

EFFECT OF SELECTIVE COLLISIONS ON THE VELOCITY DISTRIBUTION OF ATOMS AND ON NONLINEAR INTERFERENCE EFFECTS

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The effect of elastic collisions on the velocity distribution of excited atoms located in an external electromagnetic field is investigated. A collision integral is employed which simultaneously takes into account strong collisions and small-angle scattering (selective scattering). It is shown that along with the familiar Bennett dip in the velocity distribution, there may exist a collision dip due to selective scattering. The dependence of its parameters on pressure differs from that for the Bennett dip. The shape of the Lamb dip and of the amplification line of a weak wave interacting with a strong field is analyzed for either the same transition or a shifted one. Collisions involving a change of velocity manifest themselves differently in two- and three-level systems.

1. INTRODUCTION

IN most papers on nonlinear spectroscopy of gases and on gas lasers, the experimental data are interpreted within the framework of the relaxation-constant model, in which no account is taken of the change in the velocity  $v$  of the atoms upon collision. An analysis of the influence of the scattering on the spectral characteristics of gas lasers is presented in<sup>[1]</sup>, where two semiphenomenological models of collisions (strong and weak) are considered. The model of strong collisions describes well the resonant exchange of excitation<sup>[2-4]</sup> and the dragging of resonant radiation<sup>[5,6]</sup>, and is adequate for a description of a number of experimental situations<sup>[5]</sup>. The situation is less clear with respect to the weak-collision model. On the one hand, both general considerations and an analysis of the collision integrals for concrete interaction potentials of the colliding particles<sup>[7]</sup> offer evidence that small changes of  $v$  should play no less a role than the randomization of the phase of the atomic oscillator (the Weisskopf broadening mechanism). On the other hand, there are practically no experimental investigations aimed at clarifying the role of weak collisions (with the exception of<sup>[8-10]</sup>), and the broadening of the spectral structures produced as a result of nonlinear phenomena is interpreted as a consequence of the phase randomization.

In the present paper we analyze effects of diffusion in velocity space, in the simultaneous presence of strong and selective collisions. To describe selective collisions, we use a model with a difference kernel with subsequent solution of the integral equation. Such an approach gives more general results than the "weak collision model" (the Fokker-Planck approximation), since it does not presuppose that the change in velocity has a diffusion character<sup>1)</sup>. Principal atten-

tion is paid to an analysis of the velocity distribution of the excited atoms; this analysis is the basis for the treatment of the nonlinear phenomena in gases. In sections 6-9 we consider the contour of the Lamb dip and of the spectral structure due to nonlinear interference effects (NIE). To simplify the problem and to highlight the role of the change of the velocity, we disregard level degeneracy.

We start from the equations for the density matrix  $\rho$ <sup>[1]</sup>:

$$\begin{aligned} \left(\frac{\partial}{\partial t} + v\nabla + \Gamma_j\right)\rho_{jj}(\mathbf{r}, \mathbf{v}, t) &= \pm 2 \operatorname{Re}[iV_{nm}(t, \mathbf{r})\rho_{mn}(\mathbf{r}, \mathbf{v}, t)] \\ &+ g_j + S_j \quad j = m, n; \\ \left(\frac{\partial}{\partial t} + v\nabla + \Gamma\right)\rho_{mn}(\mathbf{r}, \mathbf{v}, t) &= iV_{nm}(t, \mathbf{r})[\rho_{mm} - \rho_{nn}] + S; \\ S_j &= -v_j\rho_{jj}(\mathbf{r}, \mathbf{v}, t) + \int A_j(\mathbf{v}', \mathbf{v})\rho_{jj}(\mathbf{r}, \mathbf{v}', t) d\mathbf{v}'; \\ S &= -v\rho_{mn}(\mathbf{r}, \mathbf{v}, t) + \int A(\mathbf{v}', \mathbf{v})\rho_{mn}(\mathbf{r}, \mathbf{v}', t) d\mathbf{v}'; \\ V_{nm}(t, \mathbf{r}) &= \frac{d_{nm}}{2\hbar} E(t, \mathbf{r}). \end{aligned} \tag{1.1}$$

Here  $\mathbf{r}$  and  $\mathbf{v}$  are the coordinate and velocity of the atom,  $g_j$  is the number of acts of excitation of the state  $(j, \mathbf{v})$  per unit time;  $A_j(\mathbf{v}', \mathbf{v})$  and  $A(\mathbf{v}', \mathbf{v})$  are the kernels of the collision integrals  $S_j$  and  $S$ , from which the spontaneous relaxation is separated (the constants  $\Gamma_j$  and  $\Gamma$ ), and  $d_{nm}$  is the matrix element of the dipole moment. The plus and minus signs pertain to  $j = m$  and  $n$ , respectively. The field  $E(t, \mathbf{r})$  is of the form of a plane monochromatic wave (traveling or standing):

$$E(t, \mathbf{r}) = E \cos(\omega t - \mathbf{k}\mathbf{r}), \quad E(t, \mathbf{r}) = E \cos \omega t \cos \mathbf{k}\mathbf{r}. \tag{1.2}$$

we neglect throughout the "phase memory" effects ( $A(\mathbf{v}', \mathbf{v}) = 0$ ), as well as the spatial inhomogeneity in  $\rho_{jj}$ , due to saturation effects (in the case of a standing wave). In addition, we assume that  $g_j$  depends only on  $v$  in accordance with

$$g_j(v) = Q_j W(v), \quad W(v) = (\pi^{1/2} \bar{v})^{-3} \exp\{-v^2/\bar{v}^2\}, \tag{1.3}$$

$$\bar{v}^2 = 2k_B T / m.$$

<sup>1)</sup>To distinguish the considered model from the model proposed in [1], the term "weak collisions" is replaced by the term "selective collisions."

Under these conditions, the equations for  $\rho_{jj}(\mathbf{r}, \mathbf{v}, t) = \rho_{jj}(\mathbf{v})$  are

$$(\Gamma_j + \nu_j) \rho_{jj}(\mathbf{v}) - \int A_j(\mathbf{v}', \mathbf{v}) \rho_{jj}(\mathbf{v}') d\mathbf{v}' = Q_j W(\mathbf{v}) \mp 2G^2 x'(\mathbf{v}) [\rho_{mm} - \rho_{nn}], \quad (1.4)$$

and  $\rho_{mn}(\mathbf{r}, \mathbf{v}, t)$  and  $x'(\mathbf{v})$  are given by the following formulas: in the case of a traveling wave

$$\rho_{mn}(\mathbf{r}, \mathbf{v}, t) = iG[\rho_{mm}(\mathbf{v}) - \rho_{nn}(\mathbf{v})] x(\mathbf{v}) \exp\{-i(\Omega t - \mathbf{kr})\}, \quad (1.5)$$

$$x(\mathbf{v}) = x'(\mathbf{v}) + ix''(\mathbf{v}) = [\Gamma + \nu - i(\Omega - \mathbf{kv})]^{-1}; \quad G = d_{mn} E / 2\hbar,$$

and in the case of a standing wave

$$\rho_{mn}(\mathbf{r}, \mathbf{v}, t) = iG[\rho_{mm} - \rho_{nn}]^{1/2} [x_+(\mathbf{v}) e^{i\mathbf{kr}} + x_-(\mathbf{v}) e^{-i\mathbf{kr}}] e^{-i\Omega t}; \quad (1.6)$$

$$x(\mathbf{v}) = 1/2[x_+(\mathbf{v}) + x_-(\mathbf{v})], \quad x_{\pm}(\mathbf{v}) = [\Gamma + \nu - i(\Omega \mp \mathbf{kv})]^{-1}.$$

Thus, an investigation of the role of the diffusion of the atom in velocity space reduces to a solution of the system of integral equations (1.4) for  $\rho_{mm}(\mathbf{v})$  and  $\rho_{nn}(\mathbf{v})$ . In the strong-collision model, the system (1.4) admits of an exact solution for any field intensity<sup>[11]</sup>.

For arbitrary kernels  $A_j(\mathbf{v}', \mathbf{v})$  it is necessary to expand in powers of  $G^2$  and it is convenient to deal (see<sup>[11]</sup>) with Green's functions, which we define as solutions of the equations

$$F_j(\mathbf{v}', \mathbf{v}) = (\Gamma_j + \nu_j)^{-1} \int A_j(\mathbf{v}'', \mathbf{v}) F_j(\mathbf{v}', \mathbf{v}'') d\mathbf{v}'' + \delta(\mathbf{v} - \mathbf{v}'). \quad (1.7)$$

The function  $F_j(\mathbf{v}', \mathbf{v})$  is obviously the stationary distribution with respect to  $\mathbf{v}$  for the atoms at the level  $j$  under the influence of a source  $\delta(\mathbf{v} - \mathbf{v}')$ . If  $F_j$  is known, then the solution of (1.4) can be found in the form

$$\rho_{jj}(\mathbf{v}) = \sum_{l=0}^{\infty} G^{2l} \rho_j^{(l)}(\mathbf{v}), \quad \rho_j^{(0)}(\mathbf{v}) = N_j W(\mathbf{v}), \quad (1.8)$$

$$\rho_j^{(l)}(\mathbf{v}) = \frac{2}{\Gamma_j + \nu_j} \int x'(\mathbf{v}') [\rho_m^{(l-1)}(\mathbf{v}') - \rho_n^{(l-1)}(\mathbf{v}')] F_j(\mathbf{v}', \mathbf{v}) d\mathbf{v}',$$

where  $N_j$  is the unsaturated population of the level  $j$ .

## 2. GREEN'S FUNCTION OF EQUATIONS (1.4)

It will be convenient in what follows to represent the kernel  $A_j(\mathbf{v}', \mathbf{v})$  in the form

$$A_j(\mathbf{v}', \mathbf{v}) = \bar{\nu}_j B_j(\mathbf{v}', \mathbf{v}), \quad \bar{\nu}_j = \int A_j(\mathbf{v}', \mathbf{v}') d\mathbf{v}', \quad \int B_j(\mathbf{v}', \mathbf{v}) d\mathbf{v} = 1, \quad (2.1)$$

where  $\bar{\nu}_j$  has the meaning of the "arrival frequency." We assume first that  $\bar{\nu}_j$  is independent of  $\mathbf{v}$ . Then the iteration method yields

$$F_j(\mathbf{v}', \mathbf{v}) = \delta(\mathbf{v}' - \mathbf{v}) + f_j(\mathbf{v}', \mathbf{v}), \quad f_j(\mathbf{v}', \mathbf{v}) = \sum_{l=1}^{\infty} [\bar{\nu}_j / (\Gamma_j + \nu_j)]^l B_j^{(l)}(\mathbf{v}', \mathbf{v}), \quad (2.2)$$

$$B_j^{(l)}(\mathbf{v}', \mathbf{v}) = \int B_j(\mathbf{v}', \mathbf{v}_1) B_j^{(l-1)}(\mathbf{v}_1, \mathbf{v}) d\mathbf{v}_1, \quad B_j^{(1)} = B_j, \quad \int B_j^{(l)}(\mathbf{v}', \mathbf{v}) d\mathbf{v} = 1.$$

The function  $F_j(\mathbf{v}', \mathbf{v})$  contains two parts:  $\delta(\mathbf{v}' - \mathbf{v})$  describes those atoms which experience no collisions during the lifetime at the level  $j$ ; the regular part  $f_j(\mathbf{v}', \mathbf{v})$  characterizes the diffusion in velocity space. The quantities  $B^{(l)}(\mathbf{v}', \mathbf{v})$  in (2.2) are the probabilities of the velocity change  $\mathbf{v}' \rightarrow \mathbf{v}$  as a result of  $l$  collisions. From the expression

$$\int F_j(\mathbf{v}', \mathbf{v}) d\mathbf{v} = 1 + \sum_{l=1}^{\infty} \left[ \frac{\bar{\nu}_j}{\Gamma_j + \nu_j} \right]^l = 1 + n_j, \quad (2.3)$$

$$n_j = \bar{\nu}_j / \tilde{\Gamma}_j, \quad \tilde{\Gamma}_j = \Gamma_j + \nu_j - \bar{\nu}_j,$$

we see that the effective number of terms in the series is determined by the parameter  $n_j$ , which is the average number of collisions during the total lifetime  $\tilde{\Gamma}_j^{-1}$ . If  $n_j \ll 1$ , then  $\bar{\nu}_j / (\Gamma_j + \nu_j) \ll 1$ , and we can confine ourselves to the first corrections for the collision effects:

$$F_j(\mathbf{v}', \mathbf{v}) = \delta(\mathbf{v}' - \mathbf{v}) + \frac{\bar{\nu}_j}{\Gamma_j + \nu_j} B_j(\mathbf{v}', \mathbf{v}), \quad (2.4)$$

and the field effects can be easily taken into account<sup>2)</sup> in any approximation in  $G^2$ :

$$\rho_{jj}(\mathbf{v}) = N_j W(\mathbf{v}) \mp \frac{2G^2 N}{\Gamma_j + \nu_j} \left\{ \frac{x'(\mathbf{v}) W(\mathbf{v})}{1 + 2G^2 \tau_1 x'(\mathbf{v})} + \frac{\bar{\nu}_j}{\Gamma_j + \nu_j} \int \frac{x'(\mathbf{v}') W(\mathbf{v}') B_j(\mathbf{v}', \mathbf{v})}{1 + 2G^2 \tau_1 x'(\mathbf{v}')} d\mathbf{v}' \right\}, \quad (2.5)$$

$$\tau_1 = \frac{1}{\Gamma_m + \nu_m} + \frac{1}{\Gamma_n + \nu_n}, \quad n_j \ll 1, \quad N = N_m - N_n.$$

This case, however, is of no interest from the point of view of investigating collisions, since the second term in the curly brackets, which is due to collisions, is always small in comparison with the first, which describes the usual Bennett dip. In the opposite limiting case,  $n_j \gg 1$ , collisions are experienced by a major fraction  $n_j / (1 + n_j)$  of the atoms and diffusion effects are easily noticeable. Many terms of the series of (2.2) are significant here, the series is not convenient for use, and it is desirable to use a convolution that calls for special assumptions concerning the kernel  $B_j(\mathbf{v}', \mathbf{v})$ .

## 3. BASIC MODEL

We assume that the kernel  $B_j(\mathbf{v}', \mathbf{v})$  consists of two parts: the first (selective) part has a width  $\sigma_j \ll \bar{\nu}_j$ ; the second part corresponds to the strong-collision model

$$A_j(\mathbf{v}', \mathbf{v}) = \bar{\nu}_{1j} B_{1j}(\mathbf{v}', \mathbf{v}) + \bar{\nu}_{2j} W(\mathbf{v}), \quad \bar{\nu}_j = \bar{\nu}_{1j} + \bar{\nu}_{2j}. \quad (3.1)$$

The choice of the kernel in the form (3.1) is justified by the results of<sup>[7]</sup>, where  $A_j(\mathbf{v}', \mathbf{v})$  are considered for a number of concrete particle interaction potentials. In addition, (3.1) is also of interest for spectroscopic applications.

The Green's function for the kernel (3.1) is given by the formula

$$F_j(\mathbf{v}', \mathbf{v}) = \delta(\mathbf{v}' - \mathbf{v}) + \sum_{l=1}^{\infty} \left[ \frac{n_{1j}}{1 + n_{1j}} \right]^l B_{1j}^{(l)}(\mathbf{v}', \mathbf{v}) + n_{2j} W(\mathbf{v}), \quad (3.2)$$

$$n_{1j} + n_{2j} = n_j;$$

$$\int F_j(\mathbf{v}', \mathbf{v}) d\mathbf{v} = 1 + n_{1j} + n_{2j}, \quad n_{1j} = \frac{\bar{\nu}_{1j}}{\tilde{\Gamma}_{1j}}, \quad n_{2j} = \frac{\bar{\nu}_{2j}}{\tilde{\Gamma}_j} \frac{\Gamma_j + \nu_j}{\tilde{\Gamma}_{1j}},$$

$$\tilde{\Gamma}_{1j} = \Gamma_j + \nu_j - \bar{\nu}_{1j}.$$

According to (3.2), the average number of selective states  $n_{1j}$  is determined not by the total lifetime  $\tilde{\Gamma}_j^{-1}$ , but by the lifetime  $\tilde{\Gamma}_{1j}^{-1}$  with respect to spontaneous decay, quenching, and strong collisions. In other words, strong collisions serve as quenching collisions relative to the selective ones, and decrease  $n_{1j}$ . In the limiting case of high pressures and small quenching we

<sup>2)</sup>In practice this is conveniently done with the aid of (1.4), taking the integral terms into account in the first approximation (in  $\bar{\nu}_j / (\Gamma_j + \nu_j)$ ).

have  $n_{ij} \sim \tilde{v}_{ij}/\tilde{v}_{2j}$ , and this is the maximum value of  $n_{ij}$ . According to [7], we can have  $n_{ij} \sim 10$  for a Lenard-Jones potential. The presence in (3.2) of the equilibrium component  $n_{2j}W(v)$  indicates that such a component should be present also in  $\rho_{jj}(v)$ . It is therefore convenient to seek  $\rho_{jj}(v)$  in the form

$$\rho_{jj}(v) = R_j W(v) + \rho_j(v), \quad (3.3)$$

and from (1.4) we get for  $\rho_j(v)$  and  $R_j$  the system of equations

$$R_j = N_j + \frac{\tilde{v}_{2j}}{\tilde{\Gamma}_j} \int \rho_j(v) dv, \quad (3.4)$$

$$\rho_j(v) = \frac{\tilde{v}_{1j}}{\Gamma_j + v_j} \int B_{ij}(v', v) \rho_j(v') dv' \mp \frac{2G^2 x'(v)}{\Gamma_j + v_j} [(R_m - R_n)W(v) + \rho_m(v) - \rho_n(v)].$$

If the condition

$$G^2/\tilde{\Gamma}_j(\Gamma + v) \ll 1 \quad (3.5)$$

is satisfied then the selective part of  $\rho_j(v)$  is small, and (3.4) yields

$$R_j = N_j \mp \frac{2G^2 \tau_j^{(2)} XN}{1 + 2G^2 \tau_j X}, \quad X = \int x'(v) W(v) dv, \quad N = N_m - N_n; \quad (3.6)$$

$$\rho_j(v) = \frac{n_{1j}}{1 + n_{1j}} \int B_{ij}(v', v) \rho_j(v') dv' \mp \frac{2G^2 N W(v)}{1 + 2G^2 \tau_j X} \frac{x'(v)}{\Gamma_j + v_j};$$

$$\tau_j = \tau_m^{(2)} + \tau_n^{(2)}, \quad \tau_j^{(2)} = \frac{n_{2j}}{\Gamma_j + v_j} = \frac{\tilde{v}_{2j}}{\tilde{\Gamma}_j \tilde{\Gamma}_{1j}} = \frac{1}{\tilde{\Gamma}_j} - \frac{1}{\tilde{\Gamma}_{1j}}.$$

The equilibrium part  $R_j W(v)$  in  $\rho_{jj}(v)$  is of the same form as in the case of the pure strong-collision model<sup>[11]</sup>. Equation (3.6) for  $\rho_j(v)$  contains only the selective part of the kernel, and the "source" is the usual Bennett dip decreased by the "homogeneous" saturation<sup>3)</sup>.

We note that  $x'(v)$  contains only the projection of  $v$  on  $k$ . The selectiveness of the kernel  $B_{ij}(v', v)$  and the constancy of  $\tilde{v}_{1j}$  and  $\tilde{v}_{2j}$  enable us to change over from (3.6) to the one-dimensional problem:

$$\rho_j(v) = r_j(v) W(v_{\perp}), \quad v = v_{\perp} + kv/k, \quad (3.7)$$

$$W(v_{\perp}) = (\pi \bar{v}^2)^{-1} \exp\{-v_{\perp}^2/\bar{v}^2\};$$

$$r_j(v) = \frac{n_{1j}}{1 + n_{1j}} \int B_{ij}(v', v) r_j(v') dv' + y_j(v);$$

$$B_{ij}(v', v) = \int B_{ij}(v', v) W(v_{\perp}') dv_{\perp}';$$

$$y_j(v) = \mp \frac{x'(v)}{\Gamma_j + v_j} \frac{2G^2 N W(v)}{1 + 2G^2 \tau_j X}.$$

In the next section we analyze the Green's function  $F_j(v', v)$  of Eq. (3.7), with the aid of which we can calculate  $r_j(v)$ :

#### 4. GREEN'S FUNCTION OF ONE-DIMENSIONAL PROBLEM

The fact that  $B_{ij}(v', v)$  does not have the singularity possessed by the three-dimensional kernel<sup>[7]</sup> enables us to operate with the lucid concept of the width of the kernel. We consider the model kernel (see<sup>[12,7]</sup>)

$$B_{ij}(v', v) = [\pi^{1/2} \bar{v} \sqrt{1 - \gamma^2}]^{-1} \exp\left\{-\frac{(v - \gamma v')^2}{(1 - \gamma^2) \bar{v}^2}\right\}, \quad (4.1)$$

$$B_{ij}^{(0)}(v', v) = [\sqrt{\pi} \bar{v} \sqrt{1 - \gamma^2}]^{-1} \exp\left\{-\frac{(v - \gamma^l v')^2}{(1 - \gamma^{2l}) \bar{v}^2}\right\}.$$

<sup>3)</sup>In spite of the condition (3.5), the term  $2G^2 \tau_j X$  may not be small if  $(\Gamma + v)X\tilde{v}_{2j}/\tilde{\Gamma}_j \sim (\Gamma + v)\tilde{v}_{2j}/\tilde{\Gamma}_j k\bar{v} \sim 1$ .

The parameter  $\gamma$  characterizes both the width  $\sigma_j = \bar{v}\sqrt{1 - \gamma^2}$  of the kernel, and the "memory" of the velocity  $v'$  prior to collision. The kernels  $B_{ij}^{(l)}(v', v)$  have the same structure, but their widths increase with increasing  $l$ , while the "memory" decreases ( $\gamma^2 < 1$ ). In the case  $n_{1j} \gg 1$  of interest to us, the series in (3.2) can be replaced by the integral

$$F_j(v', v) = \delta(v' - v) + \frac{1}{\pi^{1/2} \bar{v}} \int_0^{\infty} \exp\left\{-\frac{l}{L_j} - \frac{(v - \gamma^l v')^2}{(1 - \gamma^{2l}) \bar{v}^2}\right\} \frac{dl}{\sqrt{1 - \gamma^{2l}}}$$

$$= \delta(v' - v) + \frac{n_{1j}}{\pi^{1/2} \bar{v}} \int_0^{\infty} \exp\left\{-\tilde{\Gamma}_j \tau - \frac{[v - \exp(-\mu_j \tau) v']^2}{[1 - \exp(-2\mu_j \tau)] \bar{v}^2}\right\} \frac{\tilde{\Gamma}_j d\tau}{[1 - \exp(-2\mu_j \tau)]^{1/2}}, \quad (4.2)$$

$$\tilde{\Gamma}_j = \Gamma_j + v_j - \bar{v}_{1j}, \quad \mu_j = \Gamma_j L_j \ln [1 - \sigma_j^2/\bar{v}^2]^{-1/2},$$

$$L_j = n_{1j} + 1/2 - 1/12n_{1j} + \dots$$

The integral with respect to  $\tau$  in (4.2) is the Green's function for the collision integral in the diffusion approximation

$$S_j = -(v_{1j} - \bar{v}_{1j}) F_j + \mu_j \left\{ \frac{\partial}{\partial v} [v F_j] + \frac{\bar{v}^2}{2} \frac{\partial^2}{\partial v^2} F_j \right\}.$$

In (4.2),  $\delta(v' - v)$  describes the atoms that have avoided elastic collisions. The fraction of these atoms is  $(1 + n_{1j})^{-1}$ , but the  $\delta$ -function cannot be discarded even when  $n_{1j} \gg 1$ , since it can lead to a much more abrupt structure in  $r_j(v)$  than  $f_j(v', v)$ .

Formula (4.2) enables us to trace the transition from the selective scattering to the model of strong collisions. If

$$\mu_j/\tilde{\Gamma}_j = L_j \ln [1 - \sigma_j^2/\bar{v}^2]^{-1/2} \cong 1/2 n_{1j} \sigma_j^2/\bar{v}^2 \ll 1, \quad (4.3)$$

$$n_{1j} \gg 1, \quad \sigma_j^2/\bar{v}^2 \ll 1,$$

then we can obtain from (4.2)

$$F_j(v', v) = \delta(v' - v) + \frac{n_{1j}}{\sigma_j \sqrt{1 + n_{1j}}} \exp\left\{-2 \frac{|v - v'|}{\sigma_j \sqrt{1 + n_{1j}}} - \frac{v^2 - v'^2}{2\bar{v}^2}\right\}. \quad (4.4)$$

The second term in (4.4) describes a slightly asymmetrical (owing to the factor  $\exp[-(v^2 - v'^2)/2\bar{v}^2]$ ) and relatively narrow distribution with characteristic width  $\sigma_j \sqrt{1 + n_{1j}} \ll \bar{v}$ . Owing to the introduction of unity in  $\sqrt{1 + n_{1j}}$ , formula (4.4) has the correct value also in the limit  $n_{1j} \ll 1$ , i.e., it can serve as a good interpolation also when  $n_{1j} \sim 1$ . If  $\mu_j = 2\tilde{\Gamma}_j$ , then formula (4.2) can be recast in the form

$$F_j(v', v) = \delta(v' - v) + \frac{n_{1j}}{\pi^{1/2} \bar{v}} \int_0^1 \exp\left\{-\frac{(v - z^2 v')^2}{(1 - z^2) \bar{v}^2}\right\} \frac{dz}{\sqrt{1 - z^2}}. \quad (4.5)$$

The integral in (4.5), which is considered in<sup>[5]</sup> (it describes the kernel of the collision integral owing to the dragging of the radiation), differs little from a Maxwellian distribution. In the limit  $\mu_j \gg \tilde{\Gamma}_j$  we get from (4.2)

$$F_j(v', v) = \delta(v' - v) + n_{1j} W(v). \quad (4.6)$$

Thus, the strong-collision model is applicable for

$$\frac{\mu_j}{\tilde{\Gamma}_j} = 1/2 \ln \left[1 - \frac{\sigma_j^2}{\bar{v}^2}\right] / \ln \left[\frac{n_{1j}}{1 + n_{1j}}\right] \gg 1. \quad (4.7)$$

This condition is satisfied either for  $\sigma_j \sim \bar{v}$  (which is trivial) or for a sufficiently large number of collisions  $n_{1j}$ , even if  $\sigma_j \ll \bar{v}$ .

According to<sup>[7]</sup>, when  $\sigma_j \ll \bar{v}$  the kernel  $B_{ij}(v', v)$  depends on  $v - v'$  ("difference kernel"). If the follow-

ing more stringent condition is satisfied (the dispersion of the velocity due to the selective collisions is much smaller than  $\bar{v}^2$ )

$$(1 + n_{ij})\sigma_j^2 \ll \bar{v}^2, \quad (4.8)$$

then  $F_j(v', v)$  is also a difference kernel and is determined by the quadrature

$$F_j(v' - v) = \delta(v' - v) + f_j(v' - v), \quad (4.9)$$

$$f_j(\eta) = \frac{n_{ij}}{2\pi} \int_{-\infty}^{\infty} \frac{B_{ij}(\kappa) e^{-i\eta\kappa}}{1 + n_{ij}[1 - B_{ij}(\kappa)]} d\kappa;$$

$$B_{ij}(\kappa) = \int_{-\infty}^{\infty} B_{ij}(\eta) e^{i\eta\kappa} d\eta, \quad n_{ij} = \frac{v_{ij}}{\Gamma_{ij}} = \frac{\bar{v}_{ij}}{\Gamma_j + v_j - \bar{v}_{ij}}.$$

In the case  $n_{ij} \gg 1$  we can obtain useful asymptotic formulas from (4.9). In this connection, an important role is played by the behavior of  $B_{ij}(\kappa)$  at small  $\kappa$ :

$$B_{ij}(\kappa) = 1 - \alpha_j \sigma_j |\kappa| - \beta_j \sigma_j^2 \kappa^2 + \dots, \quad (4.10)$$

where  $\sigma_j$  is the width of the kernel  $B_{ij}(\eta)$ , and the numerical factors  $\alpha_j$  and  $\beta_j$  depend on the form of the kernel and on the behavior of  $B_{ij}(\eta)$  as  $\eta \rightarrow \infty$ . Obviously,  $\alpha_j$  is positive, and  $\beta_j$  can have an arbitrary sign. Either the linear or the quadratic term can be decisive in the expansion (4.10). For example, for the kernel<sup>[8]</sup>

$$B_{ij}(\eta) = \pi^{-1} \sigma_j (\sigma_j^2 + \eta^2)^{-1}, \quad B_{ij}(\kappa) = \exp\{-\sigma_j |\kappa|\}, \quad (4.11)$$

$\alpha_j = 1$  and the linear term is the principal one. Therefore the width of the function  $f_j(\eta)$  becomes proportional to  $n_{ij}$ . On the other hand, if  $B_{ij}(\eta)$  decreases more rapidly than  $\eta^{-2}$  as  $\eta \rightarrow \infty$ , then  $\alpha_j = 0$ ,  $\beta_j > 0$ , and

$$f_j(\eta) = \frac{n_{ij}}{2\pi} \int_{-\infty}^{\infty} \frac{e^{-i\eta\kappa} d\kappa}{1 + \beta_j \sigma_j^2 (1 + n_{ij}) \kappa^2} = \frac{n_{ij}}{2\sigma_j \sqrt{\beta_j (1 + n_{ij})}} \times \exp\left\{-\frac{|\eta|}{\sigma_j \sqrt{\beta_j} \sqrt{1 + n_{ij}}}\right\}. \quad (4.12)$$

The width of the function  $f_j(\eta)$  varies in this case in proportion to  $\sqrt{1 + n_{ij}}$ . Which of the noted situations does actually take place is a very important question, and should be answered by experiment. We confine ourselves below to an examination of some of them. Its attraction lies in the fact that there is agreement with the law of large numbers (when  $n_{ij}$  is large the width of the distribution is proportional to  $\sqrt{n_{ij}}$ ), and in addition, (4.12) "joins" well the Green's function for the weak-collision model.

The asymptotic formula (4.12) differs from (4.4) only in the factor  $\sqrt{\beta_j}$ , the possible values of which are clear from the following figures:

$B_{ij}(\eta)$ :	$\frac{1}{2\sigma_j} \exp\left\{-\frac{ \eta }{\sigma_j}\right\}$	$\frac{1}{\sqrt{\pi}\sigma_j} \exp\left\{-\frac{\eta^2}{\sigma_j^2}\right\}$	triangular shape	rectangular shape
$\sqrt{\beta_j}$ :	1	0.500	0.457	0.408

Thus,  $\sqrt{\beta_j}$  changes, but not more than by a factor of two. We notice that (4.12) is an exact Green's function for an exponential kernel. We shall henceforth put  $\beta_j = 1$ , meaning an exponential kernel or else inclusion of  $\sqrt{\beta_j}$  in  $\sigma_j$ .

## 5. DISTRIBUTION OF POPULATIONS BY VELOCITIES

With the aid of the Green's function (4.12) we express the population  $\rho_{jj}(v)$  of the level  $j$  in terms of tabulated functions:

$$\rho_{jj}(v) = W(v) \left\{ N_j \mp \frac{2G^2 N}{1 + 2G^2 \tau_2 X} [\tau_j^{(2)} X + \tau_j x'(v) + \tau_j^{(1)} Y_j(v)] \right\},$$

$$Y_j(v) = \int_{-\infty}^{\infty} \frac{x'(v') dv'}{2\sigma_j \sqrt{1 + n_{ij}}} \exp\left\{-\frac{|v - v'|}{\sigma_j \sqrt{1 + n_{ij}}}\right\},$$

$$\tau_{ij} = \frac{1}{\Gamma_j + v_j}, \quad \tau_j^{(1)} = \frac{\bar{v}_{ij}}{(\Gamma_j + v_j) \Gamma_{ij}}. \quad (5.1)$$

For concreteness, we choose a traveling wave, so that

$$k\sigma_j \sqrt{1 + n_{ij}} Y_j(v) = \text{Re}\{ci(z_j) \sin z_j - si(z_j) \cos z_j\}, \quad (5.2)$$

$$z_j = z_j' + iz_j'' = \frac{\Gamma + v + i(\Omega - kv)}{k\sigma_j \sqrt{1 + n_{ij}}}.$$

Here  $si(z_j)$  and  $ci(z_j)$  are the integral sine and cosine functions.

The structure of the distribution (5.5) is the following (see Fig. 1). The usual Bennett dip (peak) (the term  $\tau_{ij} x'(v)$  and "collision dip" (peak) described by the function  $Y_j(v)$  are present on the Maxwellian curve, which is decreased (or increased) as the result of the "homogeneous" saturation band (the term proportional to  $X$ ). They are "tied in" with the resonant velocity  $v = \Omega/k$ ; the integrals of the three indicated terms with respect to  $v$  are related as

$$\tau_{ij} : \tau_j^{(1)} : \tau_j^{(2)}, \quad \tau_{ij} + \tau_j^{(1)}$$

$$+ \frac{\tau_j^{(2)}}{\tau_j^{(1)}} = \left[ \frac{\Gamma_j + v_j - \bar{v}_{ij}}{-\bar{v}_{ij}} \right]^{-1} = \bar{\Gamma}_{ij}^{-1}.$$

$\tau_{ij}$  is the lifetime with respect to spontaneous decay, quenching, and collisions of any kind;  $\tau_j^{(1)}$  is the lifetime after the first selective collision up to the instant when either spontaneous decay, or quenching, or strong collision takes place;  $\tau_j^{(2)}$  is the lifetime at the level  $j$  after the first strong collision. Thus, the total lifetime of the atom at the level  $j$  breaks up effectively into three intervals, each corresponding to a distinct character of the interaction with the field. During the time  $\tau_{ij}$ , the atom velocity remains unchanged, and the distribution with respect to the velocities retains the non-equilibrium structure (the Bennett dip) produced by the field. During the time  $\tau_j^{(1)}$ , only selective collisions take place; the indicated structure becomes "smeared" but remains non-equilibrium ("collision dip"). As soon as the strong collision takes place, the atom acquires an equilibrium velocity distribution and the distribution remains the same, no matter whether strong or selective collisions follow. Consequently, during the time  $\tau_j^{(2)}$  the atom interacts with the field via the "homogeneous" saturation channel. The interaction via each channel is stronger the longer the corresponding time. It is therefore also clear that strong collisions act as quenching collisions with respect to the selective ones (an increase of  $\nu_{2j}$  decreases  $\tau_j^{(1)}$ ).

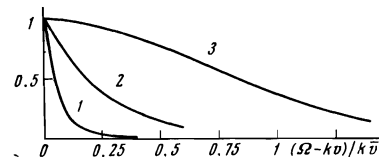


FIG. 1. Nonlinear increment to the velocity distribution at the level  $m$ : 1—Bennett dip, 2—collision dip, 3—homogeneous saturation "band". The relation between the parameters is  $\Gamma_{mn} = 0.2k\sigma_m \sqrt{1 + n_{mn}} = 0.05 k\bar{v}$ . The plots are normalized to unity at the maximum.

To determine the width and the amplitude of the "collision dip," let us analyze the function  $Y_j(v)$ . We can readily see two limiting cases:

$$Y_j(v) = x'(v) - 2[k\sigma_j\sqrt{1+n_{ij}}x'(v)]^2[4x'(v) - 3/(\Gamma + \nu)], \tag{5.3}$$

$$Y_j(v) = \frac{\pi}{2k\sigma_j\sqrt{1+n_{ij}}} \exp\left\{-\frac{|\Omega - kv|}{k\sigma_j\sqrt{1+n_{ij}}}\right\}, \tag{5.4}$$

In the first case, the "collision dip" is a Bennett dip slightly broadened by collisions. It has a near-dispersion form with width  $(\Gamma + \nu)\{1 + 3[k\sigma_j\sqrt{1+n_{ij}}/(\Gamma + \nu)]^2\}$ . In the approximation (5.4), the influence of impact broadening is practically excluded; the width of the dip  $\sigma_j\sqrt{1+n_{ij}}$  is proportional here to the width of the kernel of the collision integral. The ratio of the amplitudes of the "collision dip" to the Bennett dip in the approximation (5.4) is

$$\frac{\pi}{2} \frac{\Gamma + \nu}{k\sigma_j\sqrt{1+n_{ij}}} n_{ij}, \tag{5.5}$$

i.e., it can be sufficiently large if  $n_{ij} \gg 1$ .

A change of the ratio of the parameters  $\Gamma + \nu$  and  $k\sigma_j\sqrt{1+n_{ij}}$  changes the dispersion form of  $Y_j(v)$  into an exponential one. It will be convenient in what follows to approximate  $Y_j(v)$  by a dispersion curve with minimal errors in the area, half-width at half-height, or in the amplitude. We can use for this purpose the function

$$Y_j(v) = \frac{\delta_j}{\delta_j^2 + (\Omega - kv)^2}, \quad \delta_j = \Gamma + \nu + \frac{3k\sigma_j\sqrt{1+n_{ij}}}{\mathcal{A}}, \tag{5.6}$$

$$\mathcal{A} = \{A_j^2 + [(\Gamma + \nu)/k\sigma_j\sqrt{1+n_{ij}}]^2\}^{1/2}.$$

The constant  $A_j$  ranges from  $\sqrt{12}$  to  $3/\ln 2$ , and must be chosen by calculating the minimum error depending on the range of variation of the parameter  $(\Gamma + \nu)/k\sigma_j\sqrt{1+n_{ij}}$ . For example, if  $A_j = 4$ , then the error in the determination of the width does not exceed 9% anywhere, and the error in the amplitude does not exceed 15%.

Let us consider the variation of the parameters of the "collision dip" with pressure. Expression (5.6) for  $\delta_j$  contains two pressure-dependent quantities: the linear function  $\Gamma + \nu$ , and  $n_{ij} = \tilde{\nu}_{ij}/(\Gamma_j + \tilde{\nu}_{2j})$ , which has the form of a curve with saturation. At low pressures, so long as the spontaneous relaxation constant prevails over the frequencies of the quenching and strong collisions,  $n_{ij}$  varies linearly with pressure:  $n_{ij} \cong \tilde{\nu}_{ij}/\Gamma_j$ . At high pressures,  $n_{ij}$  tends to a constant value. The large number of parameters contained in  $\delta_j$  leads to a great variety of plots of  $\delta_j$  against pressure. We shall stop to discuss the most general properties of the plots of  $\delta_j$  and present examples for certain important particular cases.

The plot of  $\delta_j$  always lies above the straight line  $\Gamma + \nu$  characterizing the width of the Bennett dip. This is perfectly clear, since collisions with change of velocity can only broaden the Bennett dip. In the limiting case of low pressures

$$\delta_j = \Gamma + 3k\sigma_j \times [A_j^2 + (\Gamma/k\sigma_j)^2]^{-1/2}, \quad v \ll \Gamma, n_{ij} \ll 1, \tag{5.7}$$

which can be much larger than  $\Gamma$ . At high pressures, the  $\delta_j$  approaches asymptotically the straight line  $\Gamma + \nu$ :

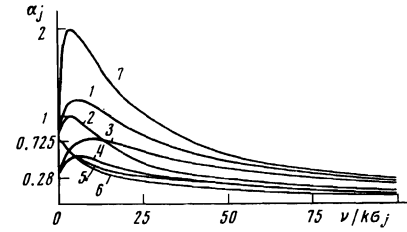


FIG. 2. Plots of the quantity  $\alpha_j = [\delta_j - \Gamma - \nu]/k\sigma_j$ , which characterizes the difference between the widths of the "collision dip" and the Bennett dip:  $1 - \eta_j \equiv (\nu_j - \nu_{ij})/\nu_j = 0.1, a_j \equiv \nu_j k\sigma_j/\Gamma_j \nu = 1, \gamma_j \equiv \Gamma/k\sigma_j = 1; 2 - \eta_j = 0.2, a_j = 1, \gamma_j = 1; 3 - \eta_j = 0.1, a_j = 1, \gamma_j = 10; 4 - \eta_j = 0.2, a_j = 1, \gamma_j = 10, 5 - \eta_j = 0.1, a_j = 0.1, \gamma_j = 1; 6 - \eta_j = 0.2, a_j = 0.1, \gamma_j = 1; 7 - \eta_j = 0.1, a_j = 10, \gamma_j = 1$ .

$$\delta_j = \Gamma + \nu + 3(k\sigma_j\sqrt{1+n_{ij}})^2 \times (\Gamma + \nu)^{-1}, \quad \Gamma + \nu \gg k\sigma_j\sqrt{1+n_{ij}}. \tag{5.8}$$

This result is the consequence of the fact that the Green's function has more rapidly decreasing "wings" than the dispersion curve: above a certain pressure, the width of the Green's function becomes less than the Bennett dip, and the width of the convolution approaches the width of the latter. The "collision dip" cannot be separated from the Bennett dip on the asymptotic plot of (5.8). This asymptotic plot is reached quite smoothly, so that in a limited range of pressures the plot of  $\delta_j$  is a straight line. Thus, at medium pressures we have

$$\delta_j = \Gamma + \nu + 3k\sigma_j\sqrt{1+n_{ij}}/A_j, \quad \Gamma + \nu < A_j k\sigma_j\sqrt{1+n_{ij}}. \tag{5.9}$$

If at the same time  $n_{ij} = \text{const}$ , then (5.9) is a straight line passing at a distance  $3A_j^{-1}k\sigma_j\sqrt{1+n_{ij}}$  above  $\Gamma + \nu$ . Its point of intersection with the ordinate axis can lie much higher than the initial point (5.7). Finally, the maximum deviation of the  $\delta_j$  curve from  $\Gamma + \nu$ , as seen from (5.6) and (5.9), is larger the larger  $k\sigma_j\sqrt{1+n_{ij}}$ . Figure 2 shows several examples of the  $\delta_j$  curves.

The ratio of the amplitudes of the "collision dip" and the Bennett dip is

$$c_j = n_{ij} \frac{\Gamma + \nu}{\delta_j} = n_{ij} \left\{ 1 + \frac{3k\sigma_j\sqrt{1+n_{ij}}}{(\Gamma + \nu)\mathcal{A}} \right\}^{-1}. \tag{5.10}$$

We see therefore that at very low pressures ( $n_{ij} \ll 1$ ) the "collision dip" is not noticeable against the background of the Bennett dip. At large pressures we have  $c_j \cong n_{ij}$ .

Worthy of particular attention is the case when the dips cannot be separated and give a single "regular" contour (their widths do not differ much from each other). The half-width of the summary contour is given by the expression

$$\Delta_j = \frac{1}{\sqrt{2}} \left\{ \left( [\delta_j^2 - (\Gamma + \nu)^2]^2 \left[ \frac{c_j - 1}{c_j + 1} \right]^2 + 4\delta_j^2(\Gamma + \nu)^2 \right)^{1/2} + [\delta_j^2 - (\Gamma + \nu)^2] \frac{c_j - 1}{c_j + 1} \right\}^{1/2}. \tag{5.11}$$

When the pressure is changed,  $\Delta_j$  changes from  $\Delta_j = \Gamma$  (low pressures) to  $\Delta_j = \Gamma + \nu$  (high pressures). In the intermediate region, the  $\Delta_j$  curve lies above the line  $\Gamma + \nu$ . We note also other limiting cases that lead to a simplification of (5.11):

$$\Delta_j = (\Gamma + \nu) \left[ 1 + c_j \frac{\delta_j^2 - (\Gamma + \nu)^2}{\delta_j^2 + (\Gamma + \nu)^2} \right], \quad c_j \ll 1; \tag{5.12}$$

$$\Delta_j = \delta_j \left[ 1 - \frac{1}{c_j} \frac{\delta_j^2 - (\Gamma + \nu)^2}{\delta_j^2 + (\Gamma + \nu)^2} \right], c_j \gg 1; \quad (5.13)$$

$$\Delta_j = \nu \delta_j / (\Gamma + \nu), \quad c_j = 1. \quad (5.14)$$

## 6. THE LAMB DIP IN THE PLOT OF THE WORK OF THE FIELD

The atom velocity distribution itself is not observed in experiment, but it can be assessed from an analysis, say, of the shape of the gain (absorption) line in the presence of two opposing waves. The expression for the work of the field in our case (standing wave) is

$$\begin{aligned} \mathcal{P} \approx \mathcal{E} \left\{ 1 - \frac{G^2/2}{1 + G^2 \tau_2 X/2} \sum_{j=m,n} \left[ \frac{\bar{\nu}_j^{(2)}}{k\bar{\nu}} \mathcal{E} + \frac{\tau_{1j}}{\Gamma + \nu} \left( 1 + \frac{(\Gamma + \nu)^2}{(\Gamma + \nu)^2 + \Omega^2} \right) \right. \right. \\ \left. \left. + \tau_j^{(1)} \int_{-\infty}^{\infty} \left[ \frac{4(\Gamma + \nu)}{4(\Gamma + \nu)^2 + k^2 \eta^2} + \frac{4(\Gamma + \nu)}{4(\Gamma + \nu)^2 + (2\Omega - k\eta)^2} \right] f_j(\eta) d\eta \right] \right\}, \\ \mathcal{E} = \exp \left[ - \frac{\Omega^2}{(k\bar{\nu})^2} \right]. \quad (6.1) \end{aligned}$$

The calculations were performed with allowance for the approximations  $(\Gamma + \nu) \ll k\bar{\nu}$ ,  $\sigma_j \sqrt{1 + n_{1j}} \ll \bar{\nu}$ . A plot of the work  $\mathcal{P}$  of the field against the frequency  $\Omega$  has the form of a Doppler contour with a "homogeneous" saturation band and with dips. One of them is the well known Lamb dip, and the two others are "collision dips" (the integral terms in (6.1) that depend on  $\Omega$ ) pertaining to the levels  $m$  and  $n$ . We note that the parameters of the "collision dips" can differ strongly from each other, depending on the character of the diffusion in the states  $m$  and  $n$ . The "collision dips" in the work of the field and in the velocity distribution (for the level  $j$ , cf. (3.8)), accurate to the substitutions  $\Gamma + \nu \rightarrow 2(\Gamma + \nu)$  and  $\Omega - k\nu \rightarrow 2\Omega$ . This result is a consequence of the difference Green's functions and, from the point of view of the experiment, is of great significance: the velocity distribution (in this case, for the population difference) is obtained by simple recalculation of the experimental plots of  $\mathcal{P}(\Omega)$ .

For the selective scattering model and for the approximation (5.6), we obtain the following final expression for the work of the field:

$$\begin{aligned} \mathcal{P} \approx \mathcal{E} \left\{ 1 - \frac{G^2/2}{1 + G^2 \tau_2 X/2} \sum_{j=m,n} \left[ \frac{\bar{\nu}_j \tau_j^{(2)}}{k\bar{\nu}} \mathcal{E} \right. \right. \\ \left. \left. + \frac{\tau_{1j}}{\Gamma + \nu} \left( 1 + \frac{(\Gamma + \nu)^2}{(\Gamma + \nu)^2 + \Omega^2} \right) + \frac{\tau_j^{(1)}}{\delta_j} \left( 1 + \frac{\delta_j^2}{\delta_j^2 + \Omega^2} \right) \right] \right\}. \quad (6.2) \end{aligned}$$

Here the expression for  $\delta_j$  is determined by formula (5.6), in which  $k$  must be replaced by  $k/2$ . In all other respects, the "collision dip" is determined by the same parameters as (5.6), and its relation to the Lamb dip is the same as that to the Bennett dip in the velocity distribution. Thus, everything stated above is applicable also to the plot of the work of the field.

In experimental studies of the dependence of the width of the Lamb dip on the pressure one usually obtains a plot whose slope is connected with the cross section of the Weisskopf broadening, and the intercept on the ordinary axis is connected with the natural line width  $\Gamma$ . In the presence of selective collisions, the situation can change strongly if the "collision dip" and the Lamb dip produce one "regular" contour of width

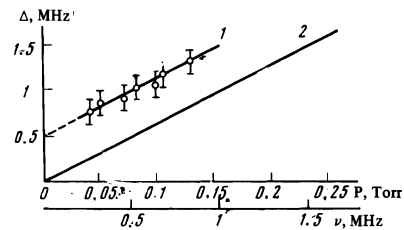


FIG. 3. Width of the Lamb dip vs. pressure in CO<sub>2</sub> laser with absorbing cell: 1—linear approximation of experimental data [9], 2—plot of  $\Delta = \nu$ .

$\Delta_j$ . The pressure dependence of  $\Delta_j$  is generally speaking nonlinear, but in a limited region of pressures the experimental points can lie, within the limits of errors, on a straight line whose parameters depend now on the pressure range in which the results are obtained, and on the characteristics of the selective scattering. For example, if the limiting case (5.13) is realized in conjunction with (5.9), then the slope is connected by the previous relation with the cross section of the Weisskopf broadening, but linear extrapolation to zero pressure yields

$$\Delta_j = \Gamma + 1/2 \ln 2 k \sigma_j \sqrt{1 + n_{1j}}, \quad (6.3)$$

where the second term can be much larger than the first. At lower pressures, when the influence of the relaxation constant  $\Gamma$  comes into play, both the slope and the ordinate of the linear extrapolation to zero pressure change. Only in the region of sufficiently high pressures (approximation (5.8)) do we again obtain the previous results. Thus, for an unequivocal determination of the causes of the broadening of the dip it is unnecessary to perform the measurements in a sufficiently wide range of pressures, so as to catch all the possible nonlinearities of the curve. Unfortunately, there are still no experimental data on this subject.

Let us discuss the experimental paper [9], which has in our opinion some bearing on the question considered here. The plot given in [9] for the dependence of the width of the Lamb dip on the pressure for a CO<sub>2</sub> laser with absorbing cell (Fig. 3), when linearly extrapolated to zero pressure, gives an analogously large value of the width (0.5 MHz), which cannot be accounted for by the natural line width ( $\Gamma \sim 10^3$  sec<sup>-1</sup>). It is natural to assume that in this case an important role is played by broadening due to selective collisions. In the pressure region where the experimental data of [9] were obtained, one can neglect the spontaneous relaxation in quenching constants, so that  $n_{1j}$  does not depend on the pressure and is equal to  $n_{1j} = \tilde{\nu}_{1j} / \tilde{\nu}_{2j}$ . The Lennard-Jones potential model gives  $n_{1j} \cong 8$  [7]. We assume furthermore that scattering in states  $m$  and  $n$  occurs in equal manner  $\sigma_m = \sigma_n \equiv \sigma$ ,  $n_{1m} = n_{1n} \equiv n$ ,  $\Delta_m = \Delta_n \equiv \Delta$ . From Fig. 3 and from the estimate of  $n$  it follows that, within the limits of the experimental curve, the amplitude of the "collision dip" is larger than the amplitude of the Lamb dip ( $c > 4$ ), and the following approximation holds for the half-width  $\Delta$  of the total dip:

$$\Delta = \nu + 1/2 \ln 2 k \sigma \sqrt{1 + n}. \quad (6.4)$$

Extrapolation of (6.4) to zero pressure yields

$$\Delta_0 = 1/2 \ln 2 \cdot k \sigma \sqrt{1 + n} = 0.50 \text{ MHz}, k \sigma = 0.48 \text{ MHz} \quad (6.5)$$

Let us compare this result with the estimate of  $k\sigma$  in accordance with the formula for the quantum uncertainty of the scattering angle:

$$\vartheta = \lambda / d \sim \sigma / \bar{v}, \quad \lambda = \hbar / \sqrt{2m\bar{v}}, \quad (6.6)$$

where  $d$  is a certain characteristic dimension of the  $\text{CO}_2$  molecule and  $\lambda$  is the de Broglie wavelength for the average relative velocity. For  $d = 4 \text{ \AA}$  (see<sup>[7]</sup>) we have ( $T = 1000^\circ\text{K}$ , radiation wavelength  $\lambda = 10^{-3} \text{ cm}$ )

$$k\sigma = 0.25 \text{ MHz}. \quad (6.7)$$

Thus, the value of  $k\sigma$  obtained on the basis of the selective-scattering model agrees in order of magnitude with the estimate from formula (6.6). Unfortunately, these are so far the only data, and this coincidence cannot be assigned too great an importance, for it can be accidental. However, if the agreement between the results of (6.6) and (6.7) is not accidental, then this is evidence of a noticeable role played by forward scattering, which is determined mainly by diffraction phenomenon. To resolve this problem it is necessary to perform special experiments aimed at revealing the role of diffusion in velocity space due to selective collisions. Principal attention in these experiments should be paid to low pressures, where it is precisely here that the distinguishing features of the phenomena can come into play, e.g., the nonlinear plot of the width of the Lamb dip against pressure, etc.

### 7. THREE-LEVEL SYSTEM

Let the external field be represented by two plane monochromatic waves, each of which interacts with only one transition in a three-level system (Fig. 4a):

$$\begin{aligned} V_{mn}(t, \mathbf{r}) &= G \exp\{-i(\Omega t - \mathbf{k}\mathbf{r})\}, \\ V_{nl}(t, \mathbf{r}) &= G_\mu \exp\{-i(\Omega_\mu t - \mathbf{k}_\mu \mathbf{r})\}; \\ G &= d_{mn}E / 2\hbar, \quad G_\mu = d_{nl}E_\mu / 2\hbar, \\ \Omega &= \omega - \omega_m, \quad \Omega_\mu = \omega_\mu - \omega_{nl}. \end{aligned} \quad (7.1)$$

Here  $\omega$ ,  $\omega_\mu$ ,  $\mathbf{k}$ , and  $\mathbf{k}_\mu$  are the frequencies and wave vectors of the waves. We assume the field  $E_\mu$  to be weak (it does not change the average level populations), and calculate the gain  $\alpha_\mu$  of the weak field. If there is no "phase memory" for the transitions  $n-l$  and  $m-l$ , then  $\alpha_\mu$  takes the following standard form<sup>[13,14]</sup>:

$$\alpha_\mu \propto \text{Re} \left\langle \frac{[\Gamma_{nl} + i(\Omega_\mu' - \Omega')](\rho_{mm} - \rho_{ll}) - iV_{mn} \rho_{nm}}{[\Gamma_{nl} + i(\Omega_\mu' - \Omega')][\Gamma_{ml} + i\Omega_\mu'] + G^2} \right\rangle, \quad (7.2)$$

$$\Omega' = \Omega - k\bar{v}, \quad \Omega_\mu' = \Omega_\mu - k_\mu \bar{v}, \quad \Gamma_{ij} = (\Gamma_i + \Gamma_j) / 2 + \nu_{ij}$$

$\rho_{mm}$  and  $\rho_{nm}$  are taken from the solution of Eqs. (1.4) and from (1.5),  $\rho_{ll}$  is the unsaturated population of the level  $l$ , and the angle brackets denote averaging over the velocities  $\mathbf{v}$ . Expression (7.2) consists of two terms that differ in nature: the first is proportional to the population difference  $\rho_{mm} - \rho_{ll}$ ; the second, proportional to  $\rho_{nm}$ , is due to nonlinear interference effects (NIE) and is called the interference term<sup>[13,14]</sup>.

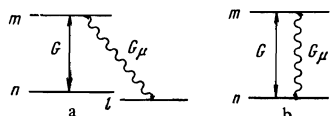


FIG. 4 Transition schemes for three-level (a) and two-level (b) systems.

All the new results of diffusion in velocity space can be traced by taking into account the first nonlinear corrections in the strong field, so that we shall henceforth confine ourselves to this approximation. Formula (7.2) is transformed into

$$\alpha_\mu \propto \frac{\sqrt{\pi}}{k\bar{v}} (N_m - N_l) \mathcal{E}_\mu \quad (7.3)$$

$$+ \text{Re} \left\langle \frac{\rho_m^{(1)}}{\Gamma_{ml} + i\Omega_\mu'} - \frac{G^2(N_m - N_n)W(\mathbf{v})}{[\Gamma_{nl} + i(\Omega_\mu' - \Omega')][\Gamma_{ml} + i\Omega_\mu'][\Gamma_{mn} - i\Omega']} \right\rangle,$$

$\rho_m^{(1)}$  is the correction for the saturation in the population of the level  $m$ , while  $\mathcal{E}_\mu$  is obtained from  $\mathcal{E}$  of (6.1) by making the substitution  $\Omega \rightarrow \Omega_\mu$ . In the derivation of (7.3) it is assumed that the following usual condition is satisfied:

$$\Gamma_{ij} \ll k\bar{v}, \quad k_\mu \geq k. \quad (7.4)$$

The first term in (7.3) describes the line of the unsaturated gain, the second describes the part of the nonlinear increment due to the change of the level population under the influence of the strong field (the "population" term), and the third is the interference term. Collisions with change of velocity of the atom manifest themselves only in the explicit dependence of  $\rho_m^{(1)}$  on  $\mathbf{v}$ ; in the interference term, on the other hand, these processes lead only to a change of the quantities  $\Gamma_{ij}$  with changing pressure, and its explicit form is the same as in the relaxation-constant model. Thus, in the weak-saturation approximation and in the absence of phase memory ( $A_{ij} = 0$ ), elastic collisions affect only the structure of the "population" term.

We shall henceforth confine ourselves to two mutual orientations of the vectors  $\mathbf{k}$  and  $\mathbf{k}_\mu$  ( $\mathbf{k} \uparrow \uparrow \mathbf{k}_\mu$  and  $\mathbf{k} \uparrow \downarrow \mathbf{k}_\mu$ ), and assume the model of Sec. 4, using a one-dimensional difference kernel and expression (5.1) for  $\rho_{jj}$ . If the waves have equal directions ( $\mathbf{k}_\mu \uparrow \uparrow \mathbf{k}$ ), substitution of (5.1) in (7.3) yields

$$\alpha_\mu \propto \mathcal{E}_\mu \left\{ N_m - N_l - 2G^2(N_m - N_n) \text{Re} \left[ \left( \frac{1}{\Gamma_m + \nu_m} + \frac{1}{\Gamma_{nl}'' + i(\Omega_\mu - \Omega')} \right) \times \frac{1}{\Gamma_{ml} + \Gamma_{mn}'' + i(\Omega_\mu - \Omega')} + \frac{\tau_m^{(1)}}{2\sigma_m \sqrt{1 + n_{lm}}} \right] \right\} \quad (7.5)$$

$$\times \int_{-\infty}^{\infty} \frac{\exp[-|\zeta|/\sigma_m \sqrt{1 + n_{lm}}]}{\Gamma_{ml} + \Gamma_{mn}'' + i(\Omega_\mu - \Omega' - k\zeta)} d\zeta + 2\tau_m^{(2)} \frac{\sqrt{\pi}}{k\bar{v}} \mathcal{E} \left. \right\};$$

$$\Gamma_{mn}'' = \frac{k_\mu}{k} \Gamma_{mn}, \quad \Gamma_{nl}'' = \Gamma_{nl} + \frac{k_\mu - k}{k} \Gamma_{mn}, \quad \Omega'' = \frac{k_\mu}{k} \Omega.$$

The coefficient  $\alpha_\mu$  as a function of  $\Omega_\mu$  has the form of a Doppler contour with dips due to the change of the population under the influence of the strong field and the NIE. The interference term (which is proportional to  $[\Gamma_{nl}'' + i(\Omega_\mu - \Omega'')]^{-1}$ ) and the part of the "population" term (proportional to  $1/(\Gamma_m + \nu_m)$ ) have the same structure as in the relaxation-constant model<sup>[13,14]</sup>. The remaining part of the nonlinear increment to (7.5) is due to diffusion in velocity space and represents a "collision dip" (integral term) and a Doppler band due to strong collisions.

In the case of opposing waves ( $\mathbf{k}_\mu \uparrow \downarrow \mathbf{k}$ ), the interference term drops out and in the remaining part of the (7.5) it is necessary to make the substitution  $\Omega'' \rightarrow -\Omega''$ .

It is easy to note that the "population" term in (7.5) is formally similar to the expression for the field in-

crement to  $\rho_{mm}$  (see (5.1)) following the substitutions  $\Omega_\mu \rightarrow k\nu$ ,  $\Gamma_{mn}'' + \Gamma_{ml} \rightarrow \Gamma_{mn}$ ,  $\Omega'' \rightarrow \Omega$ . With respect to the "collision dip," this effect is due to the use of the difference kernel  $B_m(\nu - \nu')$ . When  $k_\mu \uparrow k$ , the nonlinear increment to  $\alpha_\mu$  is wholly equivalent to the velocity distribution (5.1) following the substitutions  $\Omega_\mu \rightarrow -k\nu$ ,  $\Omega'' \rightarrow \Omega$ ,  $\Gamma_{mn}'' + \Gamma_{ml} \rightarrow \Gamma_{mn}$ . The noted distinguishing features are quite useful in experimental investigations of the processes of diffusion in velocity space, since the velocity distribution is obtained by simply recalculating the plot for the line shape of the weak-field gain. We note also that, unlike investigations of the Lamb dip, the experiments in which  $\alpha_\mu$  is measured yield information on elastic scattering only in one state  $m$ .

## 8. TWO-LEVEL SYSTEMS

Two-level systems (Fig. 4b) are described by Eqs. (1.1), where  $V_{mn}(t, \mathbf{r})$  takes the form

$$V_{mn}(t, \mathbf{r}) = G \exp[-i(\Omega t - k\mathbf{r})] + G_n \exp[-i(\Omega_\mu t - k_\mu \mathbf{r})],$$

$$\Omega_\mu = \omega_\mu - \omega_{nn}. \quad (8.1)$$

It is convenient to seek the solution of (1.1) in the form

$$\rho_j = N_j W(\nu) + |G|^2 R_j + 2\text{Re} \{G^* G_n \bar{r}_j \exp[-i(\epsilon t - q\mathbf{r})]\}, \quad j = m, n;$$

$$\rho_{mn} = R \exp[-i(\Omega t - k\mathbf{r})] + r \exp[-i(\Omega_\mu t - k_\mu \mathbf{r})] \quad (8.2)$$

$$+ \bar{r} \exp[-i(2\Omega - \Omega_\mu)t - (2k_\mu - k)\mathbf{r}], \quad \epsilon = \Omega_\mu - \Omega, \quad q = k_\mu - k,$$

where the amplitudes  $\bar{r}_j$ ,  $R_j$ ,  $r$ , and  $\bar{r}$  do not depend on the coordinates and on the time. The NIE in two-level system reduce to oscillations of the population at the difference frequency. The quantity  $G^* G_n \bar{r}_j$  is the amplitude of these oscillations. The correction for the saturation  $|G|^2 R_j$  is due only to the strong field. In the presence of weak and strong fields on the  $m - n$  transition, polarization is induced in the atom at the frequency of each of them as well as at the combination frequency ( $2\Omega - \Omega_\mu$ ). Under the same assumptions as in Sec. 7 (first nonlinear corrections in  $|G|^2$ , absence of "phase memory" in collisions), the gain  $\alpha_\mu$  of the weak field is proportional to the following expression:

$$\alpha_\mu \propto \frac{\sqrt{\pi}}{k\nu} \mathcal{E}_\mu + |G|^2 \text{Re} \left\langle \frac{R_m - R_n + \bar{r}_m - \bar{r}_n}{\Gamma_{mn} - i(\Omega_\mu - k_\mu \nu)} \right\rangle. \quad (8.3)$$

The first term in (8.3) corresponds to the linear approximation. The increment proportional to  $|G|^2$  breaks up into two parts, namely,  $R_m - R_n$  is due to the change of the population as a result of the strong field ("population" term), and  $\bar{r}_m - \bar{r}_n$  is due to nonlinear interference effects. The values of  $R_j$  and  $\bar{r}_j$  are obtained from the equations

$$\Gamma_j R_j = S_j(R_j) \mp N_0 W(\nu) \{[\Gamma_{mn} - i(\Omega - k\nu)]^{-1} + [\Gamma_{mn} + i(\Omega - k\nu)]^{-1}\}; \quad (8.4)$$

$$[\Gamma_j - i(\epsilon - q\nu)] \bar{r}_j = S_j(\bar{r}_j) \mp N_0 W(\nu) \{[\Gamma_{mn} - i(\Omega_\mu - k_\mu \nu)]^{-1} + [\Gamma_{mn} + i(\Omega - k\nu)]^{-1}\};$$

$$S_j(R_j) = -\nu_j R_j + \int A_j(\nu', \nu) R_j(\nu') d\nu'.$$

For opposing waves,  $k_\mu \uparrow k$ , the interference term in (8.3) drops out. With respect to the remaining "population" term, we can repeat the analyses of the preceding section, the only difference being that the population of one level  $m$  is replaced here by the population difference  $R_m - R_n$ . New effects arise when the waves

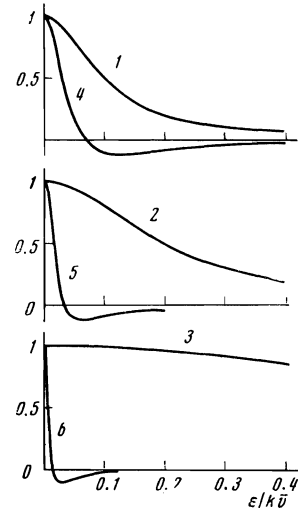


FIG. 5. Nonlinear increments to the gain  $\alpha_\mu$ , pertaining to the level  $j$  (2-level system): 1—"population" dip due to the Bennett dip; 2—"collision dip"; 3—part of Doppler "band"; 4, 5, 6—corresponding interference structures;  $\Gamma_{mn} = 0.05 k\nu$ ,  $\delta_j = 4\Gamma_{mn}$ ,  $\Gamma_j + \nu_j = \Gamma_{mn}$ ,  $\Gamma_j + \nu_j - \nu_j = 0.2$  ( $\Gamma_j + \nu_j$ ),  $\Gamma_{ij} = 0.5$  ( $\Gamma_j + \nu_j$ ). The plots are normalized to unity at the maximum.

have equal directions ( $k_\mu \uparrow k$  or  $q = 0$ ). The difference between the resonant terms  $[\Gamma_{mn} - i(\Omega - k\nu)]^{-1}$  and  $[\Gamma_{mn} - i(\Omega_\mu - k_\mu \nu)]^{-1}$  in (8.4) can be neglected within the framework of the condition (7.4). The equation for  $\bar{r}_j$  is then obtained from the equation for  $R_j$  by means of the substitution  $\Gamma_j \rightarrow \Gamma_j - i\epsilon$ . We can then use the results of the preceding section to find  $\bar{r}_j$ . At  $\epsilon = 0$  we have  $R_j = \bar{r}_j$ , and the interference term in (8.3) is equal to the "population" term. It is remarkable that in a two-level system the elastic collisions influence also the interference term<sup>4)</sup>. In the employed collision model we obtain for  $\alpha_\mu$  the expression ( $q = 0$ )

$$\alpha_\mu \propto \mathcal{E}_\mu \left\{ 1 - 2|G|^2 \text{Re} \sum_{j=m,n} \left[ \frac{1}{2\Gamma_{mn} - i\epsilon} \left( \frac{1}{\Gamma_j + \nu_j} + \frac{1}{\Gamma_j + \nu_j - i\epsilon} \right) + \frac{\tau_j^{(1)}}{2\sigma_j \sqrt{1 + n_{1j}}} \int_{-\infty}^{\infty} \frac{\exp[-|\xi|/\sigma_j \sqrt{1 + n_{1j}}]}{2\Gamma_{mn} - i(\nu - k\xi)} d\xi \right. \right.$$

$$\left. \left. + \frac{\tau_j^{(1)}(\epsilon)}{2\sigma_j \sqrt{1 + n_{1j}}(\epsilon)} \int_{-\infty}^{\infty} \frac{\exp[-|\xi|/\sigma_j \sqrt{1 + n_{1j}}(\epsilon)]}{2\Gamma_{mn} - i(\epsilon - k\xi)} d\xi \right] \right\},$$

$$n_{1j}(\epsilon) = \frac{\bar{\nu}_{1j}}{\Gamma_j + \nu_j - \bar{\nu}_{1j} - i\epsilon}, \quad \tau_j^{(1)}(\epsilon) = \frac{n_{1j}(\epsilon)}{\Gamma_j + \nu_j - i\epsilon}.$$

The nonlinear increment to (8.5) has a much more complicated structure than in three-level systems. The reason is that the interference term is also sensitive to collisions, and, in addition, the scattering in both states  $m$  and  $n$  is appreciable. Processes that differ in the character of the relaxation introduce in (8.5) nonlinear increments having the same structure as the given process. The interaction with the field up to the first collision leads to the result characteristic of the relaxation-constant model<sup>[15]</sup> (the term in the first line). The diffusion in velocity space gives rise to new incre-

<sup>4)</sup>An analogous influence is possible also in a three-level when the "phase memory" at the forbidden transition is taken into account.



ments: The second and third lines in (8.5) are due to relaxation via the selective-collision channel, and the fourth line is due to relaxation for a Maxwellian velocity distribution of the level populations. Each of these increments breaks up in turn into "population" and interference terms. At  $\epsilon = 0$  both terms are equal. The contour of the  $\alpha_\mu$  line contains thus an entire set of "population" and interference structures. The former include a dip with a half-width  $2\Gamma_{mn}$ , two "collision dips," pertaining to the levels  $m$  and  $n$ , and a band due to "homogeneous" saturation. As is readily seen, there are altogether six interference structures. The set of spectral structures corresponding to the level  $j$  is shown in Fig. 5. Naturally, they do not manifest themselves in equal degrees under all conditions: sometimes certain of them can predominate, and other times others. The choice of the corresponding condition and comparison of experiment with theory afford many opportunities of verifying the employed collision model.

The structure of the "population" terms in (8.5) is the same as in three-level systems, and we therefore do not stop to analyze them. The NIE in a two-level system is connected with population beats at the difference frequency  $\epsilon$  and is therefore determined by the character of the relaxations of the populations themselves, and not of the polarization as in the case of a three-level system. Each relaxation process introduces its own spectral structure with a width determined by the duration of this process. In the first channel, the interference structure has a width  $\sim\Gamma_j + \nu_j$  in accordance with the fact that  $1/(\Gamma_j + \nu_j)$  is the lifetime to the first collision. The relaxation of the total population of the level  $j$  is the relaxation process with maximum duration. It corresponds to a time  $1/\tilde{\Gamma}_j$ , during which the atom has time to acquire an equilibrium velocity distribution. The spectral structure with width  $\tilde{\Gamma}_j$  therefore appears in the last channel. The relaxation process connected with selective collisions has a relaxation time  $1/(\Gamma_j + \nu_j - \tilde{\nu}_{ij})$ , during which the nonequilibrium velocity distribution is preserved. The parameters of the corresponding interference structure therefore contains the quantity  $\Gamma_j + \nu_j - \tilde{\nu}_{ij}$ . If  $k\sigma_j\sqrt{1+n_{ij}} \gg 2\Gamma_{mn}$ , the interference structure in the selective-collision channel at the level  $j$  takes the form

$$2|G|^2 \text{Re} \frac{\pi}{2} \frac{1}{\Gamma_j + \nu_j - i\epsilon} \frac{1}{(\Gamma_j + \nu_j - i\epsilon)(\Gamma_j + \nu_j - \tilde{\nu}_{ij} - i\epsilon)} \quad (8.6)$$

The width of its narrow part is  $\sim\Gamma_j + \nu_j - \tilde{\nu}_{ij}$ . A decrease of  $k\sigma_j\sqrt{1+n_{ij}}$  leads to a change in the form of this term, from (8.6) to

$$2|G|^2 \text{Re}(\Gamma_j + \nu_j - i\epsilon)^{-1}(\Gamma_j + \nu_j - \tilde{\nu}_{ij} - i\epsilon)^{-1} \quad (8.7)$$

in such a way that the width of the narrow part always remains  $\sim\Gamma_j + \nu_j - \tilde{\nu}_{ij}$ .

If the condition

$$2\Gamma_{mn} = \Gamma_m + \Gamma_n + \nu_m + \nu_n \quad (8.8)$$

characteristic of spontaneous relaxation is satisfied, then we have in the first channel

$$\sum_{m,n} \frac{1}{2\Gamma_{mn} - i\epsilon} \left( \frac{1}{\Gamma_j + \nu_j} + \frac{1}{\Gamma_j + \nu_j - i\epsilon} \right)$$

$$= \frac{1}{(\Gamma_m + \nu_m)(\Gamma_n + \nu_n - i\epsilon)} + \frac{1}{(\Gamma_n + \nu_n)(\Gamma_m + \nu_m - i\epsilon)} \quad (8.9)$$

This means that in the relaxation-constant model, under the condition (8.8), there are only two dips of equal amplitude and with widths  $\Gamma_m + \nu_m$  and  $\Gamma_n + \nu_n$ . The relation (8.8) should be regarded as a condition for a sort of coherence of the atomic states. Violation of (8.8) as a result of collisions leads to the appearance of a third "dip" (with width  $2\Gamma_{mn}$ ), which can be ascribed to an additional relaxation process.

The ratios of the amplitudes of the interference increments in (8.5) (just as those of the "population" increments) are

$$1 : n_{ij} f(z_j) : \sqrt{\pi} \frac{\Gamma_{mn}}{k\bar{v}} (1 + n_{ij}) n_{2j}; \quad (8.10)$$

$$f(z) = z[\text{ci}(z) \sin z - \text{si}(z) \cos z], \quad z_j = 2\Gamma_{mn} / k\sigma_j \sqrt{1+n_{ij}};$$

$$f(z) \rightarrow 1/2\pi z \quad (z \rightarrow 0), \quad f(z) \rightarrow 1 \quad (z \rightarrow \infty).$$

In the general case, any of these structures can predominate. If e.g., "homogeneous" saturation plays an important role in the system, then the amplitude of the narrowest structure becomes noticeable. In the case when the number of collisions  $n_1$  is large enough, the interference structure due to selective collisions can prevail.

The general conclusions of the present section remains in force also in the presence of inelastic collisions, of the type of exchange of rotational energy in molecular systems. We illustrate this using the following simple model of the collision-integral kernel:

$$A_j(\nu', J'; \nu, J) = \bar{v}_j W(\nu) W_B(J),$$

$$W_B(J) = (2J+1) \exp\left[-\frac{B}{k_B T} J(J+1)\right] w, \quad (8.11)$$

$$w = \left[ \sum_J W_B(J) \right]^{-1},$$

where  $B$  is the rotational constant in the vibrational state  $j$ . The model (8.11) denotes that after each collision the molecule acquires a Maxwellian distribution with respect to the velocities and a Boltzmann distribution  $W_B(J)$  over the rotational levels  $J$ . Assume that two fields, strong and weak, act on one vibrational-rotational transition  $m, J_m \rightarrow n, J_n$ ; then at  $k_\mu = k$  we have

$$a_\mu \propto \mathcal{E}_\mu \left\{ 1 - 2|G|^2 \text{Re} \sum_{j=m,n} \left[ \frac{1}{2\Gamma_{mn} - i\epsilon} \left( \frac{1}{\Gamma_j + \nu_j} \right. \right. \right. \quad (8.12)$$

$$\left. \left. \left. + \frac{1}{\Gamma_j + \nu_j - i\epsilon} \right) + \frac{\sqrt{\pi}}{k\bar{v}} \mathcal{E} \left( \frac{\bar{v}_j W_B(J_j)}{\tilde{\Gamma}_j(\Gamma_j + \nu_j)} \right. \right. \right.$$

$$\left. \left. \left. + \frac{\bar{v}_j W_B(J_j)}{(\tilde{\Gamma}_j - i\epsilon)(\Gamma_j + \nu_j - i\epsilon)} \right) \right] \right\}, \quad \tilde{\Gamma}_j = \Gamma_j + \nu_j - \bar{v}_j.$$

The width of the narrowest interference structure is determined here by the total lifetime  $1/\tilde{\Gamma}_j$  and the vibrational state  $j$ , in spite of the fact that inelastic collisions took place during that time. Our example shows particularly clearly the physical cause of the narrow interference of structures. After the first collision, the molecule goes from the level  $J_m$ , stays at other rotational levels (on the average) for a time  $[\tilde{\nu}_j W_B(J_j)]^{-1}$ , returns to  $J_m$ , where it stays a time  $(\Gamma_j + \nu_j)^{-1}$ , leaves the level again, etc. Thus, the probability of being at the level  $J_m$  has the form of short "spikes" with width  $(\Gamma_j + \nu_j)^{-1}$ , separated by a time  $[\tilde{\nu}_j W_B(J_j)]^{-1}$ . The effective number of spikes is

obviously  $\tilde{\nu}_j W_B(J_j)/\tilde{\Gamma}_j$ , and their total duration  $\tilde{\nu}_j W_B/\tilde{\Gamma}_j(\Gamma_j + \nu_j)$  specifies the amplitude of the structure (in accordance with (8.12)). Since the phase relations are not important for the population beats, the width of the structure is determined by the total time during which the "spikes" can occur, i.e., by the time  $\tilde{\Gamma}_j^{-1}$ . The other spectral structures can be analogously interpreted.

## 9. DRAGGING OF THE RADIATION

The application of the foregoing results to the particular case of dragging of resonant radiation leads to a curious and somewhat unexpected effect, namely the narrowing of the "spectral line" with increasing pressure. If one of the working levels (say,  $m$ ) is optically coupled with the ground state (0), then, at a sufficiently high gas density, dragging of the radiation takes place on the transition  $m - 0$ . In atomic systems, where spontaneous relaxation predominates, this effect is particularly strong. Let us examine the gain of the weak field on the transition  $m - n$  under conditions of total dragging for  $m - 0$ . For simplicity, we neglect the collisions in which the velocity is changed and the phase is randomized. The expression (8.5) for  $\alpha_\mu$  takes the form

$$\alpha_\mu \propto \mathcal{E}_\mu \left\{ 1 - 2|G|^2 \operatorname{Re} \left[ \frac{1}{\Gamma_m(\Gamma_n - i\epsilon)} + \frac{1}{\Gamma_n(\Gamma_m - i\epsilon)} \right] + \frac{\sqrt{\pi}}{k\bar{v}} \mathcal{E} \left( \frac{\Gamma_{m0}}{(\Gamma_m - \Gamma_{m0})\Gamma_n} + \frac{\Gamma_{m0}}{(\Gamma_m - \Gamma_{m0} - i\epsilon)(\Gamma_m - i\epsilon)} \right) \right\}, \quad (9.1)$$

$$\Gamma_{mn} = \frac{\Gamma_m + \Gamma_n}{2}.$$

The constant  $\Gamma_{m0}$  is the probability of spontaneous decay from the level  $m$  to the ground state. The terms of the first line in (9.1) described dips with widths  $\Gamma_m$  and  $\Gamma_n$  on the gain line contour. The term proportional to  $\Gamma_{m0}$  is due to the dragging of the radiation and specified the "band" of the homogeneous saturation and the interference structure, the width of the narrow part of which is  $\Gamma_m - \Gamma_{m0}$ , i.e., less than the natural width of the level  $m$ . If the spontaneous decay proceeds predominantly to the ground state, then the relation  $\Gamma_m - \Gamma_{m0} \ll \Gamma_m, \Gamma_n$ , can be realized, and the dip due to dragging may turn out to be much narrower than all others. The ratio of the amplitude of the  $(\Gamma_m - \Gamma_{m0})$  dip to the amplitude of the  $\Gamma_m$  dip is

$$c = \frac{\sqrt{\pi}}{k\bar{v}} \frac{\Gamma_m \Gamma_n}{\Gamma_m - \Gamma_{m0}}. \quad (9.2)$$

In some systems, the factor (9.2) can be large enough, and the narrow dip can become perfectly noticeable.

Let us illustrate our results with the  $2s_2 - 2p_4$  ( $1.15 \mu$ ) of the Ne atom as an example. In accordance with<sup>[5,16,17]</sup> we have the following values of the required parameters:

$$\Gamma_m = 26 \text{ MHz} \quad \Gamma_n = 8.3 \text{ MHz} \quad \Gamma_m - \Gamma_{m0} = 1.6 \text{ MHz} \\ k\bar{v} = 400 \text{ MHz} \quad c \approx 0.6.$$

Thus, the width of the interference dip due to dragging is much smaller here than the natural widths  $\Gamma_n$  and  $\Gamma_m$  and the relative amplitude of the narrow dip is large enough.

Formula (9.1) is applicable, generally speaking, at pressures such that the dragging can be regarded as complete<sup>5)</sup>. At low pressures there is no dragging, and the last term of (9.1) drops out. For neon this means that the contour of the  $\alpha_\mu$  line will contain a superposition of dips with widths 26 MHz and 8.3 MHz. The radiation dragging should reveal itself experimentally in the fact that when the pressure is increased the summary dip first becomes effectively narrower, after which a fine structure with width 1.6 MHz appears.

<sup>5)</sup>At these pressures, the spontaneous approximation used in (9.1) may still be perfectly valid.

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ELECTROMAGNETIC RADIATION IN A MOVING MEDIUM

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The radiation intensity has been determined from an arbitrary source in an unbounded medium moving with a velocity greater than the velocity of light in the medium. All types of waves propagating in the moving medium are discussed, and drag on the light by the medium is taken into account when the latter is moving faster than the velocity of light.

WHEN a medium is moving faster than the velocity of light, in addition to ordinary electromagnetic waves with an index of refraction  $N_1$  there appears a second type of electromagnetic waves with an index of refraction  $N_2$ . The latter arise as the result of drag on the light by the moving medium. The regions of propagation of waves with refractive indices  $N_1$  and  $N_2$  are two coaxial cones whose solid angles face the direction of motion of the medium.

The appearance of the new waves substantially affects the radiation of an arbitrary source around which the moving medium flows. If the velocity of the medium relative to the source is greater than the phase velocity of light in the medium at rest, the radiation intensity depends strongly on the relative velocity of this motion and the structure of the source. Here a multipole expansion of the radiation field, as a rule, loses its meaning. The motion of the medium also affects the angular distribution of the radiation intensity.

According to the Umov-Poynting theorem<sup>[1,2]</sup> the rate of change of electromagnetic energy in an unbounded moving medium is equal to the power expended by external currents<sup>1)</sup>,

$$d\mathcal{E}/dt = - \int J_\alpha(x, t) E_\alpha(x, t) dV, \tag{1}$$

where  $\mathbf{E}$  is the intensity of the electric field produced by external currents with volume density  $\mathbf{J}$ . For a monochromatic source

$$J_i(x, t) = j_i(x, \omega) e^{i\omega t} + j_i^*(x, \omega) e^{-i\omega t}$$

the radiation intensity (1) averaged over a period is

$$\frac{d\mathcal{E}}{dt} = - \frac{\omega}{4\pi^2} \text{Im} \int j_i^*(\mathbf{k}, \omega) j_n(\mathbf{k}, \omega) L_{im}(\mathbf{k}, \omega) d\mathbf{k}. \tag{2}$$

In the case of an isotropic medium moving with velocity  $\mathbf{v}$ , the Green's function  $L_{im}$  takes the form

$$L_{im} = \frac{4\pi}{c^2} \frac{u_i u_m \kappa / (\kappa + 1) - g_{im} + K_{im}}{k_n^2 + \kappa (k_n u_n)^2 + i\delta},$$

$$K_{im} = \frac{1}{\kappa + 1} \left( \frac{k_i u_m + k_m u_i}{k_n u_n} - \frac{k_i k_m}{(k_n u_n)^2} \right), \tag{3}$$

where  $\kappa = \epsilon - 1$ ,  $\epsilon$  is the dielectric permittivity of the medium at rest,  $c$  is the velocity of light in vacuum,  $\mathbf{k}_i$

is the 4-dimensional wave vector,  $g_{im}$  is the metric tensor, and  $u_i$  is the 4-velocity of the medium. The latter two quantities are determined by the relations

$$g_{00} = 1, \quad g_{\alpha\beta} = -\delta_{\alpha\beta}, \quad g_{0\alpha} = g_{\alpha 0} = 0;$$

$$u_0 = \frac{1}{(1 - v^2/c^2)^{1/2}}, \quad u_\alpha = \frac{v_\alpha}{c(1 - v^2/c^2)^{1/2}}.$$

The dielectric permittivity  $\epsilon$  describes the electric and magnetic properties, and therefore the magnetic permeability  $\mu$  of the medium at rest is set equal to unity<sup>[1]</sup>. The bypassing of the poles in the Green's function (3) is determined by the infinitely small imaginary term  $i\delta$ . For  $\omega > 0$  we have  $\delta < 0$ . The value of  $\delta$  changes sign together with the frequency  $\omega$ . The frequency distribution of the energy  $d\mathcal{E}(\omega)$  radiated during the entire time of action of the arbitrary source  $J_i(\mathbf{x}, t)$  is given by the right-hand side of Eq. (2), multiplied by  $d\omega/2\pi$ . In this case the quantity  $j_i(\mathbf{k}, \omega)$  in the variable arguments  $\mathbf{k}$  and  $\omega$  is the Fourier component of the external four-dimensional current  $j_i(\mathbf{x}, t)$ .

Equation (2) is the relativistic generalization of the radiation intensity in a medium at rest<sup>[3]</sup>. The integrand in Eq. (2) is simplified if we use the charge conservation law  $\mathbf{k}_i j_i(\mathbf{k}, \omega) = 0$ .

We will write the denominator of the Green's function (3) in the form

$$k_n^2 + \kappa (k_n u_n)^2 = (kc - \omega N_{k\omega}) (kc + \omega N_{-k\omega}) [\kappa (ku/k)^2 - 1],$$

where we have introduced the designation

$$N_{k\omega} = \frac{[1 + \kappa u_0^2 - \kappa (ku/k)^2]^{1/2} - \kappa u_0 (ku/k)}{1 - \kappa (ku/k)^2},$$

$$\kappa = \epsilon - 1 \equiv N^2 - 1. \tag{4}$$

In the presence of dispersion the refractive index of the medium at rest  $N' = N'(\mathbf{k}', \omega')$  depends on the wave vector  $\mathbf{k}'$  and the frequency  $\omega'$  of the radiation in the medium at rest. The quantities  $\mathbf{k}'$  and  $\omega'$  are expressed in terms of the wave vector  $\mathbf{k}$  and frequency  $\omega$  in the moving medium by the Lorentz transformation equations for the wave 4-vector.

The first pole

$$kc - \omega N_{k\omega} = 0 \tag{5}$$

of the Green's function (3) in the moving medium describes radiation of direct waves with vector  $\mathbf{k}$  and refractive index  $N_1 = N_1(\theta)$ , where

$$N_1(\theta) = \frac{[1 + \kappa u_0^2 (1 - \beta^2 \cos^2 \theta)]^{1/2} - \kappa u_0^2 \beta \cos \theta}{1 - \kappa u_0^2 \beta^2 \cos^2 \theta}. \tag{6}$$

<sup>1)</sup>The Greek vector indices take on values 1, 2, and 3, and the Latin indices values 0, 1, 2, and 3. The index 0 denotes the time component of the 4-vector. The following summation rule is adopted:  $a_\alpha b_\alpha = a_1 b_1 + a_2 b_2 + a_3 b_3$ ,  $a_i b_j = a_0 b_0 - a_i b_i$ .

Here  $\cos \theta = \mathbf{k} \cdot \mathbf{v}/kv$  and  $\beta = v/c$ . The refractive index  $N'$  of the medium at rest, which enters into  $\kappa$ , is taken for the corresponding fixed direction, if the medium is dispersive or anisotropic. At the point of the pole the function (4) is equal to the refractive index (6) of the moving medium, so that Eq. (5) goes over to the form  $kc = \omega N_1$ .

If  $\epsilon > 1$  and the velocity  $v$  of motion of the medium is less than the critical velocity  $v^2 < c^2/\epsilon$ , then the refractive index (6) is finite and positive. For velocities greater than light  $v^2 > c^2/\epsilon$  there exist directions of the wave vector  $\mathbf{k}$  for which  $N_1 = \infty$ , if we neglect the dispersion of the medium at rest. The angle  $\theta_0$  for which  $N_1(\theta_0) = \infty$  is  $\theta_0 = \arccos(-1/\sqrt{\kappa u_0 \beta})$ . In this case the refractive index (6) is positive in the range of angles  $\theta_0 > \theta \geq 0$ .

The second pole

$$kc + \omega N_{-\kappa\omega} = 0 \quad (7)$$

of the Green's function (3) refers to backward waves, if  $v^2 < c^2/\epsilon$ . The second pole (7) contributes to the radiation intensity (2) for a velocity of the medium greater than light  $v^2 > c^2/\epsilon$ . In this case it describes the radiation of direct waves with wave vector  $\mathbf{k}$  and refractive index

$$N_2 = -N_1(\pi - \theta), \quad (8)$$

where the function  $N_1(\theta)$  for any  $\theta$  is given by Eq. (6), and the refractive index  $N_2 = N_2(\theta)$  is determined and positive in the region  $\pi - \theta_0 > \theta \geq 0$ . Waves with refractive index (8) have anomalous Doppler frequencies<sup>[4,5]</sup>. At the pole Eq. (7) becomes  $kc = \omega N_2$ .

After the remarks which have been made, the radiation intensity of transverse waves (2) is easy to convert to a form convenient for applications,

$$\begin{aligned} \frac{d\mathcal{E}}{dt} = & \int \frac{\omega^2 N_1^2 (\kappa |j_{iu}|^2 / (\kappa + 1) - |j_i|^2)}{c^3 [1 + \kappa u_0^2 (1 - \beta^2 \cos^2 \theta)]^h} \sin \theta d\theta \\ & + \int \frac{\omega^2 N_2^2 (\kappa |j_{iu}|^2 / (\kappa + 1) - |j_i|^2)}{c^3 [1 + \kappa u_0^2 (1 - \beta^2 \cos^2 \theta)]^h} \sin \theta d\theta, \end{aligned} \quad (9)$$

where  $\theta$  is the angle between  $\mathbf{k}$  and  $\mathbf{v}$ . The modulus of the wave vector in  $\mathbf{j}_1 = \mathbf{j}_1(\mathbf{k}, \omega)$  is  $k = \omega N_1/c$  in the first interval and  $k = \omega N_2/c$  in the second. The integration is performed over the region of angles where the refractive indices  $N_1$  and  $N_2$  of the moving medium are deter-

mined and positive. The second term in Eq. (9) arises only for motion of the medium faster than light.

In the presence of dispersion in the medium at rest, it is necessary to represent the expression  $\kappa = N'^2 - 1$  as a function of the angle of integration  $\theta$ , using for this the formulas for the aberration of light. In this case inclusion of dispersion in the refractive index  $N'$  of the medium at rest removes the infinity in the refractive indices  $N_1$  and  $N_2$  of the moving medium. Existence of dispersion can also lead to the result that several branches of the waves correspond to a given frequency  $\omega$ <sup>[4,5]</sup>. Then the radiation intensity (9) will consist of integrals taken individually for each branch of the radiated waves.

As follows from Eqs. (9), (6), and (8), there exist directions of the radiated waves for which the refractive indices (6) and (8) become rather large and the corresponding wavelengths small. This leads to the result that the ordinary multipole expansion of the radiation loses its meaning in this case, since the next term of the multiple expansion may turn out to be larger than the preceding term. In this case the radiation intensity depends strongly on the structure of the radiator.

In a magnetized plasma the refractive index  $N'$  (of the medium at rest) sometimes assumes extremely large numerical values, and therefore the effects found above can appear for nonrelativistic velocities of the plasma as a whole.

<sup>1</sup>V. P. Silin and A. A. Rukhadze, *Élektromagnitnye svoïstva plazmy i plazmopodobnykh sred* (Electromagnetic Properties of Plasma and Plasma-Like Media), Atomizdat, 1961.

<sup>2</sup>V. M. Agranovich and V. L. Ginzburg, *Kristallogoptika s uchetom prostranstvennoï dispersii i teoriya éksitonov* (Crystal Optics With Inclusion of Spatial Dispersion and the Theory of Excitons), Nauka, 1965.

<sup>3</sup>A. I. Alekseev and V. V. Yakimets, *Zh. Eksp. Teor. Fiz.* 51, 1569 (1966) [*Sov. Phys.-JETP* 24, 1057 (1967)].

<sup>4</sup>I. M. Frank, *Izv. AN SSSR, ser. fiz.* 6, 3 (1942).

<sup>5</sup>V. L. Ginzburg, *Usp. Fiz. Nauk* 69, 537 (1959) [*Sov. Phys.-Uspekhi* 2, 874 (1960)].

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