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Translational nonequilibrium of a gas in a resonant optical field

I. V. Krasnov and N. Ya. Shaparev

Computing Center, Siberian Branch of the Academy of Science of the USSR, Krasnoyarsk
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It is shown that a traveling electromagnetic wave can give rise to a strong translational nonequilibrium of a resonant impurity gas. Allowance is made for two nonequilibrium mechanisms, one of which is associated with a resonant radiation pressure and the other with the difference between the cross sections for elastic collisions of excited and normal atoms with a buffer gas. A study is made of kinetic, macroscopic, and optical manifestations of an induced translational nonequilibrium.

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1. INTRODUCTION

A translational nonequilibrium of a gas due to a resonant radiation pressure[1] has been investigated by us earlier.[2-4] A study of the diffusion of resonant atoms in the field of a traveling electromagnetic wave is reported in Ref. 4 with calculations of the macroscopic fluxes of atoms resulting in an inhomogeneous distribution of particles in a bounded region. It is assumed there implicitly that the elastic scattering frequencies are the same for excited and normal atoms. Gel'mukhanov and Shalagin[5] and Kalyazin and Sazonov[6] were the first to demonstrate the appearance of a flux of resonant atoms due to the difference between the scattering cross sections and to carry out calculations[5] under conditions such that the velocity distribution function of resonant particles differs little from the equilibrium form (homogeneous broadening).

We shall take into account each of these nonequilibrium factors, consider strongly nonequilibrium states of a gas (inhomogeneous braodening), and study their macroscopic manifestations.

2. DISTRIBUTION FUNCTION

In the quasiclassical limit the state of a resonant gas in the field of a traveling electromagnetic wave can be described by the kinetic equation for the density matrix in the mixed representation $\rho(\mathbf{r},\mathbf{v},t)$. We shall assume that the gas is sufficiently dilute and that the elastic scattering frequencies at the upper and lower levels ν_i are considerably less than the longitudinal and transverse relaxation rates $\nu_i \ll \gamma, \gamma_\perp$. In the case when the mass of a resonant atom is much greater than the mass of a buffer gas atom ($m \gg M$) the collision integrals can be written in the form

$$\mathrm{St}(\rho_{ii})=\nu_i\left[\mathrm{div}_v \mathbf{v}\rho_{ii}+\frac{u_0^2}{2}\frac{\partial^2\rho_{ii}}{\partial \mathbf{v}^2}\right]=\nu_i\hat{L}\rho_{ii}, \tag{1}$$

where u_0 is the most probable velocity of the resonant gas atoms. Then, the equations for the density matrix under steady-state conditions obtained ignoring the terms $\sim(\hbar k/mv)^2 \ll 1$ give the following kinetic equation for the distribution function of the atomic velocities $f = \mathrm{Tr}(\rho)$:

$$\mathbf{v}\frac{\partial f}{\partial \mathbf{r}}+\frac{\partial}{\partial v_x}\frac{F(v_x)}{m}f-\nu^+\hat{L}\left(\left[1-\frac{\nu^-}{\nu^+}\mathscr{L}(v_x)\right]f\right)=0, \tag{2}$$

$$F(v_x)=\frac{2\hbar k\gamma_\perp d^2|E_0|^2}{\hbar^2[(kv_x-\Delta)^2+\gamma_\perp{}^2+4\eta d^2|E_0|^2\hbar^{-2}]},\quad \nu^-=\frac{\nu_2-\nu_1}{2},\quad \nu^+=\frac{\nu_1+\nu_2}{2}, \tag{3}$$

$$\mathscr{L}(v_x)=\frac{(kv_x-\Delta)^2+\gamma_\perp{}^2}{(kv_x-\Delta)^2+\gamma_\perp{}^2+4\eta d^2|E_0|^2\hbar^{-2}},\qquad \eta=\frac{\gamma_\perp}{\gamma}, \tag{4}$$

where $\Delta=\omega-\omega_{21}$; ω and ω_{21} are, respectively, the field and transition frequencies; E_0 is the amplitude of the electric field; d is the matrix element of the dipole moment; $\mathbf{k}=\omega c^{-1}\mathbf{e}_x$ is the wave vector. The second term in Eq. (2) is associated with the resonant radiation pressure and the third term is due to collisions with the buffer gas.

In the case of a weak spatial inhomogeneity, the solution of Eq. (2) is

$$f(v_x)=\frac{c_1}{\varphi(v_x)}\exp\left\{-\frac{v_x{}^2}{u_0{}^2}+\frac{4\hbar k\gamma_\perp d^2|E_0|^2}{u_0{}^2m(\nu^+-\nu^-)\hbar^2ka}\operatorname{arctg}\frac{(kv_x-\Delta)}{a}\right\}, \tag{5}$$

where

$$\varphi(v_x)=1-\frac{\nu^-}{\nu^+}\mathscr{L}(v_x),\qquad a^2=\gamma_\perp{}^2+\frac{\nu^+}{\nu^+-\nu^-}\frac{d^2|E_0|^2}{\hbar^2}4\eta,$$

and c is the normalization constant.

3. DRIFT OF ATOMS

The radiation pressure force $F(v_x)$ and the inequality of the elastic collision frequencies generally result in the anisotropy of the distribution function producing a drift of atoms. In the case when $\nu^-/\nu^+\ll 1$ and

$$\frac{2\pi\hbar k\gamma_\perp}{m\nu^+}\frac{d^2|E_0|^2}{\hbar^2ka}\ll u_0{}^2$$

the distribution function $f(v_x)$ is nearly Maxwellian and we can represent it in the form

$$f=f_0+f_1,\qquad f_1\ll f_0=\frac{1}{\pi^{1/2}u_0}\exp\left(-\frac{v_x{}^2}{u_0{}^2}\right). \tag{6}$$

The velocity of the drift of atoms along the x axis is

$$u=u_1+u_2, \tag{7}$$

$$u_1=\pi^{1/2}\frac{2d^2|E_0|^2\hbar k\gamma_\perp H([\gamma_\perp{}^2/(ku_0)^2+4\eta d^2|E_0|^2/\hbar^2(ku_0)^2]^{1/2},\Delta/ku_0)}{m\nu^+\hbar^2ku_0(\gamma_\perp{}^2+4\eta d^2|E_0|^2\hbar^{-2})^{1/2}}, \tag{8}$$

$$u_2=-\frac{\nu^-}{\nu^+}\frac{1}{\pi^{1/2}u_0}\int\limits_{-\infty}^{\infty}\frac{4\eta d^2|E_0|^2\hbar^{-2}v_x}{(kv_x-\Delta)^2+\gamma_\perp{}^2+4\eta d^2|E_0|^2\hbar^{-2}}\exp\left(-\frac{v_x{}^2}{u_0{}^2}\right)dv_x, \tag{9}$$

where $H(\alpha,\beta)$ is the Voigt function[7] and u_0 is governed by the resonant radiation pressure. Radiation excites atoms with a definite projection of the velocity and the difference between the frequencies of elastic collisions ($\nu_1\neq\nu_2$) gives rise to a difference between the forces of friction acting on excited and normal particles moving across the buffer gas, the final result being a directional motion of the resonant gas at a velocity u_2.

We shall now consider the limiting values u_1 and u_2. In the case of weak fields and inhomogeneous broadening ($a\ll ku_0$) we have respectively

$$u\approx u_1\left(1-\eta\frac{\Delta 2\nu^-m}{\gamma_\perp\hbar k^2}\right),\qquad u_1\approx 2\frac{\hbar k\gamma_\perp d^2|E_0|^2\pi^{1/2}}{\hbar^2m\nu^+ku_0a}\exp\left(-\frac{\Delta}{ku_0}\right)^2. \tag{10}$$

In the case of strong fields ($a\gg ku_0,\Delta$) and homogeneous broadening, we obtain

$$u\approx u_1\left(1-2\eta\frac{\nu^-}{\gamma_\perp}\frac{mu_0}{\hbar k}\frac{\Delta ku_0}{a^2}\right),\qquad u_1=\frac{\hbar k\gamma_\perp}{m\nu^+}\frac{2d^2|E_0|^2}{\hbar^2a^2}. \tag{11}$$

If $\eta=1/2$, $\Delta=ku_0$, $\gamma_\perp=10^8$ sec^{-1}, $T_g=300$ °K, $n(\sigma_2-\sigma_1)/2=0.2$ cm^{-1}, $\lambda=0.5$ μ, $\nu^+/\nu^-=10$, $d^2|E_0|^2\hbar^{-2}=\gamma_\perp^2$, $ku_0=10^{10}$ sec^{-1}, the drift velocities are $u_1=10^2$ cm/sec and $u_2=u_1$. In strong fields we have $d^2|E_0|^2\hbar^{-2}=5(ku_0)^2$ and the velocities are $u_2=10^{-1}u_1$ and $u_1=10^{-1}u_0$.

4. PHOTOKINETIC BLEACHING OF A GAS

Attenuation of radiation in a gas can be found from the Maxwell equations and the equations for the density matrix using

$$\frac{1}{4\pi}\frac{d|E_0|^2}{dx}=k\,\mathrm{Im}(\langle P_0^*E_0\rangle), \tag{12}$$

where P_0^* is the amplitude of the polarization of an atom and the angular brackets describe averaging over the distribution function. We shall consider the case when $\Delta=0$ and ignore the influence of the radiation pressure. Then, according to Eq. (5) the averaging in Eq. (12) should be carried out in the following function:

$$f(v_x)=c_1\exp\left[-\frac{v_x{}^2}{u_0{}^2}\right]\left\{1-\frac{M^2-G^2}{(kv_x)^2+M^2}\right\}$$

$$M^2=\frac{G^2-(\nu^-/\nu^+)\gamma_\perp{}^2}{\varepsilon}, \tag{13}$$

$$G^2=\gamma_\perp{}^2+4\eta d^2|E_0|^2\hbar^{-2},\quad \varepsilon=1-\frac{\nu^-}{\nu^+}.$$

In the case when $(ku_0)^2\gg M^2\gg G^2(\varepsilon\ll 1)$ and $c_1\approx(\pi^{1/2}u_0)^{-1}$, we can easily see that the distribution function acquires a deep and narrow dip. Figure 1 shows the distribution function $f(v_x)$ for $G^2=10^{-2}ku_0$ and $M^2=10^{-1}ku_0$. This form of the distribution of the atomic velocities corresponding to weak fields and a large difference between the collision frequencies can be understood as follows. The action of a resonant magnetic field produces a dip in the velocity distribution at the lower level and a peak at the upper level, so that the following fluxes of atoms appear in the velocity space:

$$j_i=\left(-\frac{u_0{}^2}{2}\frac{\partial\rho_{ii}}{\partial v_x}-v_x\rho_{ii}\right)\nu_i, \tag{14}$$

which disperse these nonequilibrium structures. Since the rate of collapse of the peak and dip ("hole") are very different, the distribution function f exhibits a dip or a peak. In fact, if $\Delta\rho_{ii}$ is the characteristic height of a nonequilibrium structure in the distribution function at the i-th level, then bearing in mind the predominance of the diffusion terms in Eq. (14), we find that at low velocities ($|v_x|\ll u_0$) under steady-state conditions ($j_1+j_2=0$) we obtain

$$\Delta\rho_{11}\approx-(\nu_2/\nu_1)\Delta\rho_{22}, \tag{15}$$

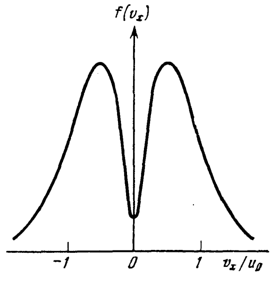

FIG. 1.

and, consequently,

$$\Delta f \approx \Delta\rho_{22}(1-\nu_2/\nu_1).$$

Thus, for $\nu_2 > \nu_1$, $\Delta f < 0$ the distribution function has a dip whereas for $\nu_2 < \nu_1$ it has a peak ($\Delta f > 0$).

In the latter situation where $\nu_2 \ll \nu_1$, the width of the narrow peak in the distribution function can be $M/k \ll G/k$.

We shall now carry out the averaging in Eq. (12) over the distribution function (13). Under the conditions given above and for

$$4\eta d^2|E_0|^2/\hbar^2 \ll \gamma_\perp^2 \ll M^2$$

the absorption coefficient is

$$k_f = k_0\left(\varepsilon + \frac{\gamma_\perp^2}{M^2}\right), \qquad k_0 = {}^3/_2\pi\lambda^2\frac{\gamma n}{\pi^{1/2}ku_0}, \tag{16}$$

where n is the concentration of normal atoms and k is the classical absorption coefficient. It follows from the above expression that the absorption coefficient decreases strongly. This weakening in the absorption coefficient (in the absence of saturation when $dE_0 \ll \hbar\gamma$) is not surprising and it is associated with the presence of a dip in the distribution function. If $\Delta \neq 0$, we can expect bleaching of the gas and formation of a flux. In Ref. 4 we also demonstrated the special properties of absorption of radiation when allowance is made for the radiation pressure.

We shall conclude by noting that these effects allow us to extend the use of coherent resonant light in control of the translational degrees of freedom of a gas. The deep narrow dip in the distribution function, induced by resonant light, may prove very useful in high-resolution spectroscopy.

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Photoionization of a hydrogenlike atom in a homogeneous electric field

V. D. Kondratovich and V. N. Ostrovskiĭ

Leningrad State University

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The dependence of the ionization cross section of a hydrogenlike atom in a constant homogeneous electric field on the frequency and on the polarization of the light wave is considered in a quasiclassical approximation. For light polarized parallel to the homogeneous field, the calculated cross section oscillates smoothly as a function of the light frequency at a photon energy close to the ionization potential of the atom, whereas in the case of perpendicular polarization there is practically no structure. The oscillations are not connected with above-barrier reflection or below-barrier resonances, but are due to the singularities of the electron motion in the final state. The oscillations are qualitatively interpreted in terms of the classical trajectories of the electron following the excitation. The positions of the minima of the structure agree with recently obtained experimental data.

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1. INTRODUCTION

The study of the stark effect on a hydrogenlike atom was one of the first problems of quantum mechanics[1,2] and has been the subject of many theoretical papers even in recent years (see, e.g., Refs. 3–5 and the bibliographies cited therein). Nonetheless, even certain qualitative aspects of the problem remain unclear. Thus, no theoretical investigation of the photoionization of an atom in a uniform electric field has been carried out to this day, and the oscillatory structure of the frequency dependence of this process was observed in experiment only recently.[6,7] Much interest in paid in general of late to the study of the stark effect on highly excited states, in view of the use of this effect for the study of Rydberg atoms, as well as of in view of the interesting fundamental features of this phenomenon (see, e.g., Ref. 8).

It is known that in the presence of a time-constant uniform electric field of intensity $\mathscr{E}$ the energy spectrum of the atom becomes continuous and extends over