Spectra of an atom broadened by its own pressure in an electromagnetic field

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Absorption and scattering of strong resonant radiation in a medium consisting of identical atoms is considered, with account taken of the dependence of the collisional relaxation characteristics on the frequency ω and on the amplitude E_0 of the electromagnetic field strength. Expressions are obtained for the absorbed and scattered incident-radiation power, as well as of the absorbed (amplified) weak-signal power. The validity of the balance approach in a basis of "dressed" particles is demonstrated; expressions for the collision widths in terms of the scattering matrix are obtained.

1. INTRODUCTION

This paper deals with the spectra of light scattered or absorbed by a medium of identical atoms, with allowance for the optical-collisional (OC) nonlinearity. This nonlinearity is due to the effect of the magnetic field on the collision dynamics.[1–4] The spectra of an atom in a strong field, with account taken of OC effects, were considered in greatest detail in Refs. 5–11 for the case of broadening by the structureless particles of a buffer gas. Nonlinear OC effects in collisions of identical atoms were investigated in Ref. 2, where the absorbed power was determined by a balance approach in a "two atoms + field" basis, i.e., in a basis of "dressed" states.

In this paper the weak-signal scattering and absorption spectra are calculated for a gas of identical atoms in a strong field with allowance for the nonlinear OC effects. In addition, in contrast to Ref. 2, the problem of the absorbed strong-field power is solved on the basis of the Boltzmann kinetic equation for the density matrix. This confirms the validity of the balance approach 2 in the case of "well separated levels"[1,7] in a basis of dressed states, when $\Omega \gg \gamma$, where

$$\Omega = [\Delta\omega^2 + 4D^2E_0^2/\hbar^2]^{1/2}$$

is the Rabi frequency ($\mathbf{D}$ is the dipole moment of the atom, $\Delta\omega = \omega - \omega_0$ is the detuning of the resonant-radiation frequency) and γ is the reciprocal of the atom's phase-relaxation time.

Section 2 deals with the kinetic equation for the atom density matrix with a collision integral that takes into account the possibility of light absorption directly in the act of collision of identical particles, and the influence of the field both on the level-population kinetics and on the collision dynamics. In Sec. 3 are obtained expressions for the absorbed strong-radiation power with allowance for the nonlinear OC effects in the basis of the eigenstates of the atom. A transition is effected from the usual basis to the basis of dressed states and the balance relations[2] are obtained. A connection is established (in the new basis) between the collisional characteristics and the scattering matrix.

Expressions for the probing-signal power absorption (amplification and scattering are given in Sec. 4. It is shown that the collisional charactertistics of these spectra do not coincide with those of the absorption spectrum. In the dressed-state basis in the "shock" regions[7]

$$|\Delta\omega_0| \ll \Omega_W, \quad |\Delta\omega_0 \pm \Omega| \ll \Omega_W$$

(Ω_W is the Weisskopf frequency, see, e.g., Ref. 1; $\Delta\omega_0 = \omega_{sc} - \omega$, where ω_{sc} is the frequency of scattered photon), they are expressed in terms of the scattering matrix.

2. EQUATION FOR THE ATOM DENSITY MATRIX IN A HOMOGENEOUS GAS

The interaction of a field with a medium is determined by the polarization P of the latter or by the absorbed power Q. To determine them we must find the density matrix ρ of the absorbing atom.

The general expression for the density matrix of an individual atom in a field $\mathbf{E}$ is

$$(d\rho/dt)_{sp} = -\hat{D}\rho\hat{D}^+ + {}^1/_2(\hat{D}\hat{D}^+\rho + \rho\hat{D}\hat{D}^+), \tag{1}$$

where H_0 is the Hamiltonian of the isolated atom and $V = \mathbf{D}\cdot\mathbf{E}$ is the energy of interaction of the atom with the field. The second term is the change of ρ because of the spontaneous radiative transitions, and the third is the collision integral.

The expression for $(d\rho/dt)_{sp}$, which is valid also for a degenerate system, can be rewritten in the form 12

$$d\rho/dt = -i[H_0 + V, \rho] + (d\rho/dt)_{sp} + (d\rho/dt)_{\text{coll}}, \tag{2}$$

where

$$\hat{D} = (3\pi\hbar c^3)^{-1/2} \sum_{ij} \omega_{ij}^{1/2} D_{ij} |i\rangle\langle j|,$$

$$\hat{D}^+ = (3\pi\hbar c^3)^{-1/2} \sum_{ij} \omega_{ij}^{1/2} D_{ij} |j\rangle\langle i|, \quad \varepsilon_j > \varepsilon_i,$$

with

$$(\hat{D}\hat{D}^+)_{ii} = (\hat{D}^+\hat{D})_{jj} = A_{ij},$$

and A_{ij} is the probability of the spontaneous $j \to i$ radiative transition.

The entire gist of the broadening by its own pressure is contained in the collision integral. Since the nonlinear OC effects manifest themselves in the quasistatic region $\Omega > \Omega_W$ (Refs. 1, 2) the collision integral should take into account the evolution of both atoms in the field directly in the collision act. Without discussing the detailed derivation, which is similar to that of Refs. 8 and 9, we present an expression for the collision integral in the binary approximation. It is obtained under the assumption that the colliding atoms move

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classically in a straight line, and generalizes Ref. 10 to the case of resonant excitation transfer:

$$\left(\frac{d\rho}{dt}\right)_{\text{coll}}=\left\langle\int_{-\infty}^{\infty}d\tau\,\mathrm{Sp}'\left[\hat{U},-i\hat{\rho}\times\hat{\rho}'(t)+\int_{-\infty}^{t}S^{+}(t,t')\times[\hat{U}(t'),\rho\times\rho']S(t,t')\,dt'\right]\right\rangle. \tag{3}$$

The multiplication sign $\times$ denotes here the direct product; the angle brackets denote averaging over the velocities and the impact parameters of the broadening particle; τ is the instantaneous collision time; ρ and ρ' are density matrices that satisfy one and the same equation (1), but describe generally speaking the states of atoms with differing velocities; as a result, (3) has a quadratic dependence on ρ; the symbol Sp′ stands for the trace over the states of the "primed" atom; S^+ and S are the evolution operators of both atoms, and are determined by the equations

$$\begin{aligned}iS^{+}&=(H_0\times 1'+V\times 1'+1\times H_0'+1\times V'+U)S^{+},\\ iS&=-S(H_0\times 1'+V\times 1'+1\times H_0'+1\times V'+U),\\ S^{+}(t,t)&=S(t,t)=1\times 1',\end{aligned} \tag{4}$$

where V and U are the operators for the interaction of the atoms with the fields and with one another.

The interaction energy of like atoms takes in the dipole approximation the form

$$\hat{U}=\frac{1}{R^3}[\mathbf{DD}'-3(\mathbf{Dn})(\mathbf{D}'\mathbf{n})], \tag{5}$$

where $\mathbf{n}$ is a unit vector in the direction of the axis joining the nuclei, and R is the distance between the atoms.

S^+, S, and U are two-particle operators; to label their matrix elements it is convenient to introduce the states

$$|\bar{1}\rangle=|1\rangle|1'\rangle,\quad |\bar{2}\rangle=|2\rangle|1'\rangle,$$

$$|\bar{3}\rangle=|1\rangle|2'\rangle,\quad |\bar{4}\rangle=|2\rangle|2'\rangle.$$

The elements $U_{\bar{2}\bar{3}}$ and $U_{\bar{3}\bar{2}}$ that determine the resonant excitation transfer differ from zero in this representation.

The collision integral (3) contains both kinetic (t) and dynamic (τ) times. This means that the broadening collisions are not equivalent in an arbitrary field at times t and t' for which $|t-t'|\gg\Omega_W^{-1}$. If, however,the radiation-pulse duration is long compared with Ω_W^{-1}, there is nothing to distinguish between the broadening collisions, and the dependence on the kinetic time in (3) should be contained only in the density matrix that determines the "initial" state of the system prior to the collision.

To prove this we introduce the kinetic time T of closest approach of the atoms at a fixed collision, and define it by the relations $t=T+\tau$ and $t'=T+\tau'$. Since the atom interaction energy changes substantially over times $\sim\Omega_W^{-1}$, we have $U(t)=U(t-T)=U(\tau)$. We obtain next from (4)

$$\begin{aligned}S^{+}(t,t')_{ij}^{kl}&=S^{+}(\tau,\tau')_{ij}^{kl}\exp[i\omega_{ij}T+i\omega_{kl}T],\\ S(t,t')_{ij}^{kl}&=S(\tau,\tau')_{ij}^{kl}\exp[i\omega_{ij}T+i\omega_{kl}T],\\ \omega_{ij}&=\varepsilon_i/\hbar+\omega-\varepsilon_j/\hbar,\quad \varepsilon_i<\varepsilon_j,\quad \omega_{ii}=0.\end{aligned}$$

The indices i and j pertain here to the unprimed atom, and k and l to the primed; thus, e.g.,

$$U_{12}^{21}=\langle 1,2'|U|2,1'\rangle=U_{\bar{2}\bar{3}}.$$

In the resonance approximation, the solution of (1) is

$$\rho_{ij}(t)=\bar{\rho}_{ij}\exp(i\omega_{ij}t),$$

where $\bar{\rho}_{ij}$ is a slowly (over times Ω_W^{-1}) varying function, which can be regarded as constant in (3).

Substitution of the presented relations in (3), with allowance for the fact that the nonzero U_{ij}^{kl} are those for which $\omega_{ij}+\omega_{kl}=0$, yields

$$\left(\frac{d\bar{\rho}}{dt}\right)_{\text{coll}}=\left\langle\int_{-\infty}^{\infty}d\tau\,\mathrm{Sp}'\left[U(\tau),-i\bar{\rho}(t)\times\bar{\rho}'(t)+\int_{-\infty}^{\tau}S^{+}(\tau,\tau')[U(\tau'),\hat{F}(\tau,\tau')\bar{\rho}(t)\times\bar{\rho}'(t)]S(\tau,\tau')\,d\tau'\right]\right\rangle. \tag{6a}$$

In the calculation of the matrix element of (6a), the action of the operator $\hat{F}$ is the following:

$$(\hat{F}(\tau,\tau')\bar{\rho}\times\bar{\rho}')_{i'j'}^{lm}=\exp[-i\omega_{ij}\tau+i(\omega_{i'j'}+\omega_{lm})\tau']\bar{\rho}_{i'j'}\bar{\rho}_{lm}'. \tag{6b}$$

It is easily seen that the matrix elements (6a) reduce to the form

$$(d\bar{\rho}/dt)_{\text{coll}}^{ij}=\langle\langle(\Gamma_{ij})_{i'j'}^{lm}\rangle_b\bar{\rho}_{lm}(t,\mathbf{v}')\rangle_{v'}\bar{\rho}_{i'j'}(t,\mathbf{v}) \tag{6c}$$

(with summation over the dummy indices). The quantities Γ do not depend explicitly on the time,and it is natural to refer to them as relaxation constants.

The condition $\dot{E}_0/E_0\ll\Omega_W$ needed to introduce these constants (E_0 is the field amplitude) has a more profound meaning. We indicate without proof that for an atom in an arbitrary bichromatic (and all the more, polychromatic) field with $|\omega_1-\omega_2|\gtrsim\Omega_W$ one cannot introduce the relaxation constants Γ in the basis of its eigenstates.[11] They can be introduced only in an "atom + bichromatic field" basis that has an infinite number of states, and Γ can differ substantially for the different states. This is reflected, in particular, in the fact that the probing-signal absorption and scattering spectra differ from Γ in (6), see Sec. 4. The analogy with the condition for the applicability of (6) can be easily perceived by recalling that at $\dot{E}_0/E_0\gtrsim\Omega_W$ the radiation constitutes a set of monochromatic waves with a wave spread that exceeds the Weisskopf frequency.

3. ABSORBED POWER. BALANCE RELATIONS

Equation (1), jointly with (2) and (6), determines completely the evolution of an atom broadened by its own pressure in a field having an arbitrary frequency detuning and amplitude (the latter varies quite slowly over a time Ω_W^{-1}), with allowance for the OC nonlinearities.

We consider below the limit $\dot{E}_0/E_0\ll\gamma_{in}$, where γ_{in} is the characteristic time of inelastic relaxation of the atom. In this case we can put $\bar{\rho}_{ij}=0$ in (2). Solution of (1) is made difficult by the nonlinear terms even in this case. We confine ourselves to two cases: a) weak field; b) the absorption-line wing ($\Delta\omega\gg\Delta\omega_D$, where $\Delta\omega_D$ is the Doppler width).

a) In a weak field all the ρ matrix elements but $\rho_{11}\sim 1$ are small. Therefore, when finding the off-diagonal element ρ_{12}, which determines the power absorbed per unit volume of the medium

$$Q \propto \omega_0 \langle \mathrm{Re}[iV^* \bar{\rho}_{12}(\mathbf{v})] \rangle_v,$$

it suffices to retain in the integral of (6) the terms proportional to $\bar{\rho}_{12}\bar{\rho}_{11}$, after which, by solving the simple integral equation, we get

$$Q \propto \omega_0 |V|^2 \left\langle \frac{1}{\delta^2} \frac{(\gamma_{oc}+A/2)k_1+(\Delta\omega+\omega_0 v/c)k_2}{k_1^2+k_2^2} \right\rangle_v,$$

$$k_1 = 1 + \left\langle \frac{1}{\delta^2}\left(\gamma_{oc}+\frac{A}{2}\right)\Delta_{oc} \right\rangle_v, \qquad (7)$$

$$k_2 = \left\langle \frac{1}{\delta^2}\left(\Delta\omega+\omega_0\frac{v}{c}\right)\Delta_{oc} \right\rangle_v,$$

$$\delta^2 = (\Delta\omega+\omega_0 v/c+\Delta_{oc})^2+(\gamma_{oc}+A/2)^2,$$

with

$$\gamma_{oc}+i\Delta_{oc} = (\Gamma_{12})_{12}{}^{11}+(\Gamma_{12})_{11}{}^{12}, \qquad (8)$$

$$\gamma_{oc} = \frac{N}{4}\left\langle \left| \int_{-\infty}^{\infty} U_0(\tau)\exp\left[-i\Delta\omega\tau + i\int_{-\infty}^{\tau} U_0(\tau')\,d\tau'\right] d\tau \right|^2 \right\rangle,$$

where $U_0 \approx D_{12}^2/2R^3$. In the impact region

$$\gamma_{oc}(\Delta\omega \ll \Omega_W) = \gamma_{sh} \approx 1.5\pi e^2 f N/m\omega_0$$

(f is the transition oscillator strength), which coincides with well-known results.[13,14]

We note that the frequency dependence of Q differs noticeably from that of Voigt. In the absence of Doppler broadening, the absorption spectrum has a Lorentz profile of width γ_{OC}.

b) In the far wing of the line we can neglect the nontrivial dependence of the ρ matrix elements on the velocity and set ρ and ρ' equal. It can then be easily verified that $\Gamma_{ii} = 0$. This result is natural, for in the absence of thermal motion excitation exchange is in the kinetic sense an elastic process.

Recognizing that the power absorbed per unit volume of a medium consisting of closed two-level systems coincides with the total scattered power $Q \propto A\rho_{22}$, we obtain

$$Q = -\hbar\omega AN \frac{\alpha+A+2\gamma_{oc}-\gamma_{int}}{4\gamma_{int}} \times \left\{ \left[1+\frac{8(A/2+\gamma_{oc})\gamma_{int}}{\alpha+A+2\gamma_{oc}-\gamma_{int}} \right]^{1/2} - 1 \right\}. \qquad (9)$$

Here

$$\alpha = A\Delta\omega^2/2V^2, \quad \gamma_{oc} = \mathrm{Re}[(\Gamma_{21})_{21}{}^{11} - (\Gamma_{12})_{21}{}^{11}],$$
$$\gamma_{int} = \mathrm{Re}[(\Gamma_{21})_{21}{}^{22} - (\Gamma_{12})_{21}{}^{22}] - \gamma_{oc}. \qquad (10)$$

Expression (9) goes over into the known Karplus-Schwinger equation[1,6] in the absence of saturation ($\alpha \gg A + 2\gamma_{OC} - \gamma_{\mathrm{int}}$), and also in the impact region $\Omega \ll \Omega_W$, with $\gamma_{\mathrm{int}} \to 0$.

In the general case, the more complicated form of Q, due to the nonlinear terms in the collision integral, is a reflection of the evolution of the "broadening" atom in the field. Since atoms having like states do not interact, at $\Delta\omega > \Omega_W$ the interaction is "turned on and off" $\Delta\omega/\Omega_W$ times in one collision. For this effect to manifest itself, we need rather strong fields $\rho_{22} \sim \rho_{11}$, i.e., nearly saturating fields.

In the quasistatic region $\Delta\omega > \Omega_W$ and in an intense field[1] we have

$$E_0 > E_{0\,cr} = \hbar^{7/6} v^{1/2} \Delta\omega^{2/3}/D^{4/3}$$

(see also Ref. 2) the collision widths decrease with increasing E_0:

$$\gamma \approx \hbar^{1/2} N v^{1/2} (\hbar\Delta\omega)^2/D^2 E_0{}^3.$$

The nonlinear OC effect leads to a decrease of the power absorbed per unit volume.

Calculation of Γ in (6) calls for determination of the evolution operator $S(t)$, followed by a complicated integration. It is much simpler to calculate Γ in the basis of dressed states, i.e., "atom + field" states.

The connection between density matrices in different bases is given by the unitary transformation $\tilde{\rho} = Y\rho Y^+$, where Y is the quasistationary solution for the atom evolution operator in the field[1,6,10]:

$$i\dot{Y} = -Y(H_0+V). \qquad (11)$$

The equation for $\tilde{\rho}$ follows directly from (1):

$$i\dot{\tilde{\rho}} = (d\tilde{\rho}/dt)_{sp} + (d\tilde{\rho}/dt)_{\mathrm{coll}}. \qquad (12)$$

The transformation (3) is carried out with allowance for the commutativity of Y and Y':

$$\left.\frac{d\tilde{\rho}}{dt}\right|_{\mathrm{coll}} = \left\langle \int_{-\infty}^{\infty} d\tau\, \mathrm{Sp}'\left[\bar{U}, -i\tilde{\rho}\times\tilde{\rho}' + \int_{-\infty}^{t} \bar{S}^+(t,t') \times[\bar{U}(t'), \tilde{\rho}\times\tilde{\rho}'(t')]\bar{S}(t,t')\,dt' \right] \right\rangle, \qquad (13)$$

where

$$\bar{U} = (Y\times Y')\,U\,(Y^+\times Y^{+\prime}),$$

and the evolution operators are defined by the equations

$$i\dot{\bar{S}}^+ = \bar{U}\bar{S}^+, \quad i\dot{\bar{S}} = -\bar{S}\bar{U}. \qquad (14)$$

The "dressing"' procedure (13), (14) corresponds to a transition to a separate new basis for each atom. In the two-level approximation for each atom we have

$$\bar{U} = U_0 \begin{vmatrix} \alpha_2 & \alpha_3\lambda & \alpha_3\lambda & -\alpha_2\lambda^2 \\ & -\alpha_2 & \alpha_1 & -\alpha_3/\lambda \\ & \text{c.c.} & -\alpha_2 & -\alpha_3/\lambda \\ & & & \alpha_2 \end{vmatrix}, \qquad (15)$$

where

$$U_0 = U_{\bar{2}\bar{3}} = U_{\bar{3}\bar{2}}, \quad \alpha_1 = {}^1/_2(1+\Delta\omega^2/\Omega^2), \quad \alpha_2 = 2V^2/\Omega^2,$$
$$\alpha_3 = V\Delta\omega/\Omega_1{}^2, \quad \lambda = \exp(i\Omega t).$$

The introduction of Γ in (13) is carried out, just as in the usual basis, with allowance for the fact that $U_0 = U_0(\tau)$, but the right-hand side of (15) is an explicit function of the kinetic time.

The density-matrix elements are of the form

$$\tilde{\rho}_{kk} = \bar{\tilde{\rho}}_{kk}, \quad \tilde{\rho}_{kl} = \bar{\tilde{\rho}}_{kl}\exp[i(-1)^l\Omega t].$$

We separate in the product $\tilde{\rho}\times\tilde{\rho}'$ the slowly varying (over times Ω_W^{-1}) components $\widetilde{M}\times\widetilde{M}'$. The simple transformation (13) with the aid of (14) shows that Γ for the components M can be calculated from the impact collision integral

$$\Gamma_M=\langle \mathrm{Sp}'(\tilde{S}_\infty^+\bar{\tilde{M}}\times\bar{\tilde{M}}'\tilde{S}_\infty-\bar{\tilde{M}}\times\bar{\tilde{M}}')\rangle, \tag{16}$$

where $\tilde{S}_\infty=\tilde{S}(\infty)$ is the scattering matrix. In the impact limit[2)] expression (16) is valid for the complete product $\tilde{\rho}\times\tilde{\rho}'$. For $(\Gamma_{ij})^{II}_{mm}$ it can be used also at $\Omega>\Omega_W$. The latter indicates already a possible equivalence of the approach described here and the balance approach.

A relation that follows from (16) is

$$\sum_p \langle (\tilde{S}_\infty^+)^{pl}_{ii'}(\tilde{S}_\infty)^{mp}_{j'j}-\delta_{ii'}\delta_{jj'}\delta_{lm}\delta_{pm}\rangle$$
$$=\sum_{p'}\Big(\Big\langle\int_{-\infty}^{\infty}d\tau\Big[\tilde{U}(\tau),-i\tilde{A}+\int_{-\infty}^{\tau}\tilde{S}^+(\tau,\tau')\times[\tilde{U}(\tau'),\tilde{A}]\tilde{S}(\tau,\tau')d\tau'\Big]\Big\rangle\Big)^{p'p'}, \tag{17}$$

where $\tilde{A}=\sigma_{i'j'}\sigma_{lm}$, σ_{ks} are projection operators:

$$(\sigma_{ks})_{rt}=\delta_{kr}\delta_{st},$$

and $\tilde{U}(\tau)$ is determined by (15) with the substitution $t\to\tau$.

In the dressed-states basis the absorbed power is given by

$$Q\propto\omega_0 V\,\mathrm{Im}\,\bar{\tilde{\rho}}_{12}.$$

To determine it we must resort to (12):[3)]

$$\partial\bar{\tilde{\rho}}_{22}/\partial t=(\Gamma_{22})_{ij}^{km}\bar{\tilde{\rho}}_{ij}\bar{\tilde{\rho}}_{km}+\tilde{\mathscr{T}}_{km}^{22}\bar{\tilde{\rho}}_{km}, \tag{18}$$
$$\partial\bar{\tilde{\rho}}_{12}/\partial t=-[i\Omega+(\Gamma_{12})_{12}^{km}\bar{\tilde{\rho}}_{km}]\bar{\tilde{\rho}}_{12}+(\Gamma_{12})_{ij}^{km}\bar{\tilde{\rho}}_{ij}\bar{\tilde{\rho}}_{km}+\tilde{\mathscr{T}}_{km}^{12}\bar{\tilde{\rho}}_{km}.$$

The off-diagonal matrix elements in (18) are

$$\bar{\tilde{\rho}}_{ij}\propto(-i\Omega+\gamma_1)\gamma_2/(\Omega^2+\gamma^2),$$

where γ_1 and γ_2 are some combinations of the relaxation constants. Clearly, in the case $\Omega\gg\gamma_1,\gamma_2$ the contribution of the off-diagonal elements is small ($\sim\gamma/\Omega$). Neglecting them, we obtain in the quasistationary approximation the simple balance relations

$$(\Gamma_{22})_{ii}^{mm}\bar{\tilde{\rho}}_{ii}\bar{\tilde{\rho}}_{mm}+\tilde{\mathscr{T}}_{mm}^{22}\bar{\tilde{\rho}}_{mm}=0,$$
$$Q\propto(\omega_0V/\Omega)[(\Gamma_{12})_{ii}^{mm}\bar{\tilde{\rho}}_{ii}\bar{\tilde{\rho}}_{mm}+\tilde{\mathscr{T}}_{mm}^{12}\bar{\tilde{\rho}}_{mm}]. \tag{19}$$

All the Γ in (19) are expressed in terms of the scattering matrix (16).

The equation for the populations reduces, by symmetrizing the states $|2\rangle$ and $|3\rangle$ and by using the properties of the scattering matrix, to the form

$$A(1+\Delta\omega/\Omega)^2\tilde{N}_2/4-A(1-\Delta\omega/\Omega)^2\tilde{N}_1/4-{}^1/_2(\tilde{N}_1-\tilde{N}_2)\{\tilde{N}_2W_{\tilde{3}\tilde{2}}+\tilde{N}_1W_{\tilde{1}\tilde{2}}+2NW_{\tilde{1}\tilde{3}}\}=0,$$

which is the result of Ref. 2 with the velocities W_{32} and W_{21} interchanges. Here

$$W_{\tilde{i}\tilde{j}}=\langle|\tilde{S}_\infty(v,\,b)_{\tilde{i}\tilde{j}}|^2\rangle_{v,b},$$
$$|\tilde{1}\rangle=|\bar{1}\rangle,\quad |\tilde{2}\rangle=(|\bar{2}\rangle+|\bar{3}\rangle)/\sqrt{2},\quad |\tilde{3}\rangle=|\bar{4}\rangle.$$

The disparity with Ref. 2 is due to an error in the choice of the necessary OC transition rates, and not to the principle underlying the balance equations.

It follows from the connection between $\tilde{N}_2$ and the density matrix in the usual basis that

$$\gamma_{oc}=2\Omega^2(W_{\tilde{1}\tilde{2}}+2W_{\tilde{1}\tilde{3}})/V^2,\quad \gamma_{inl}=2\Omega^2(W_{\tilde{1}\tilde{2}}-W_{\tilde{2}\tilde{3}})/V^2. \tag{20}$$

Relations (20) can be proved directly with the aid of (17).

The balance relations for the absorbed power are not expressed explicitly in terms of the OC transition rates W. The determination of Q in terms of the rates W requires the use of (17) and of cumbersome calculations, after which the results reduce to Eq. (5.18) of Ref. 2. This proves the validity of the balance approach and its full equivalance to that described here in the limit $\Omega\gg\gamma_{OC}+A/2$.

4. WEAK SIGNAL SCATTERING AND ABSORPTION (AMPLIFICATION) SPECTRA

Rayleigh scattering, resonant fluorescence, and weak-signal absorption are limiting cases of interaction of an atom with a bichromatic field. Nonetheless, since the interaction with the second field is weak, it is possible to introduce relaxation constants that are generally speaking different from Γ in (6).

To calculate the spectra it is convenient to use the probing-field method, which is the first order of perturbation theory. The corrections to the density matrix ρ_p for the interaction with the weak field or for spontaneous emission are defined by the equation

$$\dot{\rho}_p=-i[H_0+V_\omega,\rho_p]+(d\rho_p/dt)_{\mathrm{cn}}+(d\rho_p/dt)_{\mathrm{coll}}-iA_\omega^{(k)}, \tag{21}$$

where V_ω is the operator of interaction with the strong field, the second and third terms are given by Eqs. (2) and (3) with $\rho=\rho_p$, and the operator $A_\omega^{(k)}$ is of the form

$$A_\omega^{(1)}=[\mathbf{D}\times\mathbf{E}_p,\rho_\omega] \tag{22a}$$

for interaction with the weak field $\mathbf{E}_p$ ($\rho\omega$, the atom density matrix, is the solution of Eq. (1) with $\mathbf{E}_p\equiv0$) and

$$A_\omega^{(2)}=\hat{D}^+\rho_\omega-\rho_\omega\hat{D} \tag{22b}$$

for spontaneous radiative transitions [$\hat{D}+$ and $\hat{D}$ are defined in (2)].

The absorbed or scattered power is given for a two-level system by the expression

$$Q_1\propto-\omega_0N\,\mathrm{Re}[i\mathbf{D}_{21}\mathbf{E}_p\exp(-i\omega_0t)\rho_{12}^p],$$
$$Q_2\propto-\omega_0N\,\mathrm{Re}[i(3\pi\hbar c^3)^{-1/2}\omega_0^{1/2}D_{21}\exp(-i\Delta\omega_pt)\rho_{12}^p], \tag{23}$$

where $\Delta\omega_p=\omega_p-\omega_0$ and ω_p is the frequency of the weak field or of the reradiated photon.

The form of the collision integral follows from the expansion in (3):

$$\left(\frac{d\rho^p}{dt}\right)_{\mathrm{coll}}=\Big\langle\int_{-\infty}^{\infty}d\tau\,\mathrm{Sp}'\Big[U,-i(\rho_\omega\times\rho_p'+\rho_p\times\rho_\omega')+\int_{-\infty}^{t}S^+(t,t')[U,\rho_\omega\times\rho_p'+\rho_p\times\rho_\omega']S(t,t')dt'\Big]\Big\rangle. \tag{24}$$

The equation for the corrections ρ_p is thus linear, but the Γ depend on the matrix elements ρ_ω.

In the quasistationary ($\dot{E}_0/E_0\ll\gamma$) approximation the solution of (21) is of the form[11,16]

$$\rho_p=\exp(i\Delta\omega_0t)\hat{R}+\exp(-i\Delta\omega_0t)\bar{R}, \tag{25}$$

where $\Delta\omega_0 = \omega_p - \omega$. In the presence of thermal motion,

$$\Delta\omega_0 = \omega_p - \omega + \omega_0 \mathbf{v}(\mathbf{n}_p - \mathbf{n})/c,$$

where $\mathbf{n}_p$ and $\mathbf{n}$ are the propagation directions of the ω_p and ω photons. The time variation of $\hat{R}$ and $\hat{K}$ coincides with the time variation of ρ_ω. In the calculations it suffices to determine $\hat{R}$, in which case

$$Q \propto \mathrm{Re}[iV_p \exp(-i\Delta\omega_0 t) R_{12}].$$

Γ is introduced only for $\hat{R}$ (or $\hat{K}$, but not for ρ_p), just as in the case of a strong field:

$$\left(\frac{d\bar{R}}{dt}\right)_{\text{coll}} = \left\langle \int_{-\infty}^{\infty} d\tau \Big[U(\tau), -i(\bar{\rho}_\omega \times \bar{R}' + \bar{R} \times \bar{\rho}_\omega') + \int_{-\infty}^{\tau} S^+(\tau, \tau') [U(\tau'), F_p(\tau, \tau') (\bar{\rho}_\omega(t) \times \bar{R}'(t) + \bar{R}(t) \times \bar{\rho}_\omega'(t))] S(\tau, \tau') d\tau' \Big] \right\rangle. \tag{26}$$

The action of the operator $F_p(\tau, \tau')$ takes in the calculation of the matrix element of (26) the form

$$(F_p^{ij}(\tau, \tau') \bar{\rho}_\omega \times \bar{R}')_{i'j'}^{lm} = \exp[-i\omega_{ij}\tau + i(\omega_{i'j'} + \omega_{lm})\tau' + i\Delta\omega_0(\tau' - \tau)] \rho_{i'j'}^{\omega} R_{lm}'. \tag{27}$$

The relaxation constants that follow from (26) depend on the frequency ω_p, it is therefore more accurate to define them as relaxation rates. In the absence of thermal motion $(d\bar{R}/dt)_{ii} = 0$, and since the matrices R and ρ_ω are not equivalent the matrix elements $(d\bar{R}/dt)_{ij}$ contain terms proportional to $\bar{\rho}_{ij} \bar{R}_{ll}$.

Turning to Eq. (21), however, it can be easily seen that in the case $\Omega \gg \gamma$ we have

$$iV_\omega \bar{R}_{ll} \gg \Gamma \bar{\rho}_{ij} R_{ll}.$$

Neglect of the terms proportional to R_{ll} reduces (26) to the integral (6) in the region $|\Delta\omega_0| \ll \Omega_W$.

The weak-signal scattering and absorption spectra of a two-level system in the case of own-pressure broadening constitute triples with frequences $\omega_p = \omega$ and $\omega_p = \omega \pm \Omega$, just as in the case of broadening by buffer-gas atoms,[12] since the number of components is determined only by the external field and by the character of the system degeneracy.

In the absence of thermal motion, with allowance for the equivalence of the relaxation constants in (26) and (6) in the region $|\Delta\omega_0| \ll \Omega_W$, we obtain for the power of coherent Rayleigh scattering by a closed two-level system

$$Q_{cR} \propto \pi\omega |V_p|^2 (1-\theta) \delta(\omega_p - \omega) V_\omega^2 \frac{\Delta\omega^2}{(\Delta\omega^2 + 4V_\omega^2\eta)^2}, \tag{28a}$$

and for incoherent scattering and absorption

$$Q_{0R} \propto \omega |V_p|^2 \frac{V_\omega^2}{\Omega^2} \left[1 - \Omega^2 (1-\theta) \frac{\Delta\omega^2}{(\Delta\omega^2 + 4V_\omega^2\eta)^2} \right] \frac{\Gamma_0}{\Delta\omega_0^2 + \Gamma_0^2}. \tag{28b}$$

Here $\theta = 0$ for scattering, $\theta = 1$ for absorption, and

$$\eta = (1 + 2\gamma_{oc} + 2\gamma_{int}\rho_{22}^{\omega})/2A,$$

where γ_{OC} and γ_{int} are defined by Eqs. (20),

$$\rho_{22}^{\omega} = Q_\omega/\hbar\omega NA,$$

and Q_ω is given in (9). We note that $W_{0R} > 0$ for all $\Delta\omega$ and V_ω.

The width of the spectral distribution of incoherent Rayleigh scattering Q_{0R} is of the form

$$\Gamma_0 = A[\Delta\omega^2 + 4V_\omega^2\eta]/\Omega^2. \tag{29}$$

In the impact region $\Omega \ll \Omega_W$ expressions (28) and (29) go over into the known results of Ref. 12, with

$$\eta = (1 + 2\gamma_{sh})/2A.$$

In the region $\Omega > \Omega_W$ and of insufficiently strong fields, η decreases with increasing intensity in proportion to E_0^{-3}. the spectrum (28) can in this case narrow down to the collisionless width. Since usually the coefficient $\eta \gg 1$, its decrease leads to an increase of the fraction of the coherent Rayleigh scattering Q_{cR}.

In the case of absorption $Q_{cR} = 0$. Nonlinear effects are manifest only in a narrowing of the spectral distribution Q_{0R}.

It is convenient to calculate the resonant fluorescence and absorption spectra of a weak signal $\omega_p \sim \omega \pm \Omega$ in a dressed-state basis. The collision integral in this basis is similar to (26), with $F_p \to \tilde{F}_p$, and

$$(\tilde{F}_p^{ij} \tilde{\rho} \tilde{R}')_{i'j'}^{lm} = \exp[-i\tilde{\omega}_{ij}^{p}\tau + i(\tilde{\omega}_{i'j'}^{p} + \tilde{\omega}_{lm})\tau'] \tilde{\rho}_{i'j'} \tilde{R}_{lm},$$

$$\tilde{\omega}_{12} = \Omega = -\tilde{\omega}_{21}, \quad \tilde{\omega}_{ij}^{p} = \Delta\omega_0 + \tilde{\omega}_{lm}. \tag{30}$$

Assuming $\Omega \gg \gamma$ and neglecting the thermal motion of the atoms, calculations (see Ref. 11) of the intensities Q_+ of the resonant fluorescence and Q_- of the three-photon process, as well as of the absorbed power ($\theta = 1$) at $|\Delta\omega_0 \pm \Omega| \ll \Omega_W$, lead to the expressions

$$Q_+ \propto \omega N |V_p|^2 \frac{\Gamma_+}{(\Delta\omega_0 + \Omega)^2 + \Gamma_+^2} \frac{1}{2\Omega^2(\Delta\omega^2 + 4V_\omega^2\eta)} \times \{(\Omega + \Delta\omega)^2 [V_\omega^2\eta(1+\theta) - \theta(\Delta\omega^2/2 + 2V_\omega^2\eta)] - V_\omega^2 \Delta\omega(\Omega + \Delta\omega)\}, \tag{31a}$$

$$Q_- \propto \omega N |V_p|^2 \frac{\Gamma_-}{(\Delta\omega_0 - \Omega)^2 + \Gamma_-^2} \frac{1}{2\Omega^2(\Delta\omega^2 + 4V_\omega^2\eta)} \times \{(\Omega - \Delta\omega)^2 [V_\omega^2\eta(1+\theta) - \theta(\Delta\omega^2/2 + 2V_\omega^2\eta)] + V_\omega^2 \Delta\omega(\Omega - \Delta\omega)\}. \tag{31b}$$

It is easily seen that at $\theta = 0$ (scattering) expressions (31) are strictly positive. At $\theta = 1$ (absorption) we have $Q_+ < 0$ for all $\Delta\omega$ and V_ω, but $Q_- < 0$ for $V_\omega < \Delta\omega/\sqrt{2}$ and $Q_- > 0$ (corresponding to amplification) when this inequality does not hold.

The widths of the spectral distributions of Q_+ and Q_- are equal ($\Gamma_+ = \Gamma_-$) are are given by

$$\Gamma_\pm = {}^1/_2 A (1 + 2V_\omega^2/\Omega^2) + \langle (\bar{\Gamma}_{12})_{12}^{lm} + (\bar{\Gamma}_{12})_{lm}^{12} \rangle \bar{\bar{\rho}}_{lm}. \tag{32}$$

Since the off-diagonal elements $\bar{\bar{\rho}}_{ij} \sim \gamma/\Omega$, it is necessary to retain in (32) the collisional terms proportional to the diagonal elements. The quantities

$$(\Gamma_{12})_{12}^{ll} + (\Gamma_{12})_{ll}^{12}$$

are determined by the integral (26) in the basis of dressed

states with $\tilde{F}_p$ as defined in (30). Substituting the values of $\tilde{\omega}_{ij}$ in (30) we readily verify that relation (17) holds in the regions $|\Delta\omega_0 \pm \Omega| \ll \Omega_W$, and consequently the collisonal relaxation constants are expressed in terms of the scattering matrix (16):

$$(\Gamma_{ij})_{ij}^{il} = \left\langle \sum_{m=1}^{2} (\tilde{S}_\infty^{+})_{ii}^{ml} (\tilde{S}_\infty)_{jj}^{lm} - 1 \right\rangle. \quad (33a)$$

In the shock region $\Omega \ll \Omega_W$ we have

$$\Gamma_\pm = {}^1/_2 A (1+2V_\omega^2/\Omega^2) + (1-4V_\omega^2\Delta\omega^2/\Omega^4)\gamma_{sh} \approx \gamma_{sh}. \quad (33b)$$

In the quasistatic region $\Omega > \Omega_W$ at $E_0 > E_{0cr}$ the collision constants decrease with increasing E_0. Owing to the specific dependence of the widths of the spectra (31) on the field, the narrowing can manifest itself more strongly than in (28). In the general case the widths of the scattering and absorption spectra depend on the population $\tilde{\rho}_{22}$. To determine the latter we must use Eq. (19).

5. CONCLUSION

The atom spectra (9), (28), and (31) broadened in a magnetic field by the atom's own pressure differ from the analogous spectra broadened by a buffer gas only in the collision widths, which are connected by the simple relation (16) with the scattering matrix in the dressed-state basis. The effects of the OC nonlinearity, which manifest themselves by a decrease of the collisional widths in an intense field, are automatically taken into account here.

The experimentally observed scattering spectra can be considerably distorted by thermal motion and dragging of the radiation by the medium. The influence of thermal motion in weak-signal absorption spectra can be significantly decreased by aligning the propagation directions of the strong and weak fields ($\mathbf{n} = \mathbf{n}_p$). Since it is easy to allow for the drag in this case, the absorption spectrum is the most convenient for the observation of OC nonlinearity effects.

A stringent condition that limits the applicability of the results of this paper is the requirement that the collisions be binary:

$$N(v/\Omega_W)^3 = N(1{,}5e^2 f/m_e\omega_0 v)^{3/2} \ll 1. \quad (34)$$

Assuming $\omega_0 \approx 2\cdot 10^{15}$ s^{-1}, $j \approx 1$, and $v \approx 10^5$ cm/s we have $N \ll 10^{18}$ cm^{-3}. The Doppler broadening does not exceed the collisional one at $N \gtrsim 3\cdot 10^{16}$ cm^{-3}.

The situation is somewhat better for the absorption-line wing $\Delta\omega \gtrsim \Omega_W$, since the Doppler broadening is insignificant and the binarity condition is noticeably relaxed. The last statement is valid only for the absorption of the most intense radiation, but not for the scattering and absorption spectra of the probing signal. The binarity condition for these coincides with (34).

We note an important factor that influences the possibility of observing the effects of OC nonlinearity in a homogeneous gas. This is multiphoton ionization followed by breakdown in the gas. For alkali-metal atoms this is a two-photon process whose cross sections are not small. The use of more complicated atoms such as yttrium, which has a three-photon threshold, may possibly improve the situation. Otherwise the observation of the OC nonlinearity in metal vapors will be possible only if short radiation pulses are used.

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[1)] Assuming $v \approx 10^5$ cm/s, $D = 5$ a.u., and $\Delta\omega \approx 10^{12}$ s^{-1}, we get $E_0 \gtrsim 104$ W/cm.

[2)] At $\Omega \ll \Omega_W$ it is easy to calculate $\tilde{S}_\infty$ by putting $\lambda = \exp(i\Omega\tau) = 1$ in (15).

[3)] It is convenient to calculate the radiative relaxation constants $\tilde{\mathscr{T}}$ in (2) with the aid of the projection operators σ_{kl} (Ref. 11).

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