Resonance broadening in a strong light field

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Collisions of identical atoms in a strong resonance radiation field $E_0\cos\omega t$ for which the period of oscillation of an atom in the field is comparable to the collision time of the atom are considered. The problem of the absorption of light in such collisions is reduced to the problem of inelastic transitions in a three-level compound system "two atoms + electromagnetic field." Probabilities and cross sections for inelastic scattering are calculated for transitions between levels of such a system in two limiting cases—the impact case (rapid collisions) and the static case (slow collisions). In the general case these cross sections depend nonlinearly on the intensity of the field E_0. For values of E_0 which are small compared to a certain critical value E_0^* the results agree with the well-known linear theory of resonance broadening. For $E_0 \gg E_0^*$ a decrease in absorption (with increasing E_0) is observed in the wing of the line—the medium becomes "more transparent." The kinetics of absorption of light in a medium of identical atoms are considered taking into account collisions and inelastic spontaneous relaxation. The principal characteristic feature of the kinetics is the nonlinearity with respect to the occupancy by the atoms of the levels of the compound system. Stationary solutions are investigated. The results obtained are of direct interest for the observation of nonlinear effects both in the kinetics of light absorption, and also in the scattering of atoms, since these effects appear already in moderate fields $E_0^* \sim 10^4$–10^5 V/cm.

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§1. INTRODUCTION

The problem of resonance broadening of spectral lines and the problem associated with it of collisions of identical atoms has been repeatedly discussed in the literature (cf., for example, the review[1]). From the most recent papers on this subject we draw attention to articles[2,3] in which the cross sections for resonance scattering and the line shape of the radiation in a medium of identical atoms have been obtained in the most consistent manner.

The process of resonance collision and light absorption are usually discussed separately: at first the scattering cross sections are calculated and they are then utilized in the equations for the kinetics of light absorption in the medium. Such an approach is justified, as can be easily understood, for sufficiently weak electromagnetic (EM) fields which do not affect the process of atomic collisions. In strong fields this approximation breaks down. We estimate the order of magnitude of the EM field E_0^* at which the events of absorption and collision can no longer be separated. In order to do this we compare the characteristic atomic collision time $\tau \sim \rho_{eff}/v$ with the period T of oscillations of the atom in the EM field (ρ_{eff} is the effective impact parameter for the collision, v is the velocity of the atoms). In the case of a dipole–dipole interaction between atoms $V \sim d^2/\rho^3$ (d is the dipole moment for the transition) the characteristic collision radius is equal to $\rho_{eff} \sim d/\sqrt{v}$ (the Weisskopf radius[1,4]) from which we have $\tau \sim d/v^{3/2}$. Comparing τ with the period of oscillation of the atom in the EM field $T \sim (dE_0)^{-1}$ we obtain the magnitude[1)] of E_0^*:

$$E_0^* \sim v^{3/2}/d^2. \tag{1.1}$$

Assuming (in atomic units) $d \sim 1$, $v \sim 10^{-4}$, we obtain $E_0^* \sim 10^{-6} - 10^4$ V/cm—a value easily attainable in laser physics.

Taking into account the effect of the EM field on the process of atomic collisions means that we are dealing essentially with a nonlinear theory of resonance broadening in which the atomic scattering cross sections depend on the intensity E_0 of the EM field. For structureless perturbing particles the nonlinear theory of broadening was developed in[5–8]. The distinctive features of the case under investigation of identical particles are associated with the existence of "double resonance": in the system "atom + EM field" and in the system of two atoms. The fact that both colliding atoms possess internal structure brings the given problem closer to the so-called radiation collisions.[9] This means, in particular, that there is no sharp distinction between broadening (optical) and radiation collisions.

Below we consider collisions of identical atoms in a monochromatic (laser) field $E_0\cos\omega t$ the frequency ω of which is close to the frequency ω_0 of a transition in the atom so that

$$|\omega - \omega_0| \equiv |\Delta\omega| \ll \omega_0. \tag{1.2}$$

The effective ranges of interaction ρ_{eff} and the velocities of the atoms are such that in the collision process one can neglect the spontaneous relaxation of atoms and the spatial inhomogeneity of the field which is legitimate when

$$\rho_{eff}\gamma/v \ll 1, \quad \rho_{eff} \ll \lambda, \tag{1.3}$$

where γ is the radiation width of the upper level, λ is the wavelength of the radiation.

The atomic collisions are assumed to be binary (pair) and this is valid under the condition

$$N\rho_{eff}^3 \ll 1, \tag{1.4}$$

where N is the density of atoms. Within the framework

of the limitations indicated above only effects linear in the density of the broadening particles N are taken into account. This means, in particular, that the quasistatic distribution of intensity[1,4] can be realized only in the remote (binary) wing of the line.

In this paper we shall be interested in the main qualitative regularities of resonance broadening in a strong light field. Therefore at the basis of the present investigation lies the simplest two-level model of an atom widely used in laser physics.[10] In actual fact at least the excited state of atoms is degenerate with respect to the component of angular momentum. A generalization of the corresponding results taking such degeneracy into account does not encounter any difficulties in principle, even though it does significantly complicate quantitative calculations. Therefore we shall take it into account only in those cases when it leads to new qualitative consequences (cf., § 4). In other cases we shall neglect it.

§2. THE BASIC SYSTEM OF EQUATIONS FOR ATOMIC COLLISIONS IN AN EM FIELD

The wave function Ψ for a pair of interacting atoms X and Y in an EM field satisfies the Schrödinger equation with the Hamiltonian

$$\hat{H}(t)=\hat{H}_{0X}+\hat{H}_{0Y}+\hat{V}_{FX}+\hat{V}_{FY}+\hat{V}_{XY}(t), \tag{2.1}$$

where $\hat{H}_{0X}$ and $\hat{H}_{0Y}$ are the Hamiltonians for the free atoms X and Y, $\hat{V}_{FX}$ and $\hat{V}_{FY}$ are their interactions with the EM field, $\hat{V}_{XY}(t)$ describes the interaction of the atoms with each other.

Restricting ourselves to the dipole approximation we have

$$V_{FX}=-\mathbf{d}_X\mathbf{E}_0\cos\omega t, \quad V_{FY}=-\mathbf{d}_Y\mathbf{E}_0\cos\omega t, \tag{2.2}$$

$$\hat{V}_{XY}(t)=[\mathbf{d}_X\mathbf{d}_Y R^2(t)-3(\mathbf{d}_X\mathbf{R}(t))(\mathbf{d}_Y\mathbf{R}(t))]/R^5(t), \tag{2.3}$$

where $R^2(t)\equiv R_{XY}^2=\rho^2+v^2t^2$ is the distance between the atoms, while $\mathbf{d}_X$ and $\mathbf{d}_Y$ are the operators for the dipole moments of the atoms.

When the condition (1.2) is satisfied the wave function $\Psi(t)$ can be sought in the form

$$\Psi(t)=c_{00}\psi_{0X}\psi_{0Y}+c_{10}\psi_{1X}\psi_{0Y}+c_{01}\psi_{0X}\psi_{1Y}+c_{11}\psi_{1X}\psi_{1Y}. \tag{2.4}$$

We have introduced the notation X and Y distinguishing between the atoms only for the convenience of the argument. In what follows one should have in mind that X and Y are identical atoms. The functions ψ_{0X} and ψ_{1X} correspond respectively to the lower and the upper states of the atom.

It is convenient to introduce the coefficients

$$c_1=c_{00}e^{-i\Delta\omega t}, \quad c_2=\frac{c_{10}+c_{01}}{\sqrt{2}}, \quad c_3=c_{11}e^{i\Delta\omega t}, \quad c_4=\frac{c_{10}-c_{01}}{\sqrt{2}}. \tag{2.5}$$

The coefficients c_2 and c_4 correspond (after being substituted into (2.4)) respectively to the even and the odd combination of the atomic wave functions $\psi_{0X}\psi_{1Y}$ and $\psi_{1X}\psi_{0Y}$.

Substituting (2.4) and (2.5) into the Schrödinger equation we obtain a system of equations for the coefficients c_i:

$$\begin{aligned}&\dot{c}_1=-i\Delta\omega c_1+2^{1/2}iV_0c_2,\ \dot{c}_2=2^{1/2}i(V_0^*c_1+V_0c_3)-iV(t)c_2,\\ &\dot{c}_3=i\Delta\omega c_3+2^{1/2}iV_0^*c_2,\ \dot{c}_4=iV(t)c_4,\end{aligned} \tag{2.6}$$

where $V_0\equiv\mathbf{d}_{01}\cdot\mathbf{E}_0/2$ and $V(t)=2|\mathbf{d}_{01}|^2/R^3(t)$.

It can be seen that the amplitude of the odd state c_4 is not connected with the other amplitudes. The system (2.6) corresponds to the model of three levels with energies $+\Delta\omega$, 0, $-\Delta\omega$, which are described respectively by the amplitudes c_1, c_2, c_4, and c_3. The levels c_1 and c_3 are coupled to c_2 by the electromagnetic interaction V_0, while the interatomic interaction $V(t)$ is diagonal.

If we assume that the interaction $V(t)\equiv 0$, then we obtain a system of equations with constant coefficients the eigenvalues of which determine the levels of the compound system "two atoms + EM field." We now formulate the main assertion: transitions between these new levels of the compound system are responsible for light absorption. These transitions appear when the interaction $V(t)$ is switched on between the atoms, i.e., they are brought about by optical collisions (OC) of the atoms atoms. The assertion which we have just made can be easily verified by calculating the change in the average energy of the atoms in the EM field before and after a collision, cf., [7] and § 3.

We obtain the basic system of equations which describes transitions between levels of the compound system brought about by collisions. Setting $V(t)\equiv 0$, we find the matrix $\hat{A}(E_0, \Delta\omega)$, which diagonalizes the system (2.6) and which realizes the transformation to the compound system "atom + EM field":

$$\hat{A}=\begin{vmatrix}\frac{1}{2}\left(1-\frac{\Delta\omega}{\Omega}\right) & -\frac{V_0\sqrt{2}}{\Omega} & -\frac{1}{2}\left(1+\frac{\Delta\omega}{\Omega}\right)\\ \frac{V_0\sqrt{2}}{\Omega} & -\frac{\Delta\omega}{\Omega} & \frac{V_0\sqrt{2}}{\Omega}\\ \frac{1}{2}\left(1+\frac{\Delta\omega}{\Omega}\right) & \frac{V_0\sqrt{2}}{\Omega} & -\frac{1}{2}\left(1-\frac{\Delta\omega}{\Omega}\right)\end{vmatrix}. \tag{2.7}$$

It can be easily verified that the matrix $\hat{A}$ is unitary: $\hat{A}\hat{A}^+=1$. The energy eigenvalues $\mathscr{E}_i$ of the compound system are equal to

$$\mathscr{E}_i=0, \pm\Omega, \ \Omega\equiv(\Delta\omega^2+4V_0^2)^{1/2}. \tag{2.8}$$

We now transform the complete system (2.6) with the aid of the matrix $\hat{A}$ introducing the new amplitudes b in accordance with the relation

$$\hat{b}=\hat{A}^+\hat{c}, \ \hat{c}=\hat{A}\hat{b} \ (b_4=c_4). \tag{2.9}$$

Substituting (2.9) into (2.6) we obtain after some uncomplicated transformations

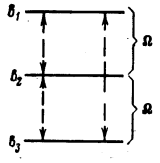

FIG. 1. The level scheme for the compound system "two atoms + EM field." Arrows indicate possible collision transitions.

$$\dot{\hat{b}}=i\begin{vmatrix}\Omega & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & -\Omega\end{vmatrix}\hat{b}+i\,\frac{V(t)}{1+\beta^2}\begin{vmatrix}-\frac{\beta^2}{2} & \frac{\beta}{\sqrt{2}} & -\frac{\beta^2}{2}\\ \frac{\beta}{\sqrt{2}} & -1 & \frac{\beta}{\sqrt{2}}\\ -\frac{\beta^2}{2} & \frac{\beta}{\sqrt{2}} & -\frac{\beta^2}{2}\end{vmatrix}\hat{b}, \qquad (2.10)$$

where $\beta\equiv 2|V_0/\Delta\omega|$,

$$b_i(t)=b_i(-\infty)\exp\left(i\int_{-\infty}^{t}V(\tau)\,d\tau\right). \qquad (2.11)$$

Equations (2.10) and (2.11) are the basic equations on the basis of which we shall be able to calculate the probabilities of transitions and, consequently, the absorption of light in the OC process. Thus, the problem of absorption (emission) of light in a collision is reduced to the problem of inelastic transitions in a three-level compound system "two atoms + EM field." We note that in the case of broadening by external (structureless) particles the corresponding system was a two-level one.[7] The scheme of levels of the system (2.10) and the possible transitions within it is shown in Fig. 1.

§3. THE SCATTERING MATRIX IN A COMPOUND SYSTEM. CHANGE IN ENERGY AS A RESULT OF A COLLISION.

We introduce the evolution operator $\hat{S}(-\infty, t)$ in the compound system:

$$\hat{b}(t)=\hat{F}(t)\hat{S}(-\infty, t)\hat{b}(-\infty). \qquad (3.1)$$

Here $\hat{b}(-\infty)$ are the amplitudes prior to the switching on of the interaction $V(t)$ ($V(-\infty)=0$). The matrix $\hat{F}(t)$ is equal to

$$\hat{F}(t)=\begin{vmatrix}e^{i\Omega t} & 0 & 0\\ 0 & 1 & 0\\ 0 & 0 & e^{-i\Omega t}\end{vmatrix}. \qquad (3.2)$$

The matrix S is unitary ($SS^+=1$) and satisfies the initial conditions: $S(-\infty, -\infty)=1$.

The squares of the corresponding nondiagonal matrix elements of the scattering matrix $S(-\infty, +\infty)$ define, evidently, the probabilities of inelastic transitions between the levels of the compound system. In order to relate these probabilities to absorption of light it is necessary to calculate the change in the energy of the atoms in the EM field after a collision.

We shall be interested in the internal (electronic) energy $\bar{\mathscr{E}}$ of the atoms averaged over a period of oscillation of an atom in the EM field $2\pi\Omega^{-1}$. It is not difficult to express it in terms of the initial coefficients c_i (2.5), and then, utilizing (2.9) in terms of the coefficients b_i:

$$\bar{\mathscr{E}}=\mathscr{E}_0+\mathscr{E}_1+\frac{\Delta\omega}{\Omega}(\mathscr{E}_1-\mathscr{E}_0)(|b_1|^2-|b_3|^2), \qquad (3.3)$$

where $\mathscr{E}_0$ and $\mathscr{E}_1$ are the energies of the atom in the lower and upper states ($\mathscr{E}_1-\mathscr{E}_0=\omega_0$).

The energy $\Delta\mathscr{E}$ absorbed (or emitted) in a collision is equal to the difference between average (over a period) energies $\bar{\mathscr{E}}$ after and before a collision:

$$\Delta\mathscr{E}=\bar{\mathscr{E}}(+\infty)-\bar{\mathscr{E}}(-\infty). \qquad (3.4)$$

Expressing $\bar{\mathscr{E}}$ in terms of the amplitudes b from (3.3) and taking into account the relationship $\hat{b}(+\infty)=\hat{S}(-\infty, +\infty)\hat{b}(-\infty)$ we can express $\Delta\mathscr{E}$ in terms of the elements of the scattering matrix S and the initial amplitudes $b(-\infty)$ for atoms in an EM field. We now take into account the fact that the atoms X and Y begin to interact with the EM field at random times t_X and t_Y (since they are situated randomly in the gas). Only the initial amplitudes $b_i(-\infty)$ depend on the instants of switching on t_X and t_Y. In order to obtain this simple dependence it is sufficient to compare the expressions for the wave function $\Psi(t)$ expressed on the one hand in terms of the coefficients $b_i(t)$, and on the other hand in terms of the products $\psi_X(t, t_X)$; $\psi_Y(t, t_Y)$ of the wave functions of an atom in an EM field that has been switched on respectively at the instants t_X and t_Y (cf., [11], the problem associated with § 40). Then by averaging the quantity $\Delta\mathscr{E}$ over the instants of switching on t_X and t_Y we obtain[2)]

$$\Delta\mathscr{E}=\omega\Omega^{-1}\Delta\omega\{|S_{12}|^2(b_2^2-b_1^2)+|S_{32}|^2(b_3^2-b_2^2)+2|S_{13}|^2(b_3^2-b_1^2)\}. \qquad (3.5)$$

The physical meaning of (3.5) is obvious: the absorbed energy is proportional to the transition probability (for example, $|S_{12}|^2$) multiplied by the difference in the populations of the initial and the final states (for example, $b_2^2-b_1^2$).

With the aid of (3.5) it is not difficult to write the expression for the power Q_{OC} absorbed in collisions. Indeed, the quantities $|b_i|^2$ are the probabilities of finding the compound system in corresponding states (Fig. 1). If we express these probabilities in terms of the densities N_I and N_{II} of a single atom in an EM field in levels I and II with the aid of the relations

$$|b_1|^2=N_{II}^2/N^2,\ |b_2|^2=N_IN_{II}/N^2,\ |b_3|^2=N_I^2/N^2, \qquad (3.6)$$

then the energy absorbed by the atoms per unit volume per unit time because of OC will be obtained by multiplying (3.5) by N^2v and by averaging over all velocities and impact parameters:

$$Q_{OC}=\omega\Omega^{-1}\Delta\omega(N_I-N_{II})\{W_{12}N_{II}+W_{32}N_I+2W_{13}N\}. \qquad (3.7)$$

Here N_I and N_{II} are the densities in the levels of the isolated atom in an EM field, while W_{ik} are the rates of the corresponding transitions:

$$W_{ik}=\left\langle v\int 2\pi\rho\,d\rho|S_{ik}(\rho, v)|^2\right\rangle_v=\langle v\sigma_{ik}\rangle_v \qquad (3.8)$$

($\langle\ldots\rangle_v$ denotes averaging over the velocity).

We note that, just as in[5-9], the cross sections σ_{ik} (and the rates W_{ik}) for the transitions depend not only on the characteristics of the colliding atoms but also on the characteristics of the external laser field (E_0 and $\Delta\omega$).

The fact that Q_{OC} vanishes as $\Delta\omega \to 0$ does not mean that there is no absorption at resonance. As was shown in[7] (cf., also § 5) the total absorbed power Q is made up of two terms: Q_{OC} and Q_{nr}—absorption due to inelastic (spontaneous) relaxation. For $\Delta\omega = 0$ and $V_0 \gg \gamma$ (γ is the maximum width of the levels) the medium becomes saturated, the role of absorption Q_{OC} associated with elastic relaxation diminishes and the absorbed power is determined entirely by the inelastic relaxation, cf.,[10].

§4. THE IMPACT AND THE QUASISTATIC APPROXIMATION. NONLINEAR EFFECTS IN SCATTERING

The system (2.10) represents a typical three-level system in the theory of atomic collisions. There exists no general solution of it and, therefore, we restrict ourselves to an investigation of two basic limiting cases: rapid and slow collisions.

The rapid collisions correspond to the criterion

$$\rho_{\text{eff}}\,\Omega/v \ll 1, \tag{4.1}$$

which serves as the basis of the impact approximation in the theory of broadening.[4,7] This criterion indicates that during the collision time the atom "does not have time" to oscillate in the EM field. This means in turn that the events of collision and absorption here can be separated. Therefore the cross sections for scattering in the impact approximation do not depend on the parameters of the EM field and coincide with the corresponding results of the usual impact theory.[4] This can be easily verified if one neglects in the solution of (2.10) the terms containing the quantity Ω.

Of the greatest interest (from the point of view of the effects under discussion here) is the case of slow collisions which correspond to the criterion inverse to (4.1). Here, however, one must take into account the nature of the space quantization of the atoms. Indeed, in the case of a slow collision of atoms in an EM field there are two defined directions in space: the vector of the EM field $\mathbf{E}_0$ and the vector of the interatomic distance $\mathbf{R}$. Below we shall consider the case

$$|\Delta\omega| \gg V_0,\ \beta \ll 1. \tag{4.2}$$

The criterion for the slowness of collisions has the form

$$\Delta\omega \gg v^{1/2}/d. \tag{4.3}$$

The principal feature of the problem when the conditions (4.2), (4.3) are satisfied can be easily discerned directly from the system (2.10). Indeed, for $\beta \equiv |V_0/\Delta\omega| = 0$ and with a gradual increase in V the two levels, 1 and 3, of the compound system (Fig. 1) remain unchanged, while level 2 is linearly displaced with increasing V. It is clear that for a certain value of $V \sim \Omega \approx \Delta\omega$ a point of intersection occurs between level 2 and level 1 (or 3). These points according to the Landau–Zener[11] theory of inelastic transitions are the factor responsible for transitions in the compound system.

As has been noted already, in the case of slow collisions the nature of the quantization of the atom is significant, and, therefore, it is necessary to generalize the initial system (2.6) taking into account the degeneracy of the states with respect to the component of angular momentum. We set the angular momentum L of the upper state to be equal to unity, and of the lower one to zero. Then the amplitudes c_i in (2.6) will depend on the quantum numbers of the components of angular momentum. Thus, for example, c_4^{mk} denotes the amplitude of the state in which the atom X has the component of angular momentum m, while the atom Y has the component k.

Introducing further the symmetric and the antisymmetric combinations of the amplitudes[3)]

$$\begin{aligned} c_\pm^m &= (c_2^m \pm c_3^m)/\sqrt{2}, \\ c_{4\pm}^{mk} &= (c_4^{mk} \pm c_4^{km})/\sqrt{2}, \end{aligned} \tag{4.4}$$

We obtain instead of (2.6) two systems of equations—the symmetric one:

$$\begin{aligned} \dot c_+^m &= 2^{1/2} i V_0^m c_1 + i V_0^k c_{4+}^{mk} - i V_{mm'} c_+^{m'}, \\ \dot c_1 + i\Delta\omega c_1 &= 2^{1/2} i V_0^m c_+^m, \\ \dot c_{4+}^{mk} - i\Delta\omega c_{4+}^{mk} &= i V_0^{k*} c_+^m + i V_0^{m*} c_+^k, \end{aligned} \tag{4.5}$$

and the antisymmetric one:

$$\begin{aligned} \dot c_-^m &= i V_0^k c_{4-}^{mk} + i V_{mm'} c_-^{m'}, \\ \dot c_{4-}^{mk} - i\Delta\omega c_{4-}^{mk} &= i V_0^{k*} c_-^m - i V_0^{m*} c_-^k. \end{aligned} \tag{4.6}$$

The matrix of the interatomic interaction in the system of coordinates with the Oz axis$\parallel$R has the form

$$V_{mm'}(t) = \frac{|d_{01}|^2}{R^3(t)} \begin{vmatrix} -2 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{vmatrix}. \tag{4.7}$$

When condition (4.2) is satisfied in order to obtain the levels of the quasimolecule composed of the pair of atoms we set in (4.5), (4.6) the interaction with the EM field V_0 equal to zero. Then the levels of the compound system as functions of V will have the form shown in Fig. 2. For $\Delta\omega < 0$ the levels c_1 and $c_{4\pm}^{mk}$ change places.

We obtain the transition probabilities w at the points of intersection by means of the Landau–Zener formula[11]

$$w = e^{-p},\ p = 2\pi |V_0|^2 / |\dot V(t_k)|. \tag{4.8}$$

Here V_0 is the nondiagonal matrix element for the transition between the levels under consideration; $\dot V(t_k)$ is the derivative of the potential at the point of intersection t_k determined by the condition

$$V(t_k) = \frac{|d_{01}^m|^2}{R_k^3} = \Delta\omega, \quad R_k = \left(\frac{|d_{01}^m|^2}{\Delta\omega}\right)^{1/3}, \quad t_k = \frac{1}{v}(R_k^2 - \rho^2)^{1/2}. \tag{4.9}$$

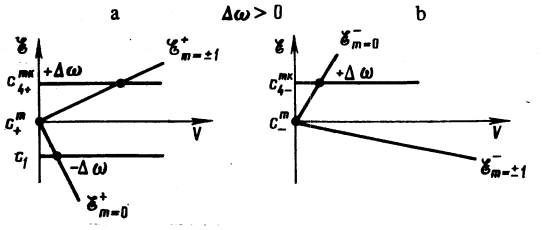

FIG. 2. Intersection of levels of a compound system.

The values of the nondiagonal matrix elements of V_0 for the components of angular momentum $m=0$ and $m=\pm 1$ are equal to (Oz||R)

$$|V_0^{m=0}|^2=\frac{|d_{01}|^2}{3}E_{0z}^2,\quad |V_0^{m=\pm 1}|^2=\frac{|d_{01}|^2}{3}\frac{E_{0x}^2+E_{0y}^2}{2} \tag{4.10}$$

When the level $\hat{c}$ intersects with the system of degenerate levels c_k it can be easily shown that the nondiagonal matrix element should be replaced by the effective matrix element

$$|(V_0)_{\text{eff}}|^2=\sum_k |V_0^k|^2.$$

We consider the geometry of the collision of the atoms in the EM field (Fig. 3). The field $\mathbf{E}_0$ is at an arbitrary angle θ to the normal n to the collision plane defined by the vectors ρ and v. The condition for the intersection of the levels (4.9) is realized at two points: $\mathbf{R}_k$ when the atoms approach each other and $\mathbf{R}_{k+1}$ when they are separating. The angles θ', θ'', formed by $\mathbf{E}_0$ with $\mathbf{R}_k$ and $\mathbf{R}_{k+1}$ are different, and, consequently, the nondiagonal matrix elements (and together with them the transition probabilities) are different at the two points. Therefore the total transition probability in passing through the two points of intersection is equal to[12]

$$w=w_k(1-w_{k+1})+w_{k+1}(1-w_k), \tag{4.11}$$

where w_k is determined by (4.8).

Taking the geometry of the collision into account the matrix elements (4.10) can be written in the form

$$|V_0^{m=0}|^2=\frac{|d_{01}|^2}{3}E_0^2\cos^2\begin{pmatrix}\theta'\\ \theta''\end{pmatrix}$$
$$=\frac{|d_{01}|^2}{3}E_0^2\sin^2\theta\frac{(\rho\cos\varphi\pm vt_k\sin\varphi)^2}{R_k^2}, \tag{4.12}$$

$$|V_0^{m=\pm 1}|^2=\frac{|d_{01}|^2}{6}E_0^2\sin^2\begin{pmatrix}\theta'\\ \theta''\end{pmatrix}$$
$$=\frac{|d_{01}|^2}{3}\frac{E_0^2}{2}\left[1-\sin^2\theta\frac{(\rho\cos\varphi\pm vt_k\sin\varphi)^2}{R_k^2}\right], \tag{4.13}$$

where the signs ± correspond to the points $\mathbf{R}_k$ and $\mathbf{R}_{k+1}$ (Fig. 3).

In order to obtain the total transition probability we have to average (4.11) over all the angles θ', θ'' (or θ, φ), and also over the impact parameters ρ. The quantities averaged in this manner obviously determine the cross sections for inelastic transitions between sublevels of the compound system. Thus, for a transition between the energy levels $\mathcal{E}_{m=0}^+$ and c_1 in Fig. 2 we have

$$\sigma_{m=0}^+=\pi(R_k^{m=0})^2\Lambda_1(2\pi E_0^2R_k^4/9v), \tag{4.14}$$

$$\Lambda_1(z)\equiv\int_0^1 dt\int_{-1}^{+1}dx\left\{\exp\left[-\frac{zx^2}{\sqrt{1-t}}\right]\right.$$
$$\left.-\frac{1}{2\pi}\int_0^{2\pi}d\varphi\exp\left[-\frac{2z(1-x^2)\,(t\cos^2\varphi+(1-t)\sin^2\varphi)}{\sqrt{1-t}}\right]\right\}. \tag{4.15}$$

Similarly for the cross sections $\sigma_{m=0}^-$ and $\sigma_{m=\pm 1}^+$ we find

$$\sigma_{m=0}^-=\pi(R_k^{m=0})^2\Lambda_2\left(\frac{2\pi E_0^2R_k^4}{9v}\right),\quad \sigma_{m=\pm 1}^+=\pi(R_k^{m=\pm 1})^2\Lambda_3\left(\frac{4\pi E_0^2R_k^4}{9v}\right). \tag{4.16}$$

The functions $\Lambda_1(z)$, $\Lambda_2(z)$ and $\Lambda_3(z)$ behave alike for $z\ll 1$:

$$\Lambda_1(z)\approx{}^4/_3z,\ \Lambda_2(z)\approx{}^8/_3z,\ \Lambda_3(z)\approx{}^{20}/_3z. \tag{4.17}$$

For $z\gg 1$ the function $\Lambda_1(z)$ plays the principal role:

$$\Lambda_1(z)\approx 4(\pi/z)^{1/2}/\pi,\ z\gg 1, \tag{4.18}$$

while the functions $\Lambda_2(z)$ and $\Lambda_3(z)$ are of a higher order of smallness ($\Lambda_2(z)\propto 1/z$ and $\Lambda_3(z)\propto e^{-z}$).

As follows from (4.14)–(4.18), the cross section for an inelastic transition between levels of the compound system (and thereby also the absorption of light) undergoes a qualitative change for fields E_0 greater than a certain critical field E_0^* which is equal, according to (4.14) and (4.16), to

$$E_0^*\sim v^{1/2}/R_k^2\sim v^{1/2}\Delta\omega^{2/3}/|d_{01}|^{4/3}. \tag{4.19}$$

Setting $\Delta\omega\sim v/\rho_{\text{eff}}\sim\tau^{-1}$ we obtain the estimate (1.1) given above.

In weak fields $E_0\ll E_0^*$ the absorption of light is of the usual nature, which can be easily obtained with the aid of (4.17):

$$\sigma\propto E_0^2R_k^6/v\propto|d_{01}|^2E_0^2|d_{01}|^2/\Delta\omega^2v, \tag{4.20}$$

i.e., the absorption is proportional to the intensity of light (E_0^2) and has the usual spectral distribution for resonance broadening in the wing of a line ($\propto 1/\Delta\omega^2$[11]).

For resonance broadening the distribution in the "negative" region ($\Delta\omega<0$) is of the same nature as in the region $\Delta\omega>0$. For other broadening mechanisms the line can have a sharp asymmetry associated with the exponential falling off of the "negative" wing (cf.,[6] and the literature quoted there). We note that this falling off is of the same nature as the exponential falling

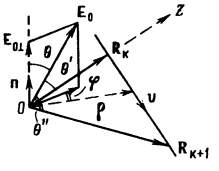

FIG. 3. Geometry of collision of atoms in an EM field.

off of the transition probability between two nonintersecting levels in the case of slow collisions (i.e., in the case of a large value of the Massey parameter $\rho\Delta\omega/v$). This result was obtained in[6] within the framework of the single-particle Spitzer approximation. A more rigorous calculation[13,14] shows that the terms neglected in this case are of a higher order of smallness in terms of the binary parameter $N\rho_{\text{eff}}^3 \ll 1$.

For strong fields $E_0 \gg E_0^*$ the absorption diminishes with increasing E_0. Indeed, with the aid of (4.18) we obtain

$$\sigma_{m=0}^{+} \propto v^{1/5}/E_0. \tag{4.21}$$

The dependence $\sigma \propto 1/E_0$ is typical for the behavior of the nonlinear cross section in the wing of the line.[12,8] It is of interest to note that (4.21) does not depend on the frequency detuning $\Delta\omega$. The specific nature of the resonance form of the interaction manifests itself in this.

§5. KINETICS OF ABSORPTION OF LIGHT IN A MEDIUM

In going over to the calculation of the kinetics of light absorption we note the following. Our investigation is based, following[7], on the elementary equations describing the balance of populations in the compound system "atom + EM field" utilizing transition probabilities, the absorbed energy and other quantities averaged over a period of oscillation of the atom in the EM field. Indeed, the time evolution of an atom is of a more complex nature associated with the quasiperiodic variation of the populations of the levels of the atom in the field. In the case of an analogous problem in NMR this was the first shown in[15]. A detailed analysis of the time evolution of a two-level system in an EM field has been carried out in[16]. Therefore in what follows we restrict ourselves to an investigation of the rougher characteristics, and in particular of the stationary absorbed power in a medium of identical atoms.

In order to obtain a stationary picture it is necessary to include in our investigation the processes of inelastic (for example, radiation) relaxation between the levels of an atom. Let the rates of transition between these levels be characterized by the relaxation constants γ_{01} and γ_{10} (for spontaneous transitions $\gamma_{01} \equiv 0$). As has been pointed out already in (1.3), we shall neglect relaxation in the process of collision. For the evaluation of absorbed power it is necessary to know the densities N_{I} and N_{II} associated with the levels I and II of the compound system "atom + EM field." These densities are determined by the balance between transitions brought about by inelastic relaxation and by collisions. For the inelastic relaxation we have the equation[7]

$$dN_{\text{I}}/dt = \gamma_{\text{II I}} N_{\text{II}} - \gamma_{\text{I II}} N_{\text{I}}, \tag{5.1}$$

where $\gamma_{\text{II I}}$ and $\gamma_{\text{I II}}$ are the rates of inelastic transition in the compound system "atom + EM field":

$$\gamma_{\text{II I}} = {}^1/_4(1+\Delta\omega/\Omega)^2\gamma_{10} + {}^1/_4(1-\Delta\omega/\Omega)^2\gamma_{01}, \tag{5.2}$$

$$\gamma_{\text{I II}} = {}^1/_4(1+\Delta\omega/\Omega)^2\gamma_{01} + {}^1/_4(1-\Delta\omega/\Omega)^2\gamma_{10}. \tag{5.3}$$

In order to be able to state the rates of transition due to collisions we note the following. We know the transition probabilities in the system "two atoms + EM field," while in kinetics it is necessary to know the transitions in the system "one atom + EM field." The problem is solved with the aid of the connection between the amplitudes of the system "two atoms + EM field" with the densities N_{I} and N_{II} characterizing the levels of the system "atom + EM field." For example, the population N_{I}, as can be easily seen from Fig. 1, is equal to

$$N_{\text{I}} = N[|b_3|^2 + {}^1/_2|b_2|^2 + {}^1/_2|b_4|^2] = {}^1/_2N[1+|b_3|^2-|b_1|^2]. \tag{5.4}$$

Formula (5.4) can be easily verified by the substitution of (3.6) and taking into account the equation

$$N_{\text{I}} + N_{\text{II}} = N. \tag{5.5}$$

Starting from (5.4) we obtain for the change ΔN_{I} in the population of the lower state as a result of one collision event (OC) we obtain

$$\Delta N_{\text{I}} = -{}^1/_2(N_{\text{I}} - N_{\text{II}})\{N_{\text{II}}|S_{12}|^2 + N_{\text{I}}|S_{32}|^2 + 2N|S_{13}|^2\}, \tag{5.6}$$

while the rate of change in the population of the level I is equal to

$$dN_{\text{I}}/dt = -{}^1/_2(N_{\text{I}} - N_{\text{II}})\{N_{\text{II}}W_{12} + N_{\text{I}}W_{32} + 2NW_{13}\}, \tag{5.7}$$

where W_{ik} are the rates of transition:

$$W_{ik} \equiv \langle v|S_{ik}|^2\rangle_{\rho,v} = \langle v\sigma_{ik}\rangle_v \tag{5.8}$$

(the symbol $\langle \ldots \rangle_{\rho,v}$ denotes averaging over the distribution of velocities of the particles and the impact parameters).

Combining (5.1) and (5.7) we obtain the desired equation for the balance of the occupancies in the system "atom + EM field" which takes into account inelastic relaxation and collisions:

$$dN_{\text{I}}/dt = 0 = \gamma_{\text{II I}}N_{\text{II}} - \gamma_{\text{I II}}N_{\text{I}} - {}^1/_2(N_{\text{I}} - N_{\text{II}})\{N_{\text{II}}W_{12} + N_{\text{I}}W_{32} + 2NW_{13}\}. \tag{5.9}$$

Equation (5.9) in the general case is nonlinear in contrast to the case of broadening by structureless particles.[7] The nonlinearity of the equation is associated with the very formulation of the problem: the transfer of excitation between atoms depends on the density of the excited atoms, i.e., on the absorption which, in its turn, is determined by the collision transfer of excitation. Such a formulation differs from the usual treatment of resonance broadening where the density of emitting excited atoms is essentially an independent parameter.

In the case when the transition probabilities in the system "two atoms + EM field" (Fig. 1) W_{12} and W_{32} are equal, Eq. (5.9) becomes linear:

$$dN_I/dt=\gamma_{II\,I}N_{II}-\gamma_{I\,II}N_I$$
$$-{}^1/_2N(N_I-N_{II})\{W_{12}+2W_{13}\}=0. \quad (5.10)$$

We investigate the stationary solution (5.9) which corresponds to the case $dN_I/dT=0$. In the impact limit (§ 4) the rates of transitions W_{12} and W_{32} are equal and we arrive at linear kinetics (5.10). This case differs from the case of broadening by structureless particles[7] only by a redefinition of the frequency of optical collisions. Therefore the corresponding results coincide with[7] (§ 4) and we shall not dwell upon them here.

We examine in detail the quasistatic limit corresponding to conditions (4.2), (4.3). Here in accordance with the results of § 4 the rate of transition W_{13} vanishes while the rates W_{12} and W_{32} in the general case are not equal to each other. We introduce two characteristic dimensionless parameters:

$$\lambda=W_{12}/W_{32}\geqslant 0, \quad (5.11)$$

characterizing the degree of nonlinearity of the kinetics, and

$$\varepsilon=\gamma_{10}/NW_{32}>0, \quad (5.12)$$

characterizing the ratio of inelastic relaxation to collision broadening. Below we shall consider the case of radiation relaxation when $\gamma_{01}=0$. When condition (4.2) is satisfied it follows from (5.2), (5.3) that for the case under consideration

$$\gamma_{II\,I}\approx\gamma_{10}, \quad \gamma_{I\,II}\approx\gamma_{01}=0.$$

Taking (5.11) and (5.12) into account the stationary case of equation (5.9) can be written in the following form

$$n_I^2(\lambda-1)-n_I\left(\frac{3\lambda-1}{2}+\varepsilon\right)+\varepsilon+\frac{\lambda}{2}=0, \quad (5.13)$$

where $n_I=N_I/N$ is the density in level I in units of density of perturbing particles N.

The solution of (5.13) has the form

$$n_I=\frac{1}{2}\left\{\frac{3\lambda-1}{2}+\varepsilon-\left[\left(\frac{3\lambda-1}{2}+\varepsilon\right)^2-4(\lambda-1)\left(\varepsilon+\frac{\lambda}{2}\right)\right]^{1/2}\right\}(\lambda-1)^{-1}. \quad (5.14)$$

The sign before the square root in (5.14) is chosen to be the minus sign from the condition of finiteness of n_I for $\lambda=1$ and the requirement $n_I\geqslant 0$. The solution exists if the descriminant Δ in (5.14) is greater than zero, i.e.

$$\Delta=\left(\frac{3\lambda-1}{2}+\varepsilon\right)^2-4(\lambda-1)\left(\varepsilon+\frac{\lambda}{2}\right)$$
$$=\left(\varepsilon-\frac{\lambda}{2}\right)^2+\frac{\lambda}{2}+\frac{1}{4}+3\varepsilon>0, \quad (5.15)$$

and this is satisfied for all $\varepsilon>0$ and $\lambda>0$.

It is not difficult to find the power Q_{OC} (3.7) absorbed in collisions. Taking (5.9), (5.5) and (5.14) into account we have

$$Q_{OC}=2\gamma_{10}\omega Nn_{II}(\varepsilon,\lambda),$$
$$n_{II}=1-n_I=\frac{1}{2}\left\{\frac{\lambda-3}{2}-\varepsilon+\left[\left(\frac{\lambda-3}{2}-\varepsilon\right)^2+2(\lambda-1)\right]^{1/2}\right\}(\lambda-1)^{-1}. \quad (5.16)$$

For absorption due to inelastic relaxation Q_{ir} we write the expression[7]

$$Q_{ir}=\gamma_{10}\omega N[1+2n_{II}(\varepsilon,\lambda)]V_0^2/\Delta\omega^2. \quad (5.17)$$

Then the total absorbed power Q is equal to

$$Q=Q_{ir}+Q_{ir}=\gamma_{10}\omega N[2n_{II}(\varepsilon,\lambda)+V_0^2/\Delta\omega^2]. \quad (5.18)$$

We should remember that $n_{II}(\varepsilon,\lambda)$ in (5.18) depends in accordance with (5.8), (5.12), and (5.14) on the characteristics of the EM field V_0 and $\Delta\omega$.

We investigate limiting cases of formula (5.18). Let, as is usually the case, the pressure broadening be large compared to broadening brought about by spontaneous decay. Then for fields $E_0\approx E_0^*$ (cf., (4.19)) the magnitude of the cross section is according to (4.14) of the order of πR_k^2 and for the quantity $\varepsilon=\varepsilon^*$ (5.12) we obtain[4)]

$$\varepsilon^*=\gamma_{10}/Nv\pi R_k^2\ll 1. \quad (5.19)$$

In this case it follows from (5.14) and (5.15) ($\lambda\sim 1$):

$$n_I\approx n_{II}\approx{}^1/_2, \quad \varepsilon\ll 1, \quad (5.20)$$

i.e., we obtain a result corresponding to saturation of the medium.

The quantity ε can become large compared to unity in two particularly interesting cases: $E_0\ll E_0^*$ and $E_0\gg E_0^*$. In the first case utilizing (4.17) and taking into account (4.14), (4.16) and (4.9) we obtain $\lambda=9/2$. In the second case utilizing (4.18) we obtain $\lambda\ll 1$. Then expanding (5.16) in a series for $\varepsilon\gg 1$ we obtain for both cases indicated above

$$n_{II}\approx{}^1/_2\varepsilon, \quad \varepsilon\gg 1, \quad (5.21)$$

i.e., a result independent of λ.

Substituting (5.21) into (5.18), we obtain taking (5.12) into account

$$Q=\omega N[NW_{32}(\Delta\omega,E_0)+\gamma_{10}V_0^2/\Delta\omega^2]. \quad (5.22)$$

For $E_0\ll E_0^*$ utilizing (4.20) we have

$$Q\approx\omega N[a_1N|d_{01}|^2+\gamma_{10}]V_0^2/\Delta\omega^2, \quad (5.23)$$

where $a_1=32\pi^2/27$. Thus, for weak fields absorption is, as it ought to be, proportional to the intensity of light V_0^2, and the collisional and radiation widths are simply additive.

For $E_0\gg E_0^*$ utilizing (4.21) we obtain

$$Q\approx\omega N[6\sqrt{2}Nv^{1/2}/E_0+\gamma_{10}V_0^2/\Delta\omega^2]. \quad (5.24)$$

It can be seen from (5.24) that in strong fields absorption associated with collisions is reduced—the medium becomes more transparent, cf., [6-8]. The transparency effect is the principal special feature of absorption of powerful EM radiation in the wing of the line. It is important to note that the absorbed power Q for $E_0 \gg E_0^*$ is proportional to the temperature of the medium ($\propto T^{3/4}$), so that this effect can be utilized to determine T.

The latter remark concerns the nature of approach to a stationary state. The nonlinearity of stationary equations noted above indicates that the very nature of approach to a stationary state will be different than in the case of linear equations. Without claiming to describe a nonstationary solution in the general case we restrict ourselves to the case of weak fields $|\Delta\omega| \gg V_0$, cf., (4.2), where we can count on approximate validity of the balance equations.[16] The nonstationary equation for this case (cf., (5.13)) has the form

$$\frac{dn_1}{dt'} = n_1^2(\lambda-1) - n_1\left(\frac{3\lambda-1}{2}+\varepsilon\right)+\varepsilon+\frac{\lambda}{2}, \qquad (5.25)$$

where $t' = NW_{32}\,t$ is a dimensionless time.

The solution of (5.25) has the form

$$n_1(t') = \frac{1}{2(\lambda-1)}\left\{\frac{3\lambda-1}{2}+\varepsilon-\sqrt{\Delta}\,\mathrm{th}\,\frac{t'\sqrt{\Delta}}{2}\right\}. \qquad (5.26)$$

For large times ($t' \to \infty$) the solution (5.26) goes over into the stationary solution (5.14). The characteristic time to reach stationary conditions is determined by $\Delta^{-1/2}$. This time, as follows from (5.15), depends on the parameters ε and λ, i.e., on the ratio of the relaxation constants in the compound system.

It can be seen from (5.25), (5.26) that the nature of the solution changes in going over to linear kinetics ($\lambda = 1$).

§6. DISCUSSION

We examine the principal results of the above investigation. First of all we note that the kinetics of light absorption developed for a compound system (§5) together with the "dyamic" system (2.10) represent a closed formulation of a nonlinear theory of broadening for a resonant interaction. Its principal difference from the case of broadening by external particles[7] consists of the fact that the dynamic behavior of the system is described by a three-level scheme, while the kinetic equation for the absorption of light becomes nonlinear. These results refer to the simplest two-level model of an atom. Their generalization to a multilevel case taking the degeneracy of levels into account does not contain, as is clear from § 4, any difficulties of principle. However, taking degeneracy into account considerably complicates specific quantitative calculations.

We touch still another aspect of the problem, viz., the scattering of identical atoms in a resonance EM field. This aspect was practically not discussed above, although corresponding results can be obtained on the basis of system (2.10). Indeed, in (2.10) there are essentially contained potentials for the interaction of atoms with each other in the presence of an EM field. These potentials can be directly substituted, for example, into the quasiclassical scattering formula and one can obtain the corresponding cross sections dependent on the intensity of the EM field.

The problem discussed above is of direct experimental interest, since we are dealing with a very simple system—a medium consisting of unexcited identical atoms. For the lasers at present in existence apparently the most convenient media are the vapors of alkali elements (Na, Rb, Cs) for which the transition from the ground to the first excited state lies within the range of laser frequencies. Here two types of experiments are of the greatest interest. Firstly, it is possible to observe nonlinear effects in the absorption of light, particularly in the wing of the line (the effect of "the medium becoming transparent," § 4), directly when the gaseous medium is irradiated by a laser. Secondly, one can measure the scattering of atomic beams in the field of laser radiation. The required values of the intensity of the laser field can be easily estimated by means of formula (4.19). At the present time there exist experimental data of the first type obtained by A. M. Bonch-Bruevich and collaborators and reported in the review[17]. In these experiments inelastic scattering of laser light was observed in rubidium and cesium vapors. The observations were made in the far wings of resonance lines where the difference from the resonance nature of the interaction between atoms is already significant. At intensities of the laser field $E_0 \sim 5\times10^5$ V/cm a deviation from the linear dependence $Q \propto E_0^2$ in the direction of diminished absorption becomes noticeable. Although a direct comparison of these results with theory is premature, they indicate directly the existence of the effect itself—absorption of light in the process of collision of atoms.

We note that the effects of interest to us of absorption accompanying collisions manifest themselves usually in the far wings of spectral lines $\Delta\omega \gtrsim v/\rho_{eff} \sim v^{3/2}/d \sim \tau^{-1}$ (cf., (1.1)) where the Doppler broadening is no longer significant (for example, the experiments mentioned above correspond to moving out into the wing of the line by an amount of $\Delta\lambda \sim 1000$ cm^{-1}). This statement can be easily checked by comparing the magnitude of the Doppler broadening $\Delta\omega_D \sim \omega_0 v/c$ with the value of the inverse collision time $\tau^{-1} \sim v^{3/2}/d$; under gas-kinetic conditions for $v \sim 10^4 - 10^5$ cm·sec^{-1} and $\omega_0 \lesssim 10^{15}$ sec^{-1} (optical frequencies) the inequality $\Delta\omega_D\,\tau \ll 1$ is satisfied, which enables us not to take Doppler broadening into account. The simultaneous taking into account of broadening collisions and of the Doppler effect is considered in[18,19].

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[1)] We set $e = \hbar = m = 1$.

[2)] The averaging being carried out over a period of oscillation of an atom in an EM field does not allow us in the general case to investigate the detailed time behavior of the popula-

tions of the atomic levels, which are of a quasiperiodic nature. Therefore these results are utilized below for obtaining the stationary absorbed power for which the utilization of the averaged values leads, as was shown in[7], to the well-known result of Karplus–Schwinger.

[3)]To avoid misunderstandings we note that the amplitudes c_i^m introduced here differ from the amplitudes (2.5). In order to establish the correspondence between the systems of equations (2.6) and (4.5), (4.6) it is sufficient to neglect the difference in the states with different components of angular momentum. Then, evidently, c_+^m, c_-^m, c_{4+}^{mk} ($c_{4-}^{mk} \equiv 0$) from (4.4) correspond to the amplitudes c_2, c_4 and c_3 from (2.5).

[4)]Here and later we do not carry out a consistent averaging over the velocity, assuming it, as is usually done in the theory of broadening, to be equal to a certain characteristic velocity of the Maxwellian distribution.

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