

Field narrowing of spectral lines

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It is shown that an electromagnetic field of sufficient intensity affects the collision integrals. A kinetic equation derived in the impact approximation contains a collision integral which can be expressed in terms of the generalized T matrix in which the external field is taken into account. A monochromatic field and elastic scattering are considered in the non-degenerate state model. Spectroscopic effects in two and three-level systems are analyzed. One of the external effects is a reduction of impact broadening of spectral lines.

1. INTRODUCTION

In an earlier paper^[1] we called attention to the limitations on the theory of broadening of spectral lines by low magnetic-field intensities. If the external field does not succeed in changing the state of the optical electron during the collision time τ_c , then the atom scattering takes place practically as in the absence of a field. On the other hand, if the following conditions are satisfied:

$$|V|\tau_c \gg 1, \quad |V| \gg |\delta|, \quad \delta = \omega - \omega_{mn}, \quad (1.1)$$

$$V = d_{mn}E / 2\hbar,$$

(d_{mn} is the dipole-moment matrix element, and E and ω are the amplitude and frequency of the field), then after a time τ_c the field mixes the states m and n of the atom, and the scattering can be significantly altered during the collision. In this case, consequently, the collision integrals, the relaxation constants, and other characteristics depend on the field.

One of the phenomena connected with this fact is the narrowing of the spectral lines. We recall that in the case of elastic scattering, the collision broadening is due to the difference between the interaction potentials in the two combining states m and n . When the condition (1.1) is satisfied, the atom executes several transitions $m \leftrightarrow n$ during the time τ_c , and is scattered in a certain average potential. The indicated cause of the line broadening therefore disappears, i.e., the impact line width should become leveled-out by the strong field (1.1).

For $\lambda \sim 1$, $f \sim 0.1$, and $\tau_c \sim 10^{-12}$ sec, the condition $|V|\tau_c = 1$ is satisfied at intensities $\sim 10^7$ W/cm², which at present is by far not exotic.

The present article is devoted to an investigation of the described effect. In Sec. 2 we obtain a general expression for the collision integral (in the impact approximation). This expression is made more concrete in Sec. 3 for the case of a plane monochromatic wave and elastic scattering. Sections 4 and 5 contain an analysis of the spectroscopic phenomena.

2. THE COLLISION INTEGRAL

We consider below a two-component gas (particles a and b) at low pressure and calculate the collision integral for particles a as a result of interaction with particles b . We start from a chain of Bogolyubov equations terminated at the two-particle function F_{ab} , and with the Bogolyubov initial conditions

$$i\hbar \frac{\partial}{\partial t} F_a(t) = [H_a + V(t), F_a(t)] + i\hbar S, \quad S = \frac{N_b}{i\hbar} \text{Sp}_b[W, F_{ab}],$$

$$i\hbar \frac{\partial}{\partial t} F_{ab}(t) = [H_a + H_b + V(t) + W, F_{ab}(t)],$$

$$\lim_{t \rightarrow -\infty} F_{ab}(t) = F_a(t) \times F_b(t). \quad (2.1)$$

Here F_a and F_b are single-particle functions. H_a and H_b are the Hamiltonians of the isolated particles. $V(t)$ and W are the Hamiltonians of the interaction of the particles a with the field and with particles b , and N_b is the concentration of particles b .

Relations (2.1) are the net result of solution of the statistical part of the problem of calculating the collision integral S , and any further detailed description of S presupposes the use of results of the solution of the dynamic problem of the collision of the two particles a and b . We can attempt to express S directly in terms of the potential W ^[1-5]. This, however, will not yield a universal formula for S , inasmuch as the solution of the scattering problem cannot be presented in closed form at arbitrary W . The problem has therefore frequently been formulated of late in a new manner, namely, express S in terms of standard collision-theory operators (scattering amplitudes, cross sections, etc.). This approach was used in many papers¹⁾, but these assumed besides the premises of impact theory, also the existence of temporal and spatial homogeneity, which do not hold true in six strong external fields. We therefore obtain below for S an expression free of the indicated limitations.

We introduce Green's functions in accordance with the equations

$$i\hbar \frac{\partial}{\partial t} G(t, t') = [H_a + H_b + V(t)]G(t, t') + i\hbar \delta(t - t'), \quad (2.2)$$

$$i\hbar \frac{\partial}{\partial t} \mathcal{G}(t, t') = [H_a + H_b + V(t) + W]\mathcal{G}(t, t') + i\hbar \delta(t - t').$$

The solution of the initial problem for $F_{ab}(t)$ we express in terms of $\mathcal{G}(t, t')$:

$$F_{ab}(t) = \mathcal{G}(t, t') F_{ab}(t') \mathcal{G}^+(t, t'). \quad (2.3)$$

Between $F_{ab}(t')$ and $\mathcal{G}, \mathcal{G}^*$ we introduce unit matrices in the form

$$1 = G(t, t') G^+(t, t') = G^+(t, t') G(t, t')$$

and write

$$F_{ab}(t) = \Omega(t, t') G(t, t') F_{ab}(t') G^+(t, t') \Omega^+(t, t'),$$

$$\Omega(t, t') = \mathcal{G}(t, t') G^+(t, t'), \quad (2.4)$$

$$i\hbar \frac{\partial}{\partial t} \Omega(t, t') = [H_a + H_b + V(t), \Omega(t, t')] + W \Omega(t, t') + i\hbar \delta(t - t').$$

In this form, the use of the initial conditions (2.1) is elementary:

$$F_{ab}(t) = \Omega(t) (F_a(t) \times F_b(t)) \Omega^+(t), \quad \Omega(t) = \lim_{t' \rightarrow -\infty} \Omega(t, t'). \quad (2.5)$$

The operator $\Omega(t)$ is a generalization of the Moller operator $\Omega^{(\pm)}$ to the case of a time-dependent Hamiltonian.

When (2.5) is taken into account, the kinetic equation takes the form

$$\frac{\partial}{\partial t} F_a(t) = -\frac{1}{2} [\Gamma F_a(t) + F_a(t) \Gamma] + \frac{1}{i\hbar} [H_a + V(t), F_a(t)] + S, \quad (2.6)$$

$$S = \frac{N_b}{i\hbar} S_{p_b} \{ \mathcal{T}(t) (F_a \times F_b) \Omega^+(t) - \Omega(t) (F_a \times F_b) \mathcal{T}^+(t) \}, \quad \mathcal{T}(t) = W \Omega(t).$$

We have introduced here an additional term with a diagonal matrix Γ . This term describes the spontaneous damping. $\mathcal{T}(t)$ is a generalized T-matrix. If $\Omega(t)$ is represented in the form^[10]

$$\Omega(t) = 1 + K(t), \quad (2.7)$$

then S breaks up into "arrival" terms (containing $K(t)$) and "departure" terms

$$S = \frac{N_b}{i\hbar} S_{p_b} \{ \mathcal{T}(t) (F_a \times F_b) - (F_a \times F_b) \mathcal{T}^+(t) + \mathcal{T}(t) (F_a \times F_b) K^+(t) - K(t) (F_a \times F_b) \mathcal{T}^+(t) \}. \quad (2.8)$$

We emphasize that S is expressed in terms of the Moller operator, and not the scattering matrix (S matrix). This result is closely connected with the fact that collisions are equally probable on the interval $(-\infty, t)$ that precedes the instant of observation t . It is the operator $\Omega(t)$ which describes the evolution on this interval. The scattering matrix, on the other hand, describes the net result of the evolution on the interval $(-\infty, +\infty)$, and the collision is taken to occur at finite t (scattering by a target); the S matrix has therefore no direct connection with kinetic problems.

Formulas (2.6), supplemented with Eq. (2.4) for $\Omega(t, t')$, give the most general, to our knowledge, formulation of the kinetic equation within the framework of the problem (2.1). All the known expressions for S are obtained from (2.6) as particular cases. It is also easy to generalize (2.6) to include the case of collisions of identical atoms.

3. COLLISION INTEGRAL IN ELASTIC SCATTERING AND IN A STRONG MONOCHROMATIC EXTERNAL FIELD

We consider, in the resonance approximation, elastic scattering in the presence of an external field

$$V(t) = -dE \cos(\omega t - \mathbf{k} \cdot \mathbf{r}_a). \quad (3.1)$$

We denote by m and n the states of the isolated atom a , for which $\omega_{mn} \sim \omega$ ($\omega_{mn} > 0$). The similarity transformation with the matrix $P(t)$

$$P_{ij}(t) = \delta_{ij}, \quad j \neq n, \quad P_{nn}(t) = e^{i\omega t},$$

excludes from the Hamiltonian the explicit dependence on the time, and the argument of the operator $\tilde{\Omega}(t, t') = P^{-1}(t) \Omega(t, t') P(t)$ is $t - t'$. Therefore $\tilde{\Omega}(t, t') \equiv \Omega$ does not depend on the time and satisfies the equation

$$[H_a + H_b + \nabla, \Omega] + \tilde{W} \Omega = 0, \quad \tilde{V} = P^{-1}(t) V(t) P(t), \quad (3.2)$$

or the integral equation

$$\Omega = 1 + \frac{1}{i\hbar} \int_0^\infty d\tau G(\tau) \tilde{W} \Omega G^+(\tau), \quad G(\tau) = P^{-1}(t) G(t, t - \tau) P(t - \tau), \quad (3.3)$$

the forms of which are standard for the stationary scattering theory^[10]. On the other hand, the kinetic equation is

$$i\hbar \frac{\partial}{\partial t} F_a(t) = [H_a + \nabla, F_a(t)] - \frac{i\hbar}{2} [\Gamma F_a(t) - F_a(t) \Gamma] + i\hbar S;$$

$$F_a(t) = P^{-1}(t) F_a(t) P(t),$$

$$(H_a)_{nn} = E_n + \hbar\omega + p^2 / 2m_a, \quad (H_a)_{ij} = \delta_{ij} E_j + p^2 / 2m_a,$$

$$S = \frac{N_b}{i\hbar} S_{p_b} \{ \mathcal{T}(F_a(t) \times F_b(t)) \Omega^+ - \Omega(F_a(t) \times F_b(t)) \mathcal{T}^+ \}, \quad \mathcal{T} = W \Omega. \quad (3.4)$$

We assume first the simplest model with nondegenerate states n and m and with structureless perturbing particles. In the equation for Ω , we put $k = 0$ ($2\pi/k$ is much larger than the interaction radius (ρ_{int})), after which the variables of the mass center and of the relative motion of the particles a and b separate:

$$\Omega_{ij}(p_a, p_a', p_b, p_b') = \delta(p - p') \Omega_{ij}(p, p'),$$

$$\Omega_{ij}(p, p') = \delta(p - p') \delta_{ij} + K_{ij}(p, p');$$

$$P = p_a + p_b, \quad p = \mu_b p_a - \mu_a p_b, \quad P' = p_a' + p_b', \quad p' = \mu_b p_a' - \mu_a p_b', \quad (3.5)$$

$$\mu_a = \frac{m_a}{m_a + m_b} = \frac{\mu}{m_b}, \quad \mu_b = \frac{m_b}{m_a + m_b} = \frac{\mu}{m_a}, \quad \mu = \frac{m_a m_b}{m_a + m_b}.$$

The introduction of $\delta(p - p')$ is connected with the assumption $W(\mathbf{r}_a - \mathbf{r}_b)$. The system of equations for $K_{ij}(p, p')$ is

$$\hbar \lambda K_{li}(p, p') - \int W_{li}(p - p_1) K_{il}(p_1, p') dp_1 = W_{li}(p - p'), \quad l \neq m, n; \quad (3.6)$$

$$\begin{pmatrix} \lambda - W_{mn}/\hbar & V & -V^* & 0 \\ V^* & \lambda - \delta - W_{nn}/\hbar & 0 & -V^* \\ -V & 0 & \lambda + \delta - W_{mm}/\hbar & V \\ 0 & -V & V^* & \lambda - W_{nn}/\hbar \end{pmatrix} \begin{pmatrix} K_{mm} \\ K_{nm} \\ K_{mn} \\ K_{nn} \end{pmatrix}$$

$$= \begin{pmatrix} W_{mm} \\ 0 \\ 0 \\ W_{nn} \end{pmatrix}, \quad (3.7)$$

$$\lambda = (p'^2 - p^2) / 2\mu\hbar, \quad V = d_{mn} E / 2\hbar,$$

where W_{ij} in the left-hand side of (3.7) should be taken to mean the integral operator (as in (3.6)).

The interaction with the external field connects the equations for K_{ij} ($i, j = m, n$) and in this sense plays a role analogous to inelastic processes, the only essential distinguishing feature being that the "inelastic part" of the potential is constant in the elastic-interaction radius ρ_{int} . On the other hand, the field inelasticity has a specific character, and connects together four equations for K_{ij} ; the truly inelastic processes (W_{mn}) lead only to a pairwise connection of K_{mm} with K_{nn} and of K_{nn} with K_{mm} . This difference is due, in final analysis, to the fact that V is dynamic in origin and W static (see Eq. (3.2)).

The fact that the matrix of the system (3.7) is not diagonal means, in "time language," that the external field induces transitions of the atom from the state m into the state n and back, and the atom "feels" alternately first W_{mm} and then W_{nn} . This interpretation coincides with the explanation of the saturation effect, the analogy with which is obvious: the structure of the field terms in the equations for F_a and K is the same; the K_{jj} are analogous to the populations $(F_a)_{jj}$, and K_{ij} are analogous to the off-diagonal elements $(F_a)_{ij}$; finally, W_{jj} in the right-hand side of (3.7) corresponds to the rates of excitation of the levels j .

The indicated analogy breaks down at the following point: in the theory of effective saturation one usually considers a system of four first-order equations; the equations for K , on the other hand, are a system of four second-order equations (in the coordinate representation). This difference is eliminated in the eikonal approximation. In this case, in analogy with the saturation effect, we can prove the following: if

$$|V| \gg \left| \omega - \omega_{mn} - \frac{W_{mm} - W_{nn}}{\hbar} \right|, \quad |V| \tau_c \gg 1, \quad \tau_c = \frac{\mu}{p'} \rho_b, \quad (3.8)$$

then $K_{mn} = K_{nm} = 0$, $K_{mm} = K_{nn}$ and satisfies the equation

$$K_{mm}(\mathbf{p}, \mathbf{p}') = -2\pi i \delta_+ \left(\frac{p'^2 - p^2}{2\mu} \right) \overline{\mathcal{F}}(\mathbf{p}, \mathbf{p}'), \quad (3.9)$$

$$\overline{\mathcal{F}}(\mathbf{p}, \mathbf{p}') = \int \overline{W}(\mathbf{p} - \mathbf{p}_1) [\delta(\mathbf{p}_1 - \mathbf{p}') + K_{mm}(\mathbf{p}_1, \mathbf{p}')] d\mathbf{p}_1, \quad \overline{W} = \frac{W_{nn} + W_{mm}}{2}$$

The condition (3.8) coincides with (1.1) if one includes in δ the frequency shift due to the interaction.

Thus, in a strong external field (3.8), the equation for K_{mn} has the same form as at $V = 0$, but the scattering potential is now \overline{W} . From the point of view of the analogy with the saturation effect, this means "equalization of the populations" within the time τ_c of the action of the "pulsed excitation". If at least one of the conditions (3.8) (or both) is reversed, then the external field has no effect:

$$K_{jj}(\mathbf{p}, \mathbf{p}') = -2\pi i \delta_+ \left(\frac{p'^2 - p^2}{2\mu} \right) T_{jj}(\mathbf{p}, \mathbf{p}'), \quad j = m, n; \quad (3.10)$$

$$T_{jj}(\mathbf{p}, \mathbf{p}') = \int W_{jj}(\mathbf{p} - \mathbf{p}_1) [\delta(\mathbf{p}_1 - \mathbf{p}') + K_{jj}(\mathbf{p}_1, \mathbf{p}')] d\mathbf{p}_1.$$

We confine ourselves henceforth to a comparison of S_{ij} in strong and weak fields, and the intermediate case

$$|V| \sim \frac{1}{\tau_c} + \left| \delta - \frac{W_{mm} - W_{nn}}{\hbar} \right|$$

will be disregarded.

If the states m and n are degenerate (this factor was not taken into account above), then the situation becomes more complicated: some of the magnetic sublevels may not interact directly with the external field, and the latter will be effective only as a result of collision mixing. Therefore the collision integrals will depend not only on V , but also on the polarization of the field, on the type of the scattering potential, on the ratio of the angular momenta of the combining levels, etc.

Let us spell out the expressions for S_{ij} more concretely for the case of a spatially homogeneous distribution of F_b :

$$F_b(\mathbf{p}_b, \mathbf{p}_b') = \delta(\mathbf{p}_b - \mathbf{p}_b') F_b(\mathbf{p}_b), \quad \int F_b(\mathbf{p}_b) d\mathbf{p}_b = (2\pi\hbar)^3. \quad (3.11)$$

With the aid of (3.5) and (3.11) we obtain from (3.4)

$$S_{ij}(\mathbf{p}_a, \mathbf{p}_a') = -\nu_{ij}(\mathbf{p}_a) F_{aij}(\mathbf{p}_a, \mathbf{p}_a') + \int A_{ij}(\mathbf{p}_a, \mathbf{p}_{a1}) F_{aij}(\mathbf{p}_{a1}, \mathbf{p}_{a1} + \mathbf{p}_a' - \mathbf{p}_a) d\mathbf{p}_{a1}, \quad (3.12)$$

where we have left out in (3.12) all the terms except those containing $F_{aij}(\mathbf{p}_a, \mathbf{p}_a')$. The general structure of S_{ij} coincides with that postulated earlier^[11]. Depending on the field amplitude, the expressions for the departure frequencies $\nu_{ij}(\mathbf{p}_a)$ and the kernels $A_{ij}(\mathbf{p}_a, \mathbf{p}_{a1})$ will contain different functions: in the case of a strong field they contain the scattering amplitudes \mathcal{F} and $\overline{\mathcal{F}}$, which are obtained by solving (3.9):

$$\nu_{ij}(\mathbf{p}_a) = \frac{iN_b}{\hbar\mu^3} \int [\mathcal{F}_{ii}(\mathbf{p}, \mathbf{p}) - \mathcal{F}_{jj}^+(\mathbf{p}, \mathbf{p})] F_b \left(\frac{\mu\mathbf{p}_1 - \mathbf{p}}{\mu_a} \right) d\mathbf{p};$$

$$A_{ij}(\mathbf{p}_a, \mathbf{p}_{a1}) = \frac{2\pi N_b}{\hbar\mu^3} \int d\mathbf{p} d\mathbf{p}_1 F_b \left(\frac{\mu_b\mathbf{p}_{a1} - \mathbf{p}_1}{\mu_a} \right) \delta(\mathbf{p}_a - \mathbf{p}_{a1} + \mathbf{p}_1 - \mathbf{p}) \quad (3.13)$$

$$\times \left\{ \mathcal{F}_{ii}(\mathbf{p}, \mathbf{p}_1) \overline{\mathcal{F}}_{jj}^+(\mathbf{p}_1, \mathbf{p}_1) \delta_- \left(\frac{p_1^2 - p^2}{2\mu} \right) + \overline{\mathcal{F}}_{ii}(\mathbf{p}, \mathbf{p}_1) \mathcal{F}_{jj}^+(\mathbf{p}_1, \mathbf{p}_1) \delta_+ \left(\frac{p_1^2 - p^2}{2\mu} \right) \right\}$$

In the absence of a field, ν_{ij} and A_{ij} are obtained from (3.13) by replacing \mathcal{F}_{jj} and $\overline{\mathcal{F}}_{jj}$ by T_{jj} , after which the expression in the curly brackets in (3.13) becomes equal to⁽²⁾

$$T_{ii}(\mathbf{p}, \mathbf{p}_1) T_{jj}^+(\mathbf{p}, \mathbf{p}_1) \delta \left(\frac{p_1^2 - p^2}{2\mu} \right). \quad (3.14)$$

In the collision integrals S_{ij} ($l \neq m, n, j = m, n$), the frequency and the kernel take the form

$$\nu_{ij}(\mathbf{p}_a) = \frac{iN_b}{\hbar\mu_a^3} \int [T_{ii}(\mathbf{p}, \mathbf{p}) - \mathcal{F}_{jj}^+(\mathbf{p}, \mathbf{p})] F_b \left(\frac{\mu_b\mathbf{p}_a - \mathbf{p}}{\mu_a} \right) d\mathbf{p},$$

$$A_{ij}(\mathbf{p}_a, \mathbf{p}_{a1}) = \frac{2\pi N_b}{\hbar\mu_a^3} \int d\mathbf{p} d\mathbf{p}_1 F_b \left(\frac{\mu_b\mathbf{p}_{a1} - \mathbf{p}_1}{\mu_a} \right) \delta(\mathbf{p}_a - \mathbf{p}_{a1} + \mathbf{p}_1 - \mathbf{p}) \quad (3.15)$$

$$\times \left\{ T_{ii}(\mathbf{p}, \mathbf{p}_1) \overline{\mathcal{F}}_{jj}^+(\mathbf{p}_1, \mathbf{p}_1) \delta_- \left(\frac{p_1^2 - p^2}{2\mu} \right) + T_{ii}(\mathbf{p}, \mathbf{p}_1) \mathcal{F}_{jj}^+(\mathbf{p}_1, \mathbf{p}_1) \delta_+ \left(\frac{p_1^2 - p^2}{2\mu} \right) \right\}$$

In a number of problems it is convenient to use the Wigner representation^[4]

$$\rho_{ij}(\mathbf{q}, \mathbf{r}) = \int F_{ij} \left(\mathbf{q} + \frac{\mathbf{r}}{2}, \mathbf{q} - \frac{\mathbf{r}}{2} \right) \exp \left[-\frac{i}{\hbar} \mathbf{r} \cdot \mathbf{r} \right] d\mathbf{r}, \quad (3.16)$$

which we shall use below. Here $S_{ij}(\mathbf{q}, \mathbf{r})$ are given by

$$S_{ij}(\mathbf{q}, \mathbf{r}) = -\nu_{ij}(\mathbf{q}) \rho_{ij}(\mathbf{q}, \mathbf{r}) + \int A_{ij}(\mathbf{q}, \mathbf{q}_1) \rho_{ij}(\mathbf{q}_1, \mathbf{r}) d\mathbf{q}_1, \quad (3.17)$$

with $\nu_{ij}(\mathbf{q})$ and $A_{ij}(\mathbf{q}, \mathbf{q}_1)$ obtained from (3.13–3.15) after making the substitution $\mathbf{p}_a, \mathbf{p}_{a1} \rightarrow \mathbf{q}, \mathbf{q}_1$.

Let us ascertain now the changes that the external fields introduce in the physical properties S_{ij} . We shall operate mainly with the departure frequencies $\nu_{ij}(\mathbf{p}_a)$ and the arrival frequencies

$$\bar{\nu}_{ij}(\mathbf{p}_a) = \int A_{ij}(\mathbf{p}_a, \mathbf{p}_{a1}) d\mathbf{p}_{a1}. \quad (3.18)$$

It is easily deduced from (3.13) the universal relation

$$\nu_{ij}(\mathbf{p}_a) + \bar{\nu}_{ji}(\mathbf{p}_a) = \nu_{ij}(\mathbf{p}_a) + \nu_{ji}(\mathbf{p}_a) = 2\text{Re } \nu_{ij}(\mathbf{p}_a), \quad (3.19)$$

which was proved earlier^[11] in the Born approximation. Thus, the fulfillment of (3.19) does not depend on the accuracy with which the \mathcal{F}_{jj} are calculated, nor on the amplitude of the external field. We note that, generally speaking, $\nu_{ij} \neq \bar{\nu}_{ij}$. As is well known, the equalities $\nu_{jj} = \bar{\nu}_{jj}$ and $\text{Re } \nu_{ij} = \text{Re } \bar{\nu}_{ij}$ mean physically, respectively, the conservation of the number of particles (at the level j) and the conservation of the "phase memory" (or the absence of broadening due to the interaction)^[11, 12].

If $W_{mm} = W_{nn}$, then we obtain from (3.7), regardless of the value of $|V|$:

$$K_{mm} = K_{nn}, \quad K_{mn} = K_{nm} = 0, \quad \mathcal{F}_{jj} = \overline{\mathcal{F}}_{jj} = T_{jj}, \quad A_{jj} = A_{nn}, \quad (3.20)$$

i.e., at identical potentials in the states m and n , the external field does not change the collision integrals; both the numbers of the atoms and the phase memory are conserved ($\nu_{jj} = \bar{\nu}_{jj} = \nu_{mn} = \bar{\nu}_{mn}$).

Let now $W_{mm} \neq W_{nn}$. If $|V| = 0$, then $\nu_{jj} = \bar{\nu}_{jj}$ by virtue of the diagonality of the scattering potential, which is equivalent to the optical theorem

$$T_{jj}(\mathbf{p}, \mathbf{p}) - T_{jj}^+(\mathbf{p}, \mathbf{p}) = -2\pi i \int T_{jj}(\mathbf{p}, \mathbf{p}_1) T_{jj}^+(\mathbf{p}, \mathbf{p}_1) \delta \left(\frac{p_1^2 - p^2}{2\mu} \right) d\mathbf{p}_1. \quad (3.21)$$

In the case of a strong field, a relation analogous to (3.21) is satisfied for $\overline{\mathcal{F}}$, but not for \mathcal{F}_{jj} ($\overline{\mathcal{F}}$ corresponds to elastic scattering in the potential \overline{W}). Therefore in the general case $\nu_{jj} \neq \bar{\nu}_{jj}$. It can be stated, therefore, that the transitions $m \leftrightarrow n$, which are induced by the external field, lead to "inelasticity" of the collision integrals in spite of the diagonal potential. The "field inelasticity" has opposite signs for S_{mm} and S_{nn} , namely,

$$\nu_{mm} - \bar{\nu}_{mm} = -(\nu_{nn} - \bar{\nu}_{nn}), \quad (3.22)$$

meaning conservation of the number of particles at both levels m and n .

It is frequently useful to use a model in which one of the W_{jj} vanishes. Assume, for concreteness, that $W_{nn} = 0$. Then at $|V| = 0$ we have

$$S_{nn} = 0, \quad A_{mn} = 0, \quad \nu_{mn} = \bar{\nu}_{mn} = 2\text{Re } \nu_{mn}, \quad (3.23)$$

i.e., there is no arrival term in S_{mn} , and the impact half-width of the line $\text{Re } \nu_{mn}$ is equal to half the frequency of the elastic scattering in the state m [13]. In a strong field we have $\bar{\nu} = \bar{\nu}_{mm}/2$, and the quantity ν_{mm} decreases (scattering in a potential $W_{mm}/2$, but all the relations of (3.23), with the exception of $A_{mn} = 0$, remain in force; there appears in S_{mn} an arrival term with complete phase memory and with a kernel³⁾

$$A_{mn} = \frac{2\pi N_b}{\hbar\mu_a^3} \int F_b \delta \delta \bar{\mathcal{T}} \mathcal{T}^+ dp dp_1, \quad 2\text{Re } A_{mn} = A_{mm}. \quad (3.24)$$

Thus, the field mixing of the states m and n during the collision time gives rise to $A_{mn} \neq 0$ and, furthermore, annihilates completely the impact broadening of the line in the same $m - n$ transition. A similar effect exists also for the neighboring transition $n - l$, in which S_{nl} also acquires an arrival term ($A_{nl} = 0$ at $V = 0$):

$$A_{nl} = \frac{2\pi N_b}{\hbar\mu_a^3} \int F_b \delta \delta \bar{\mathcal{T}} \mathcal{T}^+ dp dp_1, \quad (3.25)$$

where $\text{Re } \bar{\nu}_{nl}$ can be either larger or smaller than $\text{Re } \nu_{nl}$.

It may seem strange that $S_{nn} = 0$ at $W_{nn} = 0$ even in the case of a strong field. Indeed, an atom colliding at the level n with momentum \mathbf{p}_{a1} may remain after the collision at the same level, but obtain $\mathbf{p}_a \neq \mathbf{p}_{a1}$, owing to the interaction W_{mm} during the time τ_c , i.e., $S_{nn} \neq 0$ seems natural. To resolve this question, we express S_{nn} in the form

$$i\hbar S_{nn} = N_b \text{Sp}_b \{ W_{nn} [\Omega(F_a \times F_b) \Omega^+]_{nn} - [\Omega(F_a \times F_b) \Omega^+]_{nn} W_{nn} \}. \quad (3.26)$$

The expression in the curly brackets has the same meaning as the combination $V_{nm} F_{mn} - F_{nm} V_{mn}$, which describes the field variation of the number of atoms at the level n and is interpreted in the following manner: in the transition $m - n$ the external field induces a polarization F_{mn} and performs work proportional to $[V, F]_{nn}$. The analogy with (3.26) is obvious. The scattering potential produces a "polarization" $[\Omega(F_a \times F_b) \Omega^+]_{nn}$, and the change of the number of atoms in the state n , \mathbf{p}_a is determined by the work $\text{Tr}_b [W_{nn}, [\Omega(F_a \times F_b) \Omega^+]_{nn}]$ of the interaction forces precisely in this state. But if $W_{nn} = 0$, then the work is equal to zero, although the polarization is finite. The foregoing reasoning, however, pertained precisely to polarization, but not to work.

A formally instructive case is $W_{mm} = -W_{nn}$. Here

$$K_{ij} = 0, \quad \bar{\mathcal{T}} = 0, \quad \mathcal{T}_{ij} = W_{ij}, \quad S_{ij} = 0, \quad A_{mn} = 0, \quad (3.27)$$

$$\nu_{mn} = -\nu_{nm} = 2iN_b(2\pi\hbar)^3 W_{mm}(0) / \hbar\mu_a^3.$$

The only nonvanishing terms are ν_{mn} and ν_{nm} , which are pure imaginary and are proportional (as in first-order perturbation theory in W) to the difference of the average interaction energies $W_{mm} - W_{nn} = 2W_{mm}$. Consequently, when $W_{mm} = -W_{nn}$, there occurs neither a change in the velocity of the atoms, nor a broadening of the lines, but only an impact shift (by ν_{mn}). This is perfectly understandable physically: the atoms "feels" alternately potentials that are equal in magnitude but are opposite in sign, and does not change its momentum. For the line shift, however, the difference $W_{mm} - W_{nn}$ is important, and it is finite. On the transitions $m - l$ and $n - l$, the collision integrals differ from zero when

$W_{mm} = -W_{nn}$, but the contribution of the levels m and n is determined only by W_{jj} :

$$\nu_{ij}(\mathbf{p}_a) = \frac{iN_b}{\hbar\mu_a^3} \int [W_{ij}(0) - T_{ij}^+(\mathbf{p}, \mathbf{p})] F_b \left(\frac{\mu_b \mathbf{p}_a - \mathbf{p}}{\mu_a} \right) dp,$$

$$A_{ij}(\mathbf{p}_a, \mathbf{p}_{a1}) = \frac{2\pi N_b}{\hbar\mu_a^3} \int W_{ij}(\mathbf{p} - \mathbf{p}_1) T_{ij}^+(\mathbf{p}, \mathbf{p}_1) \times \delta_+ \left(\frac{p_1^2 - p^2}{2\mu} \right) \delta(\mathbf{p}_a - \mathbf{p}_{a1} + \mathbf{p}_1 - \mathbf{p}) F_b dp dp_1. \quad (3.28)$$

Of course, the equality $W_{mm} = -W_{nn}$ cannot be satisfied at all $\mathbf{r}_a - \mathbf{r}_b$ (if for no other reason than that $W > 0$ for all states at small distances), but at large distances W_{mm} and W_{nn} can have opposite signs⁴⁾, and the external field will lead to a decrease of S_{ij} and of the line width, but to an increase of the relative line shift.

It is clear from the foregoing that a strong field alters radically a number of main properties of collision integrals. Each of these changes has a lucid interpretation in analogy with the saturation effect, but we shall not dwell on this obvious fact.

At high field intensities ($|V| \gg \bar{k}p_a/m_a$, and all the more $|V|\tau_c \gg 1$), the natural variables are not F_{ij} but their combinations $F_{ij} \pm F_{ij}'$. For such quantities, the collision integrals are determined by the following frequencies and kernels:

$$\nu_1 = \frac{1}{2}[\nu_{mm} + \nu_{nn}] = \frac{1}{2}[\nu_{mn} + \nu_{nm}] = \frac{iN_b}{\hbar\mu_a^3} \int [\bar{\mathcal{T}} - \bar{\mathcal{T}}^+] F_b dp,$$

$$A_1(\mathbf{p}_a, \mathbf{p}_{a1}) = \frac{1}{2}[A_{mm} + A_{nn}] = \frac{1}{2}[A_{mn} + A_{nm}] = \frac{2\pi N_b}{\hbar\mu_a^3} \int \bar{\mathcal{T}}(\mathbf{p}, \mathbf{p}_1) \bar{\mathcal{T}}^+(\mathbf{p}, \mathbf{p}_1) \times \delta \left(\frac{p_1^2 - p^2}{2\mu} \right) \delta(\mathbf{p}_a - \mathbf{p}_{a1} + \mathbf{p}_1 - \mathbf{p}) F_b \left(\frac{\mu_b \mathbf{p}_{a1} - \mathbf{p}_1}{\mu_a} \right) dp dp_1,$$

$$\nu_2' = \frac{1}{2}[\nu_{mm} - \nu_{nn}], \quad i\nu_2'' = \frac{1}{2}[\nu_{mn} - \nu_{nm}], \quad (3.29)$$

$$\nu_2 = \nu_2' + i\nu_2'' = \frac{iN_b}{\hbar\mu_a^3} \int [\mathcal{T}_{mm}(\mathbf{p}, \mathbf{p}) - \mathcal{T}_{nn}(\mathbf{p}, \mathbf{p})] F_b \left(\frac{\mu_b \mathbf{p}_a - \mathbf{p}}{\mu_a} \right) dp,$$

$$A_2 = \frac{1}{2}[A_{mm} - A_{nn}] + \frac{1}{2}[A_{mn} - A_{nm}] = \frac{2\pi N_b}{\hbar\mu_a^3} \int [\mathcal{T}_{mn}(\mathbf{p}, \mathbf{p}_1) - \mathcal{T}_{nm}(\mathbf{p}, \mathbf{p}_1)] \times \delta \left(\frac{p_1^2 - p^2}{2\mu} \right) \delta(\mathbf{p}_a - \mathbf{p}_{a1} + \mathbf{p}_1 - \mathbf{p}) F_b \left(\frac{\mu_b \mathbf{p}_{a1} - \mathbf{p}_1}{\mu_a} \right) dp dp_1.$$

According to (3.29), the symmetrical combinations of the frequencies and of the kernels are identical for the diagonal and off-diagonal elements. With respect to the departure frequencies, this is a universal property (see (3.9)). For kernels, the inequality $A_{mm} + A_{nn} = A_{mn} + A_{nm}$ takes place only in a strong field. In addition, $\bar{\nu}_1 = \nu_1$, i.e., phase memory and particle conservation exist. We note that antisymmetrical combinations are the real and imaginary parts of one and the same quantity (ν_2, A_2).

4. POPULATION AND POLARIZATION

We consider stationary population and the polarization of an atom in a strong field (3.8). We shall use the Wigner representation (3.16) and introduce the functions

$$\rho_{1,2}(\mathbf{q}, \mathbf{r}) = \rho_{mm}(\mathbf{q}, \mathbf{r}) \pm \rho_{nn}(\mathbf{q} - \hbar\mathbf{k}, \mathbf{r}), \quad V/|V| = \exp(i\varphi),$$

$$\rho_{3,4}(\mathbf{q}, \mathbf{r}) = \exp[-i(\varphi + \mathbf{k}\mathbf{r})] \rho_{mn}(\mathbf{q} - \frac{1}{2}\hbar\mathbf{k}, \mathbf{r})$$

$$\mp \exp[i(\varphi + \mathbf{k}\mathbf{r})] \rho_{nm}(\mathbf{q} - \frac{1}{2}\hbar\mathbf{k}, \mathbf{r}). \quad (4.1)$$

We neglect the recoil effect^[15]; this corresponds to $\mathbf{k} = 0$ in the arguments of ρ_{ij} . Then the system of kinetic equations takes the form

$$\Gamma\rho_1 + \gamma\rho_2 = S_1(\rho_1) + S_2'(\rho_2) + Q_1,$$

$$\Gamma\rho_2 + \gamma\rho_1 + 2i|V|\rho_3 = S_1(\rho_2) + S_2'(\rho_1) + Q_2,$$

$$\Gamma\rho_3 - i\delta\rho_4 + 2i|V|\rho_2 = S_1(\rho_3) + iS_2''(\rho_4),$$

$$\Gamma\rho_4 - i\delta\rho_3 = S_1(\rho_4) + iS_2''(\rho_3),$$

$$\begin{aligned}
Q_1 &= Q_m + Q_n, & Q_2 &= Q_m - Q_n, & 2\Gamma &= \Gamma_m + \Gamma_n, & 2\gamma &= \Gamma_m - \Gamma_n, \\
\delta &= \omega - \omega_{mn} - \mathbf{kq}/m_a, \\
S_1(\rho) &= -\nu_1 \rho + \int A_1(\mathbf{q}, \mathbf{q}_1) \rho(\mathbf{q}_1) d\mathbf{q}_1, \\
S_2'(\rho) + iS_2''(\rho) &= -\nu_2 \rho + \int A_2(\mathbf{q}, \mathbf{q}_1) \rho(\mathbf{q}_1) d\mathbf{q}_1.
\end{aligned} \tag{4.2}$$

The rates of excitation Q_m and Q_n of the levels will be regarded as Maxwellian functions. Under the conditions $|V| \gg 1/\tau_c \gg kq/m_a$ of interest to us, the field interacts practically in the uniform manner with all the atoms. If, in addition, the perturbing gas has the same temperature, then the distribution over the momenta for gas will obviously be Maxwellian. Further, taking into account the momentum conservation law $\mathbf{q} - \mathbf{q}_1 = \mathbf{p} - \mathbf{p}_1$, the following identity holds true:

$$\begin{aligned}
q_i^2 / \bar{q}^2 + [\mu_0 q_i - p_i]^2 / \mu_a^2 \bar{p}_0^2 &= q^2 / \bar{q}^2 + [\mu_0 \mathbf{q} - \mathbf{p}]^2 / \mu_a^2 \bar{p}_0^2 + [p_i^2 - p^2] / \bar{p}^2, \\
\bar{q}^2 / m_a &= \bar{p}_0^2 / m_0 = \bar{p}^2 / \mu = 2k_b T,
\end{aligned} \tag{4.3}$$

with the aid of which we can easily find from (3.29) that for Maxwellian functions

$$\begin{aligned}
S_1(\rho) &= 0, & S_2(\rho) &= -(a + ib)\rho, & a + ib &= \nu_2 - \frac{2\pi N_b}{\hbar \mu_a^3} \int [\mathcal{F}_{mm}(\mathbf{p}, \mathbf{p}_1) \\
& & & - \mathcal{F}_{nn}(\mathbf{p}, \mathbf{p}_1)] \overline{\mathcal{F}}^+(\mathbf{p}, \mathbf{p}_1) \exp\left\{-\frac{p_i^2 - p^2}{\bar{p}^2}\right\} \\
& & & \times \delta_-\left(\frac{p_i^2 - p^2}{2\mu}\right) F_b\left(\frac{\mu_0 \mathbf{q} - \mathbf{p}}{\mu_a}\right) dp d\mathbf{p}_1.
\end{aligned} \tag{4.4}$$

Relations (4.4) simplify the system (4.2), from which it follows that

$$\begin{aligned}
\rho_1 &= \frac{Q_1}{\Gamma}, & \rho_{mm} &\cong \rho_{nn} = \frac{Q_1}{2\Gamma} = \frac{Q_m + Q_n}{\Gamma_m + \Gamma_n}, \\
\rho_2 &= \rho_{mm} - \rho_{nn} \sim \frac{\Gamma^2 + (\delta - b)^2}{4|V|^2} \frac{1}{\Gamma} \left[Q_2 - \frac{\gamma + a}{\Gamma} Q_1 \right], \\
\rho_3 &= -\frac{i}{2|V|} \left[Q_2 - \frac{\gamma + a}{\Gamma} Q_1 \right] \\
&= -\frac{i}{|V|} \frac{\Gamma_m \Gamma_n}{2\Gamma} \left[\frac{Q_m}{\Gamma_m} - \frac{Q_n}{\Gamma_n} - \frac{a}{\Gamma_m \Gamma_n} (Q_m + Q_n) \right].
\end{aligned} \tag{4.5}$$

According to (4.5), the ρ_{jj} are determined by the mean rates of excitation and decay. The difference of the populations ρ_2 and ρ_3 (which determines the imaginary part of the polarizability) depends not only on Q_j and Γ_j , but also on the parameter of the "field inelasticity" a , which plays the same role as the difference $2\gamma = \Gamma_m - \Gamma_n$ of the relaxation constants.

The equation for ρ_4 in (4.2) has a collision integral with a complete phase memory S_1 , corresponding to the general considerations concerning the leveling-off of the impact broadening in a strong field (Sec. 1). There exists, however, a specific line shift connected with the term $iS_2''(\rho_3)$. If, for example, the parameter a depends weakly on \mathbf{q} , then ρ_3 is also a Maxwellian function, $S_2''(\rho_3) = -b\rho_3$, and for the usual variable $\rho = e^{i\mathbf{k}\cdot\mathbf{r}} \rho_{mn}$ we obtain from (4.2) and (4.5) the equation

$$\Gamma \rho = S_1(\rho) + \frac{1}{2} \frac{V}{|V|} [\Gamma + i(\omega - \omega_{mn} - b - \frac{\mathbf{kq}}{m_a})] \rho_3. \tag{4.6}$$

We shall find the solution of this equation in two simple models. In the Weisskopf model (the velocity of the atoms remains unchanged) we have $S_1 = 0$ and (4.6) means that the collisions lead only to a shift of the line b , but not to broadening. In this strong collision model

$$\begin{aligned}
A_1(\mathbf{q}, \mathbf{q}_1) &= \nu_1 W_m(\mathbf{q}) = \nu_1 (\gamma \bar{q})^{-3} \exp[-q^2 / \bar{q}^2], \\
\rho(\mathbf{q}) &= \frac{V}{2|V|} \left[1 + \frac{i}{\Gamma} (\omega - \omega_{mn} - b) - \frac{i}{\Gamma + \nu_1} \mathbf{kq}/m_a \right],
\end{aligned} \tag{4.7}$$

i.e., here, too, the width is purely radiative. Were we to

have $\tilde{\nu}_1 \neq \nu_1$, then the frequency dependence would be determined by the combination

$$(\omega - \omega_{mn} - b) / (\Gamma + \nu_1 - \tilde{\nu}_1),$$

i.e., it would contain an impact width (this follows from the result of an earlier article^[16]).

5. EFFECTS OF NONLINEAR SPECTROSCOPY

As is well known, a monochromatic field splits the levels of an atom into sublevels (see, e.g.,^[17,18])

$$E_{m\pm} \pm |V| + \delta/2, \quad E_{n\pm} \pm |V| - \delta/2 \tag{5.1}$$

and the emission (or absorption) of radiation at the frequencies ω_μ different from ω can be regarded as transitions between the sublevels (5.1) (the two-level problem) or between a third level l and several of the (5.1) sublevels (three-level problem). In this section we are interested in the line contours corresponding to these transitions and in their impact broadening. We start with the three-level problem, assuming, for the sake of concreteness, $\omega_\mu \sim \omega_{ml}$ and $\omega_{ml} > 0$. The radiation at this transition takes the form of a doublet with resonances at the frequencies

$$\omega_\mu = \omega_{ml} \pm |V| + \delta/2.$$

At $|V| \gg 1/\tau_c + |\delta|$ the components of the doublet do not overlap and their spectral contours can be considered independently (moreover, they must be considered separately, since the impact approximation is valid only near the maxima, within a frequency interval much larger than $1/\tau_c$).

We shall stipulate a weak field in the form of a monochromatic (ω_μ) plane traveling (\mathbf{k}_μ) wave. Then the m, l element of the Wigner function can be naturally represented in the form

$$\rho_{ml}(\mathbf{q}, \mathbf{r}, t) = \rho_{ml}(\mathbf{q}) \exp[-i(\omega_\mu t - \mathbf{k}_\mu \mathbf{r})], \tag{5.2}$$

and $\rho_{ml}(\mathbf{q})$, as can be easily shown, satisfies the equation

$$\begin{aligned}
[1/2(\Gamma_m + \Gamma_n) - i\varepsilon_\pm] \rho_{ml}(\mathbf{q}) &= S - 1/2 V_\mu [\rho_{mm}(\mathbf{q}) - \rho_{ll}(\mathbf{q})], \\
V_\mu &= d_{ml} E_\mu / 2\hbar, \quad \varepsilon_\pm = \omega_\mu - \omega_{ml} \pm |V| - 1/2(\omega - \omega_{mn}) - (\mathbf{k}_\mu - 1/2\mathbf{k}) \mathbf{q} / m_a, \\
S &= -\nu(\mathbf{q}) \rho_{ml}(\mathbf{q}) + \int A(\mathbf{q}, \mathbf{q}_1) \rho_{ml}(\mathbf{q}_1) d\mathbf{q}_1,
\end{aligned} \tag{5.3}$$

$$\nu(\mathbf{q}) = \frac{iN_b}{\hbar \mu_a^3} \int [\overline{\mathcal{F}}(\mathbf{p}, \mathbf{p}) - T_{ll}^+(\mathbf{p}, \mathbf{p})] F_b\left(\frac{\mu_0 \mathbf{q} - \mathbf{p}}{\mu_a}\right) dp,$$

$$\begin{aligned}
A(\mathbf{q}, \mathbf{q}_1) &= \frac{2\pi N_b}{\hbar \mu_a^3} \int \overline{\mathcal{F}}(\mathbf{p}, \mathbf{p}_1) T^+(\mathbf{p}, \mathbf{p}_1) \delta\left(\frac{p_i^2 - p^2}{2\mu}\right) \delta(\mathbf{q} - \mathbf{q}_1 + \mathbf{p}_1 - \mathbf{p}) \\
&\quad \times F_b\left(\frac{\mu_0 \mathbf{q}_1 - \mathbf{p}_1}{\mu_a}\right) dp d\mathbf{p}_1.
\end{aligned}$$

The singularities of the spontaneous width $(\Gamma_m l + \Gamma_n l)/2$, of the shift of the line maximum by $\pm |V| + (\omega - \omega_{mn})/2$, of the Doppler shift $(\mathbf{k}_\mu - \mathbf{k}/2)\mathbf{q}/m_a$, and of the right-hand side in Eq. (5.3) for $\rho_{ml}(\mathbf{q})$ are ensured already at $|V| \gg k_\mu \bar{q}/m_a$, $|\delta|$ ^[16]. The stronger condition $|V| \gg 1/\tau_c + |\delta|$ determines the specifics of the collision integral S , which contains $\overline{\mathcal{F}}$ (scattering in \overline{W}). If

$$k\bar{q}/m_a + |\delta| \ll |V| \ll 1/\tau_c + |\delta|,$$

then ν and A would contain in place of $\overline{\mathcal{F}}$ the combination $(T_{mm} + T_{nn})/2$, and each of the T_{jj} corresponds to scattering in the potential W_{jj} . Thus, the impact broadening on the transition $m - l$ will change. We denote by γ_1 and γ_2 the impact widths ($\text{Re}(\nu - \tilde{\nu})$) at $|V| \gg 1/\tau_c + |\delta|$ and

$k\bar{q}/m_a + |\delta| \ll |V| \ll 1/\tau_c + |\delta|$, respectively. From (5.3) we easily get

$$\gamma_1 - \gamma_2 = \frac{\pi N_b}{\hbar \mu_a^2} \text{Re} \int [2|\bar{\mathcal{F}} - T_{ll}|^2 - |T_{mm} - T_{ll}|^2 - |T_{nn} - T_{ll}|^2] \times \delta \left(\frac{p_1^2 - p^2}{2\mu} \right) F_0 dp dp_1. \quad (5.4)$$

In the Born approximation we have $\bar{\mathcal{F}} = \bar{W}$ and $T_{jj} = W_{jj}$, and a direct check shows that $\gamma_2 > \gamma_1$, i.e., a strong field (in the sense of (3.8)) decreases the broadening (due to the interaction) on the transition $m-l$. For another neighboring transition $n-l$, the collision frequency and the kernel are given by the same formulas (5.3), and everything said concerning the $m-l$ transition automatically holds for the $n-l$ transition.

In the three-level problem, to calculate the line contour it is necessary to know the function

$$\rho_{mn}(\mathbf{q}, \mathbf{r}, t) = \rho_{mn}(\mathbf{q}) \exp(i\mathbf{k}\mathbf{r}) + r_{mn}(\mathbf{q}) \exp[-i(\omega_\mu - \omega)t + i\mathbf{k}_\mu \mathbf{r}] + \bar{r}_{mn}(\mathbf{q}) \exp[i(\omega_\mu - \omega)t - i(\mathbf{k}_\mu - 2\mathbf{k})\mathbf{r}]. \quad (5.5)$$

The quantities $\rho_{mn}(\mathbf{q})$ and $r_{mn}(\mathbf{q})$ determine the dipole moment at the frequencies ω and ω_μ , while $\bar{r}_{mn}(\mathbf{q})$ determine them at the combination frequency $2\omega - \omega_\mu$ [19]. The resonant frequencies for the emission (absorption) of a weak field are $\omega_\mu = \omega$ and $\omega_\mu = \omega \pm 2|V|$. Near these frequencies, the equations for r_{mn} take the form (if (3.8) holds)

$$\begin{aligned} (\Gamma - i\varepsilon)r_{mn} &= S_1(r_{mn}) - 1/2iV_\mu \rho_{mn}, \\ [\Gamma - i(\varepsilon \pm 2|V|)]r_{mn} &= S_1(r_{mn}) - 1/4iV_\mu \rho_{mn}, \\ \varepsilon &= \omega_\mu - \omega - (\mathbf{k}_\mu - \mathbf{k})\mathbf{q}/m_a, \end{aligned} \quad (5.6)$$

The solution (5.6) enables us, using the known prescription [17], to calculate the spectral density of the spontaneous emission on the transition $m-n$. The dielectric constant is determined with the aid of r_{mn} , which satisfies an analogous equation, but with a different right-hand part:

$$\begin{aligned} (\Gamma - i\varepsilon)r_{mn} &= S_1(r_{mn}) - \frac{i}{2}V_\mu \frac{\varepsilon}{V^*} \rho_{nm}, \\ [\Gamma - i(\varepsilon \pm 2|V|)]r_{mn} &= S_1(r_{mn}) \mp \frac{i}{2}V_\mu \frac{|V|}{V^*} \rho_{nm}. \end{aligned} \quad (5.7)$$

Equations (5.6) and (5.7) contain a collision integral S_1 without impact broadening (complete phase memory). This property is most clearly manifest in the case of "observation forward" ($\mathbf{k}_\mu \parallel \mathbf{k}$), when ε does not depend on \mathbf{q} and the mean value $\langle r_{mn} \rangle_{\mathbf{q}}$ is obtained from (5.6) and (5.7) in elementary fashion, since $\langle S_1 \rangle_{\mathbf{q}} = 0$:

$$\begin{aligned} \langle r_{mn} \rangle_{\mathbf{q}} &= -\frac{i}{2} \frac{V_\mu \langle \rho_{mn} \rangle_{\mathbf{q}}}{\Gamma - i(\omega_\mu - \omega)}, \quad \langle r_{mn} \rangle_{\mathbf{q}} = -\frac{i}{4} \frac{V_\mu \langle \rho_{mn} \rangle_{\mathbf{q}}}{\Gamma - i(\omega_\mu - \omega \pm 2|V|)}, \\ \langle r_{mn} \rangle_{\mathbf{q}} &= -\frac{i}{2} \frac{V_\mu (\omega_\mu - \omega) \langle \rho_{nm} \rangle_{\mathbf{q}}}{V^* \Gamma - i(\omega_\mu - \omega)}, \quad \langle r_{mn} \rangle_{\mathbf{q}} \\ &= \mp \frac{i}{2} \frac{|V|V_\mu}{V^*} \frac{\langle \rho_{nm} \rangle_{\mathbf{q}}}{\Gamma - i(\omega_\mu - \omega \pm 2|V|)} \end{aligned} \quad (5.8)$$

The widths of all the spectral contours (5.8) are determined only by the spontaneous decay, and the collisions do not come into play at all. If \mathbf{k}_μ is not parallel to \mathbf{k} , then ε contains \mathbf{q} , and the line contour will depend on the concrete type of S_1 . In any case, however, S_1 describes the change of velocity without a phase collapse, and the collisions will only narrow down the line contour (the Dicke effect). We can use here the results of [12]

(where the wave vector should be taken to mean the difference $\mathbf{k}_\mu - \mathbf{k}$, which depends on the angle between \mathbf{k}_μ and \mathbf{k}).

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¹From among the latest papers, we mention, e.g., [6-9], where additional references can be found.

² S_{ij} can be expressed also in terms of the scattering amplitudes f_{kk} , which are connected with the T-matrix elements in the normalization assumed here, by the relation

$$f_{kk}(\mathbf{p}, \mathbf{p}_1) = -(2\pi)^2 \mu \hbar^2 \mathcal{F}_{kk}(\mathbf{p}, \mathbf{p}_1).$$

³For the sake of brevity, we shall not indicate the arguments of the functions where there is no danger of misunderstanding.

⁴By way of example we indicate the difference between the signs of W_{ij} in the ground and metastable states of helium [14].

¹É. G. Pestov and S. G. Rautian, Zh. Eksp. Teor. Fiz. 56, 902 (1969) [Sov. Phys.-JETP 29, 488 (1969)].

²N. N. Bogolyubov, Problemy dinamicheskoy teorii v statisticheskoy fizike (Problems of Dynamical Theory in Statistical Physics), Gostekhizdat (1946).

³K. P. Turov, Osnovaniya kineticheskoy teorii (Principles of Kinetic Theory), Nauka (1966).

⁴V. P. Silin, Vvedenie v kineticheskuyu teoriyu gazov (Introduction to the Kinetic Theory of Gases), Nauka (1971).

⁵T. L. Andreeva, Zh. Eksp. Teor. Fiz. 54, 641 (1968) [Sov. Phys.-JETP 27, 342 (1968)].

⁶V. A. Alekseev, T. L. Andreeva, and I. I. Sobel'man, Zh. Eksp. Teor. Fiz. 62, 614 (1972) [Sov. Phys.-JETP 35, (1972)]; FIAN Preprint No. 124 (1971).

⁷A. Tip, Physica 53, 183; 493, 1971.

⁸E. Smith, J. Cooper, W. R. Chappell and T. Dillon, J. Quant. Spectr. Radiative Transf., 11, 1547, 1567, 1971.

⁹E. Smith, J. Cooper, W. R. Chappell and T. Dillon, J. Stat. Phys., 3, 401, 1971.

¹⁰R. G. Newton, Scattering Theory, Benjamin, 1964.

¹¹S. G. Rautian, Zh. Eksp. Teor. Fiz. 51, 1176 (1966) [Sov. Phys.-JETP 24, 788 (1967)].

¹²S. G. Rautian and I. I. Sobel'man, Usp. Fiz. Nauk 90, 209 (1966) [Sov. Phys.-Uspekhi 9, 701 (1967)].

¹³I. I. Sobel'man, Vvedenie v teoriyu atomnykh spektrov (Introduction to the Theory of Atomic Spectra), Fizmatgiz (1963).

¹⁴J. Hirschfield, C. Curtiss, and R. Bird, Molecular Theory of Gases and Liquids, Wiley, 1964.

¹⁵A. P. Kol'chenko, S. G. Rautian, and R. I. Sokolovskii, Zh. Eksp. Teor. Fiz. 55, 1864 (1968) [Sov. Phys.-JETP 28, 986 (1969)].

¹⁶A. P. Kol'chenko and S. G. Rautian, Zh. Eksp. Teor. Fiz. 54, 959 (1968) [Sov. Phys.-JETP 27, 511 (1968)].

¹⁷S. G. Rautian, Trudy FIAN 43, 3 (1968).

¹⁸T. Ya. Popova, A. K. Popov, S. G. Rautian, and A. A. Feoktistov, Zh. Eksp. Teor. Fiz. 57, 444 (1969) [Sov. Phys.-JETP 30, 243 (1970)].

¹⁹T. I. Kuznetsova, Trudy FIAN 43, 116 (1968).

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