

KINETIC EQUATION FOR THE DENSITY MATRIX

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A two-component mixture of multi-level particles in the gaseous state is examined. Generalized kinetic equations are obtained by the Bogolyubov method for single-particle density matrices with collision integrals defined both by the motion of center of gravity of the particles and by their internal states. The possibility of various types of energy exchange between the colliding particles, including the transformation of kinetic energy into internal and vice versa, is taken into account. The derived equations are used to analyze excitation in gas-mixture lasers. A correct description of excitation processes in integral form (with respect to momenta) is found. The effect of inelastic collisions on the polarization of the gas is discussed. The emission-line broadening and shift due to collisions with impurity particles are examined.

1. INTRODUCTION

IN spectroscopy one encounters, as a rule, three types of kinetic problems, namely the broadening of spectral lines, the excitation of interatomic states, and the interaction of the atomic system with an external electromagnetic field. Each of these problems has its own history, its own methods and procedures for theoretical analysis, its own formalism, etc. In the study of the broadening of spectral lines, the most developed is the approach based on the theory of random functions and correlation analysis (see, for example,<sup>[1]</sup>). Incidentally, successful attempts were made recently to formulate the problem on the basis of the kinetic-equation method<sup>[2-6]</sup>.

For the analysis of the excitation of atoms, it is assumed that elementary considerations concerning the balance of the excitation and de-excitation acts are sufficient<sup>[1]</sup>. Recently, in connection with the appearance of lasers and intense electromagnetic-radiation fluxes, interest arose in the problem of the interaction of a strong field with an atomic system. The mathematical difficulties here are so great, that the relaxation model is chosen frequently only from computational considerations.

It is perfectly clear that the indicated kinetic problems can be regarded as particular solutions of a general kinetic equation. In spite of the obvious advantage of such an approach, the kinetic equation has at present no rigorous derivation whatever capable of providing a unified point of view for different spectroscopic problems. Strange as it seems, the universally accepted Bogolyubov scheme (see, for example,<sup>[7]</sup>) is used very rarely under conditions that are canonical for spectroscopy. Indeed, spectroscopic objects have stationary states with a clearly pronounced discrete spectrum, and the main interest attaches to changes of the interatomic motions (rotations and vibrations of molecules, motion of electrons in molecules and atoms) under the influence of external actions—collisions with other particles or interactions with the external electromagnetic field. Bogolyubov's scheme, on the other hand, deals with structureless particles and with mo-

tion of their inertia centers<sup>[7]</sup>. An exception is the paper by Andreeva<sup>[6]</sup>, in which, however, a very particular problem (adiabatic relaxation, stationary structureless perturbing particles) is considered.

In the present paper we derive, on the basis of Bogolyubov's methods, a kinetic equation for the single-particle density matrix. Our main purpose is to include in the analysis all three kinetic problems of spectroscopy.

2. GENERAL EXPRESSIONS

We consider a closed system consisting of particles of two kinds in the gaseous state. We denote by  $x_k$  ( $k = 1, 2, \dots, N$ ) and  $y_\alpha$  ( $\alpha = 1, 2, \dots, M$ ) the coordinates of the centers of inertia of the particles, and by  $\xi_k$  and  $\eta_\alpha$  the aggregate of their internal coordinates. The total density matrix of the system  $F$  satisfies the equation

$$\partial F / \partial t = [\mathcal{H}, F], \quad \mathcal{H} = \mathcal{H}_a + \mathcal{H}_b + \mathcal{H}_{ab}, \quad (2.1)$$

where  $\mathcal{H}$  is the total Hamiltonian of the system, consisting of the Hamiltonians of the two kinds of particles ( $\mathcal{H}_a, \mathcal{H}_b$ ) and the operator of their interaction energy:

$$\begin{aligned} \mathcal{H}_a &= \sum_{k=1}^N \mathcal{H}_a(k) + \frac{1}{2} \sum_{k \neq j} U(k, j), & \mathcal{H}_b &= \sum_{\alpha=1}^M \mathcal{H}_b(\alpha) + \frac{1}{2} \sum_{\alpha \neq \beta} G(\alpha, \beta), \\ \mathcal{H}_a(k) &= \frac{p_k^2}{2m_a} + \mathcal{H}_a(\xi_k), & \mathcal{H}_b(\alpha) &= \frac{p_\alpha^2}{2m_b} + \mathcal{H}_b(\eta_\alpha), \\ \mathcal{H}_{ab} &= \sum_{k=1}^N \sum_{\alpha=1}^M W(k, \alpha), & W(k, \alpha) &= W(x_k, y_\alpha, \xi_k, \eta_\alpha), \\ U(k, j) &= U(x_k, x_j, \xi_k, \xi_j), & G(\alpha, \beta) &= G(y_\alpha, y_\beta, \eta_\alpha, \eta_\beta). \end{aligned} \quad (2.2)$$

Here  $\mathcal{H}_a(\xi_k)$  and  $\mathcal{H}_b(\eta_\alpha)$  denote the Hamiltonians of the internal degrees of freedom of the particle;  $p_k^2/2m_a$  and  $p_\alpha^2/2m_b$  are the operators of the kinetic energy;  $W(k, \alpha)$ ,  $U(k, j)$ , and  $G(\alpha, \beta)$  are the operators of the pair-interaction energies.

The formulation of our problem differs from the initial premises of the kinetic theory of Bogolyubov and Gurov<sup>[7]</sup> in that the internal coordinates  $\xi_k$  and  $\eta_\alpha$  are explicitly introduced. This circumstance is

important (from the formal point of view) for the following reason: As is well known, the case most thoroughly investigated in kinetic theory is the spatially-homogeneous case. We shall likewise make this assumption (at a definite stage) with respect to the coordinates of the center of inertia. For the internal variables, on the other hand, this assumption or its analogs is in no way a satisfactory approximation and is not admissible. In addition, in spectroscopic problems it is necessary to take into account the dependence of the energy of the pair interaction on the internal states of the colliding particles. It is in these respects that the kinetic equation obtained in the present section differs from the results of<sup>[7,8]</sup>. Nonetheless, the subsequent calculations are performed in accordance with a scheme close to that of<sup>[7]</sup> (see Sec. 10).

We introduce single- and two-particle density matrices:

$$F_1(k) = F_1(t, \mathbf{x}_k, \xi_k; \mathbf{x}_k', \xi_k') = V \text{Sp}_{[k]} F,$$

$$F_2(k, \alpha) = F_2(t, \mathbf{x}_k, \mathbf{y}_\alpha, \xi_k, \eta_\alpha; \mathbf{x}_k', \mathbf{y}_\alpha', \xi_k', \eta_\alpha') = V^2 \text{Sp}_{[k, \alpha]} F, \quad (2.3)$$

where the indices  $[k]$  and  $[k, \alpha]$  indicate the particles over whose coordinates no averaging takes place;  $V$  is the volume of the system. We shall seek the matrices  $F_2$  in the form of a sum of correlation matrices  $g$  and the product of single particle matrices:

$$F_2(k, \alpha) = F_1(k)F_1(\alpha) + g(k, \alpha). \quad (2.4)$$

Relations of the form (2.3) and (2.4) are satisfied also by the matrices  $F_1(\alpha)$ ,  $F_2(k, j)$ , and  $F_2(\alpha, \beta)$ .

From the general expression (2.1) follows a well known chain of equations for  $F_1$  and  $F_2$ , containing also the three-particle matrices  $F_3$ . From these equations, using the representation (2.4), we obtain differential equations for the correlation matrices  $g(k, \alpha)$ ,  $g(k, j)$ , and  $g(\alpha, \beta)$ . It is easy to ascertain that allowance of terms proportional to the concentrations  $M/V$  and  $N/V$  in the equations for the correlation matrices gives rise to terms quadratic in the concentration in the equations for the single-particle density matrices. Since we deal with limiting case of rarefied gases, we shall not take into account these terms in the equations for  $g$ .

In addition, we shall assume the interaction energy to be small and omit terms containing products of the correlation matrices and of the operators  $U$ ,  $G$ , and  $W$ . Thus, the equations for the correlation matrices assume the relatively simple form:

$$\frac{\partial g(k, \alpha)}{\partial t} = [\mathcal{H}_a(k) + \mathcal{H}_b(\alpha), g(k, \alpha)] + [W(k, \alpha), F_1(k)F_1(\alpha)]. \quad (2.5)$$

In the derivation of (2.5) we omitted, for simplicity, the symmetrization operators, i.e., quantum exchange effects were disregarded.

To solve (2.5), we go over to a momentum representation (with respect to the variables of the inertia center) and an energy representation (with respect to the internal variables), i.e., we put

$$F_1(k) = \sum_{mn} \Psi_m(\xi_k) \Psi_n^*(\xi_k') \exp\{-i\omega_{mn}t\} (2\pi\hbar)^{-3} \int d\mathbf{p}_k d\mathbf{p}_k' F_{mn}(\mathbf{q}_k, \mathbf{p}_k') \times \exp\left\{\frac{i}{\hbar} \left( \mathbf{x}_k \mathbf{p}_k - \mathbf{x}_k' \mathbf{p}_k' - \frac{p_k^2 - p_k'^2}{2m_a} \right)\right\}$$

$$g(k, \alpha) = \sum_{mnm\nu} \Psi_m(\xi_k) \Psi_\mu(\eta_\alpha) \Psi_n^*(\xi_k') \Psi_\nu^*(\eta_\alpha') \exp\{-i(\omega_{mn} + \omega_{\nu\mu})t\} \times (2\pi\hbar)^{-6} \int d\mathbf{p}_k d\mathbf{p}_k' d\mathbf{p}_\alpha d\mathbf{p}_\alpha' g_{mnm\nu}(\mathbf{p}_k, \mathbf{p}_k', \mathbf{p}_\alpha, \mathbf{p}_\alpha') \times \exp\left\{\frac{i}{\hbar} \left( \mathbf{x}_k \mathbf{p}_k + \mathbf{y}_\alpha \mathbf{p}_\alpha - \mathbf{x}_k' \mathbf{p}_k' - \mathbf{y}_\alpha' \mathbf{p}_\alpha' - \frac{p_k^2 - p_k'^2}{2m_a} - \frac{p_\alpha^2 - p_\alpha'^2}{2m_b} \right)\right\} \quad (2.6)$$

and analogously for  $F(\alpha)$ ,  $g(k, j)$ , and  $g(\alpha, \beta)$ . As before, Latin indices ( $k, j, m, n, \dots$ ) number the states of the particles of the first kind, and Greek indices ( $\alpha, \beta, \mu, \nu, \dots$ ) the states of particles of the second kind. Going over from (2.5) to equations for  $g_{mnm\nu}$  etc., and solving them, we obtain

$$g_{mnm\nu}(\mathbf{p}_k, \mathbf{p}_k'; \mathbf{p}_\alpha, \mathbf{p}_\alpha', t) = g_{mnm\nu}(\mathbf{p}_k, \mathbf{p}_k'; \mathbf{p}_\alpha, \mathbf{p}_\alpha', t_0) + \int_{t_0}^t \langle m\mu | \mathbf{p}_k \mathbf{p}_\alpha | [W(k, \alpha), F_1(k)F_1(\alpha)] | n\nu \mathbf{p}_k' \mathbf{p}_\alpha' \rangle dt'. \quad (2.7)$$

Analogous expressions are obtained also for the matrix  $g(k, j)$  and  $g(\alpha, \beta)$ .

We use Bogolyubov's initial condition<sup>[8]</sup>

$$\lim_{t_0 \rightarrow -\infty} g(\mathbf{p}_k, \mathbf{p}_k'; \mathbf{p}_\alpha, \mathbf{p}_\alpha', t_0) = 0,$$

which, as is well known, leads to the appearance of irreversibility in the kinetic theory. In accordance with Bogolyubov's main idea, we seek also solutions of (2.5) having only an implicit time dependence, via the time dependence of the single-particle matrices. In addition, we assume as usual that the single-particle matrices change to a considerable degree only over times that greatly exceed the collision times. As a result we should put<sup>1)</sup>  $t' = t$  in the matrix elements  $F_{mn}$  and  $F_{\mu\nu}$ , which appear in the integrand of (2.7). We can thus obtain

$$g_{mnm\nu}(\mathbf{p}_k, \mathbf{p}_k'; \mathbf{p}_\alpha, \mathbf{p}_\alpha', t) = \frac{i}{\hbar} \sum_{\rho} \int d\mathbf{p}_1 d\mathbf{p}_2 \times \left\{ W_{m\mu\lambda}(\mathbf{p}_k, \mathbf{p}_1; \mathbf{p}_\alpha, \mathbf{p}_2; t) \delta_+^* \left( \omega_{m\lambda} + \omega_{\mu\lambda} + \frac{p_k^2 - p_1^2}{2m_a\hbar} + \frac{p_\alpha^2 - p_2^2}{2m_b\hbar} \right) \times F_{in}(\mathbf{p}_1, \mathbf{p}_k', t) F_{\lambda\nu}(\mathbf{p}_2, \mathbf{p}_\alpha', t) - F_{mi}(\mathbf{p}_k, \mathbf{p}_1, t) F_{\mu\lambda}(\mathbf{p}_\alpha, \mathbf{p}_2, t) \times W_{in\lambda\nu}(\mathbf{p}_1, \mathbf{p}_k'; \mathbf{p}_2, \mathbf{p}_\alpha'; t) \delta_+^* \left( \omega_{in} + \omega_{\lambda\nu} + \frac{p_1^2 - p_k^2}{2m_a\hbar} + \frac{p_2^2 - p_\alpha^2}{2m_b\hbar} \right) \right\}, \quad (2.8)$$

where

$$\delta_+(x) = \int_0^\infty e^{ix\tau} d\tau = \frac{1}{2} \delta(x) + \frac{i}{2\pi x}, \quad (2.9)$$

$$W_{m\mu\lambda}(\mathbf{p}_k, \mathbf{p}_1; \mathbf{p}_\alpha, \mathbf{p}_2; t) = (2\pi\hbar)^{-6} \exp\left\{i \left( \omega_{m\lambda} + \omega_{\mu\lambda} + \frac{p_k^2 - p_1^2}{2m_a\hbar} + \frac{p_\alpha^2 - p_2^2}{2m_b\hbar} \right) t\right\} \times \int d\xi_k d\eta_\alpha d\mathbf{x}_k d\mathbf{y}_\alpha \Psi_m^*(\xi_k) \Psi_\mu(\eta_\alpha) \Psi_k^*(\xi_k) \Psi_\lambda(\eta_\alpha) \times W(k, \alpha) \exp\left\{-\frac{i}{\hbar} [\mathbf{p}_k - \mathbf{p}_1] \mathbf{x}_k + (\mathbf{p}_\alpha - \mathbf{p}_2) \mathbf{y}_\alpha\right\} \quad (2.10)$$

We shall not write out the expressions for the matrix elements of  $g(k, j)$  and  $g(\alpha, \beta)$ . They are obtained from (2.8) and (2.10) with the aid of the obvious substitution of Greek indices for Latin ones and vice versa (and accordingly  $\psi \rightleftharpoons \varphi$ ).

The succeeding steps raise no fundamental difficulties, since it is possible to calculate with the aid of

<sup>1)</sup> It is precisely because of this fact that we chose in the expansion (2.6) a representation in which the factors  $\exp[-iE_m t/\hbar]$ , which are connected with the internal motions, are separated in explicit form.

(2.8) and (2.4) the collision integrals in the kinetic equations for the single-particle density matrices  $F_i$ . We present the final results for the spatially-homogeneous problem, when

$$\begin{aligned} F_{mn}(\mathbf{p}_k, \mathbf{p}_k', t) &= (2\pi\hbar)^3 \delta(\mathbf{p}_k - \mathbf{p}_k') \rho_{mn}(\mathbf{p}_k, t), \\ F_{\mu\nu}(\mathbf{p}_\alpha, \mathbf{p}_\alpha', t) &= (2\pi\hbar)^3 \delta(\mathbf{p}_\alpha - \mathbf{p}_\alpha') f_{\mu\nu}(\mathbf{p}_\alpha, t). \end{aligned} \quad (2.11)$$

We assume, in addition, that the interaction between the particles depends only on the difference of their coordinates  $(\mathbf{x} - \mathbf{y})$ . In this case the matrix elements of the interaction operators can be written in the form

$$\begin{aligned} W_{m|\mu\lambda} &= \bar{W}_{m|\mu\lambda}(\mathbf{p}_k - \mathbf{p}_k') \delta(\mathbf{p}_k + \mathbf{p}_\alpha - \mathbf{p}_k' - \mathbf{p}_\alpha') \\ &\times \exp\left\{i\left(\frac{p_k^2 - p_k'^2}{2m_a\hbar} + \frac{p_\alpha^2 - p_\alpha'^2}{2m_b\hbar}\right)t\right\}, \end{aligned} \quad (2.12)$$

and analogously for the matrix elements  $U$  and  $G$ .

Taking the foregoing considerations into account during the course of the transformations, we obtain the equations

$$\partial\rho_{mn}/\partial t = S_{mn} + \sigma_{mn}, \quad \partial f_{\mu\nu}/\partial t = S_{\mu\nu} + \sigma_{\mu\nu}. \quad (2.13)$$

Here  $S_{mn}$  and  $S_{\mu\nu}$  are the integrals of collisions with particles of their own kind, and  $\sigma_{mn}$  and  $\sigma_{\mu\nu}$  are integrals of collisions with particles of the other kind (impurity particles):

$$\begin{aligned} \sigma_{mn}(\mathbf{p}_k) &= \sum_w \int A_{mw'n}(\mathbf{p}_k, \mathbf{p}) \rho_{w'}(\mathbf{p}) d\mathbf{p} \\ &- \sum_l [B_{ml}(\mathbf{p}_k) \rho_{ln}(\mathbf{p}_k) + \rho_{ml}(\mathbf{p}_k) B_{nl}^*(\mathbf{p}_k)]; \end{aligned} \quad (2.14)$$

$$\begin{aligned} A_{mw'n}(\mathbf{p}_k, \mathbf{p}) &= \frac{M}{V} \frac{1}{\hbar^2} \sum_{\lambda\mu\nu} \int d\mathbf{p}_1 d\mathbf{p}_2 \delta(\mathbf{p}_k - \mathbf{p} + \mathbf{p}_2 - \mathbf{p}_1) \bar{W}_{m|\mu\lambda}(\mathbf{p}_k - \mathbf{p}) \\ &\times f_{\lambda\nu}(\mathbf{p}_1) \bar{W}_{l'nv\mu}(\mathbf{p} - \mathbf{p}_k) \left\{ \delta_+^* \left( \omega_{ml} + \omega_{\mu\lambda} + \frac{p_k^2 - p^2}{2m_a\hbar} + \frac{p_2^2 - p_1^2}{2m_b\hbar} \right) \right. \\ &\left. + \delta_+^* \left( \omega_{l'\nu} + \omega_{\nu\mu} + \frac{p^2 - p_k^2}{2m_a\hbar} + \frac{p_1^2 - p_2^2}{2m_b\hbar} \right) \right\}; \end{aligned} \quad (2.15)$$

$$\begin{aligned} B_{ml}(\mathbf{p}_k) &= \frac{M}{V} \frac{i}{\hbar} \sum_{\mu\nu} \bar{W}_{ml'\nu\mu}(0) \int f_{\mu\nu}(\mathbf{p}_\alpha) d\mathbf{p}_\alpha \\ &+ \frac{M}{V} \frac{1}{\hbar^2} \sum_{l'\lambda\mu\nu} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p} \delta(\mathbf{p}_k - \mathbf{p} + \mathbf{p}_2 - \mathbf{p}_1) \\ &\times \delta_+^* \left( \omega_{l'\nu} + \omega_{\lambda\nu} + \frac{p^2 - p_k^2}{2m_a\hbar} + \frac{p_1^2 - p_2^2}{2m_b\hbar} \right) \\ &\times \bar{W}_{l'\mu\lambda}(\mathbf{p}_k - \mathbf{p}) \bar{W}_{l'\nu\mu}(\mathbf{p} - \mathbf{p}_k) f_{\nu\mu}(\mathbf{p}_2); \end{aligned} \quad (2.16)$$

$$\begin{aligned} S_{mn}(\mathbf{p}_k) &= \sum_{l'ss'} \int d\mathbf{p}_1 d\mathbf{p}_2 a_{ml'ns's'}(\mathbf{p}_k, \mathbf{p}_1, \mathbf{p}_2) \rho_{l's'}(\mathbf{p}_1) \rho_{ss'}(\mathbf{p}_2) \\ &- \sum_{l'ss'} \int d\mathbf{p} [b_{ml'ss'}(\mathbf{p}_k, \mathbf{p}) \rho_{ln}(\mathbf{p}_k) \rho_{ss'}(\mathbf{p}) + \rho_{ml}(\mathbf{p}_k) \rho_{ss'}(\mathbf{p}) b_{nl's's}^*(\mathbf{p}_k, \mathbf{p})], \end{aligned} \quad (2.17)$$

where in turn

$$\begin{aligned} a_{ml'ns's'}(\mathbf{p}_k, \mathbf{p}_1, \mathbf{p}_2) &= \frac{N}{V} \frac{1}{\hbar^2} \sum_q \int d\mathbf{p} \delta(\mathbf{p}_k - \mathbf{p}_1 + \mathbf{p} - \mathbf{p}_2) \\ &\times \bar{U}_{ml'qs'}(\mathbf{p}_k - \mathbf{p}_1) \bar{U}_{l'ns'q}(\mathbf{p}_1 - \mathbf{p}_k) \left\{ \delta_+^* \left( \omega_{ml} + \omega_{qs'} \right. \right. \\ &\left. \left. + \frac{p_k^2 - p_1^2}{2m_a\hbar} + \frac{p^2 - p_2^2}{2m_a\hbar} \right) + \delta_+^* \left( \omega_{l'n} + \omega_{sq} + \frac{p_1^2 - p_k^2}{2m_a\hbar} + \frac{p_2^2 - p^2}{2m_a\hbar} \right) \right\}, \end{aligned} \quad (2.18)$$

$$\begin{aligned} b_{ml'ss'}(\mathbf{p}_k, \mathbf{p}) &= \frac{N}{V} \left\{ \frac{i}{\hbar} \bar{U}_{ml'ss'}(0) + \frac{1}{\hbar^2} \sum_{l'q} \int d\mathbf{p}_1 d\mathbf{p}_2 \delta(\mathbf{p}_k - \mathbf{p}_1 + \mathbf{p} - \mathbf{p}_2) \right. \\ &\left. \times \bar{U}_{ml's'q}(\mathbf{p}_k - \mathbf{p}_1) \bar{U}_{l'iqs}(\mathbf{p}_1 - \mathbf{p}_k) \delta_+^* \left( \omega_{l'n} + \omega_{qs} + \frac{p_1^2 - p_k^2}{2m_a\hbar} + \frac{p_2^2 - p_1^2}{2m_a\hbar} \right) \right\}. \end{aligned} \quad (2.19)$$

The collision integrals  $S_{\mu\nu}$  and  $\sigma_{\mu\nu}$  are obtained respectively from  $S_{mn}$  and  $\sigma_{mn}$  by replacing the indices of the particles of the first kind by the indices of the particles of the second kind and vice versa; in addition, we make the substitutions  $N \rightleftharpoons M$ ,  $U \rightarrow G$ , etc. Within the framework of the employed scheme, it is easy to take into account also the quantum effects connected with the identity of the particles.

So far we have considered the kinetic-equation terms that are due to collisions. To solve spectroscopic problems it is necessary to take into account the interaction with the external electromagnetic field and with the thermostat. In many cases this can be done by adding corresponding additive terms without changing the collision integrals. Indeed, if the external field is not too intense, then the processes occurring during the collisions do not depend on whether the field is present or not. Further, the external field produces a spatial inhomogeneity, the scale of which is the wavelength. If, however, the wavelength greatly exceed the correlation radius (this is satisfied in the optical region), then it is easy to show that the collision integral will have practically the same form as in the spatially homogeneous problem, which was considered above. In this weakly-inhomogeneous problem<sup>[7]</sup>, the density matrices should be regarded as functions of the coordinates. If, in addition, we go over to a representation different from (2.6), without introducing factors of the type  $\exp[-ip_k^2/2m_a\hbar]$ , then convective terms  $im_a^{-1}p_k \nabla_k \rho_{mn}$  and  $im_b^{-1}p_\alpha \nabla_\alpha f_{\mu\nu}$  appear in the left sides of the kinetic equations. Thus, under the assumptions made, inclusion of the external field means addition of ordinary "dynamic" terms in the kinetic equations without practically changing the statistical or collision parts. The same considerations are valid, obviously, also with respect to the interaction between the atoms and the thermostat.

The kinetic equations thus assume the final form

$$\begin{aligned} \left( \frac{\partial}{\partial t} + \frac{p_k}{m_a} \nabla \right) \rho_{mn} &= [V', \rho(k)]_{mn} + S_{mn} + \sigma_{mn} + \Gamma_{mn}, \\ \left( \frac{\partial}{\partial t} + \frac{p_\alpha}{m_b} \nabla \right) f_{\mu\nu} &= [V', f(\alpha)]_{\mu\nu} + S_{\mu\nu} + \sigma_{\mu\nu} + \Gamma_{\mu\nu}, \end{aligned} \quad (2.20)$$

where  $V'$  is the operator of interaction of the particles with the electromagnetic field, and  $\Gamma_{mn}$  and  $\Gamma_{\mu\nu}$  are due to the interaction with the thermostat (see, for example,<sup>[9,10]</sup>

The kinetic equations (2.20) with the collision integrals (2.14) and (2.17) can serve as a basis for the analysis of those basic problems of spectroscopy, referred to in Sec. 1. We shall consider below certain problems in the theory of excitation of atoms (Sec. 3) and broadening of spectral lines (Sec. 4).

In concluding this section, we note the following. If we take into consideration the approximations made in<sup>[6]</sup>, and if the impurity particles are assumed to be immobile and structureless, then we obtain in lieu of (2.14) expressions for  $\sigma_{mn}$  which coincide with those obtained in<sup>[6]</sup>.

### 3. EXCITATION IN A MIXTURE OF GASES

In the widely used scheme for describing the excitation of atoms, one introduces in the right side of the equation for the diagonal element  $\rho_{jj}$  a certain term

$q_{jj}(\mathbf{p}_k)$ , with the meaning of the number of excitation acts per unit time (see, for example, <sup>[9,12,13]</sup>). It is also assumed usually that the same processes have no great influence on the nondiagonal elements  $\rho_{mn}$ , and that there are no corresponding terms in the equations for  $\rho_{mn}$  <sup>[13,14,11]</sup>. Such a scheme is physically quite clear. However, insofar as we know, it has not been rigorously proved, and the limits of its applicability are not clear.

We shall use the results of the preceding section to clarify this question, and consider first the equations for the diagonal elements. We assume that the particles of the first kind are excited only by collisions with particles of the second kind. The processes of the excitation of the level  $m$  are described, obviously, by the "inelastic part" of the arrival term in the collision integral (2.14):

$$q_{mm}(\mathbf{p}_k) = \sum_{l'} \int A_{ml'l'm}(\mathbf{p}_k, \mathbf{p}) \rho_{l'}(\mathbf{p}) d\mathbf{p}, \quad (3.1)$$

The prime at the summation sign denotes that  $l \neq m$  and  $l' \neq m$ . From (2.12) and (2.15) we see that  $A_{ml'l'm} \sim \exp[i\omega_{ll'}t]$ . For transitions lying in the visible region of the spectrum, these will be rapidly oscillating terms and can be neglected. Therefore

$$q_{mm}(\mathbf{p}_k) = \sum_l \int A_{mlml}(\mathbf{p}_k, \mathbf{p}) \rho_{ll}(\mathbf{p}) d\mathbf{p}. \quad (3.1')$$

Formula (3.1') states that the number of acts of excitation of the level  $m$  per unit time consists of the transitions  $l \rightarrow m$  and  $\mathbf{p} \rightarrow \mathbf{p}_k$  with probabilities  $A_{mlml}(\mathbf{p}_k, \mathbf{p})$ . In other words, the kinetic equations (2.13) corresponds fully to the aforementioned intuitive representations, so long as we are dealing with populations. Thus, this scheme is applicable if conditions are satisfied under which our derivation of the kinetic equation is valid, and in addition if it is possible to disregard terms with  $l \neq l'$  in (3.1). The most important conditions for the applicability of Eqs. (2.13) are smallness of the collision time in comparison with the free path time, and smallness of the interaction energy. The general structure of the collision integral is apparently determined by the first of these assumptions, whereas the second determines the explicit form of the kernels  $A_{ml'l'n}$ .

In the theory of gas quantum generators, the term  $q_{mm}(\mathbf{p}_k)$  is usually approximated by a given function  $\mathbf{p}_k$  (for example a Maxwellian distribution  $W_M(\mathbf{p}_k)$  <sup>[11,13]</sup>). This indeed takes place in the so-called model of strong collisions <sup>[3]</sup>, in which it is assumed that

$$A_{mlml}(\mathbf{p}_k, \mathbf{p}) = A_{ml} W_M(\mathbf{p}_k). \quad (3.2)$$

In this case

$$q_{mm}(\mathbf{p}_k) = Q_m W_M(\mathbf{p}_k), \quad Q_m = \sum_l A_{ml} \int \rho_{ll}(\mathbf{p}) d\mathbf{p}. \quad (3.3)$$

An analogous situation arises in the opposite limiting case, when the collision does not change the velocity (model of weak collisions, excitation by electrons).

Here

$$A_{mlml} = A_{ml} \delta(\mathbf{p}_k - \mathbf{p}), \quad q_{mm}(\mathbf{p}_k) = \sum_l A_{ml} \rho_{ll}(\mathbf{p}_k), \quad (3.4)$$

and if the distributions  $W(\mathbf{p}_k)$  of the atoms with respect to the momenta are identical at all levels, then

formula (3.3) remains in force.

As is well known, excitation of atoms in atom-atom collisions in gas-kinetic conditions is effective only for resonant processes <sup>[15]</sup>. In our formulas, this is reflected in the arguments of the  $\delta_+$  functions in (2.15), (2.16), (2.18), and (2.19): the kernel  $A_{ml'l'm}$ , for example, will have an appreciable value if  $\omega_{ml} \approx \omega_{\lambda\mu}$ , i.e., the transition  $m \rightarrow l$  of the atom of the first kind is accompanied by a transition  $\lambda \rightarrow \mu$  of the atom of the second kind between the levels with approximately the same energy difference. In the case of resonance and  $m_a \approx m_b$ , the model of strong collisions is realized <sup>[4]</sup>, but if  $|\omega_{ml} - \omega_{\lambda\mu}| \hbar \sim kT$ , then the distribution with respect to the velocities of the excited atoms has an appreciable probability of differing greatly, from equilibrium, since the difference of the energies of the internal degrees of freedom  $\hbar(\omega_{ml} - \omega_{\lambda\mu})$  is transferred to the translational motion <sup>[2]</sup>.

We now turn to the equations for the nondiagonal elements. These equations include terms analogous to (3.1):

$$q_{mn}(\mathbf{p}_k) = \sum_{l'} \int A_{ml'l'n}(\mathbf{p}_k, \mathbf{p}) \rho_{l'l'}(\mathbf{p}) d\mathbf{p}. \quad (3.5)$$

We can see from (2.15) that  $q_{mn}$  has an appreciable magnitude only if

$$\omega_{ml} + \omega_{\mu\lambda} \approx 0, \quad \omega_{n'l'} + \omega_{\mu\nu} \approx 0. \quad (3.6)$$

If the particles of the second kind are not polarized ( $f_{\lambda\mu} = \delta_{\lambda\nu} f_{\lambda\lambda}$ ), then both conditions (3.6) are satisfied simultaneously only when  $\omega_{ml} = \omega_{n'l'}$ , i.e., it is necessary that there exist levels  $l$  and  $l'$  equally remote from the levels  $m$  and  $n$  respectively, with  $\rho_{l'l'} \neq 0$ . Certain interference phenomena occurring in this situation are discussed in <sup>[18,19]</sup>. On the other hand, if  $\rho_{l'l'} \sim \delta_{l'l'}$ , or else  $\omega_{ml} \neq \omega_{n'l'}$ , then the nondiagonal elements are "not excited," although in the same collisions the populations are produced quite effectively.

Thus, the collision integral (2.14) includes in the form of particular cases also the "incoherent" excitation of the atomic system (the case  $q_{mn} = 0$ ) and the polarization transfer in collisions.

#### 4. BROADENING AND SHIFT OF SPECTRAL LINE

In this section we consider the question of broadening of a spectral line in the canonical formulation, i.e., for the transition between nondegenerate states and in the resonant approximation. We confine ourselves, furthermore, to broadening by extraneous particles. In this case it is necessary to retain only  $\sigma_{mn}$  in Eq. (20) for  $\rho_{mn}$ , and only the terms containing  $\rho_{mn}$  should be retained in  $\sigma_{mn}$ :

$$\begin{aligned} \left( \frac{\partial}{\partial t} + \frac{\mathbf{p}_k}{m_a} \nabla \right) \rho_{mn} &= -[\Gamma_1 + i\Delta_1 + v(\mathbf{p}_k)] \rho_{mn} \\ &+ \int A(\mathbf{p}_k, \mathbf{p}) \rho_{mn}(\mathbf{p}) d\mathbf{p} + iV_{mn}(\rho_{mm} - \rho_{nn}), \\ v(\mathbf{p}_k) &= B_{mm} + B_{nn}^*, \quad A(\mathbf{p}_k, \mathbf{p}) = A_{mmnn}(\mathbf{p}_k, \mathbf{p}); \\ \left( \frac{\partial}{\partial t} + \frac{\mathbf{p}_k}{m_a} \nabla \right) \rho_{jj} &= -[\Gamma_{1j} + v_j(\mathbf{p}_k)] \rho_{jj} \end{aligned} \quad (4.1)$$

<sup>2)</sup> Analogous effects are characteristic of the occurrence of atoms in dissociation of molecules (photodissociation <sup>[16]</sup>, dissociation in collisions <sup>[17]</sup>).

$$\begin{aligned}
 & + \int A_j(\mathbf{p}_k, \mathbf{p}) \rho_{jj}(\mathbf{p}) d\mathbf{p} \pm 2 \operatorname{Re} [i \rho_{mn} V_{mn}^*], \\
 v_j(\mathbf{p}_k) & = 2 \operatorname{Re} B_{jj}, \quad A_j(\mathbf{p}_k, \mathbf{p}) = A_{jjjj}(\mathbf{p}_k, \mathbf{p}). \quad (4.2)
 \end{aligned}$$

The terms  $\Gamma_{ij}$  and  $\Gamma_1 + i\Delta_1$  are due to the interaction with the thermostat. The collision integrals written out in (4.1) and (4.2) coincide in the form with those used in<sup>[3,11]</sup>. By the same token, the conclusions drawn in<sup>[3,11]</sup> acquire a physical meaning, for in the indicated papers the equations with such collision integrals were only postulated, but now they are the result of the application of the general Bogolyubov method. This makes it possible to explain in greater detail the structure of the functions  $\nu$ ,  $\nu_j$ ,  $A$ , and  $A_j$  introduced phenomenologically in<sup>[11]</sup>, and to establish important relations between them.

It is seen from (2.15) and (2.16) that the quantities of interest to us depend, generally speaking, on all the matrix elements  $f_{\mu\nu}$ , including the nondiagonal ones. In other words, the obtained formulas contain unique interference effects connected with the possible coherence of the states of the perturbing particles. In our paper, however, we shall not consider these phenomena, and assume that the matrix  $f_{\mu\nu}$  is diagonal. In this case

$$\begin{aligned}
 B_{jj}(\mathbf{p}_k) & = \frac{M}{V} \left\{ \frac{i}{\hbar} \sum_{\mu} W_{jj\mu\mu}(0) \langle f_{\mu\mu} \rangle + \frac{1}{\hbar^2} \sum_{l,\mu} \int d\mathbf{p}_l d\mathbf{p}' d\mathbf{p} \right. \\
 & \times \delta(\mathbf{p}_k - \mathbf{p}_l + \mathbf{p}' - \mathbf{p}) \delta_{\mu\alpha} \left( \omega_{lj} + \omega_{\mu\lambda} + \frac{p_l^2 - p_k^2}{2m_a\hbar} + \frac{p^2 - p'^2}{2m_b\hbar} \right) \\
 & \left. \times |W_{jj\mu\mu}(\mathbf{p}_k - \mathbf{p})|_{\lambda\mu}^2 \langle f_{\mu\mu} \rangle \right\}, \quad \langle f_{\mu\mu} \rangle = \int f_{\mu\mu}(\mathbf{p}_\alpha) d\mathbf{p}_\alpha; \quad (4.3) \\
 A_{jjj'j'}(\mathbf{p}_k, \mathbf{p}) & = \frac{M}{V} \frac{1}{\hbar^2} \sum_{\lambda,\mu} \int d\mathbf{p}_1 d\mathbf{p}_2 \delta(\mathbf{p}_k - \mathbf{p} + \mathbf{p}_2 - \mathbf{p}_1) \\
 & \times \delta \left( \omega_{\lambda\mu} + \frac{p_k^2 - p^2}{2m_a\hbar} + \frac{p_2^2 - p_1^2}{2m_b\hbar} \right) W_{jj\lambda\mu}(\mathbf{p}_k - \mathbf{p}) W_{j'j'\mu\lambda}(\mathbf{p} - \mathbf{p}_k) f_{\mu\mu}(p_1).
 \end{aligned}$$

In writing down (4.3) we used the fact that

$$W_{j\mu\lambda}(\mathbf{p}) = W_{j\lambda\mu}^*(-\mathbf{p})$$

in view of the Hermitian character of the operator  $W$ .

The structure of expressions (4.3) is quite clear. In our problem, the perturbing particles have a discrete spectrum of states; the terms of the series in  $\mu$  in (4.3) determine the contribution of the population  $f_{\mu\mu}$  of the level  $\mu$  to the broadening of the line in the transition  $m \rightarrow n$ , and to the probability of quenching of the levels  $j = m, n$ . The term linear in  $W$  determines the addition to the energy resulting from the correlation between the particles in the self-consistent-field approximation. In the case of structureless particles, this term is of no interest, since it means a shift of the energy reference point (see<sup>[7]</sup>, p. 235). In our problem, on the other hand, it makes a definite contribution to the shift (but not the width) of the line, since the renormalization of the energy turns out, generally speaking, to be different for different levels  $j$ .

Terms quadratic in  $W_2$  can be interpreted in the following manner:  $|W_{j\lambda\lambda\mu}|^2$  determines the probability of the transition  $j \rightarrow l$  of a particle of the first kind with a particle of the second kind located at the level  $\lambda$  and undergoing the transition  $\lambda \rightarrow \mu$ . In each act, the momenta of the particles can also change in accordance with the scheme  $\mathbf{p}_k \rightarrow \mathbf{p}'_k, \mathbf{p}_\alpha \rightarrow \mathbf{p}'_\alpha$ , a fact regu-

lated by the laws of the conservation of the total energy and of the total momentum (the  $\delta$  and  $\delta_+$  functions in (4.3)).

On the whole, the real part of  $B_{jj}$  (and consequently also of  $\nu_j$ ) gives the total probability per unit time of the departure of a particle located at the level  $j$  with momentum  $\mathbf{p}_k$  to other points of momentum space. The terms  $|W_{jl\lambda\mu}|^2, j \neq l$ , correspond in this case to the simultaneous transition  $j \rightarrow l$  and  $\lambda \rightarrow \mu$ , i.e., to quenching, and the terms with  $|W_{jj\lambda\mu}|^2$  are connected with quaselectric processes—a particle of the first kind remains at the level  $j$ , and the internal state of the particle of the second kind may change. It is easy to verify that the integral with respect to  $\mathbf{p}_k$  of the elastic part (in the indicated sense) of the collision integral  $\sigma_{jj}$  vanishes. This means that only the velocity changes, but not the level, i.e., the interpretation presented above is confirmed.

The integral terms in (4.1) and (4.2) describe the “arrival” of particles with momentum  $\mathbf{p}_k$  from other parts of momentum space. It is seen from (4.3) that in the discussed approximation they are determined entirely by the quasielastic processes, and only processes in which the particle does not lead either the level  $m$  or the level  $n$  contribute to the nondiagonal element  $A_{mmnn}$ .

Let us compare the diagonal and nondiagonal collision integrals. We conclude from (4.1) and (4.2) that the “departure frequencies” are connected by the simple relation

$$2\operatorname{Re} v = \nu_m + \nu_n. \quad (4.4)$$

With respect to the arrival terms, we can only setup the inequality

$$A_m(\mathbf{p}_k, \mathbf{p}) + A_n(\mathbf{p}_k, \mathbf{p}) \geq 2\operatorname{Re} A(\mathbf{p}_k, \mathbf{p}), \quad (4.5)$$

which is equivalent, by virtue of the definitions (4.1)–(4.3), to the obvious inequalities

$$|W_{mm\lambda\mu} - W_{nn\lambda\mu}|^2 \geq 0. \quad (4.6)$$

The inequality (4.5) means that the rate of arrival in the populations is not smaller than the rate of arrival for the nondiagonal element. The equal sign in (4.5) is reached only when  $W_{mm\lambda\mu} = W_{nn\lambda\mu}$ , i.e., for identical perturbations of the combining levels  $m$  and  $n$ . In such collisions, the phase of the equivalent atomic oscillator does not collapse. Consequently, it can be stated that the inequality (4.5) is connected with the fact that the lifetime of the nondiagonal element is due not only to quenching processes, but also to the “phase memory” of the oscillator.

In certain simplest cases, the quantities  $\Gamma_{ij}, \Gamma_1, \Delta, \nu$ , and  $\nu_j$  determine directly the width and the shift of the levels, but in the general case this cannot be said without specifying concretely the form of the kernels  $A$  and  $A_j$ . Let us therefore consider first the general problem of the power absorbed (or emitted) by the atomic system. We start from the work performed by the field:

$$P = -2\operatorname{Re} [iV_{mn}^* \rho_{mn}]. \quad (4.7)$$

We assume that the external field is weak, i.e., the populations  $\rho_{jj}$  of the levels  $j = m, n$ , are practically



independent of the field. Then the term with  $iV_{mn}(\rho_{mm} - \rho_{nn})$  in (4.1) can be regarded as a specified function of  $\mathbf{p}_k, \mathbf{r}_k,$  and  $t,$  and the problem reduces to finding a solution of the inhomogeneous equation (4.1). We express this solution in terms of the Green's function  $f(\mathbf{r}, \mathbf{p}, t | \mathbf{r}_0, \mathbf{p}_0, t_0)$  of the corresponding homogeneous equation:

$$\rho_{mn}(\mathbf{r}, \mathbf{p}, t) = i \int f(\mathbf{r}, \mathbf{p}, t | \mathbf{r}_0, \mathbf{p}_0, t_0) \cdot V_{mn}(\mathbf{r}_0, t_0) N(\mathbf{r}_0, \mathbf{p}_0, t_0) d\mathbf{r}_0 d\mathbf{p}_0 dt_0;$$

$$\left(\frac{\partial}{\partial t} + \frac{\mathbf{p}_k}{m_a} \nabla + \Gamma_1 + i\Delta + \nu\right) f - \int A(\mathbf{p}_k, \mathbf{p}) f d\mathbf{p} \quad (4.8)$$

$$= \frac{1}{\pi} \delta(t - t_0) \delta(\mathbf{r} - \mathbf{r}_0) \delta(\mathbf{p} - \mathbf{p}_0). \quad (4.9)$$

We shall assume, as usual, that  $N, A, \Gamma_1, \Delta,$  and  $\nu$  do not depend on  $\mathbf{r}$  or  $t.$  It is then natural to go over to the space-time Fourier transform of the Green's function, after which (4.7) can be rewritten in the form

$$P = 2\text{Re} \{ \int F(\mathbf{k}, \mathbf{p}, \Omega | \mathbf{p}_0) N(\mathbf{p}_0) d\mathbf{p}_0 d\mathbf{k} d\Omega \times \exp[-i\Omega(t - t_0) + ik(\mathbf{r} - \mathbf{r}_0)] V_{mn}^*(\mathbf{r}, t) V_{mn}(\mathbf{r}_0, t_0) d\mathbf{r}_0 dt_0 \}, \quad (4.10)$$

where the function  $F(\mathbf{k}, \mathbf{p}, \Omega | \mathbf{p}_0)$  is a solution of the equation<sup>3)</sup>

$$\left[ \nu + \Gamma_1 - i \left( \Omega - \Delta - k \frac{\mathbf{p}}{m_a} \right) \right] F - \int A F d\mathbf{p} = \frac{1}{\pi} \delta(\mathbf{p} - \mathbf{p}_0). \quad (4.11)$$

The factor in the second line of (4.10) determines obviously the intensity of the field concentrated in the plane monochromatic wave with frequency  $\Omega$  and wave vector  $\mathbf{k}.$  Consequently, the Fourier transform of the Green's function  $F(\mathbf{k}, \mathbf{p}, t | \mathbf{p}_0)$  determines the absorption (emission) line shape for atoms having a velocity  $\mathbf{v} = \mathbf{p}/m_a,$  and of excited atoms having a velocity  $\mathbf{v}_0 = \mathbf{p}_0/m_a.$  The absorption of energy from the plane wave by the ensemble of atoms is described by the function

$$I = \int d\mathbf{p} d\mathbf{p}_0 F(\mathbf{k}, \mathbf{p}, \Omega | \mathbf{p}_0) w(\mathbf{p}_0), \quad (4.12)$$

where  $w(\mathbf{p}_0)$  is the distribution of the atoms with respect to the velocities, at an excitation normalized to unity. Under such a normalization, as can be readily shown, we have  $\text{Re} \int I(\Omega) d\Omega = 1.$

Formulas (4.11) and (4.12) solve the problem of the shape of the absorption line in general form, without specifying concretely the form of a kernel  $A(\mathbf{p}_k, \mathbf{p}).$  The principal role is played here by the Fourier transformation of the Green's function of the kinetic equation (2.20).

Let us consider the so-called Lorentz-Weisskopf case, when the change of the velocity by the collision is neglected, i.e.,  $A \sim \delta(\mathbf{p} - \mathbf{p}').$  Then

$$F(\mathbf{k}, \mathbf{p}, t | \mathbf{p}_0) = \frac{\pi^{-1} \delta(\mathbf{p} - \mathbf{p}_0)}{\Gamma - i(\Omega - k\mathbf{p}/m - \Delta)};$$

$$\Gamma = \Gamma_1 + \nu' - \tilde{\nu}', \quad \Delta = \Delta_1 + \nu'' - \tilde{\nu}'', \quad \tilde{\nu} = \tilde{\nu}' + i\tilde{\nu}'' = \int A(\mathbf{p}, \mathbf{p}') d\mathbf{p}'. \quad (4.13)$$

The width  $\Gamma$  and the shift  $\Delta$  of the line are made up of components due to the interaction with the thermostat ( $\Gamma_1, \Delta_1$ ) and the particles of the second kind. We are now interested in the latter components:

$$\Gamma_2 \equiv \nu' - \tilde{\nu}' = \text{Re} \left\{ B_{mm}^* + B_{nn}^* - \int A(\mathbf{p}, \mathbf{p}') d\mathbf{p}' \right\},$$

$$\Delta_2 \equiv \nu'' - \tilde{\nu}'' = \text{Im} \left\{ B_{mm}^* + B_{nn}^* - \int A(\mathbf{p}, \mathbf{p}') d\mathbf{p}' \right\}. \quad (4.14)$$

From (4.14) and (4.5) we can see that

$$2\Gamma_2 \geq \Gamma_{2m} + \Gamma_{2n}; \quad \Gamma_{2j} = \nu_j - \tilde{\nu}_j, \quad \tilde{\nu}_j = \int B_{jj} d\mathbf{p}. \quad (4.15)$$

The quantities  $\Gamma_{2j}$  represent the rates of quenching of the levels  $j = m, n.$  The inequality (4.15) signifies that the line width is determined not only by the quenching of the levels (Lorentz mechanism), but also by the quasielastic processes which appear as a result of the collapse of the phase of the atomic oscillator (the Weisskopf mechanism).

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<sup>3)</sup>We hope that the symbols  $F(\mathbf{k}, \mathbf{p}, t | \mathbf{p}_0)$  and  $N(\mathbf{p}_0), V_{mn}, f(\mathbf{r}, \mathbf{p}, t | \mathbf{r}_0, \mathbf{p}_0, t_0),$  adopted to be consistent with [11], will not be confused with  $F, F(\mathbf{k}),$  etc.