THE EFFECT OF COLLISIONS ON SPECTRAL CHARACTERISTICS OF GAS LASERS

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Stimulated processes occurring in the field of a monochromatic standing wave are considered within the framework of impact theory. The collision integral in the density matrix equation includes atom diffusion in velocity space and atomic-oscillator phase shifts that occur simultaneously with changes in the velocity of the atomic center of mass. When collisions of this kind are considered, the gas laser power becomes an asymmetric frequency function, in agreement with experimental results.[2,3]

1. INTRODUCTION

BENNETT and others[1] observed an asymmetry of the neon line, $\lambda = 3.39\ \mu$, due to collision perturbations of the emitting atoms. The same cause can apparently be ascribed to the asymmetric frequency dependence established by Javan and others.[2,3]

The results of the above research are of definite interest in relation to many viewpoints, and primarily to the problem of spectral line broadening. The point is that the experimental conditions of that research have a priori satisfied certain relevance criteria of the impact theory of line broadening (see[4], for example). According to the usual variants of this theory, however, the line contour should be symmetric owing to both Doppler broadening (if the velocity distribution of atoms is symmetric) and interaction broadening. Such a clear discrepancy between the above conclusion of the impact theory and the results of[1-3] has caused lively discussions[5-7]. Fork and Pollack[5] stated that changes in the velocity of an atom in collisions cannot in principle result in asymmetry of line contour, because of the even velocity distribution of atoms; consequently, the phenomena observed in[1-3] should be ascribed to interaction broadening. Javan and Szöke[6] adopted essentially the same view.

It should be emphasized that the search for causes of the asymmetry is hardly a problem in alternatives. The point is that concurrent action of two causes of broadening (Doppler and collision broadening in this case) can result in an asymmetric line contour even if each cause acting separately produces a symmetrical contour. It all depends on whether these two causes are statistically dependent or independent. If the broadening mechanisms under consideration are statistically independent, the resulting contour will be symmetrical. If the reverse is true, there will be no symmetry in general. This situation follows from the general theorems of Fourier analysis and is well known from the theory of fluctuation modulation; similar phenomena have been studied in certain microwave circuits (see[8], for example).

No such phenomena seem to have been previously observed in spectroscopy, although there are some indications of a statistical dependence of the broadening due to interaction and Doppler effect.[4] The first discussion in relation to spectroscopic problems was published by the author and Sobel'man[9], who postulated a general theory of Doppler broadening of spectral lines, taking collisions into account. It was found that the line contour is generally asymmetric in situations of practical interest involving broadening due to atom-atom collisions. Consequently, it seems to us, the explanation of the results obtained in[1-3] should be based on a concurrent consideration of the Doppler and collision broadening, taking their statistical dependence into account. The present paper deals with the theoretical analysis of this problem.

Let us emphasize that such an interpretation means in essence that the collision integral in the density matrix equation cannot be reduced to the usual relaxation terms with some relaxation constants. Such an approximation of the collision integral leads to a shifted but symmetrical line contour. In this connection, Sec. 2 of this paper deals with a collision integral that describes atomic diffusion in velocity space and takes into account the statistical dependence of the perturbations of internal motions of the atom and of its translational motion.

Subsequent sections deal with the saturation effect in a standing-wave field for two model equations of the collision integral (Sects. 3 and 4), and with the computation of a gas laser power (Sec. 5).

2. GENERAL EQUATIONS

The present paper deals with the case when the external field spectrum $E(t, \mathbf{r})$ is concentrated near the transition frequency ω_{mn} between two excited states m and n of an isolated atom, so that the two-level approximation can be used. We start with the following system of equations for the elements of the density matrix $\hat{\rho}$:

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho_{jj} = \pm 2\operatorname{Re}[iV_{mn}{}^*\rho_{mn}] + q_j + S_j,$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho_{mn} = iV_{mn}[\rho_{mm}-\rho_{nn}] + S,$$

$$S_j = -\nu_j\rho_{jj} + \int A_j(\mathbf{v}', \mathbf{v})\rho_{jj}(\mathbf{r}, \mathbf{v}', t)\,d\mathbf{v}' \quad (j = m, n),$$

$$S = -\nu\rho_{mn} + \int A(\mathbf{v}', \mathbf{v})\rho_{mn}(\mathbf{r}, \mathbf{v}', t)\,d\mathbf{v}'. \tag{2.1}$$

Here, $\mathbf{r}$ and $\mathbf{v}$ are respectively the coordinate and velocity in the c.m.s. of the atom, q_j is the excitation rate of atoms in the states j, $\mathbf{v}$, and $\hbar V_{mn} = p_{mn} e^{i\omega_{mn}t} E(t, \mathbf{r})$ is the matrix element of interaction with the external field; the ± signs correspond to j = m and n, and $A_j(\mathbf{v}', \mathbf{v})$ and $A(\mathbf{v}', \mathbf{v})$ are the kernels of collision integrals S_j and S.

In the absence of inelastic processes, collisions do not affect the total number of particles at each level, i.e., integrals with respect to $\mathbf{v}$, of the diagonal collision integrals should turn to zero. This can take place if

$$\Gamma_j = \nu_j - \tilde{\nu}_j = \nu_j - \int A_j(\mathbf{v}, \mathbf{v}')\,d\mathbf{v}' \equiv 0.$$

In this case, $A_j(\mathbf{v}', \mathbf{v})$ represents the probability (per unit time) of the change in velocity $\mathbf{v}' \to \mathbf{v}$. When quenching collisions are taken into account, the Γ_j are positive and determine the time in which the atoms with velocity $\mathbf{v}$ relax from the levels j = m,n. In the limiting case, when each collision leads to quenching (the Lorentz model), $\tilde{\nu}_j = 0$ and $\Gamma_j = \nu_j$.

The kernel $A(\mathbf{v}', \mathbf{v})$ of the nondiagonal collision integral is in general complex, reflecting the level shift due to collision (the Weisskopf broadening mechanism), or, in classical terms, the phase shift of the atomic oscillator.[1)] The difference

$$\nu - \int A(\mathbf{v}, \mathbf{v}')\,d\mathbf{v}' = \Gamma + i\Delta \tag{2.2}$$

determines the impact width Γ and the shift Δ of the spectral line.

From now on it will be convenient to distinguish two kinds of collisions. In collisions of the first kind the atom velocity remains unchanged but a phase shift can occur. This obviously includes collisions with electrons. In collisions of the second kind both the atom velocity and the phase of the atomic oscillator can change (collisions with heavy particles, such as atoms, ions, and molecules). Collisions of the first and second kind can be considered statistically independent and thus the kernels can be represented in the following form:

$$A_j(\mathbf{v}', \mathbf{v}) = \tilde{\nu}_{1j}\delta(\mathbf{v}-\mathbf{v}') + A_{2j}(\mathbf{v}', \mathbf{v}), \quad \nu_j = \nu_{1j} + \nu_{2j};$$

$$A(\mathbf{v}', \mathbf{v}) = \tilde{\nu}_1\delta(\mathbf{v}-\mathbf{v}') + A_2(\mathbf{v}', \mathbf{v}), \qquad \nu = \nu_1 + \nu_2. \tag{2.3}$$

Substituting (2.3) in (2.1), we obtain

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho_{jj} = -\Gamma_{1j}\rho_{jj} - \left\{\nu_{2j}\rho_{jj} - \int A_{2j}(\mathbf{v}', \mathbf{v})\rho_{jj}(\mathbf{r}, \mathbf{v}', t)\,d\mathbf{v}'\right\} \pm 2\operatorname{Re}[iV_{mn}{}^*\rho_{mn}] + q_j,$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho_{mn} = -(\Gamma_1 + i\Delta_1)\rho_{mn} - \left\{\nu_2\rho_{mn} - \int A_2(\mathbf{v}', \mathbf{v})\rho_{mn}(\mathbf{r}, \mathbf{v}', t)\,d\mathbf{v}'\right\} + iV_{mn}(\rho_{mm} - \rho_{nn});$$

$$\Gamma_{1j} = \nu_{1j} - \tilde{\nu}_{1j}, \quad \Gamma_1 + i\Delta_1 = \nu_1 - \tilde{\nu}_1. \tag{2.4}$$

The quantities Γ_{ij}, Γ_1, and Δ_1 determine the contributions of collisions of the first kind to the quenching probability of the j = m, n levels, to the collision width, and to the line shift. The corresponding relaxation terms are of the standard form. The collision-integral term determined by collisions of the second kind has a different structure. One could arbitrarily break it up to form terms

1)We note that ρ_{mn} is analogous to the Fourier transform $\tilde{f}(\mathbf{r},\mathbf{v},t)$ (with respect to the phase variable φ) of the classical oscillator distribution function $f(\mathbf{r},\mathbf{v},\varphi,t)$ introduced in [9]. On the other hand, kernel $A(\mathbf{v}',\mathbf{v})$ corresponds to the Fourier transform $\tilde{A}(\mathbf{v}',\mathbf{v})$ of the probability (per unit time) of the discontinuity $\mathbf{v}' \to \mathbf{v}$, $\varphi' \to \varphi$.

$$\Gamma_{2j}\rho_{jj} = (\nu_{2j} - \tilde{\nu}_{2j})\,\rho_{jj} = \left(\nu_{2j} - \int A_{2j}(\mathbf{v}, \mathbf{v}')\,d\mathbf{v}'\right)\rho_{jj},$$

$$(\Gamma_2 + i\Delta_2)\,\rho_{mn} = (\nu_2 - \tilde{\nu}_2)\,\rho_{mn} = \left(\nu_2 - \int A_2(\mathbf{v}, \mathbf{v}')\,d\mathbf{v}'\right)\rho_{mn}, \tag{2.5}$$

interpreting Γ_{2j}, Γ_2, and Δ_2 as the quenching, width, and shift of the lines due to collisions of the second kind. However, the remaining part of the collision integral would be dependent upon both velocity and phase changes. The impossibility of separating the collision integral into two terms each of which defines only a single process, is clearly due to the fact that these processes occur simultaneously in every collision act and, therefore, are statistically dependent.

Quenching due to spontaneous transitions has not been considered in the above. As we know, allowance for spontaneous transitions leads to an increase of Γ_{1j} and Γ_1 by $2\gamma_j$ and $\gamma_m + \gamma_n$, where $2\gamma_j$ are the probabilities of spontaneous quenching of the $j = m, n$ states. We shall henceforth assume that Γ_{1j} and Γ_1 have been suitably redefined.

The range of applicability of (2.4) is limited by a number of considerations. Let us first note the usual limitations connected with the impact approximation and the neglect of energy-level degeneracy. A specific case is the linearity of the collision integral, signifying neglect of the collisions of atoms situated at levels m and n. This assumption is fully justified in the case of a large number of objects characterized by a relatively low concentration of excited atoms.

The system (2.4) will be solved by successive approximations assuming a weak interaction with the field. For this purpose, it suffices to find the Green's functions f_j and f of Eqs. (2.4) with $V_{mn} = 0$, i.e., to solve the equations

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \Gamma_{1j} + \nu_{2j}\right) f_j - \int A_{2j}(\mathbf{v}', \mathbf{v})\, f_j(\mathbf{r}, \mathbf{v}', t\,|\,\mathbf{r}_0, \mathbf{v}_0, t_0)\,d\mathbf{v}' = \delta(x - x_0),$$

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \Gamma_1 + i\Delta_1 + \nu_2\right) f - \int A_2(\mathbf{v}', \mathbf{v})\, f(\mathbf{r}, \mathbf{v}', t\,|\,\mathbf{r}_0, \mathbf{v}_0, t_0)\,d\mathbf{v}' = \delta(x - x_0), \tag{2.6}$$

where x and x_0 designate the sets of variables **r**, **v**, t and $\mathbf{r}_0$, $\mathbf{v}_0$, t_0. If the solutions of (2.6) are known, the $(2s + 1)$th approximation for ρ_{mn} and the $(2s)$th approximation for ρ_{jj} can be written as follows:

$$\rho_{mn}^{(2s+1)}(x) = i \int f(x\,|\,x_0)\, V_{mn}(x_0)\,[\rho_{mm}^{(2s)}(x_0) - \rho_{nn}^{(2s)}(x_0)]\,dx_0,$$

$$\rho_{jj}^{(2s)}(x) = \rho_{jj}^{(0)} \pm \int f_j(x\,|\,x_0)\cdot 2\,\mathrm{Re}\,[iV_{mn}^*(x_0)\,\rho_{mn}^{(2s-1)}(x_0)]\,dx_0,$$

$$\rho_{jj}^{(0)}(x) = \int f_j(x\,|\,x_0)\,q_j(x_0)\,dx_0. \tag{2.7}$$

In particular, when the first-order nonlinear corrections (to be considered below) are taken into account, we have

$$\rho_{jj}^{(2)}(x) = \rho_{jj}^{(0)}(x) \mp 2 \int f_j(x\,|\,x_1) \times \mathrm{Re}\,[V_{mn}^*(x_1)\, f(x_1\,|\,x_0)\, V_{mn}(x_0)]\, N(x_0)\,dx_0\,dx_1, \tag{2.8}$$

$$\rho_{mn}^{(3)}(x) = i \int f(x\,|\,x_0)\, V_{mn}(x_0)\, N(x_0)\,dx_0 - i \int f(x\,|\,x_2)\, V_{mn}(x_2) \sum_{j=m,\,n} f_j(x_2\,|\,x_1) \times \mathrm{Re}\,[V_{mn}^*(x_1)\, f(x_1\,|\,x_0)\, V_{mn}(x_0)]\cdot N(x_0)\,dx_0\,dx_1\,dx_2, \tag{2.9}$$

where

$$N(x) = \int \{f_m(x\,|\,x_0)\,q_m(x_0) - f_n(x\,|\,x_0)\,q_n(x_0)\}\,dx_0.$$

We are interested in the behavior of the atom in the field of a monochromatic standing wave with a frequency ω:

$$V_{mn}(t, \mathbf{r}) = Ge^{-i\Omega t}\cos \mathbf{kr}, \quad \Omega = \omega - \omega_{mn}. \tag{2.10}$$

To exclude the effect of excitation diffusion and inhomogeneous velocity distribution which is independent of the field, let us regard q_j as independent of the coordinate **r** and the time t, and assume a Maxwellian dependence on **v**:

$$q_j(x) = Q_j W_M(\mathbf{v}), \qquad W_M(\mathbf{v}) = (\sqrt{\pi}\bar{v})^{-3}\exp(-\mathbf{v}^2/\bar{v}^2). \tag{2.11}$$

Here Q_j is the total number of acts of atomic excitation to the level j in a unit volume per unit time.

3. THE STRONG COLLISION MODEL

Following Keilson and Storer[10], we consider the kernels

$$A_{2j}(\mathbf{v}', \mathbf{v}) = \frac{\tilde{\nu}_{2j}}{[\pi\bar{v}^2(1-\gamma^2)]^{3/2}} \exp\left\{-\frac{(\mathbf{v}-\gamma\mathbf{v}')^2}{\bar{v}^2(1-\gamma^2)}\right\}, \tag{3.1}$$

$$A_2(\mathbf{v}', \mathbf{v}) = \frac{\tilde{\nu}_2}{[\pi\bar{v}^2(1-\gamma^2)]^{3/2}} \exp\left\{-\frac{(\mathbf{v}-\gamma\mathbf{v}')^2}{\bar{v}^2(1-\gamma^2)}\right\}. \tag{3.2}$$

It follows from (3.1) that the ratio of the average atomic velocities after and before collision is γ; and the velocity dispersion is $(1 - \gamma^2)\bar{v}^2$. Consequently, the basic assumption of model (3.1) is that the above ratio is considered independent of $\mathbf{v}'$. The value of the constant γ should be chosen in accordance with the specific nature of the collisions. If the emitting particle is scattered by a much lighter particle, then $1 - \gamma \ll 1$ and each collision results in relatively small velocity changes. This limiting case will be called the weak-collision model and will be discussed in Sec. 4. An inverse ratio of masses of the emitting and perturbing particles, or even their equality, allows us to assume that $\gamma \approx 0$.[10,11] In this case the distribution of atoms with respect to the velocities **v** after the collision does not depend upon **v'**, and the change in velocity is of the order of $\bar{v}$. This case will be designated as the strong collision model.

Let

$$A_{2j}(\mathbf{v}', \mathbf{v}) = \tilde{\nu}_{2j} W_M(\mathbf{v}), \quad A_2(\mathbf{v}', \mathbf{v}) = \tilde{\nu}_2 W_M(\mathbf{v}). \quad (3.3)$$

The solutions of (2.6) with kernels of this type are given by

$$f(\mathbf{r}, \mathbf{v}, t | \mathbf{r}_0, \mathbf{v}_0, t_0) = \int \exp[-i\Omega(t - t_0) + i\mathbf{k}(\mathbf{r} - \mathbf{r}_0)] F(\mathbf{k}, \mathbf{v}, \Omega | \mathbf{v}_0)\, d\mathbf{k}\, d\Omega,$$

$$F(\mathbf{k}, \mathbf{v}, \Omega | \mathbf{v}_0) = \frac{1}{p + i\mathbf{k}\mathbf{v}} \left\{ \delta(\mathbf{v} - \mathbf{v}_0) + \frac{\tilde{\nu}_2}{1 - \tilde{\nu}_2 Z(p, k)} \frac{W_M(\mathbf{v})}{p + i\mathbf{k}\mathbf{v}_0} \right\}, \quad (3.4)$$

$$p = \Gamma_1 + \nu_2 - i(\Omega - \Delta_1);$$

$$f_j(\mathbf{r}, \mathbf{v}, t | \mathbf{r}_0, \mathbf{v}_0, t_0) = \int \exp[-i\Omega(t - t_0) + i\mathbf{k}(\mathbf{r} - \mathbf{r}_0)] \times F_j(\mathbf{k}, \mathbf{v}, \Omega | \mathbf{v}_0)\, d\mathbf{k}\, d\Omega,$$

$$F_j(\mathbf{k}, \mathbf{v}, \Omega | \mathbf{v}_0) = \frac{1}{p_j + i\mathbf{k}\mathbf{v}} \left\{ \delta(\mathbf{v} - \mathbf{v}_0) + \frac{\tilde{\nu}_{2j}}{1 - \tilde{\nu}_{2j} Z(p_j, k)} \times \frac{W_M(\mathbf{v})}{p_j + i\mathbf{k}\mathbf{v}_0} \right\} \quad p_j = \Gamma_{1j} + \nu_{2j} - i\Omega. \quad (3.5)$$

The following notation has been introduced here:

$$Z(p, k) = \int \frac{W_M(\mathbf{v})}{p + i\mathbf{k}\mathbf{v}} d\mathbf{v} = \frac{\sqrt{\pi}}{k\bar{v}} \exp\left[\left(\frac{p}{k\bar{v}}\right)^2\right]\left[1 - \Phi\left(\frac{p}{k\bar{v}}\right)\right], \quad (3.6)$$

where $\Phi(x)$ is the probability integral.[2)]

Since it is intended to apply the theory to the analysis of gas laser properties and to the interpretation of the results of[1-3], further computations will be carried out on the assumption that the Doppler linewidth $k\bar{v}$ is considerably larger than all constants Γ, Γ_m, Γ_n, ν, ν_m, ν_n. Therefore, the following formulas contain only the first terms of the power series resulting from an expansion of the ratios of these parameters to $k\bar{v}$.

Substituting f and f_j from (3.4) and (3.5) into (2.8) and (2.9), integrating with respect to x_j, and averaging ρ_{mn} over **v**, we obtain,

$$\rho_{mm} - \rho_{nn} = N W_M(\mathbf{v}) \times \left\{ 1 - \frac{G^2}{2} \sum_{j=m,n} \frac{1}{\Gamma_{1j} + \nu_{2j}} \left[\frac{\Gamma_1 + \nu_2}{(\Gamma_1 + \nu_2)^2 + (\Omega - \Delta_1 - \mathbf{k}\mathbf{v})^2} + \frac{\Gamma_1 + \nu_2}{(\Gamma_1 + \nu_2)^2 + (\Omega - \Delta_1 + \mathbf{k}\mathbf{v})^2} \right] \right\} \quad (3.7)$$

$$\langle \rho_{mn} \rangle_v = i\pi N V_{mn}(t, \mathbf{r}) \{ J_1(\Omega) - {}^1\!/_4 G^2 J_2(\Omega) \}, \quad (3.8)$$

$$J_1(\Omega) = \frac{1}{\pi} \frac{Z(p, k)}{1 - \tilde{\nu}_2 Z(p, k)}, \quad p = \Gamma_1 + \nu_2 - i(\Omega - \Delta_1), \quad (3.9)$$

$$J_2(\Omega) = \frac{1}{\sqrt{\pi}\, k\bar{v}} \sum_{j=m,n} \frac{1}{\Gamma_{1j} + \nu_{2j}} \left[\frac{1}{\Gamma_1 + \nu_2} + \frac{1}{\Gamma_1 + \nu_2 - i(\Omega - \Delta_1)} \right] \exp\left[-\left(\frac{\Omega - \Delta_1}{k\bar{v}} \right)^2 \right], \quad (3.10)$$

$$N = Q_m/\Gamma_m - Q_n/\Gamma_n, \quad \Gamma_j = \Gamma_{1j} + \Gamma_{2j}, \quad j = m, n. \quad (3.11)$$

The parameter N is the integral (with respect to velocities) difference of populations of levels m and n when $G = 0$. It follows from (3.7) that in the absence of the field the $\rho_{ij}(\mathbf{v})$ are proportional to the equilibrium distribution $W_M(\mathbf{v})$. The second term in the braces in (3.7) is a nonequilibrium addition due to stimulated transitions. The field interacts most effectively with atoms whose velocity projection on the **k** direction satisfies the condition

$$\mathbf{k}\mathbf{v} = \pm(\Omega - \Delta_1). \quad (3.12)$$

We emphasize that only Δ_1 appears in (3.12); Δ_2 does not, in spite of its seeming relevance. To understand this clearly, one should take into consideration two points: the specific features of the strong collision model, and the simultaneity of phase and velocity variation in collisions of the second kind. In the model under consideration, the velocity of the atom changes in a collision by an amount on the order of $\bar{v} \gg (\Gamma_1 + \nu_2)/k$. Consequently, the atom practically does not interact with the field after the collision. Therefore, the phase shift acquired in the course of the collision "does not have time" to appear.

The widths of the nonequilibrium "dips" in the velocity distribution (more correctly, in the distribution with respect to $\mathbf{k} \cdot \mathbf{v}$) of the atoms equals $\Gamma_1 + \nu_2$. In the absence of collisions, it is deter-

2)Z(p,k) differs from the function $w(p/k\bar{v})$ tabulated in [12] by the factor $\sqrt{\pi}/k\bar{v}$.

mined by the radiative width $\gamma_m + \gamma_n$. Let us note that in our model the increase in dip width is proportional to the frequencies of collisions of both kinds, i.e., it is proportional to the concentrations of the perturbing particles.

Let us consider the relative power of stimulated emission:

$$R(\Omega) = -2\hbar\omega \operatorname{Re}[iV_{mn}{}^*\langle\rho_{mn}\rangle_v] = 2\pi\hbar\omega\, G^2\cos^2\mathbf{kr}\,\operatorname{Re}\{J_1(\Omega) - {}^1/_4 G^2 J_2(\Omega)\}. \quad (3.13)$$

We note that the nonlinear dependence of $R(\Omega)$ on the field amplitude in our approximation does not lead to an optical inhomogeneity of the medium: the expression in the braces in (3.13) does not depend on $\mathbf{r}$ and the factor in front of the braces is proportional to squared field at point $\mathbf{r}$. This result is due to two causes: large Doppler line width and the approximation used in analyzing the nonlinear effects. If the atoms are fixed, the inhomogeneity of the medium is given by the terms of order G^4.[13,14] On the other hand, if the Doppler line width is much larger than all the relaxation constants, the inhomogeneity occurs only when G^6 or $[(\nu + \Gamma)/k\bar{v}]^2 G^4$ terms are taken into account.

The function $I_1(\Omega) = \operatorname{Re}J_1(\Omega)$ determines the line contour (normalized to unity in area) without taking saturation into account; the function has been studied in detail in [9].3) In the assumption that $k\bar{v} \gg \Gamma_1$ or ν_2, the only significant fact is that $I_1(\Omega)$ is an asymmetric frequency function; the center of gravity of $I_1(\Omega)$ is located at $\Omega = \Delta_1 + \Delta_2$, while the maximum of the function occurs at

$$\Omega_{1\,max} = \Delta_1 + 2\Delta_2. \quad (3.14)$$

In other words, phase shifts occurring in collisions of the second kind shift the line maximum anomalously by an amount twice as large as in the case of collision broadening, the latter being statistically independent of the Doppler broadening (Δ_1). The asymmetry can be accounted for by retaining the Δ_2 term in (3.9):

$$\operatorname{Re}J_1(\Omega) \cong \frac{1}{\sqrt{\pi}\,k\bar{v}}\exp\left[-\left(\frac{\Omega-\Delta_1}{k\bar{v}}\right)^2\right][1 + 2\Delta_2\operatorname{Im}Z(p,k)]. \quad (3.15)$$

The function $I_2(\Omega) = \operatorname{Re}J_2(\Omega)$ gives the line contour variation due to the nonequilibrium velocity distribution of the atoms in the external field. Using (3.15) and (3.10), we obtain in lieu of

$$R(\Omega) = 2\frac{\sqrt{\pi}}{k\bar{v}}NG^2\cos^2\mathbf{kr} \times \exp\left[-\left(\frac{\Omega-\Delta_1}{k\bar{v}}\right)^2\right]\Big\{1 + 2\Delta_2\operatorname{Im}Z(p,k) - \frac{G^2}{4}\frac{1}{\Gamma_1+\nu_2}\left[\frac{1}{\Gamma_{1m}+\nu_{2m}} + \frac{1}{\Gamma_{1n}+\nu_{2n}}\right] \times\left[1 + \frac{(\Gamma_1+\nu_2)^2}{(\Gamma_1+\nu_2)^2 + (\Omega-\Delta_1)^2}\right]\Big\}. \quad (3.16)$$

Since $\nu_2 + \Gamma_1 \ll k\bar{v}$, Eq. (3.16) defines a contour with a total width of the order of $k\bar{v}$, within which there is a comparatively sharp dip $\Gamma_1 + \nu_2$ wide, having a minimum at the frequency of $\Omega_{2max} = \Delta_1$ (Fig. 1) shifted by $2\Delta_2$ relative to the maximum of the function $I_1(\Omega)$. Consequently, the oscillator phase shifts that are synchronous with the atom velocity changes can produce asymmetry in $R(\Omega)$, if $\Delta_2 \neq 0$.

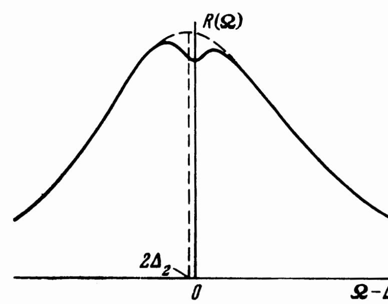

FIG. 1

The dip in the $R(\Omega)$ curve is due to the change in the distribution of the atoms with respect to v and also to the fact that the field has the form of a standing wave.[15,16] The dependence of the width and depth of the dip on the collision frequency finds an obvious intuitive interpretation in the fact that elastic collisions result in diffusion of the atoms in velocity space, in the smoothing of the non-uniformity of ρ_{jj}, and in the reduced effect of the field on $R(\Omega)$. Since the nonequilibrium part is relatively narrow $(\sim\Gamma_1/k)$, elastic collisions significantly affect the nonlinear term (with respect to G^2) if $\nu_2 \sim \Gamma_1$. The linear term of $I_1(\Omega)$, on the other hand, changes noticeably only if $\nu_2 \sim k\bar{v}$, since the effect of collisions consists in this case (given equilibrium velocity distribution) in retarding the motion of the atoms.[9]

4. THE WEAK COLLISION MODEL

We may naturally expect that the effect of diffusion on the nonlinear phenomena will be strongly dependent upon the nature of the elastic collisions.

3)The notation $J(\Omega) = J_1(\Omega)$, $I(\Omega) = I_1(\Omega)$ was used in [9].

Let us thus consider a model whose limiting case may be regarded as the opposite of the subject of Sec. 3. In the strong collision model, the velocity of the atom changed in a single collision by an amount on the order of $\bar{v}$. Now, let the velocity of the emitting atom be changed to a marked extent only after a large number of collisions, each of which contributes a small $|v - v'|$. This corresponds to the following inequality in (3.1) and (3.2):

$$1-\gamma \ll 1. \quad (4.1)$$

It can be shown [10] that the collision integrals in (2.4) assume under these conditions the Fokker-Planck form, with an accuracy to the terms of the order of $(1-\gamma)^2$:

$$S_j = -\Gamma_j\rho_{jj} + \mu_j[\mathrm{div}_{\mathbf{v}}(\mathbf{v}\rho_{jj}) + {}^1/_2\bar{v}^2\Delta_{\mathbf{v}}\,\rho_{jj}],$$
$$S = -(\Gamma+i\Delta)\rho_{mn} + \mu[\mathrm{div}_{\mathbf{v}}\,(\mathbf{v}\rho_{mn}) + {}^1/_2\bar{v}^2\Delta_{\mathbf{v}}\,\rho_{mn}], \quad (4.2)$$

where

$$\Gamma_j = \Gamma_{1j}+\Gamma_{2j}, \qquad \Gamma+i\Delta = \Gamma_1+\Gamma_2+i(\Delta_1+\Delta_2),$$
$$\mu_j = (\nu_{2j}-\Gamma_{2j})(1-\gamma), \qquad \mu = (\nu_2-\Gamma_2-i\Delta_2)(1-\gamma).$$

The parameters μ_j are the so-called dynamic friction coefficients, and $\mu_j\bar{v}^2/2$ are the diffusion coefficients (in the velocity space). It can be seen from (4.2) and (2.5) that μ_j decreases in the presence of inelastic collisions. The quantity μ is complex, reflecting a phase shift of the atomic oscillator in collision.

The solution f_j and f of (2.6) with collision integrals (4.2) can be written as

$$f_j(x|x_0) = \frac{1}{(2\pi)^3[ac-b^2]^{3/2}} \times \exp\left(-\Gamma_j\tau - \frac{c\xi^2 - 2b\xi\eta + a\eta^2}{2(ac-b^2)}\right),$$

$$a = \bar{v}^2\mu_j^{-2}[\mu_j\tau - (1-e^{-\mu_j\tau}) - {}^1/_2(1-e^{-\mu_j\tau})^2],$$
$$b = {}^1/_2\bar{v}^2\mu_j^{-1}(1-e^{-\mu_j\tau})^2, \qquad c = {}^1/_2\bar{v}^2(1-e^{-2\mu_j\tau}),$$
$$\tau = t-t_0, \qquad \xi = \mathbf{r}-\mathbf{r}_0-\mathbf{v}_0(1-e^{-\mu_j\tau})/\mu_j,$$
$$\eta = \mathbf{v}-\mathbf{v}_0e^{-\mu_j\tau}. \quad (4.3)$$

The expression for $f(x|x_0)$ will be omitted, since it is obtained from (4.3) by replacing Γ_j by $\Gamma + i\Delta$ and μ_j by μ.

Equations (4.3) show that when $\tau = t - t_0 = 0$, the function f_j becomes $\delta(\mathbf{r}-\mathbf{r}_0)\delta(\mathbf{v}-\mathbf{v}_0)$. Its width along $\mathbf{v}$, equal to $\sqrt{c}$, increases in time, approaching $\bar{v}/\sqrt{2}$ after a time interval $\sim 1/\mu_j$, and the maximum shifts at the same time towards $\mathbf{v} = 0$. We recall that the evolution of the velocity distribution was different in the strong collision model, comprising two terms in each instant of time (see (3.4)): one proportional to $\delta(\mathbf{v}-\mathbf{v}_0)$, and the other proportional to the equilibrium distribution $W_M(\mathbf{v})$. Only the relative weight of these terms changed with time (the characteristic time was $1/\nu_{2j}$).

The results of Sec. 3 show that the relatively low collision frequencies are the most interesting. Therefore, we omit the rather awkward general formulas for the functions J_1 and J_2 in (3.8), and use only corrections of first order in μ and μ_j. Let us assume that

$$\mu_j/4\Gamma_j < \Gamma/k\bar{v} \ll 1. \quad (4.4)$$

It can then be shown that

$$J_1(\Omega) \cong \frac{1}{\pi}\int_0^\infty \exp\left[-\left(\frac{k\bar{v}}{2}\right)^2x^2 + i(\Omega-\Delta)x + \frac{i}{12}(k\bar{v})^2\mu''x^3\right]dx, \quad (4.5)$$

$$J_2(\Omega) \cong \frac{1}{2\sqrt{\pi}\,k\bar{v}}\int_0^\infty\left[\frac{1/\Gamma_m}{1+a_m^2x^2} + \frac{1/\Gamma_n}{1+a_n^2x^2}\right] \times \exp\left[-\Gamma x - \frac{(k\bar{v})^2}{24}\mu'x^3\right] \times\left\{\exp\left[-\left(\frac{\Omega-\Delta}{k\bar{v}}\right)^2\right] + \exp\left[i(\Omega-\Delta)x - \frac{i}{24}(k\bar{v})^2\mu''x^3\right]\right\}dx; \quad (4.6)$$

Here,

$$\mu = \mu' + i\mu'', \qquad a_j^2 = {}^1/_8(k\bar{v})^2\mu_j/\Gamma_j, \qquad j = m,\ n.$$

It is seen from (4.5) that $I_1(\Omega)$ has a width of the order of $k\bar{v}$; the function $I_2(\Omega)$ has two terms, as in the strong collision model: one has a width $k\bar{v}$ (as does the function Ω), and the other, represented by the second term in the braces in ((4.6), is much narrower than $k\bar{v}$ (this can be readily verified by estimating with the aid of (4.4) the region x, in which the coefficients of the first line in (4.6) are substantially different from zero). Thus, if (4.4) is satisfied, the contour $R(\Omega)$ will have a "sharp dip" against a fairly wide Doppler background in the model of (4.2), too.

It can be easily shown that the function $I_1(\Omega)$ has a maximum when

$$\Omega_{1\,max} = \Delta_1 + \Delta_2 - {}^1/_2\mu''. \quad (4.7)$$

It follows from (4.2) that $\mu'' \sim -\Delta_2$. Consequently, the term $\mu''/2$ leads to an additional shift of the maximum of $I_1(\Omega)$ in the same direction as Δ_2.

The "sharp term" in $I_2(\Omega)$ is an asymmetric function of the frequency. In fact, its center of gravity lies at $\Omega = \Delta = \Delta_1 + \Delta_2$, and its maximum is displaced from that point, owing to the term $\frac{1}{24} i(k\bar{v})^2 \mu'' x^3$ in the argument of the exponential function. For example,

$$\Omega_{2\,max} = \Delta_1 + \Delta_2 + \tfrac{1}{2}(k\bar{v}/\Gamma)^2 \mu'', \qquad (4.8)$$

if

$$\mu_j/\Gamma_j \ll (\Gamma/k\bar{v})^2, \qquad \mu'' \ll \Gamma(\Gamma/k\bar{v})^2. \qquad (4.9)$$

It follows from (4.7) and (4.8) that I_1 and I_2 reach a maximum value at different frequencies. In contradistinction to the strong collision model, the difference between $\Omega_{1\,max}$ and $\Omega_{2\,max}$ is basically due to the change in $\Omega_{2\,max}$, since $k\bar{v} \gg \Gamma$. It is significant that the term $\frac{1}{2}(k\bar{v}/\Gamma)^2 \mu''$ in (4.8) has a different sign than Δ_2 and, in general, may have a different sign than Δ_1. Therefore, it is not impossible to select conditions so as to render $\Omega_{2\,max} = 0$, i.e., so that the maximum of I_2 corresponds to the unperturbed transition frequency. This case is obviously of exceptionally great interest in the problem of stabilizing the oscillation frequency.

Let us consider $R(\Omega)$ for the case when the cubic terms can be neglected in the arguments of the exponential functions in (4.5) and (4.6):

$$\mu'' = 0, \qquad \mu'/\mu_j \ll \Gamma/\Gamma_j. \qquad (4.10)$$

Under this condition, I_2 can be represented by integral exponential functions of complex argument. Numerical computations carried out with the aid of tables [17] enabled us to approximate I_2 by dispersion curves having certain effective widths. As a result it was possible to represent $R(\Omega)$ as follows:

$$R(\Omega) = \frac{\sqrt{\pi}}{k\bar{v}} NG^2 \exp\left[-\left(\frac{\Omega-\Delta}{k\bar{v}}\right)^2\right]$$
$$\times\left\{1 - \frac{G^2}{4}\sum_{j=m,n}\frac{1}{\beta_j\Gamma_j}\left[1+\frac{\beta_j^2}{\beta_j^2+(\Omega-\Delta)^2}\right]\right\},$$
$$\beta_j = \Gamma\left\{1+\frac{3a_j^2/\Gamma^2}{(1+13a_j^2/\Gamma^2)^{1/2}}\right\},$$
$$a_j^2 = \frac{(k\bar{v})^2}{8}\frac{\mu_j}{\Gamma_j} \quad (j=m,n). \qquad (4.11)$$

The "sharp dip" is determined in this case by two terms which differ from each other when $\Gamma_m \neq \Gamma_n$.

The width of the "dip" is controlled by β_m and β_n, which depend on the parameters $k\bar{v}(\mu_j/\Gamma_j)^{1/2}$. The physical significance of these parameters is quite clear. According to (4.8), the dispersion of the atom velocity is

$$\sqrt{c} = \frac{\bar{v}}{\sqrt{2}}(1-e^{-2\mu_j\tau})^{1/2} \cong \bar{v}(\mu_j\tau)^{1/2} \quad (\mu_j\tau \ll 1). \qquad (4.12)$$

Consequently, $k\bar{v}(\mu_j | \Gamma_j)^{1/2}$ is the rms change in the Doppler shift due to the diffusion of the atoms during the time $\tau = 1/\Gamma_j$ when the atom occupies the levels $j = m, n$.

According to [9], weak collisions cause a marked change in I_1 only when $\mu' \sim k\bar{v}$. The relation in

$$\mu_j \sim (\Gamma/k\bar{v})^2\Gamma_j \ll \Gamma_j \ll k\bar{v}$$

the nonlinear term of $R(\Omega)$ is a sufficient condition to ensure a fully perceptible effect of the collisions. The difference is due to the following considerations. In the linear problem the role of collisions is reduced to the retardation of the displacement of the atom over a distance of the order of $\lambda/2\pi$; this will happen if the atom suffers a substantial velocity change in flight over a distance of $\lambda/2\pi$: $\lambda/2\pi\bar{v} = 1/k\bar{v} \sim 1/\mu_j$. In the nonlinear problem, on the other hand, the atoms have a nonequilibrium velocity distribution and the main effect of the collisions is to restore the equilibrium. The width of the nonequilibrium term is of the order of Γ/k and it is quite obvious that the collisions become effective if the order of the velocity dispersion $\bar{v}\sqrt{\mu_j/\Gamma_j}$ reaches Γ/k during the lifetimes Γ_m^{-1} and Γ_n^{-1}. On the other hand, conditions (4)4 signify that the velocity dispersion does not exceed $\bar{v}$, although it may be larger than Γ/k.

According to (3.16), strong collisions will be effective in our sense if $\nu \sim \Gamma$. The change in the characteristic parameter (compared with the weak collision model) is due to the fact that in the case treated in Sec. 3 the atom may change its velocity in each collision by a quantity $\bar{v}$, so that a relatively small fraction of the atoms will fall into the velocity interval Γ/k.

Only μ_m and μ_n depend on the concentration of perturbing particles in expression (4.11) for widths β_m and β_n. If μ_j is sufficiently small, we can retain only unity under the radical sign. In that case, however, the addition to Γ, due to collisions of the second kind and proportional to μ_j, will not exceed 10%. Therefore, an experimental detection of the effects of collisions of the second kind is possible only in the opposite limiting case:

$$\beta_j \cong \Gamma + \frac{3}{2\sqrt{26}} k\bar{v}\sqrt{\frac{\mu_j}{\Gamma_j}}, \quad \frac{k\bar{v}}{\Gamma}\sqrt{\frac{\mu_j}{\Gamma_j}} > 1, \quad j = m, n. \qquad (4.13)$$

Thus, in cases of practical interest the addition to Γ is proportional to the square root of the concentration of the perturbing particles.

Let us note that the "sharp" term in (4.11) comprises one-half of I_2 at the maximum point, $\Omega = \Delta$. The same ratio holds in the case of strong collisions (see (3.16)). However, if one cannot neglect the cubic terms in the integrand of (4.6), the "sharp term" can be easily shown to be less than $\frac{1}{2} I_{2\,max}$. This effect has been noted among other experimental results in [2]. Nevertheless, a different interpretation was given in [2].

Under the conditions reported in [2], the weak collision model should apparently be more appropriate. This is due to the fact that the main role in [2] was played by the elastic scattering of neon (atomic weight 20) by He^4. Given a mass ratio of 5 : 1, the persistence of velocities in the hard sphere model amounts to about 80%. [11] In other words, the velocity of the emitting atom (neon) is changed by the collisions by not more than 20% on the average; the comparatively small change is typical of the weak collision model.

5. GENERATION POWER AS A FUNCTION OF THE FREQUENCY

The power P of a laser can be determined by equating the output flux from the working volume to the power emitted in the active medium.[13] A simple derivation yields

$$P(\Omega) = B\frac{1}{I_2(\Omega)}\left[\eta - \frac{I_{1\,max}}{I_1(\Omega)}\right]. \quad (5.1)$$

Here η is pump excess over threshold for $\Omega = \Omega_{1max}$. Since we are interested in P as a function of Ω, the coefficient B, which is practically independent of Ω, will not be specified in detail.

Let us use (3.16) to make (5.1) more specific. Assuming that $\eta - 1 \ll 1$, we get

$$P(\Omega) \infty \left[\eta - 1 - \left(\frac{\Omega - \Delta_1 - 2\Delta_2}{k\bar{v}}\right)^2\right] \times \left[1 + \frac{(\Gamma_1 + \nu_2)^2}{(\Gamma_1 + \nu_2)^2 + (\Omega - \Delta_1)^2}\right]^{-1}. \quad (5.2)$$

Figure 2 illustrates (5.2); case (a) corresponds to $\Delta_2 = 0$, when $P(\Omega)$ is symmetrical with respect to $\Omega = \Delta_1$; in case (b) the contour of $P(\Omega)$ is assymmetric and its assymetry is due to a shift of the maxima of the functions $I_1(\Omega)$ and $I_2(\Omega)$ by $2\Delta_2$ (in Fig. 2, $\Delta_2 = -(\nu_2 + \Gamma_1)/4$).

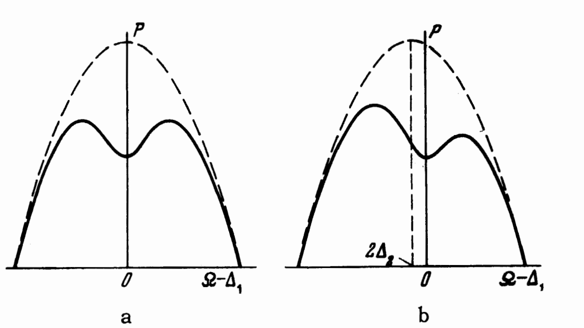

FIG. 2

In the strong collision model $P(\Omega)$ will also be asymmetric if $\Delta_2 \neq 0$. In contrast with (5.2), the asymmetry is due not only to the difference between $\Omega_{1\,max}$ and $\Omega_{2\,max}$, but also to the asymmetry of the function $I_2(\Omega)$ itself.

Thus, the impact theory provides a natural explanation of the main results of [1-3] if account is taken of the statistical dependence of the Doppler and interaction broadening. If the interpretation assumed in this work corresponds to reality (which, of course, requires a detailed experimental verification), there are interesting possibilities for the investigation of various elastic and inelastic processes accompanying atomic collisions. It is significant that the typical experimental conditions (density $\sim 10^{16}$ cm^{-3} and electron concentration $\sim 10^{11}$ cm^{-3}) are quite favorable for the identification of a comparatively small number of determining factors.

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