Optical collisions and the atomic spectrum in a strong resonant field

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The spectrum of an atom located in a strong resonant electromagnetic field and experiencing elastic collisions with the buffer gas is considered. In contrast to most previous work, in the present investigation the direct effect of the field on the dynamics of elastic collision between the working atom and buffer gas atom is taken into account. A general system of equations is obtained for the density matrix of the composite system "atom + electromagnetic field" (atom "dressed" by the electromagnetic field). The equation system takes into account spontaneous transitions and elastic collisions between the working atom and the buffer gas atoms. Equations for the relaxation characteristics are obtained by considering the dynamics of the collision between the composite system and the buffer-gas atoms. Expressions for the shapes of the triplet resonance fluorescence lines and of the doublet due to transitions to a third nonresonant level, are derived from the transport equations. The shape of the absorption lines of a weak test signal is also considered. Relaxation characteristics which depend on the intensity and frequency of the electromagnetic field in a nontrivial manner enter in the expressions obtained for the spectral lines. In particular, the effect of the strong field on the collision act leads, in principle, to a narrowing of the resonance fluorescence lines down to the collisionless limit. The customarily used phenomenological elastic-collision characteristics (which do not depend on the properties of the electromagnetic field) are valid only in the impact limit corresponding to weak frequency detuning and electromagnetic field intensities.

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1. INTRODUCTION

The properties of the atom as a radiator are significantly altered when it is placed in a strong resonant field; in particular, the spontaneous-emission spectrum of the "atom + electromagnetic field" system differs from the spectrum of the atom (for the literature see the books[1,2] and the review[3]). In the Seventies it was theoretically predicted[4,5] that a strong electromagnetic field can influence directly also the atomic collisions, changing thereby the optical properties of the medium (for details see the reviews[6,7]). This leads to an optical-collisional nonlinearity that manifests itself, for example, in a decrease of the power absorbed per unit volume with increasing intensity ($I \gtrsim 10^7$–10^9 W/cm^2) of the external electromagnetic field.[5–7] This fact was relatively recently observed in experiments by Szöke and co-workers[8] and by Bonch-Bruevich and co-workers[9] (see also Ref. 7). The report of experiments[8,9] that demonstrate directly the influence of the electromagnetic field on the atomic-collision acts makes it urgent to consider also other manifestations of such an influence.

This paper deals with the effects of the change in the line shape in the spectrum of a compound "atom + electromagnetic field" system (atom "dressed" by the field) on account of the influence of the electromagnetic field on the frequency of the collisions compound system with the broadening structureless particles (inert buffer gas). Usually the influence of collisions on the spectra of resonant fluorescence and on the absorption of a weak test signal are considered phenomenologically by introducing the corresponding collision widths[10–12] (see also Refs. 1–3). Such an analysis does not take into account the influence of the electromagnetic field on the course of the broadening collision. More consistent is an approach[5,7] in which the relaxation characteristics of the medium are obtained on the basis of an analysis of the dynamics of the collisions between the compound "atom + electromagnetic field" system and the broadening particle. This approach, however, was previously used to find only "rough" optical characteristics of the medium: the power (of the strong electromagnetic field) absorbed per unit volume and the integrated (over the spectrum) intensities of the lines of spontaneous transitions in the compound system. To find these rough characteristics it was sufficient to consider only the populations of the levels of the compound atom + electromagnetic field system.[5,7] The line-shape analysis presented here, on the other hand, calls for allowance for the correlation of the collisions, and consequently makes it necessary to resort to the density-matrix formalism.[1)]

Roughly speaking, in this paper we attempt to construct a theory of spectral-line broadening not for an atom but for a compound system. This theory has certain peculiarities. First, in contrast to the "usual" broadening theory, the properties of the investigated object depend on the parameters of the external electromagnetic field, and it is the dependence on just these parameters which is of primary interest. Second, it is impossible in principle to confine oneself here to an adiabatic model, i.e., to exclude the influence of inelastic collisions on the line shape. The point is that light absorption on account of elastic collisions between the atom and the broadening particles corresponds to inelastic transitions in the compound atom + electromagnetic field system (so-called optical collisions or OC transitions[5,7]). Therefore,

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even if the atom considered has nondegenerate levels, there will correspond to it doubly degenerate or more accurately doublet states of the compound system, and it is precisely transitions between these states which determine mainly the shapes of the lines investigated here (see below).

Let us dwell in greater detail on the formulation of the problem. Let the atom A be located in a strong monochromatic field of intensity $\mathscr{E} = \mathscr{E}_0 \cos(\omega_L t)$, where the frequency ω_L is close to the frequency $\omega_0 = (E_b - E_a)/\hbar$ of the atomic transition $a \leftrightarrow b$ (E_a and E_b are the energies of the atomic levels). The atom A collides with the structureless particles B. The interaction of the atom A, of the electromagnetic field $\mathscr{E}$, and of the particle B will be described by the Hamiltonian

$$\hat{H}=\hat{H}_0+\hat{V}_{AB}, \quad \hat{H}_0=\hat{H}_A+\hat{H}_{\mathscr{E}}+\hat{V}_{A\mathscr{E}},$$

where $\hat{H}_A$ and $\hat{H}_{\mathscr{E}}$ are the Hamiltonians of the free atom and the field, and $\hat{V}_{AB}$ and $\hat{V}_{A\mathscr{E}}$ are the interaction operators. Let $\varphi_1 = |A(a), n\rangle$ and $\varphi_2 = |A(b), n-1\rangle$ be the wave functions of the free atom and the field (the eigenfunctions of the Hamiltonian $\hat{H}_A + \hat{H}_{\mathscr{E}}$), and let n be the number of photons in the laser field ($n \gg 1$). We neglect the degeneracy of the atomic levels. The determination of the stationary states of the compound atom + electromagnetic field system[5,7] ("dressed states"[3]) reduces to consideration of a two-level problem. The wave functions of the compound system, which are the eigenfunctions of the Hamiltonian $\hat{H}_0$, are of the form

$$\psi_1 \equiv |1, n\rangle = b_1\varphi_1 + b_2\varphi_2, \quad \psi_2 \equiv |2, n\rangle = b_2\varphi_1 - b_1\varphi_2. \quad (1.1)$$

Here $b_{1,2} = 2^{-1/2}(1 \pm \Delta\omega/\Omega)^{1/2}$ are the coefficients of the expansion of the wave functions ψ_1 and ψ_2 of the compound system in terms of the wave functions φ_1 and φ_2 of the basis, which does not take interaction into account; $\Omega = (\Delta\omega^2 + V^2)^{1/2}$ is the Rabi frequency ($V = \langle b|\hat{V}_{A\mathscr{E}}|a\rangle/\hbar = D_{ba}\mathscr{E}_0/\hbar$, D_{ba} is the matrix element of the transition of the atom) and $\Delta\omega = \omega_L - \omega_0$. The Rabi frequency characterizes, in particular, the time Ω of establishment of the quasistationary states ψ_1 and ψ_2. Corresponding to the states ψ_1 and ψ_2 of the compound system are the energies

$$E_j = \hbar\omega_L(n-1) + E_b + \hbar\Delta\omega/2 - (-1)^j\hbar\Omega/2, \quad j = 1, 2. \quad (1.2)$$

We consider the scattering of a strong resonant radiation by a two-level and by a three-level system (see the figure). For a two-level system, the spectral distribution of scattered radiation has, as is well known, three maxima (triplet). The central maximum corresponds to a frequency ω equal to the frequency ω_L of the strong field (Rayleigh scattering). The two remaining maxima are shifted $\pm\Omega$, from the central maximum so that their frequencies are $\omega_\pm = \omega_L \pm \Omega$ (three-photon scattering and fluorescence). In the language of the states of the compound system, this process is directly described as spontaneous transitions of the type $|1, n\rangle \to |1, n-1\rangle$, $|2, n\rangle \to |2, n-1\rangle$ (Rayleigh scattering $\omega = \omega_L$), $|1, n\rangle \to |2, n-1\rangle$ ($\omega = \omega_+$) and $|2, n\rangle \to |1, n-1\rangle$ ($\omega = \omega_-$).[5,7,14] For scattering with transitions to the third nonresonant level c, the spectral distribution of the radiation has two maxima (doublet). The appearance of these maxima correspond to the transitions $|1, n\rangle \to |A(c), n-1\rangle$ and $|2, n\rangle \to |A(c), n-1\rangle$ (Ref. 14). The absorption of a weak test signal by an atom situated in a strong resonant field is described similarly.

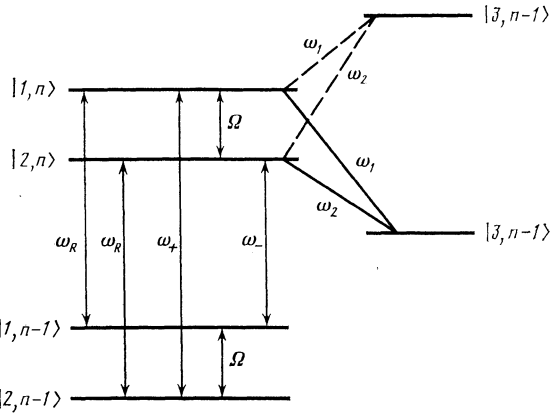

FIG. 1. Level scheme of compound atom + electromagnetic field system.

In the next section we construct a general system of transport equations for the density matrix of the compound system. This system of equations includes the relaxation characteristics obtained on the basis of an analysis of the dynamic problem—the collisions of the compound system with the broadening particles. Analysis of the dynamics of the broadening collisions (OC transitions) is given in Sec. 3 with account taken of effects of optical-collisional nonlinearity.[7] These effects arise when the matrix element of the transition in the atom becomes comparable under the influence of the field V with reciprocal collision time (the Weisskopf frequency) $\Omega_W = v/b_W$, where v is the relative velocity of the particles and b_W is the Weisskopf radius. The corresponding Weisskopf field $\mathscr{E}_W \approx \hbar v/D_{ba}b_W$ has at $b_W \approx 10$ a.u., $v \approx 10^{-4}$ a.u., and $D \approx 1$ a.u. a value $\mathscr{E}_W \approx 10^{-5}$ a.u. $\approx 10^5$ V/cm, corresponding to an intensity $I_W \approx 10^7$ W/cm^2. In Sec. 4 we consider the kinetic problem and obtain the spectra for the resonant triplet and also for the doublet corresponding to the transition to the nonresonant level. The final expressions contain the relaxation characteristics determined from the analysis of the dynamic problem and, generally speaking, dependent on the characteristics of the field (a dependence on $\Delta\omega$ and $\mathscr{E}_0$ appears at $\Omega \gtrsim \Omega_W$). The analysis is summarized in Sec. 5.

2. EQUATION FOR THE DENSITY MATRIX

We write the kinetic equation for the density matrix ρ of the compound atom + electromagnetic field system in the form

$$\frac{\partial\rho}{\partial t} = -\frac{i}{\hbar}(\hat{H}_0\rho - \rho\hat{H}_0) + \left(\frac{\partial\rho}{\partial t}\right)_{sp} + \left(\frac{\partial\rho}{\partial t}\right)_{col}, \quad (2.1)$$

where $\hat{H}_0$ is the Hamiltonian of the compound system and includes the interaction of the atom with the external monochromatic field; its eigenfunctions are of the form (1.1); $(\partial\rho/\partial t)_{sp}$ is the change of the density matrix on account of spontaneous transitions (see Ref. 14) and $(\partial\rho/\partial t)_{col}$ is the corresponding change due to other collisions with the buff-

er-gas atoms. The change of the density-matrix elements on account of the elastic collisions is represented in the form

$$\left(\frac{\partial}{\partial t}\langle i, n|\rho|j, n'\rangle\right)_{\text{col}} = -\Gamma_{ij}^{i'j'}\langle i', n|\rho|j', n'\rangle, \tag{2.2}$$

where the indices i, i', j, and j' run through the values 1 and 2. The matrix elements of the operator ρ are taken over the state of the compound system, i.e., in the basis of functions of the type (1.1); the indices n and n' describe the number of photons in the functions (1.1), i.e., specify the considered doublet (see the figure). Summation over repeated indices is implied.

We call attention to the fact that elastic collisions of an atom with a broadening particle lead to inelastic transitions in the compound system, i.e., to OC transitions.[5,7] We shall therefore call hereafter the matrix (operator) $\hat{\Gamma}$ the matrix (operator) of the rates of the OC transitions. The OC-transition rate matrix is expressed in terms of the scattering S matrix just as the collisional relaxation matrix is expressed in the theory of the broadening of atomic lines (see Ref. 15, p. 267). The only difference is that here we are dealing with broadening of not atomic levels but of levels of a compound system, so that the S matrix has a different form. Thus, for the matrix of the rates of the OC transitions we have

$$\Gamma_{ij}^{i'j'} = N\left\langle v\int_0^\infty 2\pi b\,db\,(\delta_{ii'}\delta_{jj'} - S_{ii'}S_{jj'}^*)\right\rangle, \tag{2.3}$$

where N is the density of the buffer-gas atoms; b is the impact parameter; the angle brackets denote averaging over the relative velocities v. From expression (2.3) and from the unitarity relations for the S matrix of a two-level system (see Ref. 16, p. 341 of Russ. transl.) it follows that

$$\Gamma_{ij}^{i'j'} = (\Gamma_{ji}^{j'i'})^*, \quad \Gamma_{12}^{11} = \Gamma_{12}^{22},$$
$$\Gamma_{11}^{11} = \Gamma_{22}^{22} = -\Gamma_{22}^{11} = -\Gamma_{11}^{22}, \quad \Gamma_{11}^{12} = -\Gamma_{22}^{12}. \tag{2.4}$$

In the derivation of (2.4) we used also the fact that the average kinetic energy of the relative motion (T) is much larger than the distance between the levels of the compound system ($T \gg \hbar\Omega$).

Of course, the elements of both the S matrix and the matrix of the rates of the OC transitions have in the general case a complicated dependence on the intensity $\mathscr{E}_0$ and on the frequency ω_L of the electromagnetic field. The quantity $\Gamma_{\text{OC}} \equiv \Gamma_{11}^{11} = \Gamma_{22}^{22}$ was introduced earlier,[5] and characterizes the frequency of the OC transitions $1 \leftrightarrow 2$ in the compound system under the influence of elastic collisions between the working atoms and the buffer-gas atoms, inasmuch as the quantity

$$W_{\text{OC}} = 1 - |S_{11}|^2 = 1 - |S_{22}|^2$$

gives the probability of the OC transition in one collision. It is convenient here to separate the "trivial" part of the dependence of relaxation characteristics on the parameters $\mathscr{E}_0$ and $\Delta\omega$ of the field and represent Γ_{OC} in the form[5,7]

$$\Gamma_{\text{OC}} = (V^2/2\Omega^2)\,\gamma_{\text{OC}}. \tag{2.5}$$

The quantity γ_{OC} characterizes the frequency of the elastic collisions that dephase the atom; γ_{OC} enters in the modified Lorentz formula that describes the line contour in broadening theory, and also the modified Karplus-Schwinger formula for the absorbed specific power.[7] In the general case γ_{OC} has a nontrivial dependence on $\Delta\omega$ and V. This dependence is lost only in the impact limit $\Omega \ll \Omega_W$ (Refs. 5 and 7, and below). Knowing the value of γ_{OC} we can describe the integrated characteristics of the scattered light, for example the intensities of the components of the triplet (see the figure). To construct a theory describing the line shape, however, it is necessary to know also other elements of the matrix of the rates of the OC transitions.

To find the density matrix ρ, with the aid of which we determine all the local characteristics of the medium, we must do the following: a) solve the dynamic problem and find the S matrix for the OC transitions; b) average over the impact parameter and over the relative velocities, and find the Γ matrix of the OC transitions; c) solve the transport equation (2.1) with allowance for (2.2) and (2.3). Such a procedure is possible only under various simplifying assumptions. In Secs. 4 and 5 below we use the assumption that the doublet levels of the compound system can be separated (see the figure), i.e., it is assumed that the Rabi frequency Ω is large compared with a spontaneous width γ_{sp} and with the collision widths:

$$\Omega \gg \gamma_{\text{sp}}, \quad \Omega \gg |\Gamma_{ij}^{i'j'}|. \tag{2.6}$$

In addition, in order not to take into account the inhomogeneous broadening, we put $\Omega \gg \gamma_D$, where γ_D is the Doppler width.

We note that the situation under which the conditions (2.6) are violated is of no interest for the problems considered below. The point is that the nontrivial dependence of the elements of the S matrix of the OC transitions $\Delta\omega$ and V arises only at $\Omega \gtrsim \Omega_W$ (Refs. 5 and 7). At the same time, by virtue of the binary character of the collisions we have $|\Gamma_{ij}^{i'j'}| \lesssim Nvb_W^2 \ll \Omega_W \sim v/b_W$. This enables us to construct a theory that takes, in principle, full account of the nonlinear optical-collisional effects.

3. LIMITING EXPRESSIONS FOR THE OC-TRANSITION MATRIX

Equation for the S matrix. In the basis of the functions of the compound system (1.1), the equations for the S matrix take the form[5,7]:

$$i\frac{dS}{dt} = \begin{pmatrix} U_1 & Ue^{i\Omega t} \\ Ue^{-i\Omega t} & U_2 \end{pmatrix} S, \quad S(-\infty) = 1. \tag{3.1}$$

Here $U_j = [U_b + U_a + (-1)^j \Delta\omega\Delta U/\Omega]/2$ are the level shifts of the compound system, $j = 1, 2$; $U = \Delta U V/2\Omega$ is the matrix element of the transition in the compound system, due to the elastic interaction with the broadening particle B; $U_m = \langle m|\hat{V}_{AB}|m\rangle/\hbar$, $m = a, b$ are the shifts of the atomic level by the particle B; $\Delta U = U_b - U_a$; it is assumed that the particle B causes no transitions between the atomic states: $\langle a|\hat{V}_{AB}|b\rangle = 0$ (the adiabatic approximation for the atom but not for the compound system). Equations (3.1) are valid

for binary collisions $\Omega_W \gg |\Gamma_{ij}^{i'j'}|$ and for not too large spontaneous widths $\Omega_W \gg \gamma_{sp}$.

For the nonresonant (third) state $|c\rangle$ of the atom the adiabatic approximation is valid, so that

$$S_{33}=\exp\left(-i\int_{-\infty}^{\infty} U_c\,dt\right), \tag{3.2}$$

where $U_c = \langle c|\hat{V}_{AB}|c\rangle$ is the shift of the state $|c\rangle$ by the broadening particle.

Impact approximation. Equation (3.1) admits of a general solution in the impact limit $\Omega \ll \Omega_W$ (Refs. 5 and 7). In this case it is necessary to neglect the nonlinear quantity Ωt in the arguments of the exponentials and make the change of variable

$$t \to \int_{-\infty}^{t} \Delta U dt',$$

after which (3.1) reduces to a second-order equation with constant coefficients. As a result we have

$$S=e^{-i\Phi_+}\begin{pmatrix} \cos\Phi_- + i\sin\Phi_-(\Delta\omega/\Omega) & -i\sin\Phi_-(V/\Omega) \\ -i\sin\Phi_-(V/\Omega) & \cos\Phi_- - i\sin\Phi_-(\Delta\omega/\Omega)\end{pmatrix}, \tag{3.3}$$

$$\Phi_\pm = \int_{-\infty}^{\infty} dt\,(U_b \pm U_a)/2.$$

For a power-law interaction $U_m = C_m r^{-n}$ ($m = a, b$; C_m are the interaction constants of the power-law potential; $r = (v^2t^2 + b^2)^{1/2}$ is the distance between the nuclei) we obtain

$$\Phi_\pm = (b_W/b)^{n-1}\alpha_n (C_b \pm C_a)/(C_b - C_a),$$

$$b_W = [(C_b - C_a)/v]^{1/(n-1)}, \quad \alpha_n = \sqrt{\pi}\,\Gamma\left(\frac{n-1}{2}\right)\Big/\Gamma\left(\frac{n}{2}\right) \approx 1,$$

where $\Gamma(x)$ is the gamma function. From the form of the matrix (3.3) we have in addition to relations (2.4)

$$\Gamma_{11}^{11} = -\Gamma_{21}^{12} = -\Gamma_{12}^{21},\ \Gamma_{12}^{12} = \Gamma_{11}^{12} = -\Gamma_{22}^{12} = -\Gamma_{12}^{22} = (\Gamma_{21}^{11})^*. \tag{3.4}$$

All the relaxation characteristics are then expressed in terms of the quantities

$$\begin{aligned} \Gamma_{11}^{11} &= (V^2/2\Omega^2)\gamma_{imp}, \\ \Gamma_{12}^{11} &= (V/2\Omega)(\gamma_{imp}\Delta\omega/\Omega + i\Delta_{imp}), \\ \Gamma_{12}^{12} &= \gamma_{imp}(1+\Delta\omega^2/\Omega^2)/2 + i\Delta_{imp}\Delta\omega/\Omega. \end{aligned} \tag{3.5}$$

Here

$$\gamma_{imp} = N\left\langle v\int_0^\infty 2\pi b\,db\,(1-\cos 2\Phi_-)\right\rangle \sim \pi b_W^2 N v, \tag{3.6}$$

$$\Delta_{imp} = N\left\langle v\int_0^\infty 2\pi b\,db\sin 2\Phi_-\right\rangle \lesssim \gamma_{imp}$$

determine the width and the shift of the line in the contour of the absorption of the field energy $\mathscr{E}_0$ by the atoms. In particular, the quantities (3.6) enter in the Karplus-Schwinger formula for the absorbed power.[7] In this case $\gamma_{imp} = \gamma_{OC}$ at $\Omega \ll \Omega_W$. The analysis that follows (in Secs. 4 and 5) shows that in the impact limit, when the Γ matrix of the OC transitions is determined by Eqs. (3.4)–(3.6), the results of the present paper go over into the results of Ref. 11, which are based on a phenomenological account of the collisions.

The relaxation characteristics that describe the transitions to the third level take for a power-law potential, in the impact approximation, the form

$$\begin{aligned} \Gamma_{3j}^{3j} &= |C_{cb}/C_{ba}|^{2/(n-1)}[1+(-1)^j\Delta\omega/\Omega] \\ &\quad [\gamma_{imp} - i\,\mathrm{sign}\,(C_{cb}C_{ba})\Delta_{imp}]/2 \\ &+ |C_{ca}/C_{ba}|^{2/(n-1)}[1-(-1)^j\Delta\omega/\Omega][\gamma_{imp} - i\,\mathrm{sign}(C_{ca}C_{ba})\Delta_{imp}]/2, \end{aligned} \tag{3.7a}$$

$$\begin{aligned} \Gamma_{3i}^{3j} &= (V/2\Omega)\{|C_{ca}/C_{ba}|^{2/(n-1)}[\gamma_{imp} - i\,\mathrm{sign}\,(C_{ca}C_{ba})\Delta_{imp}] \\ &\quad - |C_{cb}/C_{ba}|^{2/(n-1)}[\gamma_{imp} - i\,\mathrm{sign}\,(C_{cb}C_{ba})\Delta_{imp}]\}, \qquad i \neq j, \end{aligned} \tag{3.7b}$$

where $C_{mm'} = C_m - C_{m'}$ ($m = a, b, c$; $i, j = 1, 2$).

Weak-field approximation. If the field is assumed to be sufficiently weak (V is small), we can obtain the S matrix by perturbation-theory methods from the matrix element of the transition $U = \Delta U V/2\Omega$. With allowance for terms of first order in V, we have

$$S = \begin{pmatrix} e^{-i\Phi_a} & e^{-i\Phi_a}(-iV/2\Delta\omega)\int_{-\infty}^{\infty} dt\Delta U e^{i\eta} \\ e^{-i\Phi_b}(-iV/2\Delta\omega)\int_{-\infty}^{\infty} dt\Delta U e^{-i\eta} & e^{-i\Phi_b}\end{pmatrix}, \tag{3.8}$$

where

$$\Phi_m = \int_{-\infty}^{\infty} U_m dt, \quad m = a, b$$

is the phase that accumulates in the state of the atom m on account of the interaction with the buffer gas

$$\eta = \Delta\omega t - \int_{-\infty}^{t} \Delta U dt' \tag{3.9}$$

is the phase advance in the compound system.

For the elements of the matrix of the rates of the OC transitions, which we shall need below, we have

$$\Gamma_{11}^{11} = (V^2/2\Delta\omega^2)\gamma_{OC}(\Delta\omega),\ \Gamma_{12}^{12} = \gamma_{imp} + i\Delta_{imp} - \gamma' V^2/2\Delta\omega^2, \tag{3.10}$$

where

$$\gamma_{OC}(\Delta\omega) = N\left\langle v\int_0^\infty 2\pi b\,db\left|\int_{-\infty}^{\infty} dt\Delta U e^{i\eta(t)}\right|^2\right\rangle \tag{3.11}$$

$$\gamma' = N\left\langle v\int_0^\infty 2\pi b\,db\,e^{2i\Phi_-}\int_{-\infty}^{\infty} dt\Delta U e^{i\eta(t)}\int_{-\infty}^{t} dt'\Delta U e^{-i\eta(t)}\right\rangle.$$

To calculate γ' we shall need terms of second order in V in the S matrix. In the impact limit $|\Delta\omega| \ll \Omega_W$ we can neglect the first term in the phase shift (3.9), in which case $\gamma_{OC} = \gamma_{imp}$ and (3.10) coincides with (3.5) for $V \ll |\Delta\omega|$. We note especially that the quantities (3.11) depend on the frequency detuning of the electromagnetic field. In other words, even in the weak-field limit we cannot introduce phenomenological constants that characterize only the interaction of the

atoms A and B and are independent of the properties of the electromagnetic field.

Quasistatic region. In the quasistatic limit $\Omega \gg \Omega_W$ the situation is not as simple as in the impact limit. In particular, the results depend strongly on whether or not there exists a point of intersection of the terms $r_{\Delta\omega}$ of the compound system, specified by the condition $\Delta U(r_{\Delta\omega})\Delta\omega/\Omega = \Omega$. The behavior of the quantity γ_{OC} as a function of $\Delta\omega$ and V was investigated earlier.[5,7] We shall dwell here only on cases in which the S-matrix element are small. This takes place, in particular, for sufficiently strong fields $V \gg \Omega_W$ (for more details see Refs. 5 and 7). If we neglect the $1 \to 2$ transitions in the course of the collision, then

$$S_{ij}=\delta_{ij}\exp\left(-i\int_{-\infty}^{\infty}U_j dt\right), \quad j=1,2$$

and one characteristic

$$\Gamma_{12}^{12}=N\left\langle v\int_0^{\infty}2\pi b db\left[1-\exp\left(i\Delta\omega\Omega^{-1}\int_{-\infty}^{\infty}\Delta U dt\right)\right]\right\rangle$$
$$=(\Delta\omega/\Omega)^{2/(n-1)}[\gamma_{\text{imp}}+i\Delta_{\text{imp}}\operatorname{sign}(\Delta\omega)], \quad (3.12)$$

turns out to be different from zero. The last equation is valid for a power-law interaction (see above). Thus, in sufficiently strong fields, $V \gg \Delta\omega$ and $V \gg \Omega_W$, a decrease takes place not only in the frequency of the OC transitions $1 \leftrightarrow 2'$, but also in the other characteristics of the collisional widths.

In the same limit ($V \gg \Delta\omega, \Omega_W$), the collision widths Γ_{3j}^{3j} of the doublet for the transition to the third level $|c\rangle$ tend to finite limits, and the constant C_a of the resonant level is replaced by another quantity C_j:

$$\Gamma_{3j}^{3j}=|C_{cj}/C_{ba}|^{2/(n-1)}[\gamma_{\text{imp}}-i\Delta_{\text{imp}}\operatorname{sign}(C_{cj}C_{ba})],$$
$$C_j=[C_a+C_b+(-1)^j C_{ba}\Delta\omega/\Omega]/2, \quad j=1,2. \quad (3.13)$$

4. RESONANT FLUORESCENCE AND ABSORPTION OF TEST SIGNAL

We first obtain expressions for the spectrum of the spontaneous transitions between the states of the compound system (the spectrum of the triplet) and for spontaneous transitions from states of the compound system into a third nonresonant state $|c\rangle$ (the spectrum of the doublet). For the sake of argument we shall assume that $|a\rangle$ is the ground state of the atom and $|b\rangle$ the excited one.

Spectrum of triplet. The spectral distribution of the scattered radiation is given by the Fourier component of the correlation function (see, e.g., Ref. 14).

$$W(\omega)=\pi^{-1}\operatorname{Re} g(\omega), \quad (4.1)$$

$$g(\omega)=k\int_0^T dt'\int_0^T dt\langle\tilde{D}^+(t)\tilde{D}^-(t')\rangle\exp[-i\omega(t-t')]\theta(t-t'), \quad (4.2)$$

$D^+(t)$ and $D^-(t')$ are the operators of the dipole moment of the atom in the Heisenberg representation, having respectively positive and negative frequency terms, $\theta(t-t')$ is the Heaviside ("step") function, and T is the time of irradiation by the strong field, while the factor k is defined below [see (4.9)]. We represent the dipole moment operator $\hat{D}^{\pm}$ in the form

$$\tilde{D}^{\pm}=\sum_{ij}\tilde{D}_{ij}^{\pm}=\sum_{ij}D_{ij}^{\pm}|i,n\rangle\langle j,n\mp1|, \quad i,j=1,2, \quad (4.3)$$

where

$$D_{ij}^{\pm}=\langle i,\ n|\tilde{D}|j,\ n\mp1\rangle. \quad (4.4)$$

The correlation function $\langle\hat{D}_{ij}^+(t)\hat{D}^-(t')\rangle$ satisfies at $t>t'$ the same equations as the mean value $\langle\hat{D}_{ij}^+(t)\rangle$ (Ref. 17). According to (4.3) we have

$$\langle\tilde{D}_{ij}^+(t)\rangle=\operatorname{Sp}\rho\tilde{D}_{ij}^+=\sum_n D_{ij}^+\langle j,n-1|\rho|i,n\rangle=D_{ij}^+(\rho_{ij}^+(t))^*, \quad (4.5)$$

$$\rho_{ij}^+=\sum_n\langle i,n|\rho|j,n-1\rangle. \quad (4.6)$$

The sum of the density matrix elements that are diagonal in n will be designated ρ_{ij}:

$$\rho_{ij}=\sum_n\langle i,n|\rho|j,n\rangle. \quad (4.7)$$

Thus, the quantities $\langle\hat{D}_{ij}^+(t)\rangle/D_{ij}^+$ and $\langle\hat{D}_{ij}^+(t)\hat{D}^-(t')\rangle/D_{ij}^+$ satisfy at $t>t'$ the same equations as the elements of the matrix $(\rho_{ij}^+)^*$.

When the inequalities (2.6) are satisfied, the system of equations for the matrix elements ρ_{ij}^+ is separable. The equations for the matrix elements ρ_{ij}^+ take according to (2.1), (2.2), and (14) the form

$$\frac{\partial\rho_{ij}^+}{\partial t}=-[i(\omega_L+\omega_{ij})+(\gamma_i+\gamma_j)/2+kD_{ii}D_{jj}^+\Gamma_{ij}^{ij}]\rho_{ij}^+, \quad i\neq j, \quad (4.8)$$

where the matrix element D_{ij}^+ are defined by relation (4.4) and k is the proportionality factor between the square of the modulus of the dipole-moment matrix element $D_{ij}^{\pm}$ and the probability of the spontaneous transition γ_{ij} between the states $|j,n\rangle$ and $|j,n-1\rangle$:

$$\gamma_{ij}=k|D_{ij}^-|^2=k|D_{ij}^+|^2,$$
$$\gamma_i=\sum_{j=1}^{2}\gamma_{ji}, \quad \omega_{ij}=(E_i-E_j)/\hbar, \quad \omega_{12}=\Omega. \quad (4.9)$$

The matrix elements ρ_{11}^+ and ρ_{22}^+ satisfy the system of equations

$$\frac{\partial\rho_{ii}^+}{\partial t}=-(i\omega_L+\gamma_i)\rho_{ii}^+ +\sum_{j=1}^{2}(\gamma_{ij}-\Gamma_{ii}^{jj})\rho_{jj}^+. \quad (4.10)$$

We represent the function $g(\omega)$ (4.2) in accordance with (4.3) in the form

$$g(\omega)=g_{12}(\omega)+g_{21}(\omega)+g_c(\omega), \quad (4.11)$$

$$g_{ij}(\omega)=k\int_0^T dt'\int_0^T dt\langle\tilde{D}_{ij}^+(t)\tilde{D}^-(t')\rangle\exp[-i\omega(t-t')]\theta(t-t'), \quad i\neq j. \quad (4.12)$$

The functions $g_{12}(\omega)$ and $g_{21}(\omega)$ describe the outer lines of the triplet, and the function

$$g_c(\omega)=g_{11}(\omega)+g_{22}(\omega) \quad (4.13)$$

describe the central peak. From (4.5) and (4.9), extending the integration with respect to t in (4.12) to infinity, we obtain

$$g_{ij}(\omega)=i(\Omega_{ij}-\omega)^{-1}k\int_0^T dt'\langle\hat{D}_{ij}^{+}(t')\hat{D}^{-}(t')\rangle$$

$$=i(\Omega_{ij}-\omega)^{-1}k\int_0^T dt'\sum_l D_{ij}^{+}D_{jl}^{-}\rho_{li}, \qquad (4.14)$$

$$\Omega_{ij}=\omega_L+\omega_{ij}+i(\gamma_i+\gamma_j)/2+i\Gamma_{ij}^{ij*}.$$

The Eqs. (4.7) for the matrix elements ρ_{ij} have according to (2.1), (2.2), and Ref. 14 the same form as the corresponding equations for ρ_{ij}^{*}, (4.8) and (4.10), if we omit from the latter $i\omega_L\rho_{ij}^{+}$. On the basis of inequalities (2.6), we can neglect in (4.14) the contribution of the off-diagonal elements ρ_{li}. Assuming that the main contribution to the integral (4.14) is made by stationary solutions, we obtain

$$g_{ij}=i(\Omega_{ij}-\omega)^{-1}T\gamma_{ji}\bar{\rho}_{ii}. \qquad (4.15)$$

We obtain in an analogous manner the function $g_c(\omega)$:

$$g_c(\omega)=T[\pi\delta(\omega-\omega_L)\gamma_{11}(\bar{\rho}_{11}-\bar{\rho}_{22})^2$$
$$+\gamma_{11}(i(\omega_L-\omega)-\gamma_{12}-\gamma_{21}-2\Gamma_{11}^{11})^{-1}(1-(\bar{\rho}_{11}-\bar{\rho}_{22})^2)], \qquad (4.16)$$

where the relations (2.4) were used. The stationary values of $\bar{\rho}_{11}$ and $\bar{\rho}_{22}$ are

$$\bar{\rho}_{11}=(\gamma_{12}+\Gamma_{11}^{11})/(\gamma_{12}+\gamma_{21}+2\Gamma_{11}^{11}), \quad \bar{\rho}_{22}=1-\bar{\rho}_{11}. \qquad (4.17)$$

Expressing the matrix elements taken over the wave functions of the compound system (1.1), in terms of the matrix elements of the transitions in the atom, we get

$$\gamma_{12}=(1+\Delta\omega/\Omega)^2\gamma/4, \quad \gamma_{21}=(1-\Delta\omega/\Omega)^2\gamma/4, \quad \gamma=\gamma_1+\gamma_2, \qquad (4.18)$$

$$\gamma_{11}=\gamma_{22}=kD_{11}^{-}D_{22}^{+}=kD_{22}^{-}D_{11}^{+}=\gamma V^2/4\Omega^2, \qquad (4.19)$$

where γ is the total spontaneous-radiation probability in the transition $|b\rangle\rightarrow|a\rangle$ in the atom A. From (4.17) and (4.18) it follows that

$$\rho_{11}=[(1+\Delta\omega/\Omega)^2\gamma/4+\Gamma_{11}^{11}]/[(1+\Delta\omega^2/\Omega^2)\gamma/2+2\Gamma_{11}^{11}]. \qquad (4.20)$$

The expression for the function $W(\omega)$ of the spectral distribution takes, according to (4.1), (4.2), (4.11), (4.13), (4.15), and (4.16), the form

$$W(\omega)=\frac{T}{\pi}\left\{(\bar{\rho}_{11}-\bar{\rho}_{22})^2\pi\gamma_{11}\delta(\omega-\omega_L)+\frac{\Gamma_R\gamma_{11}(1-(\bar{\rho}_{11}-\bar{\rho}_{22})^2)}{(\omega-\omega_L)^2+\Gamma_R^2}\right.$$
$$\left.+\Gamma\left[\frac{\gamma_{21}\bar{\rho}_{11}}{(\omega-\omega_L-\Omega)^2+\Gamma^2}+\frac{\gamma_{12}\bar{\rho}_{22}}{(\omega-\omega_L+\Omega)^2+\Gamma^2}\right]\right\}, \qquad (4.21)$$

where the widths of the triplet lines are determined by the expressions (see also Ref. 13)

$$\Gamma_R=(1+\Delta\omega^2/\Omega^2)\gamma/2+2\Gamma_{11}^{11}, \qquad (4.22)$$

$$\Gamma=(1+V^2/2\Omega^2)\gamma/2+\mathrm{Re}\,\Gamma_{12}^{12}. \qquad (4.23)$$

Using expressions (4.18)–(4.21), write down in explicit form the intensities of the triplet lines. The intensities of the coherent and incoherent components of the Rayleigh scattering Q_{CR} and Q'_R are respectively

$$Q_{CR}=\gamma_{11}(\bar{\rho}_{11}-\bar{\rho}_{22})^2=\Delta\omega^2V^2\gamma[\Omega^2+\Delta\omega^2+4\Gamma_{11}^{11}\gamma^{-1}\Omega^2]^{-2},$$
$$Q_R=\gamma_{11}[1-(\bar{\rho}_{11}-\bar{\rho}_{22})^2]=\frac{V^2\gamma}{4\Omega^2} \qquad (4.24)$$
$$\times\frac{[(\Omega+\Delta\omega)^2+4\Gamma_{11}^{11}\gamma^{-1}\Omega^2][(\Omega-\Delta\omega)^2+4\Gamma_{11}^{11}\gamma^{-1}\Omega^2]}{[\Omega^2+\Delta\omega^2+4\Gamma_{11}^{11}\gamma^{-1}\Omega^2]^2}$$

The total intensity of the Rayleigh scattering Q_R (Refs. 5 and 7)

$$Q_R=Q_{CR}+Q_R'=\gamma V^2/4\Omega^2, \qquad (4.25)$$

does not depend on the collisional relaxation characteristics. The intensities Q_+ and Q_- of the outer components of the triplet are[7]

$$Q_+=\gamma_{12}\bar{\rho}_{22}=\frac{\gamma V^4\Omega^{-2}+4\Gamma_{11}^{11}(\Omega+\Delta\omega)^2}{8[\Omega^2+\Delta\omega^2+4\Gamma_{11}^{11}\Omega^2\gamma^{-1}]}$$
$$Q_-=\gamma_{21}\bar{\rho}_{11}=\frac{\gamma V^4\Omega^{-2}+4\Gamma_{11}^{11}(\Omega-\Delta\omega)^2}{8[\Omega^2+\Delta\omega^2+4\Gamma_{11}^{11}\Omega^2\gamma^{-1}]}. \qquad (4.26)$$

If the collisions are neglected ($\Gamma_{11}^{11}=\Gamma_{12}^{12}=0$), the spectral distribution (4.21) becomes symmetrical and coincides with known results.[18,19] In the impact limit $\Omega\ll\Omega_W$, using (3.5), we arrive at the results of Ref. 10. At $\Omega\ll\Omega_W$ it is necessary to use for Γ_{11}^{11} and Γ_{12}^{12} the other expressions from Sec. 3 and from Refs. 5 and 7.

Spectrum of doublet. We find the line shape of the emission connected with the transition of the atom to the third level $|c\rangle$ (see the figure, state $|3, n-1\rangle$), unperturbed by the strong field (spontaneous Raman scattering). In this case, as is well known, instead of the triplet of the resonance fluorescence a doublet will be observed whose central frequencies are respectively

$$\omega_j=\omega_{j3}=(E_{jn}-E_c-(n-1)\hbar\omega_L)/\hbar$$
$$=\omega_{bc}-(-1)^j(\Omega-(-1)^j\Delta\omega)/2. \qquad (4.27)$$

Here and below $j=1, 2$. The stationary intensities of these lines, just as for the levels of the triplet (4.21), are determined by the relations

$$Q_j=\gamma_{3j}\bar{\rho}_{jj}=\bar{\rho}_{jj}(1+(-1)^j\Delta\omega/\Omega)\gamma_{cb}/2, \qquad (4.28)$$

where γ_{31} and γ_{32}, γ_{cb} are the probabilities of spontaneous emission for the transitions $|1, n\rangle\rightarrow|3, n-1\rangle$ and $|2, n\rangle\rightarrow|3, n-1\rangle$, $|b\rangle\rightarrow|c\rangle$, but in contrast to (4.20), to calculate $\bar{\rho}_{11}$ and $\bar{\rho}_{22}$ in the stationary state for a three-level system it is necessary to introduce the probability $\mathscr{T}$ of the inelastic transition from the state $|c\rangle$ into the ground state $|a\rangle$. We then obtain from the equations for $\rho_{11}(t)$, $\rho_{22}(t)$, and $\rho_{cc}(t)$

$$\bar{\rho}_{jj}=[(1-(-1)^j\Delta\omega/\Omega)^2\gamma/4+\Gamma_{11}^{11}][2\Gamma_{11}^{11}$$
$$+(1+\Delta\omega^2/\Omega^2)\gamma/2+\gamma_{bc}\mathscr{T}^{-1}(\Gamma_{11}^{11}+\gamma V^2/4\Omega^2)]^{-1}, \qquad (4.29)$$

where γ_{ab} is the probability of the spontaneous transition $b\rightarrow a$, $\gamma=\gamma_{cb}+\gamma_{ab}$.

The line widths Γ_1 and Γ_2 are obtained in analogy with (4.23) from the equations for the matrix elements

$$\frac{\partial\rho_{3j}^{+}}{\partial t}=-i[\omega_{3j}+\gamma_j/2+\Gamma_{3j}^{3j}]\rho_{3j}^{+}, \qquad (4.30)$$

where ω_{3j} are defined by relations (4.23). We obtain accordingly

$$\Gamma_j=(1+(-1)^j\Delta\omega/\Omega)\,\gamma/4+\mathrm{Re}\,\Gamma_{3j}^{3j}. \tag{4.31}$$

In the impact limit $\Omega\ll\Omega_W$, using for Γ_{3j}^{3j} expressions (3.7), we arrive at the results of Ref. 20, and in strong fields $V\gg\Omega_W$, $\Delta\omega$ it is necessary to use (3.13). The characteristics Γ_{3j}^{3j} are symmetric here with respect to the states $|a\rangle$ and $|b\rangle$.

Line contour of test-signal absorption. Assume that a classical test field with amplitude $\mathscr{E}'$ and frequency ω', also close to the frequency ω_0 of the resonant transition, is applied to the system in addition to the strong resonant field $\mathscr{E}$. We shall assume that the test field has a low enough intensity and does not perturb the levels of the compound system (dressed states). We obtain the absorption spectrum $W'(\omega')$ of this test signal. It is determined in analogy with (4.1) and (4.2) by the Fourier component of the correlation function[10]:

$$\langle\tilde{D}^-(t)\tilde{D}^+(t')-\tilde{D}^+(t)\tilde{D}^-(t')\rangle. \tag{4.32}$$

It follows directly from this expression that the components have the same central frequencies $\omega'=\omega_L\pm\Omega$ and the same widths as the fluorescence spectrum (4.21). The absorbed specific powers $Q_\pm$ at the frequencies $\omega'=\omega_L\pm\Omega$ will be

$$\begin{aligned}Q_+&=Q(\omega_L+\Omega)=\gamma_{12}k^{-1}(\mathscr{E}')^2(\bar{\rho}_{22}-\bar{\rho}_{11}),\\ Q_-&=Q(\omega_L-\Omega)=\gamma_{21}k^{-1}(\mathscr{E}')^2(\bar{\rho}_{11}-\bar{\rho}_{22}),\end{aligned} \tag{4.33}$$

where the level populations $\bar{\rho}_{11}$ and $\bar{\rho}_{22}$ are given by expressions (4.20) and (4.17), while the spontaneous-transition probabilities γ_{ij} are given by expressions (4.18) and the factor k is defined in (4.9). After substituting (4.20), (4.18), and (4.33) we obtain

$$Q_\pm=(\mathscr{E}')^2\gamma(2k)^{-1}\frac{\Delta\omega(\Delta\omega\pm\Omega)^2}{\Omega(\Omega^2+\Delta\omega^2+4\Gamma_{11}^{11}\Omega^2\gamma^{-1})}. \tag{4.34}$$

The quantities Q_+ and Q_- have opposite signs, that one component is enhanced while the other, symmetrical to it, is absorbed. The intensity of the central component at the frequency $\omega=\omega_L$ is equal to zero in this approximation, i.e., accurate to terms of order $|\Gamma_{ij}^{ij}|/\Omega$, since the level populations of the compound system, which have different values of the quantum number n, coincide at $n\gg1$. The widths of the lines corresponding to the intensities (4.34) are given as before by relation (4.23), i.e., are determined by the relaxation characteristic Γ_{12}^{12}. At resonance ($\Delta\omega\ll V$) the populations $\bar{\rho}_{11}$ and $\bar{\rho}_{22}$ are equal and the avlues of $Q_\pm$ tend to zero. A nonzero result is obtained with the problem is solved at a higher accuracy with respect to the smallness parameter (2.6) (see Ref. 10 in this connection).

A similar procedure is used to calculate the absorption line contour of the test signal on the transition to the third nonresonant level (the Autler-Townes doublet). If this third level lies above the level b, spontaneous transitions to it are impossible, and it is populated only because of the action of the weak field $\mathscr{E}'$ at the frequency ω' close to the transition frequency ω_{cb} (see the figure). The frequencies corresponding to the maxima of the doublet components are determined as before by relations (4.27), but they must be taken with the opposite sign, if the level c lies higher than b. The intensities of the components are determined by a relation of the type (4.33) in which, however, but now the transition is between level 1 (or 2) and the nonresonant level c. The widths of the components $\Gamma_{1,2}$ will be given by relations (4.31), to which it is necessary to add the spontaneous width γ_c of the nonresonant level c:

$$\Gamma_j=(1+(-1)^j\Delta\omega/\Omega)\,\gamma_b/4+\gamma_c/2+\mathrm{Re}\,\Gamma_{3j}^{3j}. \tag{4.35}$$

In the impact limit $\Omega\ll\Omega_W$ these expressions go over into the result of Ref. 20, while in a strong field $\Omega\gg\Omega_W$ it is necessary to use (3.13).

5. DISCUSSION OF RESULTS

We have obtained in this paper the fluorescence line shape [the spectrum of the triplet (4.21)–(4.26) and of the doublet (4.28), (4.29), (4.31)], and also considered by absorption of a weak test signal [for the triplet (4.33), (4.34) and for the doublet (4.33), (4.35)]. The quantities contained in the results are determined in various limiting cases by expressions obtained by analysis of the dynamic problem of OC transitions (Sec. 3).

The main general conclusion of the paper is that the use of phenomenological characteristics of elastic collisions (Refs. 1–3, 10–12, 20) is restricted by the limits of the impact region of the Rabi frequencies, $\Omega\ll\Omega_W$. At $\Omega\gtrsim\Omega_W$ the relaxation characteristics begin to depend on the properties of the electromagnetic field even in the weak field limit $V\ll\Omega_W$. In the strong-field limit $V\gg\Omega_W$, $\Delta\omega$ the quantities Γ_{11}^{11} and Γ_{12}^{12}, which determine the line widths of the resonant fluorescence (4.22) and (4.23), begin to decrease, so that the lines can narrow down, in principle, to the collisionless limit. This effect was discussed in general form in Ref. 21. The change of the integrated intensities of the sideband lines of the triplet on account of the influence of the field on the collision act (the dependence of $\Gamma_{11}^{11}=\Gamma_{OC}$ on $\Delta\omega$ and V) was predicted in Refs. 5 and 7.

The relaxation characteristics Γ_{31}^{31} and Γ_{32}^{32}, which determine the widths of the lines of the transition to the third nonresonant level $|c\rangle$ of the working atom, also depend on the field characteristics V and $\Delta\omega$. In the strong-field limit $V\gg\Omega_W$, $\Delta\omega$, the quantities Γ_{31}^{31} and Γ_{32}^{32} turn out to be symmetrical with respect to the parameters of the states $|a\rangle$ and $|b\rangle$ of the working atom A.

We note in conclusion that in the experiments performed to date[8,9] they investigated not the spectral characteristics but the line intensities of the compound atom + electromagnetic field system. It appears that observation of the spectrum of the compound system can yield in principle more extensive information on the interaction of atomic particles, since the spectral characteristics contain then not one quantity Γ_{11}^{11}, but two of them, Γ_{11}^{11} and Γ_{12}^{12}. Experiments are also possible in which the transitions between weakly polarized states $|b\rangle$ and $|c\rangle$ are investigated when state $|b\rangle$ is mixed by the strong field with the highly polarizable ($C_a\gg C_b$, C_c) state $|a\rangle$. It is possible that in experiments of this kind it will be useful to employ methods of optical spectroscopy that single out the collisional widths of the lines against the background of the Doppler contour.[22]

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[1)]A problem similar in formulation was considered inRef. 13. There, however, the dynamic problem of finding the S matrix was not solved and the spectral intensities of the scattered radiation were not calculated.

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