Photo-induced kinetic effects in a gas [1)]

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We examine a one-component gas in resonant interaction with an electromagnetic wave, taking into account the change in the collision cross section of the particles when one of them is excited. We show that a gas can be cooled by light-stimulated energy exchange between the gas and a thermostat. We also predict that a hydrodynamic gas flow will be established under the action of the radiation.

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In this letter we examine the behavior of a one-component gas whose particles interact resonantly with an electromagnetic plane wave whose frequency satisfies the condition $|\omega - \omega_{12}| \lesssim k v_T$, where ω_{12} is the frequency of a transition from the ground state, $k = \omega/c$, and $v_T = \sqrt{2T/M}$. The essential features are the velocity-selective excitation of the particles and the change in the collision cross section when one of the colliding particles is excited. The present interest in the effects which arise under these conditions was stimulated by Ref. 1. It is shown that a non-Maxwellian distribution is established and, as a result, there is a flow of kinetic energy and momentum in the gas.

We shall seek the velocity distribution of the particles $f(v)$ as a steady-state solution of the Boltzmann equation in the weak-nonequilibrium approximation:

$$f(\mathbf{v}) = f_0(\mathbf{v}) + \delta f(\mathbf{v}) \equiv f_0(1 + \chi(\mathbf{v})), \tag{1}$$

where $f_0(\mathbf{v})$ is the local-equilibrium Maxwell distribution. The correction δf should satisfy the normalization conditions:[2]

$$\overline{\delta f} \equiv \int d^3 v \delta f = 0; \quad \overline{\epsilon \delta f} = 0; \quad \overline{\mathbf{v} \delta f} = 0; \qquad (\epsilon = M\mathbf{v}^2/2). \tag{2}$$

Let $f_2(\mathbf{v})$ and $f_1(\mathbf{v})$ be the distributions of excited and unexcited particles, respectively. The condition of weak nonequilibrium is

$$\overline{f_2} << \overline{f_1}. \tag{3}$$

We introduce the probability of population of the upper level, $\rho_{22}(\mathbf{v})$:

$$f_2(\mathbf{v}) = \rho_{22}(\mathbf{v}) f(\mathbf{v}); \qquad f_1(\mathbf{v}) = (1 - \rho_{22}(\mathbf{v})) f(\mathbf{v}). \tag{4}$$

By virtue of condition (3), ρ_{22} is defined as a functional of the equilibrium distribution $f_0(\mathbf{v})$ and does not depend on the nonequilibrium correction δf. We write the collision term in the form

$$St(f)=\int d^3v_1 d\Omega\,|v-v_1|\{\sigma(\Omega)\,(f_1(v_1')f_1(v')-f_1(v)f_1(v_1))$$

$$+\sigma_e(\Omega)(f_1(v_1')f_2(v')+f_2(v_1')f_1(v')-f_1(v)f_2(v_1)-f_2(v)f_1(v_1))\}. \tag{5}$$

Substituting Eqs. (1) and (4) into Eq. (5) and linearizing with respect to $\chi(\mathbf{v})$, we obtain the equation

$$St(f)=\int d^3v_1 d\Omega\,|\mathbf{v}-\mathbf{v}_1|\,f_o(\mathbf{v})\,f_o(\mathbf{v}_1)\,\sigma(\Omega)\,[\alpha(\mathbf{v}_1')+\alpha(\mathbf{v}')-\alpha(\mathbf{v})-\alpha(\mathbf{v}_1)]=0;$$

$$d(\mathbf{v})=\chi(\mathbf{v})+\frac{\Delta\sigma}{\sigma}\rho_{22}(1+\chi(\mathbf{v});\quad \Delta\sigma=\sigma_e-\sigma. \tag{6}$$

We seek a solution of this equation in the model of "similar cross sections": $\dfrac{\sigma_e(\Omega,v_{\rm rel})}{\sigma(\Omega,v_{\rm rel})}=$ const., where Ω and $v_{\rm rel}$ are the scattering angle and speed of the particles in the center-of-momentum system. The solution is of the form[2]

$$\alpha(\mathbf{v})=A+B\epsilon+C(\mathbf{kv}); \tag{7}$$

$$\chi(\mathbf{v})=\frac{-\dfrac{\Delta\sigma}{\sigma}\rho_{22}(\mathbf{v})}{1+\dfrac{\Delta\sigma}{\sigma}\rho_{22}(\mathbf{v})}+\frac{A+B\epsilon+C(\mathbf{kv})}{1+\dfrac{\Delta\sigma}{\sigma}\rho_{22}(\mathbf{v})}. \tag{7a}$$

The constants A, B, and C are found from conditions (2). Taking for $\rho_{22}(\mathbf{v})$ the Lorentzian shape:[3]

$$\rho_{22}=\frac{1}{2}\frac{I}{I_s}\frac{\gamma_2^2}{(\omega-\omega_{12}-\mathbf{kv})^2+\gamma_2^2\left(1+\dfrac{I}{I_s}\right)}, \tag{8}$$

we obtain

$$\chi(\mathbf{v})=-\frac{1}{2}\frac{\Delta\sigma}{\sigma}\frac{I}{I_s}\frac{\gamma_2^2}{(\omega-\omega_{12}-\mathbf{kv})^2+\gamma_2^2\left(1+\dfrac{\sigma_e+\sigma}{2\sigma}\dfrac{I}{I_s}\right)}+A+B\epsilon+C(\mathbf{kv}). \tag{9}$$

(In the second term on the right-hand side of Eq. (7a) the denominator can be considered approximately equal to unity). The velocity distribution in the case of a narrow line $\dfrac{\gamma_2}{kv_T}\left(\dfrac{I}{I_s}\right)^{1/2}\ll 1$, is shown in Fig. 1.

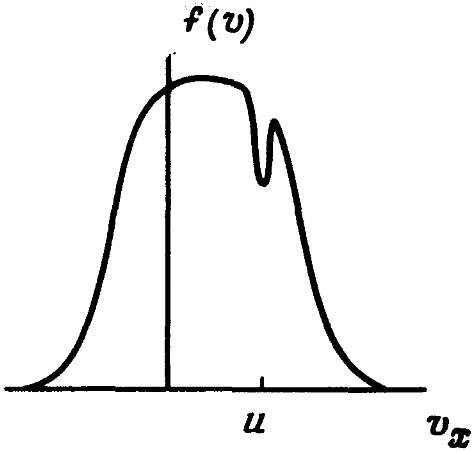

FIG. 1. Total velocity distribution of the particles. The radiation is propagating along the x axis; the velocity components v_y and v_z are arbitrary. It was assumed that $\sigma_e > \sigma$.

The "burning of a hole" through the total distribution of particles leads to additional clearing up of the transition. Defining an absorption coefficient Γ in accordance with Ref. 3, we obtain:

$$\Gamma I = -\frac{dI}{dx}, \quad \Gamma = \frac{\hbar\omega\gamma_1}{I}\int \rho_{22}(\mathbf{v}) f(\mathbf{v})\, d^3 v \approx \frac{\Gamma_{\text{unsat}}}{\left(1 + \frac{\sigma_e + \sigma}{2\sigma}\frac{I}{I_s}\right)^{1/2}} \tag{10}$$

where γ_1 is the rate of longitudinal relaxation.

We now examine the photo-induced thermoeffect, which lies in the fact that the establishment of a nonequilibrium distribution causes a flow of kinetic energy in the direction of propagation of the electromagnetic wave. Using Eqs. (1), (9), and (10), we obtain

$$\mathbf{q} \equiv \int \epsilon \mathbf{v}\, \delta f\, d^3 v = \frac{3}{2}\frac{\Delta\sigma}{\sigma}\frac{\mathbf{u}(T - T_c)}{\hbar\omega\gamma_1}\Gamma I; \qquad T_c \equiv \frac{Mu^2}{3}. \tag{11}$$

Here $\mathbf{u}$ is the resonant velocity, $\mathbf{u} \| \mathbf{k}$, $u_x = \frac{\omega - \omega_{12}}{k}$, $u_y = u_z = 0$.
This leads to a "slow" (by virtue of restriction (3) redistribution of the thermal energy over the volume occupied by the gas. The steady-state temperature distribution over the volume satisfies the equation

$$\operatorname{div}(\mathbf{q} - \kappa \nabla T) = 0, \tag{12}$$

in addition to the conditions of uniform pressure and conservation of total number of particles

$$nT = p = \text{const}, \qquad \int \frac{T_0}{T(\mathbf{r})}\frac{d^3 r}{V} = \frac{p_0}{p},$$

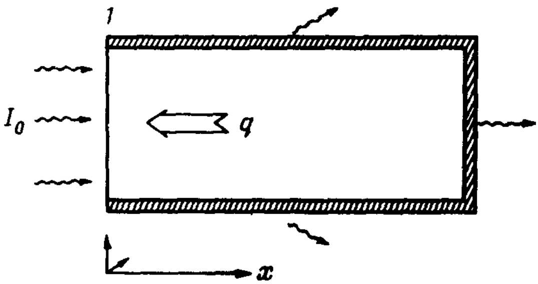

FIG. 2. Scheme for cooling the gas. End 1 is held at a constant temperature T_0; the other end and the sides are thermally insulated. All of the walls are optically transparent. $I(0)$ is the intensity of the incident radiation.

and also the boundary conditions. Here T_0 and p_0 are the initial temperature and pressure.

This effect can be used to cool the gas if one of the sides of the vessel containing it is brought into contact with a thermostat (Fig. 2). Equation (12) assumes the form

$$\frac{dT}{dx} = \lambda(T - T_c)\frac{1}{I(0)}\frac{dI}{dx}; \qquad \lambda \equiv \frac{3}{2}\frac{\Delta\sigma}{\sigma}\frac{|u|\,I(0)}{\hbar\omega\gamma_1\kappa};$$

$$T_0 > T_c; \qquad u < 0.$$

Assuming for the sake of simplicity that the thermal conductivity κ is constant, we obtain:

$$T(x) = T_c + (T_0 - T_c)\exp\left\{-\lambda\frac{I(0) - I(x)}{I(0)}\right\}. \tag{13}$$

The change in temperature will be appreciable under the condition $\lambda\Gamma L \gtrsim 1$, where L is the length of the vessel. This condition can be written in the form

$$\frac{\Delta\sigma}{\sigma_c}\frac{a\sigma}{\sigma_e + \sigma}\frac{|u|}{v_{T_0}}\left(\frac{I(0)}{I_s}\right)^{1/2}\Gamma_{\text{unsat}}L \gtrsim 1; \quad \sigma_c = \frac{\hbar\omega\gamma_1}{2I_s}; \quad \kappa \sim \frac{v_{T_0}}{\sigma}.$$

Here σ_c is the cross section for the absorption of radiation by a single particle in resonance.[3] The optimum intensity is determined by the condition $\left(\frac{I(0)}{I_s}\right)^{1/2}\frac{\gamma_2}{kv_{T_0}} \sim 1$, since for larger values of $I(0)$ the excitation is no longer velocity selective. Thus, under the conditions $\Delta\sigma/\sigma_c \sim 1, \sigma \sim \sigma_e$, and $\Gamma_{\text{unsat}}L \sim 1$, one can choose $|u| \sim \gamma_2/k$, which corresponds to a maximum attainable temperature of

$$T_{\text{lim}} \sim Mu^2 \sim Mc^2\frac{\gamma_2^2}{\omega^2}.$$

Taking $Mc^2 \sim$ 1—10 GeV, $\gamma_2 \sim 10^8$ sec^{-1}, and $\omega \sim 10^{15}$ sec^{-1}, we obtain $T_{\lim} \sim$ 0.1–1 K. Equilibrium is established in a time of the order of

$$t \sim \frac{C_v L \,|\, T - T_c \,|\, n}{|\, q(T_o)|} \sim \frac{L}{|\, u \,|} \left[\left(\frac{I}{I_s} \right)^{1/2} \frac{\gamma_2}{k v_{T_o}} \right]^{-1} .$$

Another manifestation of the nonequilibrium situation brought about by the radiation is the "photo-induced viscosity" effect, which arises in the presence of a light-intensity gradient perpendicular to the direction of **k**. If the average velocity of the excited particles is nonzero, the transport of particles in the direction of the gradient gives rise to a force which shifts layers of the gas with different light intensities relative to one another. This force arises owing to the different mean free paths of the excited and unexcited particles. This leads to the establishment of a steady-state hydrodynamic flow of the gas. In a closed volume this motion will be a closed circulation, and the total momentum of the gas will remain zero. To obtain the viscous stress tensor $\sigma'_{\alpha\beta}$, one must formally take into account terms in ρ_{22} that are due to the transport of particles. Assuming for simplicity that the conditions $I/I_s \ll 1; \gamma_2/kv_T \ll 1; \gamma_1 \tau_f \gg 1$ hold (τ_f is the average the time between collisions), we obtain

$$\rho_{22}(\mathrm{v}) = \rho_{22}^{(o)} - \gamma_1^{-1} (\mathrm{v} \nabla) \rho_{22}^{(o)} ,$$

where $\rho_{22}^{(0)}$ is given by expression (8). Evaluating the nonequilibrium correction according to Eq. (7a) in the approximation $\gamma_1^{-1} v_T |\nabla I| \ll I$, we obtain

$$\sigma'_{\alpha\beta} = - M \int v_\alpha v_\beta f(\mathrm{v})\, d^3 v ; \quad \sigma'_{xy} = - \eta' \frac{\partial \widetilde{v}}{\partial y} ; \quad \sigma'_{xz} = - \eta' \frac{\partial \widetilde{V}}{\partial z} ; \quad \sigma'_{yz} = 0;$$

$$\eta' = \gamma_1^{-1} p \frac{\Delta \sigma}{\sigma} ; \quad \widetilde{V} = \frac{1}{n} \int v_x \rho_{22}^{(o)}(v) f_o(v)\, d^3 v = u \frac{\Gamma I}{\hbar\omega\, \gamma_1 n} .$$

Under conditions of axial symmetry the Navier-Stokes equation is of the form

$$\frac{\partial V_x}{\partial t} + (\mathrm{V} \nabla)_x = - \frac{1}{\rho} \frac{\partial p_{xx}}{\partial x} - \nu' \frac{1}{r} \frac{d}{dr} \left(r \frac{d\widetilde{V}}{dr} \right) + \nu \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial V_x}{\partial r} \right).$$

Here $\nu' = \eta'/\rho$, and ν is the usual kinematic viscosity. Neglecting any inhomogeneity of the pressure, we obtain

$$V_x(r) = \frac{\nu'}{\nu} \widetilde{V}(r) + a . \tag{14}$$

The constant a is determined from the condition

$$\int \rho \cdot V_x(r)\, r\, dr = 0 \; .$$

The result (14) can be compared with the speed due to radiation pressure, namely $V_{\text{rad}} \sim \Gamma I \tau_f / cn M$:

$$\frac{V_x}{V_{\text{rad}}} \sim \frac{\Delta\sigma}{\sigma} (\gamma_1 \tau_{CB})^{-2} \frac{M\,|u|\,c}{\hbar\omega} \gtrsim 10^4,$$

if $\Delta\sigma \sim \sigma$, $\gamma_1 \tau_f \sim 1$. Thus the effect can be useful for measuring and controlling the degree of selectivity of the excitation of particles of a gas through the Doppler effect.

[1)] By request of the editorial staff, separate letters by the authors have been combined into this single article.

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