{\gamma\gamma}$  differ through the replacements  $x \rightarrow -x$ ,  $y \rightarrow 1/y$ .

Utilizing the relations (3.5) and (3.6), we can find the populations  $\rho_{ii}$  of the atomic levels:

$$\rho_{ii} = \sum_{\mu} T_{i\mu}^2 \rho_{\mu\mu}. \quad (3.7)$$

Hence if we take into account (1.9) and (1.10), the population of the upper atomic level  $|b\rangle$  is found to be

$$\rho_{\beta\beta} = -(\Gamma_{\beta\alpha} + \Gamma_{\gamma\alpha})(1 + \xi) / 2\gamma + \kappa(1 - \kappa)(1 + \xi)\delta^2. \quad (3.8)$$

We generalize these expressions for the level populations of the dressed and atomic levels, taking into account

the motion of the atoms and the spread in their velocities. For an atom moving with velocity  $\mathbf{v}$  and interacting with laser fields having wave vectors  $\mathbf{k}_1$  and  $\mathbf{k}_2$  and frequencies  $\omega_1$  and  $\omega_2$ , we must modify the expressions obtained above by writing

$$\omega_1 \rightarrow \omega_1 - \mathbf{k}_1 \cdot \mathbf{v}, \quad \omega_2 \rightarrow \omega_2 - \mathbf{k}_2 \cdot \mathbf{v}$$

and make the corresponding changes in the deviations from resonance  $\Delta_1$ ,  $\Delta_2$ , and  $\delta$  defined in (1.1). In particular,

$$\delta \rightarrow \delta + (\mathbf{k}_1 - \mathbf{k}_2) \cdot \mathbf{v}.$$

Averaging over velocities causes  $\delta^2$  to be replaced in Eqs. (3.5) and (3.8) by  $\delta_{\text{eff}}^2$ , where

$$\delta_{\text{eff}}^2 = \delta^2 + \bar{\Delta}_D^2, \quad \bar{\Delta}_D^2 = [(\mathbf{k}_1 - \mathbf{k}_2)\mathbf{u}]^2 / G^2. \quad (3.9)$$

In strong unidirectional external fields we have  $\bar{\Delta}_D^2 \ll 1$ . The quantity  $\rho_{bb}$  attains its minimum value as a function of  $\omega_1$  and  $\omega_2$  at  $\delta = 0$ , and the width of  $\delta_0$  of the resonance, i.e., the width of the black line for which the population has twice the minimum value, is

$$\delta_0 \sim (|\Gamma_{\beta\alpha} + \Gamma_{\gamma\alpha}| \gamma^{-1} + \bar{\Delta}_D^2)^{1/2} G. \quad (3.10)$$

From (3.10) we see that when (2.7) holds, the field makes the dip in the population wider. For strong fields, however, when the expressions for  $\Gamma_{\beta\alpha}$  and  $\Gamma_{\gamma\alpha}$  are given by relations (2.10), the dip becomes narrower with increasing field strength.

If the inelastic transition  $a \rightleftharpoons c$  occurs, then using (3.6) we find the correction  $\delta\rho_{\beta\beta}$  to the population (3.8):

$$\delta\rho_{\beta\beta} = (2\kappa - 1)^2 (1 + \xi) \mathcal{T} / 2\gamma. \quad (3.11)$$

The form of the expressions (3.5), (3.8), and (3.11) found for the populations implies that for  $\delta = 0$  in the strong-field limit the populations  $\rho_{\beta\beta}$  and  $\rho_{\gamma\gamma}$ , in view of (2.10), tend exponentially to a small value determined by the background Doppler broadening and inelastic collisions, whereas  $\rho_{\alpha\alpha} \rightarrow 1$ , i.e., practically speaking only the state  $|\alpha, n_1, n_2\rangle$  is populated. From (1.3) and (1.9), this means that only the atomic states  $a$  and  $c$  are populated ( $\rho_{\beta\beta} \rightarrow 0$ ), i.e., the atom is in a coherent-superposition state combining the lower states  $a$  and  $c$  (CPT effect). The values of the populations  $\rho_{\beta\beta}$ ,  $\rho_{\gamma\gamma}$ , and  $\rho_{bb}$  differ from the minimum values because of collisional-relaxation and inelastic processes.

Now let us consider a  $V$ -configuration of levels (Fig. 1b). In this case collisions are responsible for populating the  $\alpha$  level:

$$\rho_{\alpha\alpha} = -\frac{1}{4} \Gamma_{\beta\alpha} \gamma^{-1} \frac{(1-x)^2}{1+x^2} (1+\xi) - \frac{1}{4} \Gamma_{\gamma\alpha} \gamma^{-1} \frac{(1+x)^2}{1+x^2} (1+\xi) + \frac{1}{2} \kappa(1-\kappa) \xi \frac{1-x^2}{1+x^2} \delta_{\text{eff}}^2, \quad (3.12)$$

$$\delta\rho_{\alpha\alpha} = \mathcal{T} \gamma^{-1} (2\kappa - 1)^2 (1 + \xi) (1 - x^2) / 4(1 + x^2), \quad (3.13)$$

where  $\Gamma_{\beta\alpha}$  and  $\Gamma_{\gamma\alpha}$  are defined as before by Eqs. (2.7) and (2.10). For  $\delta = 0$  the value of  $\rho_{\alpha\alpha}$  attains a minimum. Note also that the populations (3.6) and (3.13) resulting from inelastic collisions vanish for  $G_1 = G_2$ , i.e.,  $\kappa = 1/2$ .

#### 4. STRENGTH OF THE FLUORESCENCE SIGNAL

As is well known, the transition to the dressed states permits us to determine the frequencies of the fluorescence

signal immediately. Thus, in our case (see Fig. 2), the fluorescence is associated with the transitions  $|\nu, n_1, n_2\rangle \rightarrow |\mu, n_1 - 1, n_2\rangle$  ( $\mu, \nu = \alpha, \beta, \gamma$ ) at the frequencies  $\omega_1 + \omega_{\nu\mu}$  and the transitions  $|\nu, n_1, n_2\rangle \rightarrow |\mu, n_2 - 1, n_2\rangle$  at frequencies  $\omega_2 + \omega_{\nu\mu}$ , i.e., for every transition with the central frequencies  $\omega_1, \omega_2$  the spectrum will have seven components. The intensity of each component is determined by the relation

$$I_{\mu\nu}(\omega_i + \omega_{\nu\mu}) \propto \gamma_{\mu\nu}^{(i)} \rho_{\nu\nu} \quad (i=1, 2), \quad (4.1)$$

where  $\rho_{\nu\nu}$  are the level populations of the dressed states and  $\gamma_{\mu\nu}^{(i)}$  are the probabilities of the spontaneous radiative transitions (1.13) and (1.14).

For  $\Lambda$  level configurations, using relations (3.5), (1.13) and the notation  $I_{\mu\nu}(\omega_i + \omega_{\nu\mu}) = I_{\mu\nu}^{(i)}$ , we have (we omit the proportionality coefficient  $\hbar\omega_i$ )

$$\begin{aligned} I_{\beta\alpha}^{(1)} &= \kappa^2 (1-\kappa) \delta^2 (1-x) \gamma_i / 2, \\ I_{\alpha\beta}^{(1)} &= (1-\kappa) (\gamma_i / 2\gamma) [-\Gamma_{\beta\alpha} - (1-x) (\Gamma_{\beta\alpha} + \Gamma_{\gamma\alpha}) (\xi/2) \\ &\quad + \kappa (1-\kappa) (1-x) \xi \gamma \delta^2], \end{aligned} \quad (4.2)$$

where  $I_{\gamma\alpha}^{(1)}$  and  $I_{\alpha\gamma}^{(1)}$  differ from  $I_{\beta\alpha}^{(1)}$  and  $I_{\alpha\beta}^{(1)}$ , respectively, through the replacements  $\Gamma_{\beta\alpha} \rightarrow \Gamma_{\gamma\alpha}$  and  $\Gamma_{\gamma\alpha} \rightarrow \Gamma_{\beta\alpha}$ , and  $x \rightarrow -x$ ,

$$I_{\beta\gamma}^{(1)} = \frac{1}{2} \frac{\kappa}{1-\kappa} I_{\alpha\gamma}^{(1)}, \quad I_{\gamma\beta}^{(1)} = \frac{1}{2} \frac{\kappa}{1-\kappa} I_{\alpha\beta}^{(1)}, \quad (4.3)$$

$$\begin{aligned} I^{(1)} &= I_{\alpha\alpha}^{(1)} + I_{\beta\beta}^{(1)} + I_{\gamma\gamma}^{(1)} = \kappa (\gamma_i / 4\gamma) [-\Gamma_{\beta\alpha} (1-x) - \Gamma_{\gamma\alpha} (1+x) \\ &\quad - (\Gamma_{\beta\alpha} + \Gamma_{\gamma\alpha}) \xi (1+x^2) + 2\kappa (1-\kappa) (1+x^2) \xi \delta^2] + \kappa (1-\kappa) \gamma_i \delta^2, \end{aligned} \quad (4.4)$$

and  $I_{\mu\nu}^{(2)}$  differs from  $I_{\mu\nu}^{(1)}$  through the replacements  $\gamma_1 \rightarrow \gamma_2$  and  $\kappa \rightarrow 1 - \kappa$ . Figure 3 shows how the intensities of the components of the fluorescence spectrum calculated from (4.1), (4.2), and (2.10) change as functions of the laser field strength.

If inelastic  $a \rightleftharpoons c$  transitions occur, then additional terms appear in (4.2)–(4.4):

$$\begin{aligned} \delta I_{\alpha\beta}^{(1)} &= (1-\kappa) (2\kappa-1)^2 (\gamma_i / 4\gamma) (1+\xi) (1-x) \mathcal{F}, \\ \delta I^{(1)} &= (1-\kappa) (2\kappa-1)^2 (\gamma_i / 2\gamma) (1+\xi) \mathcal{F}, \end{aligned} \quad (4.5)$$

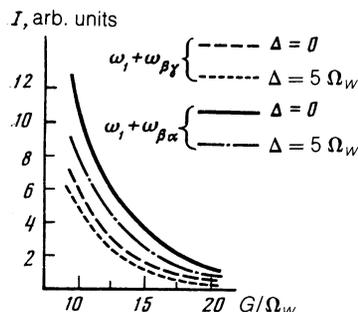


FIG. 3. Strengths of fluorescence spectrum components.

where  $\delta I_{\alpha\beta}^{(2)}$  and  $\delta I^{(2)}$  differ from (4.5) through the replacements  $\gamma_1 \rightarrow \gamma_2$  and  $\kappa \rightarrow 1 - \kappa$ , and  $\delta I_{\alpha\gamma}^{(1)}$  and  $\delta I_{\alpha\gamma}^{(2)}$  differ from  $\delta I_{\alpha\beta}^{(1)}$  and  $\delta I_{\alpha\beta}^{(2)}$  through the replacement  $x \rightarrow -x$ .

From (4.2) and (4.3) it follows that when the two-photon resonance condition  $\delta = 0$  is satisfied the fluorescence signal associated with transitions from the dressed state  $\alpha$  drops out and only five components remain in the spectrum. Strong lines associated with transitions from levels  $\beta$  and  $\gamma$  are determined by the elements of the  $\Gamma$  matrix and, as implied by (2.10), fall off exponentially with increasing field strength.

In the case of  $V$  configuration levels, the situation is reversed: for  $\delta = 0$  the fluorescence signal corresponding to transitions to level  $\alpha$  drops out. Now the components associated with the transitions  $\alpha \rightarrow \beta$  and  $\alpha \rightarrow \gamma$  are the most interesting, since it is precisely their intensities which are determined by the collisional-relaxation characteristics:

$$\begin{aligned} I_{\beta\alpha}^{(1)} &= \frac{1}{8} \frac{1-\kappa}{1+x^2} \frac{\gamma_i}{\gamma} [- (1+\xi) (1-x)^2 \Gamma_{\beta\alpha} - (1+\xi) (1+x)^2 \Gamma_{\gamma\alpha} \\ &\quad + 2\kappa (1-\kappa) \xi (1-x^2) \delta^2], \end{aligned} \quad (4.6)$$

where  $I_{\gamma\alpha}^{(1)}$  differs from  $I_{\beta\alpha}^{(1)}$  through the replacement  $x \rightarrow -x$ , and  $I_{\beta\alpha}^{(2)}$  and  $I_{\gamma\alpha}^{(2)}$  differ from  $I_{\beta\alpha}^{(1)}$  and  $I_{\gamma\alpha}^{(1)}$  through the replacements  $\gamma_1 \rightarrow \gamma_2$  and  $\kappa \rightarrow 1 - \kappa$ .

When we include inelastic  $a \rightleftharpoons c$  transitions, an additional term

$$\delta I_{\beta\alpha}^{(1)} = \frac{1}{8} (1-\kappa) (2\kappa-1)^2 (1+x) \frac{1-x^2}{1+x^2} (1+\xi) \frac{\gamma_i}{\gamma} \mathcal{F}, \quad (4.7)$$

appears, where  $\delta I_{\gamma\alpha}^{(1)}$  differs from  $\delta I_{\beta\alpha}^{(1)}$  through the replacement  $x \rightarrow -x$ , and  $\delta I_{\beta\alpha}^{(2)}$  and  $\delta I_{\gamma\alpha}^{(2)}$  differ from  $\delta I_{\beta\alpha}^{(1)}$  and  $\delta I_{\gamma\alpha}^{(1)}$ , respectively, through the replacements  $\gamma_1 \rightarrow \gamma_2$  and  $\kappa \rightarrow 1 - \kappa$ . The fluorescence spectrum for  $\delta = 0$  is shown schematically in Fig. 4.

Measuring the strength of the external laser field and observing experimentally the change in the intensities of the  $I_{\beta\alpha}$  and  $I_{\gamma\alpha}$  components compared, say, with the  $I_{\gamma\beta}$  and  $I_{\beta\gamma}$  components can enable us to determine how the collisional-relaxation characteristics change in a strong external field. When we take the remaining Doppler broadening into consideration in Eqs. (4.2)–(4.4),  $\delta^2$  must be replaced by  $\delta_{\text{eff}}^2$  given in (3.9).

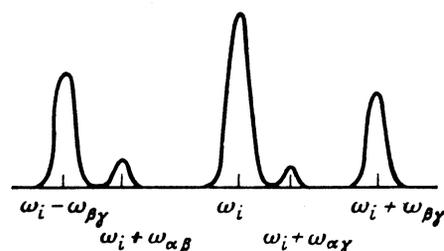


FIG. 4. General structure of the fluorescence spectrum for  $V$ -shaped configuration levels.

## 5. ABSORPTION SPECTRUM OF A TEST SIGNAL

We now assume that in addition to the strong two-frequency laser field, the system is also irradiated by a weak probe field of frequency  $\omega$ . The components  $Q_{\mu\nu}^{(i)}$  of the absorbed power corresponding to the central frequencies  $\omega_i + \omega_{\nu\mu}$  are determined by the expressions<sup>28</sup>

$$Q_{\mu\nu}^{(i)} = k\gamma_{\mu\nu}^{(i)} (\rho_{\mu\mu} - \rho_{\nu\nu}), \quad (5.1)$$

for  $i = 1, 2$ , where  $k$  is a coefficient proportional to the strength of the probe field.

Using (1.13), (1.14), and the expressions (3.5), (3.6), (3.12), and (3.13) for the populations, we obtain the components  $Q_{\mu\nu}^{(i)}$ . For  $\Lambda$ -shaped level configurations and  $\delta = 0$ , the components  $Q_{\alpha\beta}^{(i)}$ ,  $Q_{\alpha\gamma}^{(i)}$ ,  $Q_{\gamma\beta}^{(i)}$ , and  $Q_{\beta\gamma}^{(i)}$  are nonvanishing. Taking into account also that under the specified conditions we have  $\rho_{\alpha\alpha} \approx 1$  and  $\rho_{\beta\beta}, \rho_{\gamma\gamma} \ll \rho_{\alpha\alpha}$ , we find

$$Q_{\alpha\beta}^{(i)} = k\gamma_{\alpha\beta}^{(i)}, \quad Q_{\alpha\gamma}^{(i)} = k\gamma_{\alpha\gamma}^{(i)}, \quad (5.2)$$

that is, at the frequencies  $\omega_i + \omega_{\beta\alpha}$ ,  $\omega_i + \omega_{\gamma\alpha}$  the probe signal experiences absorption that does not depend on the collisional relaxation characteristics, i.e., is insensitive to the effects of optical-collisional nonlinearity. At the symmetric frequencies  $\omega_i - \omega_{\beta\alpha}$ ,  $\omega_i - \omega_{\gamma\alpha}$ , the components  $Q_{\beta\alpha}^{(i)}$  vanish. Signal amplification at these frequencies takes place only when the condition (1.7) fails:

$$Q_{\beta\gamma}^{(i)} = \frac{k\kappa(1-x)}{4(1+x)} \frac{\gamma_1}{\gamma} \{ \Gamma_{\gamma\alpha} - \Gamma_{\beta\alpha} + x(\Gamma_{\gamma\alpha} + \Gamma_{\beta\alpha}) + 2\xi x[\Gamma_{\gamma\alpha} + \Gamma_{\beta\alpha} - 2\kappa(1-\kappa)\gamma\delta^2] - 2(2\kappa-1)^2(1+\xi)x\mathcal{F} \}, \quad (5.3)$$

while  $Q_{\gamma\beta}^{(i)}$  differs from  $Q_{\beta\gamma}^{(i)}$  through the replacements  $\gamma \rightarrow \beta$ ,  $\beta \rightarrow \gamma$ , and  $x \rightarrow -x$ . Note that if absorption ( $Q_{\beta\gamma}^{(i)} > 0$ ) takes place at the frequency  $\omega_1 + \omega_{\beta\gamma}$  then amplification ( $Q_{\gamma\beta}^{(i)} < 0$ ) takes place at the frequency  $\omega_1 - \omega_{\beta\gamma}$ . The quantities  $Q_{\beta\gamma}^{(2)}$  and  $Q_{\gamma\beta}^{(2)}$  differ from  $Q_{\beta\gamma}^{(1)}$  and  $Q_{\gamma\beta}^{(1)}$  through the replacement  $\gamma_1 \rightarrow \gamma_2$  and  $\kappa \rightarrow 1 - \kappa$ .

It is noteworthy that at exact resonance  $x = 0$  ( $\Delta = 0$ ), the components  $Q_{\gamma\beta}^{(i)}$  and  $Q_{\beta\gamma}^{(i)}$  vanish, but the components (5.2) remain finite with  $Q_{\alpha\beta}^{(i)} = Q_{\alpha\gamma}^{(i)}$ . Taking into account  $\omega_{\beta\alpha} = -\omega_{\gamma\alpha} = \Omega/2$  also, we find that the spectrum for  $\Delta = 0$  contains two components corresponding to absorption at each of the frequencies  $\omega_i \pm \Omega/2$  ( $i = 1, 2$ ). For two-level systems in a single strong field we obtain a nonvanishing result for  $\Delta = 0$  only when we take into account terms to next order in the ratio  $\Gamma/\Omega$ . The spectrum is then found to be symmetric.<sup>29</sup>

For  $V$ -configurations of levels the components of the absorption spectrum contain collisional relaxation characteristics only as small corrections, which we omit.

## CONCLUSION

In this work we have calculated the intensities of the components of the fluorescence spectrum of a three-level atom in two strong laser fields and the absorption spectrum of a test signal, taking into account the optical-collisional effects of nonlinearity. We have shown that for the  $\Lambda$ -configuration of atomic levels when the exact two-photon resonance condition  $\omega_1 - \omega_2 = \omega_{ca}$  is satisfied, i.e., when the

CPT effect occurs, then in contrast with the collisionless case, the fluorescence signal does not drop out entirely. Hence the component intensities are determined by the corresponding collisional relaxation characteristics. We have computed the width of the black line and how it varies as a function of the intensity of the incident radiation. For a  $V$ -configuration of atomic levels the collisional relaxation characteristics determine only some, not all, of the components of the fluorescence spectrum. As we have shown, taking into account the influence of a strong field on the dynamics of the collisional processes causes the collisional relaxation characteristics to decrease exponentially, and as a consequence, reduces the intensities of the corresponding fluorescence and absorption spectra associated with them by the same factor. For very high fields a transition to the collisionless limit occurs.

In view of the sensitivity of this phenomenon to changes in the collisional relaxation characteristics in a strong field, it can be utilized effectively to experimentally verify theoretical deductions about the change in collisional dynamics produced by a strong external field.

<sup>1</sup>G. Orriols, *Nuovo Cim. B* **53**, 1 (1979).

<sup>2</sup>G. S. Agarwal and S. Iha, *J. Phys. B* **12**, 2655 (1979).

<sup>3</sup>P. M. Radmore and P. L. Knight, *J. Phys. B* **15**, 561 (1982).

<sup>4</sup>O. A. Kocharovskaya and Ya. I. Khanin, *Zh. Eksp. Teor. Fiz.* **90**, 1610 (1986) [*Sov. Phys. JETP* **63**, 945 (1986)].

<sup>5</sup>Yu. A. Vdovin and S. V. Traven', *Zh. Prikl. Spektrosk.* **47**, 977 (1987) [*J. Appl. Spectrosc.* **47**, 1288 (1988)].

<sup>6</sup>M. B. Gornyi, B. G. Matisov, and Yu. V. Rozhdestvenskii, *Zh. Eksp. Teor. Fiz.* **95**, 1263 (1989) [*Sov. Phys. JETP* **68**, 728 (1989)].

<sup>7</sup>H. R. Gray, R. M. Whitley, and C. R. Stroud, *Opt. Lett.* **3**, 218 (1978).

<sup>8</sup>G. Alzetta, L. Moi, and G. Orriols, *Nuovo Cim. B* **52**, 209 (1979).

<sup>9</sup>M. S. Feld, M. M. Burns, P. G. Pappas, and D. E. Muznick, *Opt. Lett.* **5**, 79 (1980).

<sup>10</sup>J. E. Thomas, P. R. Hemmer, S. Ezekiel *et al.*, *Phys. Rev. Lett.* **48**, 867 (1982).

<sup>11</sup>M. Kailova, P. Thorsen, and O. Paulsen, *Phys. Rev. A* **32**, 207 (1985).

<sup>12</sup>P. R. Hemmer, S. Ezekiel, and C. C. Leiby, *Opt. Lett.* **8**, 440 (1983).

<sup>13</sup>B. L. Dalton, R. McDuff, and P. L. Knight, *Opt. Acta* **32**, 61 (1985).

<sup>14</sup>C. P. Agarwal, *Phys. Rev. A* **24**, 1399 (1981).

<sup>15</sup>J. Mlynec, F. Mitschke, R. Deserno, and W. Lange, *Phys. Rev. A* **29**, 1297 (1984).

<sup>16</sup>V. G. Minogin and Yu. V. Rozhdestvenskii, *Zh. Eksp. Teor. Fiz.* **88**, 1950 (1985) [*Sov. Phys. JETP* **61**, 1156 (1985)].

<sup>17</sup>A. Aspect, E. Arimondo, R. Kaiser, *et al.*, *Phys. Rev. Lett.* **61**, 826 (1988).

<sup>18</sup>M. C. De Ligne and E. R. Eliel, *Opt. Comm.* **72**, 205 (1989).

<sup>19</sup>V. S. Lisitsa and S. I. Yakovlenko, *Zh. Eksp. Teor. Fiz.* **66**, 1550 (1974) [*Sov. Phys. JETP* **39**, 759 (1974)]; *Zh. Eksp. Teor. Fiz.* **68**, 479 (1975) [*Sov. Phys. JETP* **41**, 233 (1975)].

<sup>20</sup>D. S. Bakaev, Yu. A. Vdovin, V. M. Ermachenko, and S. I. Yakovlenko, *Zh. Eksp. Teor. Fiz.* **83**, 1297 (1982) [*Sov. Phys. JETP* **56**, 743 (1982)].

<sup>21</sup>K. Burnett, J. Cooper, P. D. Kleiber, and A. Ben-Reuven, *Phys. Rev. A* **25**, 1345 (1982).

<sup>22</sup>P. A. Apanasevich, S. Ya. Kilin, and A. P. Nizovtsev, *Zh. Prikl. Spektrosk.* **47**, 887 (1987) [*J. Appl. Spectrosc.* **47**, 1213 (1988)].

<sup>23</sup>A. G. Zhidkov and S. I. Yakovlenko, *Zh. Prikl. Spektrosk.* **47**, 971 (1987) [*J. Appl. Spectrosc.* **47**, 1282 (1988)].

<sup>24</sup>S. Cavalleri and K. Burnett, *J. Mol. Opt.* **35**, 1651 (1988).

<sup>25</sup>C. Cohen-Tannoudji and S. Reynoud, *J. Phys. B* **10**, 2311 (1977).

<sup>26</sup>P. R. Berman and R. Salomaa, *Phys. Rev. A* **17**, 2667 (1982).

<sup>27</sup>L. A. Vainshtein, I. I. Sobel'man, and L. P. Presnyakov, *Zh. Eksp. Teor. Fiz.* **43**, 518 (1962) [*Sov. Phys. JETP* **16**, 370 (1963)].

<sup>28</sup>C. Cohen-Tannoudji and S. Reynoud, *J. Phys. B* **10**, 2332 (1977).

<sup>29</sup>D. S. Bakaev, Yu. A. Vdovin, V. M. Ermachenko, and S. I. Yakovlenko, *Kvantovaya Elektron.* **12**, 126 (1985) [*Sov. J. Quantum Electron.* **15**, 72 (1985)].