# Kinetic phenomena in a velocity-selectively excited gas

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We consider kinetic phenomena in a single-component gas that is velocity-selectively excited by narrow-band resonant radiation. The stationary velocity distribution function is calculated for a model in which the differential cross sections for the collisions of excited and unexcited particles are similar. The stability of the stationary state to arbitrary perturbations is analyzed. The macroscopic equations of state are found. The microscopic stationary state of the gas in the entire volume of the vessel is calculated. The pressure anisotropy, the spatial temperature and density inhomogeneities, as well as the macroscopic vortical flows in such a gas are considered. The influence of the spatial inhomogeneity of the radiation and of the external force fields is analyzed.

PACS numbers: 05.20.Dd

The discovery of self-induced drift in a gas mixture<sup>1,2</sup> has stimulated research resulting in the prediction of a substantial change in the state of a singlecomponent gas under conditions of selective optical excitation<sup>3-7</sup> with account taken of the change of the gaskinetic cross section of the particles. The resultant stationary state of the gas is characterized by a nonuniform distribution of the energy over the translational degrees of freedom and, as a result of the anisotropy of the pressure and of the thermal properties.<sup>3,5</sup> Other manifestations of a system in thermodynamic disequilibrium are macroscopic stationary gas streams<sup>4</sup> and spatial inhomogeneities of the temperature and density.<sup>5-7</sup> In all these cases we have an interaction between two thermodynamic systems (the gas and the radiation), wherein their energy remains unchanged in the course of the interaction (the radiation absorbed by the gas is spontaneously emitted subsequently), but an entropy flux from the gas into the radiation exists (the light absorbed is coherent and the emitted noncoherent).

The present paper is devoted to a systematic analysis and a theoretical description of the aforementioned effects in a single-component gas. In all the cases considered in the paper we have succeeded in obtaining in explicit form the particle velocity distribution function, making it possible, besides the analysis of the phenomena themselves, to obtain simultaneously a connection (which is sometimes universal) between the macroscopic properties of the gas and its microscopic parameters. All the calculations of the distribution functions are therefore made in the first part of the article.

## 1. MOLECULE-VELOCITY DISTRIBUTION FUNCTION

In earlier articles<sup>3,5</sup> some of us advanced qualitative premises that yielded the particle stationary velocity distribution in the spatially homogeneous case under certain simplifying assumptions. We use in what follows the Boltzmann kinetic equation, which enables us to corroborate rigorously the obtained solution, to forgo certain assumptions, and to obtain the molecule velocity distribution function f(v) in the spatially inhomogeneous and the nonstationary cases as well as in the presence of external force fields, and finally to obtain an analog of the Boltzmann *H*-theorem. Confining ourselves to the two-level approximation and neglecting the inelastic collisions and the light-pressure effect, we write down the kinetic equations for the excited (e) and unexcited (g) molecules in the form

$$\frac{df_{e}}{dt} = \frac{\partial f_{e}}{\partial t} + \mathbf{v} \frac{\partial f_{e}}{\partial \mathbf{x}} + \mathbf{a} \frac{\partial f_{e}}{\partial \mathbf{v}} = \gamma \varkappa (f_{s} - f_{*}) - \gamma f_{e}$$

$$+ \int \left[ w_{ee} (f_{*}' f_{e1}' - f_{e} f_{e1}) + w_{eg} (f_{e}' f_{g1}' - f_{e} f_{g1}) \right] d^{3} \mathbf{v}_{1} d^{3} \mathbf{v}' d^{3} \mathbf{v}_{1}',$$

$$\frac{df_{g}}{dt} = \frac{\partial f_{g}}{\partial t} + \mathbf{v} \frac{\partial f_{g}}{\partial \mathbf{r}} + \mathbf{a} \frac{\partial f_{g}}{\partial \mathbf{v}} = \gamma \varkappa (f_{e} - f_{g}) + \gamma f_{e}$$

$$+ \int \left[ w_{eg} (f_{s}' f_{e1}' - f_{g} f_{e1}) + w_{gg} (f_{s}' f_{g1}' - f_{g} f_{g1}) \right] d^{3} \mathbf{v}_{1} d^{3} \mathbf{v}' d^{3} \mathbf{v}_{1}',$$
(1)

where  $f \equiv f(\mathbf{v})$ ,  $f_1 \equiv f(\mathbf{v}_1)$ ,  $f' \equiv f(\mathbf{v}')$ ,  $f_1' \equiv f(\mathbf{v}_1')$ ,  $w(\mathbf{v}', \mathbf{v}_1'; \mathbf{v}, \mathbf{v}_1)$  is the probability of a collision that changes the molecule velocities from  $\mathbf{v}$  and  $\mathbf{v}_1$  to  $\mathbf{v}'$  and  $\mathbf{v}_1'$ ;  $w_{ee}$  describes the collisions of two excited molecules,  $w_{ee}$  of an unexcited and excited one, and  $w_{ee}$  of two unexcited molecules; a is the molecule acceleration in the external force field,  $\varkappa$  is the saturation parameter:

$$\kappa(\mathbf{v}) = \int \frac{BM(\omega)}{\gamma} \frac{d\omega}{1 + [(\omega - \omega_0 - \mathbf{k}\mathbf{v})/\Delta\omega_0]^2}, \qquad (1a)$$

 $\omega$  is the frequency, k is the wave vector,  $M(\omega)$  is the spectral density of the radiation, B is the Einstein coefficient,  $\omega_0$  is the frequency of the working transition,  $\Delta\omega_0$  is the homogeneous line width of the radiation, and  $\gamma$  is the reciprocal lifetime of the molecules on the upper level. We consider a situation wherein the collisions have the following properties:

$$\frac{w_{eg}(\mathbf{v}', \mathbf{v}'_1; \mathbf{v}, \mathbf{v}_1)}{w_{gg}(\mathbf{v}', \mathbf{v}'_1; \mathbf{v}, \mathbf{v}_1)} = \beta = \text{const}, \qquad \frac{w_{eg}}{w_{eg}} = \frac{w_{eg}}{w_{sg}}.$$
 (2)

[The conditions (2) are valid, for example, for the hard-sphere model—the first for arbitrary values of  $w_{ee}$  and the second in the case  $|\beta - 1| \ll 1$ .]

We add the right- and left-hand sides of (1). The terms that describe the interaction with the electromagnetic field cancel each other, and we arrive at the equation

$$\frac{df}{dt} = \int w_{ss}(f_{eff}'_{eff} - f_{eff}f_{eff1}) d^{3}\mathbf{v}_{1} d^{3}\mathbf{v}' d^{3}\mathbf{v}_{1}', \qquad (3)$$

where  $f = f_g + f_{e^+} f_{eff} = f_g + \beta f_{e^-}$ . We note that (3) is valid also when the second condition of (2) is violated, if the

total number of excited molecules is small enough to be able to neglect their collisions with one another.

It follows from (3) that the stationary spatially-homogeneous solution at  $\mathbf{a} = 0$  is such that  $f_{eff}$  is the Maxwellian distribution  $f_0$ :

$$f_{s}+\beta f_{e}=f_{0}(N_{eff},\alpha_{eff},\mathbf{V}_{eff},\mathbf{v})=N_{eff}\left(\frac{\alpha_{eff}}{\pi}\right)^{\frac{n}{2}}\exp\{-\alpha_{eff}(\mathbf{v}-\mathbf{V}_{eff})^{2}\}.$$
 (4)

It is convenient to express the constants  $N_{eff}$ ,  $\alpha_{eff}$ , and  $V_{eff}$  in terms of the total densities of the particles (*N*), of the energy, and of the momentum:

$$\int f d^{3}\mathbf{v} = N, \quad \int \frac{mv^{2}}{2} f d^{3}\mathbf{v} = \frac{3}{2} NT + N \frac{mV^{*}}{2}, \quad \int \mathbf{v} f d^{3}\mathbf{v} = N\mathbf{V}, \tag{5}$$

where  $T = m/2\alpha = mv_T^2$  is the temperature (the average kinetic energy of the molecule in a reference frame where V = 0), V being the macroscopic velocity of the gas. With the light pressure neglected and the gas heated by the radiation, the values of N, T, and V at the initial instant coincide with their values prior to turning on the light.

Equation (4) allows us to eliminate from the system (1) one of the unknowns,  $f_e$  or  $f_e$ , and by the same token simplify substantially the calculation of the distribution function; this can be of importance, for example, when a computer and a specific model of the collisions are used. In the general case, however, the problem remains too complicated to solve analytically. Further progress is possible if the gas density is not too high, so that  $\gamma$  is large compared with the collision frequency  $\nu$ . The induced and spontaneous transitions are then much more frequent than the molecule collisions. The ratio of the populations of the (e) and (g) levels should therefore be approximately the same as in the case of an isolated molecule, i.e.,  $f_e$  and  $f_e$  satisfy, besides (4), also the relation

$$f_c/f_s = \varkappa/(1+\varkappa). \tag{6}$$

In this case  $f(\mathbf{v})$  is uniquely expressed in terms of  $f_{eff}(\mathbf{v})$ :

$$f(\mathbf{v}) = [1 - \varphi(\mathbf{v})] f_{eff}(\mathbf{v}), \qquad (7)$$

where

$$\varphi(\mathbf{v}) = \frac{(\beta - 1)\kappa}{1 + (\beta + 1)\kappa} = \frac{\beta - 1}{\beta + 1 + \kappa^{-1}}.$$
(8)

In particular, in the absence of force fields the spatially homogeneous stationary solution is of the form

$$f(\mathbf{v}) = f_c(\mathbf{v}) = [1 - \varphi(\mathbf{v})] f_0(\mathbf{v}), \qquad (9)$$

 $f = f_e + f_e$  characterizes the kinetic properties of the gas. Of interest to spectroscopy is the quantity  $f_e - f_e$ , which describes, in particular, the shape of the absorption line:

$$f_{e} - f_{g} = -f_{eff} [1 + (1 + \beta) \times]^{-1}.$$
(10)

It is seen from (1a), (9), and (10) that by suitably shaping the spectrum of the exciting radiation it is possible to vary the particle velocity distribution in a wide range, a factor of importance for the shape of the absorption line of a gas. At  $\Delta \omega$ ,  $\Delta \omega_0 \ll k v_T$  ( $\Delta \omega$  is the width of the excitingradiation spectrum) and for a gas excited by a traveling wave, the form of the distribution function  $f(v_x)$  is shown in Fig. 1.<sup>1)</sup> From the normalization conditions (5) we have in this case

$$N_{eff} = (1+\varepsilon)N, \quad \frac{1}{\alpha_{eff}} = \left\{1 + \frac{\varepsilon}{3} \left[2\alpha \left(v_0 - \frac{v_0 V}{v_0}\right)^2 - 1\right]\right\} - \frac{1}{\alpha},$$
$$V_{eff} = V + \varepsilon \left(1 - \frac{v_0 V}{v_0^2}\right) v_0, \quad (11)$$

where

$$\varepsilon = \varphi_{\circ} \psi, \quad \psi = \Delta v_{z_{0}} \left(\frac{\alpha}{\pi}\right)^{\gamma_{z}} \exp\left[-\alpha \left(v_{\circ} - \frac{v_{\circ} V}{v_{\circ}}\right)^{2}\right],$$
$$\varphi_{\circ} = \frac{1}{\Delta v_{z_{0}}} \int \varphi(v_{z}) dv_{z_{0}}, \quad (11a)$$

 $\varepsilon$  and  $\varphi_0$  are respectively the relative area and the relative depth of the dip in the distribution of the total number of particles, and

 $\mathbf{v}_0 = \mathbf{k}/(\omega - \omega_0) k^2, \ k \Delta v_{x0} = \max \{\Delta \omega, \ \Delta \omega_0\}.$ 

If the excitation is by a standing wave (that travels to some degree) and at  $V_x = 0$  the quantity  $\varphi_0$  must be replaced by  $2\varphi_s$  in the first and second equations of (11), and by  $2\varphi_a$  in the last equation ( $\varphi_s$  and  $\varphi_a$  are the amplitudes of the parts of  $\varphi(v_x)$  that are respectively symmetrical and antisymmetrical about the Doppler contour. For a purely traveling wave  $\varphi_s = \varphi_a = \varphi_0/2$ , while for a purely standing one  $\varphi_a = 0$  and  $\varphi_s = \varphi_0$ .

We note that the stationary solution (9), just as the Maxwellian distribution, is independent of the form of the function  $w(\mathbf{v}', \mathbf{v}_1'; \mathbf{v}, \mathbf{v}_1)$ , i.e., of the concrete model of the collisions [this model must satisfy only the conditions (2)]. In the presence of a spatial inhomogeneity or at  $\mathbf{a} \neq \mathbf{0}$  (or if the inequality  $\gamma \gg \nu$  is violated), this statement is incorrect for approximately the same reasons as in an ordinary spatially inhomogeneous gas (see, e.g., the calculation of the thermal conductivity and of the viscosity in Ref. 8). To find the distribution function in these cases we have used therefore the strong-collision model. It is known that one of the conditions for the applicability of this model is proximity of f to a Maxwellian distribution.<sup>9</sup> In our case it suffices that  $f_{eff}$  be close to  $f_0$ . In fact, although at  $f_{eff}$ =  $f_0$  the value of  $f_c$  can differ noticeably from  $f_0$ , the dis-

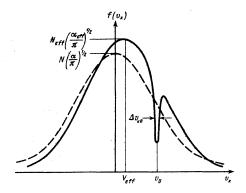


FIG. 1. Distribution of the gas particles in velocity  $(in v_x)$  under selective excitation. The dashed curve is the distribution function in the absence of excitation.

tribution of the probabilities of a collision-induced transition of a given molecule into an arbitrary velocity interval is the same, as follows from (2) and (4), as in some gas with  $\beta = 1$  and  $f = f_0$  [if (2) and (4) are satisfied, each molecule "senses" the entire remaining gas as Maxwellian].

With the foregoing taken into account, Eqs. (1) take the form

$$df_{e,g}/dt = v_{e,g}N_{e,g}'(\alpha'/\pi)^{\frac{1}{2}} \exp\{-\alpha'(\mathbf{v}-\mathbf{V}')^2\} - v_{e,g}f_{e,g} \pm \gamma \varkappa (f_g - f_e) \mp \gamma f_e.$$
(12)

Since the density, temperature, and macroscopic velocity of the aforementioned "auxiliary" gas are known beforehand, the parameters N',  $\alpha'$ , and V' should be specified beforehand as free ones and then obtained after f is determined from the conservation laws for the total particle densities, for the energy, and for the momentum.

Equations (12) yield for the distribution function at  $df_{s,s}/dt = 0$  an expression that is valid also if the condition  $\gamma \gg \nu$  is violated:

$$f(\mathbf{v}) = \left[ N_{s'} + \frac{\beta + \theta}{1 + \theta} N_{s'} - \frac{\beta - 1}{\beta + 1 + (1 + \theta)/\kappa} \left( N_{s'} + \frac{\beta - \theta}{1 + \theta} N_{s'} \right) \right] \\ \times \left( \frac{\alpha'}{\pi} \right)^{\frac{q}{2}} \exp\left\{ -\alpha' (\mathbf{v} - \mathbf{V}')^{2} \right\},$$
(13)  
$$f_{r,t} = \left( N_{s'} + \beta N_{s'} \right) (\alpha'/\pi)^{\frac{q}{2}} \exp\left\{ -\alpha' (\mathbf{v} - \mathbf{V}')^{2} \right\}.$$

where  $\theta = \nu_s / \gamma$ . With the aid of (13) we arrive at the conclusion that if  $\Delta \omega$ ,  $\Delta \omega_0 \ll k v_T$  the distribution function takes the same form (9), (11) as at  $\theta \ll 1$ , but the depth of the dip is smaller;  $\varphi(v_x)$  is determined by the expression

$$\varphi(\mathbf{v}) = \frac{\beta + 1 + (1+\theta)/\kappa_0}{\beta + 1 + (1+\theta)/\kappa_0 + 2\theta\psi} \frac{\beta - 1}{\beta + 1 + (1+\theta)/\kappa}, \qquad (14)$$

where  $\varkappa_0$  is the mean value of  $\varkappa$  in the interval  $\Delta v_{x0}$ . We see that at  $\theta \ll 1$  Eq. (14) goes over into (11a). When  $\theta$ increases, but still  $\theta \leq v_r / v_{x0}$ , its increase can be offset by suitably increasing  $\varkappa$ . (If the increase of  $\theta$  is due to a decrease of  $\gamma$ , then  $\kappa$  varies with  $\gamma$  in the required manner with practically no change in the intensity M.) Starting with  $\theta \sim v_T / \Delta v_{r0}$ , its increase lowers  $\varphi$  monotonically independently of M. This  $\varphi(\theta)$  dependence can be physically readily explained. The increase of  $\nu$ leads to a "smearing" of the dip in the gap of the distribution of f by the collisions. So long as  $\theta < v_{\tau} / \Delta v_{x0}$ , this "smearing" does not extend over the entire Maxwellian contour and it can be offset by strengthening the cause of the dip. At  $\theta \gg v_T/\Delta v_{x0}$  the radiation-excited particle is immediately "smeared" by the collisions over the entire Maxwellian contour. The excitation selectivity then practically vanishes and  $\varphi$  tends to zero with increasing  $\theta$ .

We consider now the influence of the spatial inhomogeneity and of the external force fields on the distribution function. From (3) and (7) we obtain by the same means as in Ref. 8 an equation for  $\chi = f_{eff}/f_0 - 1$  in the weak-inhomogeneity approximation  $\chi \ll 1$ :

$$I(\chi) = (1-\varphi) \left\{ \frac{d \ln N_{eff}}{dt} + \left[ \frac{3}{2\alpha_{eff}} - (\mathbf{v} - \mathbf{V}_{eff})^2 \right] \frac{d\alpha_{eff}}{dt} + 2\alpha_{eff} (\mathbf{v} - \mathbf{V}_{eff}) \left( \frac{d \mathbf{V}_{eff}}{dt} - \mathbf{a} \right) \right\} - \frac{d\varphi}{dt}, \qquad (15)$$

where

$$I(\chi) = \int w_{ss} f_{01}(\chi' + \chi_1' - \chi - \chi_1) d^3 \mathbf{v}_1 d^3 \mathbf{v}' d^3 \mathbf{v}_1'.$$

The conservation laws for the densities of the particles, of the momentum, and of the energy take the form

$$\int \chi(1-\varphi) f_0 d^3 \mathbf{v} = 0, \quad \int v \chi(1-\varphi) f_0 d^3 \mathbf{v} = 0, \quad \int v^2 \chi(1-\varphi) f_0 d^3 \mathbf{v} = 0.$$
(16)

If the optically-induced effects are weak, we can neglect when transforming the right-hand side of (15) the difference between  $N_{eff}$  and N,  $\alpha_{eff}$  and  $\alpha$ , and  $V_{eff}$  and V in all the coefficients of the derivatives and of **a**, and use the hydrodynamic equations for an ideal medium in analogy with the procedure used in Ref. 8 to calculate the viscosity and the thermal conductivity. Taking (11) into account, we have at the point V = 0

$$I(\chi) = (1-\varphi) \left\{ \left(\frac{5}{2\alpha} - v^2\right) \mathbf{v} \frac{\partial \alpha}{\partial \mathbf{r}} + \alpha \left(v_i v_j - \frac{v^2}{3} \delta_{ij}\right) \left(\frac{\partial V_i}{\partial x_j} + \frac{\partial V_j}{\partial x_i}\right) + \left[\frac{3}{2} - \alpha v_0^2 + \frac{\alpha}{3} \left(2\alpha v_0^2 - 1\right) v^2 + 2\alpha \mathbf{v}_0 \mathbf{v}\right] \left(\frac{\partial}{\partial t} + \mathbf{v} \frac{\partial}{\partial \mathbf{r}}\right) \varepsilon + 2\alpha \left[\mathbf{v}_0 - \frac{1}{3} \left(1 - 2\alpha v_0^2\right) \mathbf{v}\right] \mathbf{a}\varepsilon \right\} - \left(\frac{\partial}{\partial t} + \mathbf{v} \frac{\partial}{\partial \mathbf{r}} + \mathbf{a} \frac{\partial}{\partial \mathbf{v}}\right) \varphi.$$
(17)

where the subscripts *i* and *j* number the Cartesian components of the corresponding vectors. In contrast to the situation considered in Ref. 8, the last term of (17) allows us here to advance farther only by using the strong-collision model (we confine ourselves here and elsewhere to the case when  $\varphi$ , and hence  $\varepsilon$ , does not depend on the time):

$$\chi = \Delta_{N} + \left(\frac{3}{2} - \alpha v^{2}\right) \Delta_{\alpha} + 2\alpha \mathbf{v} \mathbf{V} + \frac{1}{v_{s}} \left(\mathbf{v} \frac{\partial}{\partial \mathbf{r}} + \mathbf{a} \frac{\partial}{\partial \mathbf{v}}\right) \varphi$$
$$+ \frac{1 - \varphi}{v_{s}} \left\{ \left(\frac{5}{2\alpha} - v^{2}\right) \mathbf{v} \frac{\partial \alpha}{\partial \mathbf{r}} + \alpha \left(v_{i}v_{j} - \frac{v^{2}}{3} \delta_{ij}\right) \left(\frac{\partial V_{i}}{\partial x_{j}} + \frac{\partial V_{j}}{\partial x_{i}}\right) \right.$$
(18)
$$+ \left(\frac{3}{2} - \alpha v_{0}^{2} + \frac{2\alpha v_{0}^{2} - 1}{3} \alpha v^{2} + 2\alpha \mathbf{v}_{0} \mathbf{v}\right) \mathbf{v} \frac{\partial \varepsilon}{\partial \mathbf{r}} + 2\alpha \left(\mathbf{v}_{0} + \frac{2\alpha v_{0}^{2} - 1}{3} \mathbf{v}\right) \mathbf{a}\varepsilon \right\},$$

where  $\Delta_N = N'/N - 1$ ,  $\Delta_{\alpha} = \alpha'/\alpha - 1$ , and  $\Delta V = V' - V$ are determined by expressions that follow from (16):

$$\Delta_{N} = 2 \frac{\Psi}{v_{g}} v_{0} \left( 2\alpha a_{x} + \frac{\partial}{\partial x} \right) \varphi_{*} \varphi_{*},$$

$$\Delta V_{x} = \frac{\Psi}{v_{g}} \left\{ \left( 1 - 2\alpha v_{0}^{2} \right) \left[ a_{x} \left( \frac{4}{3} \varphi_{*} - \varphi_{*}^{2} - \varphi_{*}^{2} \right) + \frac{3}{2\alpha} \frac{\partial \varphi_{*}}{\partial x} \right] + v_{0}^{2} \frac{\partial}{\partial x} \left( \varphi_{*}^{2} + \varphi_{*}^{2} \right) \right\},$$

$$\Delta V_{y,z} = \frac{\Psi}{v_{g}} \left[ \frac{2}{3} \left( 2\alpha v_{0}^{2} - 1 \right) \left( a_{y,z} + \frac{1}{2\alpha} \frac{\partial}{\partial y, z} \right) \varphi_{*} + \frac{1}{2\alpha} \frac{\partial}{\partial y, z} \left( \varphi_{*}^{2} + \varphi_{*}^{2} \right) \right],$$

$$\Delta_{\alpha} = 2 \frac{\Psi}{v_{g}} v_{0} \left\{ \left( \frac{2}{3} \alpha v_{0}^{2} - 1 \right) \left[ 2\alpha a_{x} (1 - \varphi_{*}) + \frac{\partial}{\partial x} \right] \varphi_{*} + \frac{1 - 2\alpha v_{0}^{2}}{3} \frac{\partial \varphi_{*} \varphi_{*}}{\partial x} \right\}.$$

At  $|v_0| + |\Delta v_{x0}| < \Delta \omega_0/k$  we must take into account the interference between the opposing waves. The case of a standing wave with the frequency at the center of the Doppler line is described by Eqs. (19) with  $v_0 = 0$ ,  $\varphi_u = 0$ , and  $\varphi_s = 2\varphi_0 \sin^2 kx$ , where  $\varphi_0$  is the amplitude of the dip produced by one of the traveling waves that make up the standing one. Before we proceed to discuss the characteristics of the gas on the basis of the obtained distribution function, we examine the stability of the obtained solutions of the kinetic equation.

#### 2. THE BOLTZMANN H-THEOREM

The problem of the stability of the stationary and equilibrium distributions  $f_0$  in an ordinary gas is known to be solved by the Boltzmann *H*-theorem.<sup>8</sup> Let us attempt to analyze the stability of the stationary solution (9). To this end we use Eq. (1), in which we express f in terms of  $f_{eff}$  with the aid of relation (5). We can then arrive by the same method as in Ref. 8 at an equation analogous to the Boltzmann *H*-theorem:

$$\frac{dS}{dt} = \frac{1}{2} \int w_{sef} f_{eff} f_{eff1} (X \ln X - 1) d^3 \mathbf{v}_1' d^3 \mathbf{v}' d^3 \mathbf{v}_1 d^3 \mathbf{v} d^3 \mathbf{r} + \int_{\mathbf{r}} \mathbf{\Phi} \, d\mathbf{\sigma} - \int \left( \frac{\partial \varphi}{\partial t} + \mathbf{v} \frac{\partial \varphi}{\partial \mathbf{r}} + \mathbf{a} \frac{\partial \varphi}{\partial \mathbf{v}} \right) f_{eff} d^3 \mathbf{v} d^3 \mathbf{r}, \qquad (20a)$$

where

$$X = \frac{f_{eff}' f_{eff}'}{f_{eff} f_{eff}}, \quad S = \int (1-\varphi) f_{eff} \ln \frac{e}{f_{eff}} d^3 \mathbf{v} d^3 \mathbf{r} = \int f \ln \frac{e(1-\varphi)}{f} d^3 \mathbf{v} d^3 \mathbf{r}$$
(20b)

plays the role of the entropy of the gas, and

$$\mathbf{\Phi} = \int \mathbf{v} (1-\varphi) f_{eff} \ln \frac{e}{f_{eff}} d^3 \mathbf{v}$$
(20c)

the role of the "entropy" flux density.

It is assumed that the force field has the property  $\partial F_i/\partial v_i = 0$  (F = ma) (this is true, for example for a Lorentz force). Therefore the right hand side of (20a) does not contain the term

$$\int (1-\varphi) f_{eff} \frac{\partial a_i}{\partial v_i} \ln \frac{e}{f_{eff}} d^3 \mathbf{v} d^3 \mathbf{r}.$$

It follows from the expressions (20) that in the case of a spatially homogeneous time-constant radiation and in the absence of an "entropy" flux through the surface  $\Sigma$  that surrounds the gas (for the latter to be valid it suffices, for example, that f and  $\varphi$  near the boundary be symmetrical about the component of v perpendicular to the boundary) the stationary solution (9) is stable to arbitrary deviations. If these conditions are not satisfied, and also if  $\gamma \leq \nu$ , the question of the stability of the stationary distribution remains open.

It should be noted that in an ordinary gas the cause of the instability of the stationary solution (9) can be not only a zero entropy flux through the walls of the vessel with the gas. In the case of selective excitation there exists a volume source of instability, namely the spatial inhomogeneity of the exciting radiation, as well as the external force fields [the last term of (20a)]. In addition, the conditions for the entropy flux through the walls to be zero become more stringent. Thus, in the model where the molecules are specularly reflected from the wall, the aforementioned flux is obviously zero in the usual situation at any vessel configuration. Under selective excitation it is necessary even in this model that  $\varphi(v_{\star})$  by a symmetric function (otherwise T and N become spatially inhomogeneous, see below), and also that the wall vessels be only perpendicular or parallel to the exciting-radiation propagation direction (in the case of oblique walls macroscopic fluxes of Nappear, see below).

### 3. MACROSCOPIC LOCAL PARAMETERS OF THE GAS

Starting from the obtained distribution function of the molecules of the selectively excited gas, we now determine its macroscopic characteristics, i.e., the fluxes of the momentum (pressure) and energy densities.

In the absence of excitation the stationary state of the gas is characterized by a uniform distribution of the energy over the degrees of freedom, Pascal's law holds, and there are no energy or particle fluxes. In our case, as follows from the form of the distribution, the differences in the dependences of f on  $v_x$ ,  $v_y$ , and  $v_z$ cause a nonuniform distribution of the energy over the degrees of freedom,  $\langle v_x^2 \rangle \neq \langle v_y^2 \rangle$ ,  $\langle v_z^2 \rangle$ . We note that, in contrast to the previously known cases, this distribution is caused only by the redistribution of the energy contained in the gas prior to the onset of the excitation, inasmuch as in the derivation of the distribution function we did not take into account any of the effects that lead to the heating of the gas by the light wave. In the upshot, the pressure  $P_{ij} = m \langle v_i v_j \rangle$  is a tensor, and because of the asymmetry of the dependence of f on  $v_0$  the heat flux  $\mathbf{q} = \frac{1}{2}m \langle \mathbf{v}v^2 \rangle$  may not be zero at  $v_0 \neq 0$  even in the absence of spatial inhomogeneity. The anisotropy of the distribution function leads apparently to anisotropy of the thermal properties of the gas, which manifests itself in the fact that the thermometer readings will depend on its material, shape, and orientation.

To calculate the macroscopic parameters of the gas we use expressions (9), (11), (18), and (19):

$$P_{xx} = NT \left\{ 1 + \frac{4}{3} b_1 \psi \varphi_z + \frac{4}{3} l \psi \left[ b_1 \frac{\partial \varphi_a \varphi_s}{\partial x} - 3 b_2 \left( \frac{\partial \varphi_a}{\partial x} + \frac{a_\pi}{v_r^2} (1 - \varphi_s) \varphi_a \right) \right] + \frac{2}{3} \frac{l}{v_r} \left( \frac{\partial V_z}{\partial x} + \frac{\partial V_y}{\partial v_r} - 2 \frac{\partial V_x}{\partial x} \right) \right\}, \quad P_{xx} + P_{yy} + P_{zz} = 3NT, \quad (21a)$$

$$P_{xy,z} = -NT \frac{l}{v_{x}} \left( 2v_{0}\psi \frac{\partial \varphi_{x} \varphi_{z}}{\partial y, z} + \frac{\partial V_{x}}{\partial y, z} + \frac{\partial V_{y,z}}{\partial x} \right), \qquad (21b)$$

$$b_{1} = 1 - \frac{v_{0}^{2}}{v_{x}^{2}} = 1 - 3\frac{T_{cr}}{T}, \quad b_{2} = 1 - \frac{v_{0}^{2}}{3v_{x}^{2}} = 1 - \frac{T_{cr}}{T}, \quad T_{cr} = \frac{mv_{0}^{2}}{3}; \quad (21c)$$

$$q_{s} = 3NTv_{0}b_{2}\psi\varphi_{a} + NTv_{r}l\psi \left[\frac{5}{3}b_{a}\frac{\partial\varphi_{*}}{\partial x} + \frac{3}{2}b_{2}\frac{v_{0}^{2}}{v_{x}^{2}}\frac{\partial(\varphi_{*}^{2} + \varphi_{a}^{2})}{\partial x}\right]$$

$$+ b_{*} \frac{a_{*}}{v_{T}^{2}} \left( 2\varphi_{*} - \varphi_{*}^{2} - \varphi_{*}^{2} \right) \left] - \frac{5}{2} N v_{T} l \frac{\partial T}{\partial x}, \qquad (22a)$$

$$q_{y,z} = NT v_{\tau} l\psi b_{1} \frac{\partial}{\partial y, z} \left( \frac{2}{3} \varphi_{s} + \frac{\varphi_{s}^{2} + \varphi_{s}^{2}}{2} \right) - \frac{5}{2} N v_{\tau} l \frac{\partial T}{\partial y, z}.$$
 (22b)

Here  $b_3 = b_1 - \frac{9}{5}b_2v_0^2/v_T^2$ ,  $b_4 = b_1 - b_2v_0^2/v_T^2$ , and *l* is the mean free path of the unexcited molecule. We recall that the terms due to  $\partial \varphi / \partial r \neq 0$  and  $a \neq 0$  were obtained in (21) and (22) only in the strong-collision model. As for the remaining terms [the second in (21a) and the first in (22a)], they are valid in any collision model that satisfies the conditions (2).

It is seen from (21) and (22) that the presence or absence of anisotropy of the pressure and of the heat flux is determined by the symmetry properties of the dependence of  $\varphi$  on  $v_x$ , meaning, at small M, also by the symmetry of the spectral distribution of the intensity relative to the center of the Doppler contour. Thus, in the spatially homogeneous case and at a = 0 the symmetric part of  $\varphi$  determines the presence of the anisotropy of the pressure, and the antisymmetric that of the heat flux. The parts of  $P_{ii}$  and q due to the weak inhomogeneity and to the external force fields have just the opposite dependence on the symmetry of  $\varphi$ . The foregoing, in principle, allows us to obtain in succession various kinetic effects by shaping the spectral and spatial characteristics of the radiation. It is physically obvious that the influence of the symmetry properties of  $\varphi$  has the same character also when the approximation  $\Delta \omega \ll k v_{\tau}$  used in Eqs. (21) and (22) does not hold.

## 4. NONLOCAL OPTICALLY INDUCED PHENOMENA IN A GAS

The considered change of the local characteristics of a gas under velocity-selective excitation leads obviously to a change of the gas macroscopic properties as a whole. Among these phenomena are the heat-flux-generated (22) stationary inhomogeneities of the temperature and density,<sup>5-7</sup> and also the macroscopic particle fluxes due to violation of Pascal's law in conjunction with the interaction with the walls or to the spatial inhomogeneity of the radiation.<sup>4,6</sup> We now consider these effects in succession.

We begin with the optically induced macroscopic fluxes. They can be arbitrarily attributed to two causes.

The first is the spatial inhomogeneity of  $P_{ij}$ , due to grad  $\varphi \neq 0$ . For the macroscopic state of a gas to be stationary at V = 0 in the entire vessel we must have div  $\mathbf{q} = 0$  and  $\partial P_{ij}/\partial x_i = 0$ . At  $\partial \varphi/\partial \mathbf{r} \neq 0$  it is generally speaking impossible to satisfy both conditions simultaneously by variation of the two scalar parameters Nand T (since  $P_{ij}$  is a tensor). This means that the gas contains macroscopic density fluxes. It is easily seen from (21) that, for example, if transverse and longitudinal radiation inhomogeneities are simultaneously present, a stationary state with  $\mathbf{V}(\mathbf{r}) = 0$  is impossible. Vortices are then produced in the gas, shown in Fig. 2 for the case  $v_0 = 0$ . From the expression for the viscosity<sup>8</sup>

$$\eta = c_{\eta}(mT)^{\nu} \sigma = c_{\eta} m v_T N l \tag{23}$$

[ $\sigma$  is the total cross section of the collisions of the unexcited molecules and  $c_{\eta}$  is a coefficient of the order of unity; in the strong-collision model  $c_{\eta} = 1$  (21b)], and from Eqs. (21) in the specular-wall models at  $L_x \gg L_y$ ( $L_x$  and  $L_y$  are respectively the longitudinal and transverse dimensions of the cell with the gas), we can obtain as an estimate the following equation for the flux velocity  $V_x(y)$ :

$$V_{x}(y) = -\frac{v_{T}}{18\varepsilon_{\eta}} \frac{L_{y}^{2}}{lL_{x}} \left(\varepsilon_{2} - \varepsilon_{1} + \varepsilon_{4} - \varepsilon_{3}\right) \left(1 - 6\left|\frac{y}{L_{y}}\right|^{2} + 4\left|\frac{y}{L_{y}}\right|^{3}\right), \quad (24)$$

where  $\varepsilon_{1,2,3,4}$  are respectively the values of  $\varepsilon$  at the points 1, 2, 3, and 4 (see Fig. 2).

The second cause of the density fluxes is the presence of off-diagonal pressure-tensor components that tend to move the gas layers in a direction parallel to the

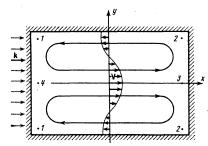


FIG. 2. Macroscopic density vortices produced in a gasfilled vessel in the presence of simultaneous longitudinal and transverse inhomogeneities of the exciting radiation. The case illustrated is  $\varepsilon_4 > \varepsilon_3 > \varepsilon_2 < \varepsilon_1 < \varepsilon_4$ ;  $|\varepsilon_4 - \varepsilon_3| > |\varepsilon_2 - \varepsilon_1|$ .

boundary between them. The decisive factor here is the influence of the vessel wall that produces a tangential force insufficient to maintain equilibrium in the layer next to the wall.<sup>2)</sup> Using the widely used approximation of specular reflection of the molecule from the vessel wall, we arrive at the conclusion that the wall acts on the adjacent gas layer only in a normal direction. Since  $P_{xy,z} \neq 0$ , a tangential force is exerted on this layer by the rest of the gas. As a result the wall layer begins to accelerate, followed by the adjacent layers, until a stationary flux is established, in which the optically induced component  $P_{xy,z}$  is cancelled by the friction force determined by the viscosity and by the V derivative perpendicular to the wall.

We consider first the case when the exciting radiation is spatially homogeneous, but certain vessel walls are oriented at an angle  $\xi \neq 0$ ,  $\pi/2$  to the light propagation direction.<sup>4</sup> Since  $P_{xx} \neq P_{yy}$ , the pressure tensor expressed in a coordinate frame connected with the wall acquires off-diagonal components

$$P_{x'y'} = -\delta P \sin \xi \cos \xi, \qquad (25)$$

where [see (21)]

 $\delta P = P_{xx} - P_{yy} = NT \left( 1 - v_0^2 / v_r^2 \right) \varepsilon_s.$ 

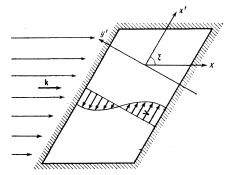
Let the vessel have the shape shown in Fig. 3. Then the condition for the equilibrium of the tangential forces acting on the gas layer parallel to the walls at the point y' takes at  $V \ll v_T$  and  $v_0 = 0$  the form

$$\eta \frac{\partial V_{\mathbf{x}'}}{\partial \mathbf{y}'} + \frac{2mN}{(2\pi)^{\gamma_{0}}} v_{\mathbf{r}} \Delta v_{\mathbf{x}0} \varphi_{\mathbf{s}} \left(1 - \frac{3}{2} \frac{V_{\mathbf{x}'}}{v_{\mathbf{r}}^{2}} \cos^{2} \xi\right) \sin \xi \cos \xi = \text{const.} \quad (26)$$

Actually (26) is the first integral of the Navier-Stokes integral in this case. From (23), (26), and the condition that the total momentum of the gas in the entire vessel be zero we obtain (see also Fig. 3)

$$V_{\mathbf{z}'}(\mathbf{y}') = -\left(\frac{3}{2}\right)^{\frac{y_{*}}{2}} \frac{v_{T}}{\cos\xi} \operatorname{th}\left(\frac{\varphi_{*}}{2\pi^{\frac{y_{*}}{2}}} \frac{\Delta v_{\mathbf{z}0}}{v_{T}} \frac{\mathbf{y}'}{l} \sin 2\xi\right).$$
(27)

As seen from (27), the macroscopic velocity V can be of the same order as the thermal. The characteristic value y' at which the velocity reaches the value  $v_T$  at  $\varphi_0 \sim 1$  is of the order of  $l/\varepsilon \sin 2\xi$ . The longitudinal dimension x' over which the selected gas volume is accelerated to a value of V of the order of the stationary velocity can be estimated at  $L_{y'}/\varepsilon \sin 2\xi$ , where  $L_{y}$  is the width of the cell. Obviously, if the cell length  $L_{x'}$  $\gg L_{y'}/\varepsilon \sin 2\xi$ , the values of V in the regions far from the end faces of the cell will be determined by (27), and



4

FIG. 3. Macroscopic gas-density fluxes resulting from oblique orientation of the vessel walls relative to the propagation direction of the exciting radiation.

stationary circulation of the gas is established in the cell. To understand the appearance of an angular momentum of the gas when the radiation is turned on, we can consider the pressure on the vessel walls (Fig. 3) at the time during which two or three collisions occur after the radiation is turned on, when Eqs. (21) are already valid, whereas the macroscopic motion in the gas has in fact not yet set in. It is easily seen then that since  $P_{xx} \neq P_{yy}$  the gas exerts on the vessel a torque that tends to rotate the vessel in a direction opposite to the circulation of V determined by (27).

We now consider the macroscopic fluxes, due to  $\partial \varphi / \partial y$  and considered qualitatively in Ref. 6. From (21b), assuming as before a specular wall, we easily obtain an equation for the dependence of the macroscopic velocity on the transverse coordinate in a rectangular or cylindrical vessel whose walls are parallel and perpendicular to the light propagation direction:

$$\frac{\partial V_x}{\partial y} = \frac{V_x - v_o}{(2\pi)^{\eta_h}} \frac{\Delta v_{x0}}{v_r} \exp\left\{-\frac{(V_x - v_o)^2}{2v_r^2}\right\} \frac{\partial \varphi_a \varphi_s}{\partial y}.$$
 (28)

In the case of cylindrical symmetry it is necessary to replace v in (28) by r. In the approximation  $|v_0 - V_x| \ll v_T$  we obtain from (28) and from the condition that the total momentum of the gas in the entire vessel be zero

$$V_{\pi}(y) = \frac{v_{\mathfrak{s}}}{(2\pi)^{1/\mathfrak{s}}} \frac{\Delta v_{\pi\mathfrak{s}}}{v_{T}} \left[ \overline{\varphi_{\mathfrak{s}} \varphi_{\mathfrak{s}}} - \overline{\varphi_{\mathfrak{s}}}(y) \varphi_{\mathfrak{s}}(y) \right], \tag{29}$$

where the bar denotes averaging over the transverse coordinate in the vessel. At  $v_0 \ge v_T$  we can regard (29) as an upper bound of  $V_x$ . We see that in contrast to the preceding case the limit of macroscopic velocity (29) is  $\sim_{\mathcal{E}} v_T \approx \Delta v_{x0}$ . It follows from (24) that the foregoing pertains to the fluxes due to the spatial inhomogeneity of the diagonal components of the pressure tensor.

We consider now the stationary state of the gas in the absence of macroscopic fluxes N. It follows from the foregoing that this state is possible if the vessel walls are only parallel or only perpendicular to the radiation propagation direction in the spatially homogeneous case, and also in the presence of only a transverse (at  $\varphi_a = 0$ ) or only a longitudinal inhomogeneity of the radiation. (We note that a stationary state with  $V(\mathbf{r}) = 0$  at  $\partial \varphi / \partial x$  $\neq 0$  or  $\partial \varphi / \partial_{y} \neq 0$  is possible only when near-wall perturbations of the distribution function are neglected.) The presence of the heat flux (22), if it depends on the coordinates, does not by itself upset the stability of the spatially homogeneous stationary state (9). Such a situation is realized, for example at  $\nabla \varphi = 0$  in a vessel with thermostatically controlled walls. However, if a thermally insulated wall is present in the path of the thermal flux (22), or if it is found to depend on the coordinates because of  $\nabla \varphi \neq 0$  even at  $\nabla N = 0$  and  $\nabla T = 0$ , the gas will obviously begin to be heated in the corresponding places, and will expand as a result. This process will continue until the opposing heat-conduction flux due to the produced temperature inhomogeneity the last term of (22)] balances the optically induced flux. A new stationary state is then established in the gas, with the temperature and density inhomogeneous. The density is determined by the equation of state (21) and by the fact that  $\partial P_{ij}/\partial x_j = 0$  in the absence of microscopic fluxes in the gas. To simplify the analysis of this state we consider separately the influence of the asymmetry of  $\varphi(v_x)$  relative to the center of the Doppler contour [the first term in the right hand side of (22a)], and also of the transverse [first term in (22b)] and of the longitudinal [second and third terms in (22a)] spatial inhomogeneities of the radiation in the external force field [fourth term in (22a)]. Taking the foregoing into account, and approximating the thermal conductivity  $\varkappa_T$  of the gas by the equation<sup>8</sup>

$$\varkappa_{r} = \frac{c_{\star}}{\sigma} \left(\frac{T}{m}\right)^{\eta_{\star}} = c_{\star} N l \upsilon_{r} \tag{30}$$

 $[c_x \text{ is a coefficient of order 1, and in the strong-collision model <math>c_x = 5/2$ , see (22)], the equation for the spatial distribution of the temperature can be written at  $\nabla \varphi = 0$  and  $\mathbf{a} = 0$  in the form

$$\frac{dT}{dx} = \frac{\varphi_a}{c_x' l_o} \frac{T_o (T_{cr} T_{min})^{\prime_a}}{T} \times \left(1 - \frac{T_{cr}}{T}\right) \exp\left\{-\frac{3}{2} \frac{T_{cr}}{T}\right\},$$
(31)

where  $c_x' = (\pi/54)^{1/2}c_x$ ,  $T_{\min} = m(\Delta v_{x0})^2$ ,  $T_0$  is the temperature, and  $l_0$  is the mean free path at the point  $x = x_0$ . The behavior of the solution of Eq. (31) is shown in Fig. 4. If the wall opposite to the direction of  $v_0$  (the right-hand wall at  $v_0 > 0$  and the left at  $v_0 < 0$ ) is thermostatically controlled, the temperature approaches  $T_{cr}$  rather than deviating from it.

At  $L_x \gg (l/\varphi_a)(T_{\min}/T_0)^{1/2}$  we have  $T \approx T_{cr}$  in the major part of the vessel with the gas, and is practically independent of  $T_0$ . Direct calculations show that a decrease of  $\partial T/\partial x$  with increasing T can be avoided by exciting the entire right (or left) half of the Doppler contour by broadband radiation. In this case the temperature increases (or decreases) linearly with increasing x.

As stated above, once the spatial distribution of the temperature is determined, the density distribution can be calculated from the condition  $\partial P_{xx}/\partial x$  using the formula [written here in a form that is valid in the case  $\nabla \varphi \neq 0$  considered below; here we must put in it  $\varepsilon_s(x) = \varepsilon_s(x_0)$ ]

$$N(x) = N_0 \frac{T_0}{T(x)} \frac{1 + \varepsilon_*(x_0)}{1 + \varepsilon_*(x)},$$
(32)

where  $N_0$  is the density at the point  $x_0$ , determined from the condition that the total number of the particles in the entire vessel be conserved:

$$\int_{x_0}^{x_0+L_x} N(x) \, dx = N_0' L_x, \tag{33}$$

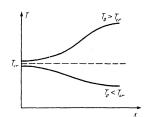


FIG. 4. Character of the coordinate dependence of the gas temperature in the case when the excitation is not symmetrical about the center of the Doppler line.

where  $N_0'$  is the gas temperature before the light is turned on.

The temperature inhomogeneity due to  $\nabla \varphi \neq 0$  and a  $\neq 0$  can be analyzed similarly with the aid of Eqs. (22). It is reasonable to restrict oneself to the case  $\varphi_a = 0$ , for otherwise macroscopic density fluxes arise at  $\partial \varphi / \partial y \neq 0$  [see (29)], and a temperature inhomogeneity unconnected with  $\partial \varphi / \partial x \neq 0$  arises at  $\partial \varphi / \partial x \neq 0$  [see (31)]. Knowing  $T(\mathbf{r})$  we can find  $N(\mathbf{r})$  from Eq. (32). In the presence of only transverse inhomogeneity we have

$$T(y, z) = T_{0} \left[ 1 + \frac{b_{1}}{5} \psi \left( \frac{4}{3} + \varphi_{*} \right) \varphi_{*} \Big|_{y_{0}, z_{0}}^{v, t} \right],$$

$$N(y, z) = N_{0} \left[ 1 + \frac{b_{1}}{5} \psi (2 - \varphi_{*}) \varphi_{*} \Big|_{y_{0}, z_{0}}^{v, t} \right].$$
(34)

In the presence of only longitudinal inhomogeneity and at

$$\frac{\Delta v_{x_0}}{v_T^3}\int (2-\varphi_s)\varphi_s a_x dx \ll 1$$

we have

$$T(x) = T_{0} \left\{ 1 + \psi \left[ \frac{2}{3} b_{s} + \frac{6}{5} b_{2} \frac{v_{0}^{2}}{v_{T}^{2}} \varphi_{s} + \frac{3}{5} b_{s} \frac{a_{x}}{v_{T}^{2}} (2 - \varphi_{s}) \right] \varphi_{s} \Big|_{\infty}^{x} \right\},$$
  

$$N(x) = N_{0} \left\{ 1 - \psi \left[ 2b_{s} + \frac{6}{5} b_{2} \frac{v_{0}^{2}}{v_{T}^{2}} \varphi_{s} + \frac{3}{5} b_{s} \frac{a_{x}}{v_{T}^{2}} (2 - \varphi_{s}) \right] \varphi_{s} \Big|_{\infty}^{x} \right\},$$
(35)

where  $b_5 = b_1 - \frac{3}{5}b_2v_0^2/v_T^2$ .

At 
$$\nabla \varphi = 0$$
,  $v_0 = 0$ , and arbitrary values of  $a_x$  we have

$$T(x)^{\eta_{1}} = T_{0}^{\eta_{1}} + \frac{9}{10(2\pi)^{\eta_{2}}} m^{\eta_{1}} \Delta v_{x0} (2-\varphi_{*}) \varphi_{*} \int_{x_{0}}^{\cdot} a_{x}(x) dx,$$
(36)

$$N(x) = N_0 \exp \left\{ \int_{x_0}^{x} \frac{T_0}{T(x')} \left[ 1 - \frac{3}{5} \left( \frac{T_0}{T(x')} \right)^{1/2} \psi(T_0) (2 - \varphi_*) \varphi_* \right] \frac{a_x(x')}{v_{x_0}^2} dx' \right\}.$$

The quantity  $N_0$  in (34)-(36), just as in the preceding case, is determined from the condition (33). We see that at  $v_0 = 0$  the gas is "pushed out" from the region of large *M* in the longitudinal direction, and "drawn" into this region in the transverse direction. With increasing deviation  $v_0$ , these effects are interchanged.

If one of the cell walls is a thermostat, then Eqs. (31) and (34)-(36) describe directly the temperature and density distributions, with  $T_0$  the temperature of the thermostat wall and  $x_0$  (or  $y_0$ ,  $z_0$ ) its coordinate. Turning on the light changes in this case not only  $P_{xx}$ , but the total pressure

$$P = \frac{1}{3}(P_{xx} + P_{yy} + P_{zz}) = NT$$

.

If the vessel is completely adiabatic, the energy of the gas in the entire vessel is not changed after the light is turned on:

$$\int N(x_{i}) T(x_{i}) dx_{i} = N_{0}' T_{0}' L_{x_{i}}$$
(37)

 $(T_0' \text{ is the gas temperature before the light is turned on) and Eqs. (33) and (37) allow us to calculate the quantities <math>N_0$  and  $T_0$  in Eqs. (31) and (34)-(36).

Let us dwell briefly on the experimental realization of the considered effects. In all the cited cases the changes in the quantities that characterize the macroscopic state of the gas (pressure, temperature, density) can be of the same order as the quantities themselves, and can be even larger in the presence of cumulative effects. The main difficulty lies in the need for optically exciting a sufficiently large number of molecules. The total number of excited particles  $N_{tote}$  is determined by the power p of the absorbed radiation and by the relaxation rate:

$$N_{tote} \approx p/\gamma \hbar \omega.$$
 (38)

We consider now the optimal arrangement of these particles, i.e., the optimal (at a given temperature) gas pressure. (We shall not touch upon the question of the optimal shape of the vessel, since it depends on the particular effect observed and on the specific variant of the experimental realization.) It follows from (14) that the depth of the dip in the particle velocity distribution is a maximum if

$$v \leq \gamma$$
, r. e.  $L^3 \geq \eta N_{tote} \sigma v_T / \gamma = \eta \rho \sigma v_T / \hbar \omega \gamma^2$ , (39)

where L is the characteristic dimension of the vessel,

$$\eta = N_{tot} / N_{tote} \approx (1 + 1/\varkappa_0) v_T / \Delta v_0$$

is the ratio of the total number  $N_{tot}$  to the number  $N_{tote}$  of the excited particles. Upon collision with the vessel walls, the excited and unexcited particles are approximately equally maxwellized. To obtain the largest possible depth of the dip in  $f(\mathbf{v})$  we must satisfy besides (39) also the conditions

$$l \leq L, \quad L^2 \leq \eta N_{tote} \sigma = \eta p \sigma / \hbar \omega \gamma. \tag{40}$$

Conditions (39) and (40) are compatible if

$$\eta N_{tote} \geq v_r^2 / \sigma \gamma^2, \quad \eta p / \hbar \omega \geq v_r^2 / \sigma \gamma.$$
(41)

We see that (41) can be satisfied at relatively small  $N_{tote}$ , for example, by defocusing the radiation so as to decrease  $\kappa_0$ , and by increasing the total amount of the gas if  $N_{tote}$  is to be preserved. The ratio  $N_{tote}/N_{tot}$  and the relative magnitude of the effects are then decreased. If (41) is not satisfied and (39) and (40) are incompatible, the second of these conditions can be dispensed with, and the vessel walls replaced by the unexcited gas. In this case, however, the influence of the spatial inhomogeneity of the radiation can not be eliminated.

It makes sense to increase the gas density to satisfy condition (40) only so long as the collision broadening does not exceed the Doppler broadening. This takes place at pressures of several torr for a vibrationalrotational transition that is at resonance with the emission of a  $CO_2$  laser.

If the conditions (39) and (40) are satisfied and  $\Delta \sigma \sim \sigma$ (the change of the cross section upon excitation from the ground state can be large<sup>10</sup>), the anisotropy of the pressure is of the order of the partial pressure  $\Delta P$  $\sim N_e T$  of the excited particles. For an allowed transition in the optical band ( $\gamma \sim 10^8 \text{ sec}^{-1}$ ) at  $p \sim 1$  W we have  $N_{\text{tote}}$  $\sim 10^{11}$ ,  $0.3 \times 10^{-2}$  cm  $< L \le 10^{-2}$  cm, and at  $T \sim 300^\circ$ ,  $\Delta P$  $\sim 1$  Torr. If we attempt to obtain the corresponding estimates for long-lived vibrational-rotational transitions in molecular gases (the real possibility of observing optically induced kinetic phenomena in this region was first pointed out in Ref. 11), we can arrive at the following. Condition (39) cannot be satisfied, inasmuch as at  $P > 10^{-3}$  Torr the value of  $\gamma$  is determined mainly by the quenching collisions, whose cross section is always smaller than the cross section for collisions that change the velocity of the molecule. At pressures on the order of several torr all the effects will therefore be decreased by a factor  $\gamma/\nu \sim 10^{-4}$ . To satisfy condition (39) we must operate with pressures  $\sim 10^{-3}$  Torr. In this region ( $\gamma \sim 10 \text{ sec}^{-1}$ ) we have:  $N_{\text{tote}} \sim 10^{20}$ ,  $L \sim 10^3$  cm and  $\Delta P \sim 10^{-6}$  Torr at  $\Delta \sigma/\sigma \sim 1$ , and  $\Delta P \sim 1$  and  $\Delta P \sim 10^{-7}$ Torr at  $\Delta \sigma/\sigma \sim 0.1$ . The use of deactivating additives can bring the ratio  $\gamma/\nu$  closer to unity also for relatively high pressures. By specifying the value  $\gamma \sim \nu$ , we have an expression for L:

$$L^{3} = \frac{\eta N_{tote} v_{\tau} \sigma}{\xi \gamma} = \frac{\eta p v_{\tau} \sigma}{\xi \gamma^{2} \hbar \omega}, \qquad (42)$$

where  $\xi = \nu/\gamma$ . The condition (40) takes the form

$$\eta N_{tote} \sigma > (v_T^2 / \xi \gamma)^2, \quad \text{i.e.,} \quad \eta p \sigma / \hbar \omega > v_T^2 / \xi^2 \gamma.$$
(43)

At  $\gamma \sim 10^6$  sec<sup>-1</sup> the condition (43) is satisfied, and we get for L and  $\Delta P$  the values  $L \sim 0.5$  cm and  $\Delta P \sim 0.1$ Torr at  $\Delta \sigma / \sigma \sim 1$ , and  $\Delta P \sim 10^{-3}$  Torr at  $\Delta \sigma / \sigma \sim 0.01$ .

<sup>1</sup> Here and elsewhere it is assumed, in cases where this is of principal importance, that the light propagates along the x axis.

<sup>2</sup> If  $P_{xy,z}$  is spatially inhomogeneous [see e.g., (21b)], the macroscopic fluxes N can arise also independently of the wall properties, for in this case the shear forces exerted on a

given gas layer by its two neighboring layers do not canel each other.

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Translated by J. G. Adashko