Macroscopic rotation of a gas by light

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The interaction of optically absorbing particles with light puts these particles in a nonequilibrium distribution among magnetic sublevels. The effect of this deviation from equilibrium on the macroscopic rotation of an absorbing gas with respect to a buffer gas is analyzed. Particles illuminated by circularly polarized light acquire a magnetic dipole moment, which gives rise to a force which rotates the gas. This force is analogous to the Magnus force in classical hydrodynamics. Linearly polarized light gives the particles a magnetic quadropole moment with an alignment axis perpendicular to the axis of the light beam. As a result, the aligned particles experience an anisotropic friction force with a nonzero curl. A system of four vortices arises in the plane perpendicular to the axis of the light beam. The vortices and the nonequilibrium increment in the density of absorbing particles decay in a power-law fashion outside the light beam.

§1. INTRODUCTION

The absorption of light results in an exchange of angular momentum between the photon and the gas particle, and the distribution of particles among magnetic sublevels deviates from equilibrium. The absorption of circularly polarized light, for example, gives the particles a magnetic dipole moment,[1] a magnetic quadrupole moment, and higher multipole moments.[2,3] We are naturally interested in how the field-induced microscopic moment of the particles is related to the macroscopic angular momentum of the gas. It is clear from conservation of angular momentum that the internal moment of the particles must act through collisions to cause a macroscopic rotation of the gas.[4] We know, however, that this effect is not described by the Boltzmann collision integral S. Kagan and Maksimov[5,6] have shown that a correlation arises between the microscopic and macroscopic moments only when the nonlocal nature of the collision integral is taken into account. The reason is that the local collision integral S conserves the macroscopic angular momentum of the gas. From momentum conservation we find that the macroscopic angular momentum is conserved:

$$\mathrm{Sp}\,([\mathbf{rp}]S)=[\mathbf{r},\,\mathrm{Sp}\,(\mathbf{p}S)], \tag{1.1}$$

where Sp denotes a trace over the quantum numbers of the particles together with an integral over the momenta **p**. The effects which stem from the nonlocal nature of the collision integral are small quantities which are smaller by an additional factor on the order of the "gaseousness" parameter d/l, where d is the interaction range, and l is the mean free path of the particles. Accordingly, if we assume that the light rotates the gas only by virtue of the nonlocal nature of the collision integral we find that the rotation of the gas is extremely weak. Specifically, the typical macroscopic rotation velocity u will be small in comparison wih the average thermal velocity of the gas absorbing the light[6]:

$$u\sim\bar{v}d/R, \tag{1.2}$$

since the ratio of d to the macroscopic scale radius $R>l$ is small (this macroscopic radius might be, for example, the cross-sectional radius of the light beam).

A two-component gas, consisting of a component which absorbs light and one which does not interact with light (a buffer gas), has a property which distinguishes it qualitatively from a single-component gas: By virtue of (1.1), when the momentum of the mixture is zero the macroscopic angular momentum of the mixture will also be zero. However, the macroscopic angular momenta of the individual components may be nonzero, since, according to (1.1),

$$\mathrm{Sp}\,([\mathbf{rp}]S_1)=[\mathbf{r},\,\mathrm{Sp}\,(\mathbf{p}S_1)]=-[\mathbf{r},\,\mathrm{Sp}\,(\mathbf{p}S_2)], \tag{1.3}$$

if the momenta of the individual components are not zero, as is the case, for example, in photoinduced diffusion[7] or photoinduced drift.[8] Here $S=S_1+S_2$, where S_i is the Boltzmann collision integral for mixture component i. We see from (1.3) that, in contrast with the rotation of the gas as a whole, a rotation of the individual components arises even in first order in the gas parameter d/l, and the corresponding rotation velocity is greater by a factor of l/d than the rotation velocity of the mixture as a whole, (1.2). In this paper we analyze the vortex motions of the components of a gas mixture which are excited by light which is resonant with one component of the mixture.

§2. DIFFUSION OF PARTICLES WITH AN EQUILIBRIUM DISTRIBUTION AMONG MAGNETIC SUBLEVELS

The macroscopic behavior of the light-absorbing gas is described by the equations

$$\frac{\partial}{\partial t}\rho(\mathbf{r})+\mathrm{div}\,\mathbf{j}=0,\quad \frac{\partial}{\partial t}\mathbf{j}+\frac{\bar{v}^2}{2}\nabla\rho(\mathbf{r})=\mathbf{F}, \tag{2.1}$$

where $\rho(\mathbf{r})$ and $\mathbf{j}$ are the mass density and mass flux density of the absorbing particles, and $(1/2)\bar{v}^2\rho(\mathbf{r})$ is the pressure. The buffer gas, which does not interact with the light, exerts a friction force on the absorbing particles, given by

$$\mathbf{F}=m\,\mathrm{Sp}\,(\mathbf{v}(S_m+S_n)).$$

Here m is the mass of the absorbing particle, and S_i is the collision integral of an absorbing particle in the state $i=m,n$ for collisions with the buffer particles.

Before we take up the effects resulting from the creation of a nonequilibrium distribution of particles among their magnetic sublevels M by the light, we wish to recall the structure of the frictional force for the case of an equilibrium distribution in M (Ref. 7). As the particles absorb light, they

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go from the ground state n to the excited state m. The absorbing particles are thus a mixture of particles of two species: n and m. We restrict the present discussion to the case of exact resonance between the light frequency ω and the frequency ω_{mn} of the m-n transition ($\omega = \omega_{mn}$). We also assume that the medium is optically thin and that the lifetime ($2/\Gamma_m$) of the excited state is considerably shorter than the time spent by an absorbing particle in the light beam. The latter assumption means that excited particles are present only in the light beam. On the other hand, there are fewer unexcited particles in the beam than outside it. We know that spatial inhomogeneity of the density gives rise to diffusion fluxes. In this case, therefore, two oppositely directed fluxes arise: a flux ($\mathbf{j}_m$) of excited particles away from the beam and a flux ($\mathbf{j}_n$) of unexcited particles into the beam. The absorbing particles experience a friction force[7]

$$\mathbf{F}_0=-\nu_m\mathbf{j}_m-\nu_n\mathbf{j}_n=-(\nu_m-\nu_n)\,\mathbf{j}_m-\nu_n\mathbf{j}. \tag{2.2}$$

Here ν_i is the rate at which the particles of species $i = m, n$ collide with the buffer particles [expression (A.1) from the Appendix], and $\mathbf{j} = \mathbf{j}_m + \mathbf{j}_n$ is the total mass flux density of the absorbing particles. If $\Gamma_m \ll \nu_m$, the flux density of excited particles in the approximation linear in the light intensity is[7]

$$\mathbf{j}_m=-\frac{\bar{v}^2\rho}{4\nu_m}\nabla\varkappa,\quad \varkappa=\frac{4}{(2J+1)\Gamma\Gamma_m}\sum_\sigma |G_\sigma|^2, \tag{2.3}$$

where ρ is the equilibrium density of absorbing particles, which we assume to be small in comparison with the density of buffer particles; $\varkappa$ is the saturation parameter for the transition between the two degenerate levels m and n with identical spins $J_m = J_n = J$; $G_\sigma = E_\sigma \langle m\|d\|n\rangle/2\sqrt{3}\hbar$; E_σ is the component of the electromagnetic field; $\langle m\|d\|n\rangle$ is the reduced matrix element of the dipole moment of the transition m-n; Γ is the homogeneous width of the absorption line, which we assume to be large in comparison with the Doppler width $k\bar{v}$; and $\mathbf{k}$ is the light wave vector. To streamline the notation we assume that the difference in collision rates $\nu_m - \nu_n$ is small in comparison with ν_n. We will accordingly retain the index of the quantum state (i) on the collision rate ν_i only in the difference $\nu_m - \nu_n$. We then find from (2.2) and (2.3)

$$\mathbf{F}_0=\left(\frac{\nu_m-\nu_n}{4\nu}\right)\rho\bar{v}^2\nabla\varkappa-\nu\mathbf{j}. \tag{2.4}$$

The first term in the expression for $\mathbf{F}_0$ is the potential function. The steady-state solution of Eqs. (2.1) with the force $\mathbf{F} = \mathbf{F}_0$ is accordingly irrotational:

$$\mathbf{j}=0,\quad \rho(\mathbf{r})=\rho_0+\left(\frac{\nu_m-\nu_n}{2\nu}\right)\rho\varkappa(\mathbf{r}). \tag{2.5}$$

It describes a diffusive attraction of particles into the light beam ($\nu_m > \nu_n$) or repulsion from the light beam ($\nu_m < \nu_n$).[7] Here ρ_0 is the mass density of the absorbing particles outside the light beam. This effect stems from the difference between the rates at which the fluxes of excited and unexcited particles are slowed ($\nu_m \neq \nu_n$).

We turn now to the basic problem of our study: determining how the deviation from an equilibrium distribution among magnetic sublevels caused by the light affects the density of the frictional force $\mathbf{F}$.

§3. DIFFUSION OF ORIENTED PARTICLES; THE MAGNUS EFFECT

As particles absorb circularly polarized light, they acquire a magnetic dipole moment, characterized by the orientation vector $\boldsymbol{\rho}$ (Refs. 2, 3, and 9). Like the excited particles, the magnetically oriented particles are present only in the light beam under the conditions specified above. By analogy with (2.3), their flux density is proportional to $\nabla\varkappa$, and their density to $\varkappa$. The oriented particles thus diffuse out of the field region at a velocity $\mathbf{u}$ given approximately by

$$\mathbf{u}=-(\bar{v}^2/2\nu)\,\nabla\ln\varkappa. \tag{3.1}$$

We know from hydrodynamics, however, that if a cylinder which is rotating at an angular velocity $\boldsymbol{\Omega}$ moves through a gas at a velocity $\mathbf{u}$ it will experience a Magnus force $\propto [\mathbf{u}\boldsymbol{\Omega}]$. There is an analogy here between the diffusing magnetically oriented particles and a rotating cylinder which is moving at a velocity $\mathbf{u}$. According to this analogy, the absorbing particles should experience a friction force in addition to that given by (2.4):

$$\delta\mathbf{F}_1=\nu_1[\mathbf{u}\boldsymbol{\rho}]=-\frac{\nu_1\bar{v}^2\xi_1\rho}{2\nu\varkappa}\,\mathrm{rot}\left(\frac{\mathbf{k}}{k}\varkappa\right). \tag{3.2}$$

The proportionality factor ν_1 in (3.2) has the meaning of a collision rate [see expression (A.2)]. The pseudoscalar $\xi_1 = \boldsymbol{\rho}\mathbf{k}/\rho k \propto \varkappa$ is proportional to the ratio of the density of the oriented particles to the total density of absorbing particles. When the direction of the circular polarization is reversed, ξ_1 changes sign. For linearly polarized light we have $\xi_1 = 0$. Solving Eqs. (2.1) with the frictional force $\mathbf{F} = \mathbf{F}_0 + \delta\mathbf{F}_1$ in the steady state, we find a distribution of the density of absorbing particles which is the same as (2.5). Since $\delta\mathbf{F}_1$ is not a potential force, however, the total flux density $\mathbf{j}$ is nonzero:

$$\mathbf{j}=-\frac{\bar{v}^2\nu_1\xi_1\rho}{2\nu^2\varkappa}\,\mathrm{rot}\left(\frac{\mathbf{k}}{k}\varkappa\right). \tag{3.3}$$

This solution describes a diffusion rotation of the absorbing gas in the plane perpendicular to $\mathbf{k}$. Momentum conservation requires that the buffer gas rotate in the opposite direction.

§4. DIFFUSION OF ALIGNED PARTICLES

How does the magnetic quadrupole moment of the particles affect the motion of the gas? To eliminate the Magnus effect, discussed above, we assume that the light is linearly polarized. In the field of linearly polarized light, particles with degenerate levels acquire a magnetic quadrupole moment or alignment.[2,3,9] From the standpoint of collisions, aligned particles behave as ellipsoids with a major axis parallel to $\mathbf{n} = \mathbf{E}/E$. The particles aligned in the light beam diffuse out of the field region at the velocity (3.1), experiencing friction with the buffer particles in the process. The motion of these oriented ellipsoids is similar to the motion of a sailboat whose keel is collinear with $\mathbf{n}$ and whose sail is perpendicular to the wind direction. In our case, the wind direction is given by the velocity **u** in (3.1). In the frictional force **F**, the term collinear with $\nabla\varkappa$, in (2.4), is thus accompanied by a term

$$\delta\mathbf{F}_2=\nu_2\xi_2\rho\mathbf{n}(\mathbf{nu})=-\frac{\nu_2\xi_2\rho\bar{v}^2}{2\nu\varkappa}\mathbf{n}(\mathbf{n}\nabla\varkappa), \qquad (4.1)$$

where $\xi_2 \propto \varkappa$ is roughly equal to the ratio of the density of aligned particles to the total density of absorbing particles. The proportionality factor ν_2 has the dimensionality of a collision rate [see (A.3)]. It can be shown that the force in (4.1) vanishes for circularly polarized light.

The density of the frictional force in (4.1), in contrast with (3.2), has a potential part as well as a rotational part. The rotational part of (4.1) describes a steady-state rotational flux of particles, while the potential part gives the anisotropic increment $\delta\rho(\mathbf{r})$ in solution (2.5) and also a small (in the limit $|\delta\mathbf{F}_2|/|\mathbf{F}_0|\ll 1$ isotropic contribution to $\rho(\mathbf{r})$. Let us examine the steady-state solution of Eqs. (2.1) with the frictional force $\mathbf{F}=\mathbf{F}_0+\delta\mathbf{F}_2$. The quantity $\delta\rho(\mathbf{r})$ obeys the following equation in a cylindrical coordinate system with z axis parallel to the axis of the axisymmetric light beam:

$$\delta\rho(\mathbf{r})=n(r)\cos(2\varphi),$$
$$\left(\frac{\partial^2}{\partial r^2}+\frac{1}{r}\frac{\partial}{\partial r}-\frac{4}{r^2}\right)n(r)=-\frac{\nu_2\xi_2\rho}{2\nu\varkappa}\left(\frac{\partial^2}{\partial r^2}-\frac{1}{r}\frac{\partial}{\partial r}\right)\varkappa, \qquad (4.2)$$

where r is the distance from the axis of the light beam to the observation point, and the angle φ is measured from the direction of **n**. Assuming that the cross-sectional radius (R) of the cell holding the gas is large in comparison with the radius (a) of the light beam, and noting that the flux density of the particles vanishes at the cell boundaries, we find the following solution of Eq. (4.2) for a Gaussian beam, $\varkappa(r)=\varkappa(0)\exp(-r^2/a^2)$:

$$n(r)=\frac{\nu_2\xi_2\rho\varkappa(0)}{2\nu\varkappa}\zeta^{-2}[1-(1+\zeta^2)\exp(-\zeta^2)],\quad \zeta=\frac{r}{a}. \qquad (4.3)$$

It can be seen from this expression that in the limit $r\to 0$ the quantity $\delta\rho(\mathbf{r})$ tends toward zero in proportion to r^2, while at $r\gg a$ it has the behavior r^{-2}; i.e., outside the light beam, $\delta\rho(\mathbf{r})$ decays by a power law (not exponentially!). This behavior of $\delta\rho(\mathbf{r})$ is qualitatively different from solution (2.5), which reproduces the transverse distribution of the light intensity.

The existence of an anisotropic, weakly damped increment $\delta\rho(\mathbf{r})$ stems from the diffusive rotational fluxes which carry particles out of the light beam. These fluxes appear because of the frictional force $\delta\mathbf{F}_2$ in (4.1), which is generated by the light and which acts on the absorbing particles. Substituting $\delta\rho(\mathbf{r})$ from (4.2) and (4.3) into the second equation in (2.1), we find expressions for the radial (j_r) and angular (j_φ) components of the total flux **j**:

$$j_r=\frac{\nu_2\xi_2\rho\varkappa(0)\bar{v}^2}{2\nu^2\varkappa a}\zeta^{-3}[1-(1+\zeta^2)\exp(-\zeta^2)]\cos(2\varphi),$$
$$j_\varphi=\frac{\nu_2\xi_2\rho\varkappa(0)\bar{v}^2}{2\nu^2\varkappa a}\zeta^{-3}[1-(1+\zeta^2+\zeta^4)\exp(-\zeta^2)]\sin(2\varphi). \qquad (4.4)$$

Outside the light beam, the flux in (4.4) behaves as r^{-3}. As

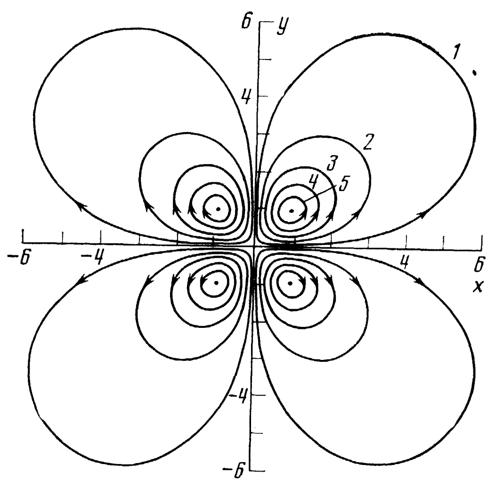

FIG. 1. Current lines. The x axis funs parallel to **n**. $\xi_2\nu_2>0$. 1—$\xi_0=0.2$; 2—$\xi_0=0.4$; 3—$\xi_0=0.6$; 4—$\xi_0=0.8$; 5—$\xi_0=1$.

the axis of the light beam is approached ($r\to 0$), the flux tends toward zero in proportion to r. We can construct current lines for (4.4):

$$dr/j_r=r\,d\varphi/j_\varphi.$$

The solution of this equation is

$$\sin 2\varphi=\Theta(\zeta_0)/\Theta(\zeta),$$
$$\Theta(\zeta)=\zeta^{-2}[1-(1+\zeta^2)\exp(-\zeta^2)]. \qquad (4.5)$$

The integration constant ζ_0 represents the minimum distance from the axis of the light beam to the current line. Current lines corresponding to Eq. (4.5) are shown in Fig. 1. The centers of the vortices are the points at which the minimum and maximum distances from the axis of the light beam to the current line are the same. These central points lie on the bisectors of the four quadrants, at distances $\zeta_0\approx 1.339$ from the axis of the light beam.

CONCLUSION

We have examined two mechanisms for the macroscopic rotation of a gas by light. In the first, the rotation of the gas, caused by a magnetic orientation of the particles induced by the light, reaches a maximum for circularly polarized light and does not occur in the case of linearly polarized light. The rotation of the gas due to the magnetic quadrupole moment of the particles, in contrast, is absent in the case of circularly polarized light and maximal for linearly polarized light.

To detect both effects, one could observe the motion of macroscopic particles (dust particles) entrained in the vortex flow of a gas. The absorbing and buffer gases, rotating in opposite directions, would exert a pressure

$$P\sim(j-\rho u_0)\bar{v}+(j_b-\rho_b u_0)\bar{v}_b$$

(the subscript b specifies the buffer gas) on dust particles with dimensions smaller than or comparable to the mean free path ($1=\bar{v}\nu$). We find the velocity (u_0) of a dust particle from the condition $P=0$, using momentum conservation, $\mathbf{j}=\mathbf{j}_b=0$:

$$u_0 = j\bar{v}\,[1-(m/m_b)^{1/2}]\,(\rho\bar{v}+\rho_b\bar{v}_b)^{-1}.$$

With $\rho\sim\rho_b$ and $|m-m_b|\sim m$ we then find $u_0/\bar{v}\sim\nu_\alpha\xi_\alpha l/\nu a$ ($\alpha=1,2$) from (3.3) and (4.4). For an atomic vapor, the degree of orientation of alignment of the particles, ξ_α, may be quite large. For our estimates we use the value $\xi_\alpha\sim10^{-2}$, which is by no means a record value. Also assuming $\nu_\alpha/\nu\sim10^{-2}$, we find (for $\bar{v}\sim10^5$ cm/s, $\nu\sim10^7$ s^{-1}, and $a\sim10^{-1}$ cm) $l\sim10^{-2}$ cm and $u_0\sim1$ cm/s. In the case of the Magnus effect, the dust particles would reverse direction when right-hand circularly polarized light was replaced by left-hand circularly polarized light.

The vortex motions caused by the alignment of the particles (Fig. 1) could also be observed indirectly, by optically detecting the spatial distributions of the density of the absorbing particles, as has recently been done, for example, in an experiment with sodium vapor,[10] where diffusive extraction was first observed.[7] If a highly anisotropic [see (4.2)], weakly damped [see (4.3)] tail is observed on the transverse distribution of the density of the absorbing particles in such an experiment, it would also be proof of the occurrence of the effect.

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APPENDIX

A rigorous solution of the kinetic equations incorporating degeneracy of the particle levels[3] leads to the following expressions for the collision rates:

$$\nu_i=\hat{L}(1-\cos\theta)\sigma_i(00\mathbf{u}_1|00\mathbf{u}),\quad i=m,\ n, \tag{A.1}$$

$$\nu_1=\hat{L}\sin\theta\,\mathrm{Im}\,\sigma(00\mathbf{u}_1|11\mathbf{u}), \tag{A.2}$$

$$\nu_2=\hat{L}[(1-\cos\theta)\sigma(00\mathbf{u}_1|20\mathbf{u})+\sqrt{3/2}\sin\theta\sigma(00\mathbf{u}_1|21\mathbf{u})], \tag{A.3}$$

where

$$\hat{L}=\frac{8\pi}{3}\left(\frac{\mu}{m\bar{v}}\right)^2 N_b\int_0^\infty du\int d\Omega u^5 W_\mu(u),\quad \mu=\frac{mm_b}{m+m_b}$$

$$\sigma_i(\varkappa q\mathbf{u}|\varkappa_1q_1\mathbf{u}_1)=\sum_{MM'}\sum_{M_1M_1'}(-1)^{M'-M_1'}\langle JMJ-M'|\varkappa q\rangle \times\langle JM_1J-M_1'|\varkappa_1q_1\rangle f_i(M\mathbf{u}|M_1\mathbf{u}_1)f_i^*(M'\mathbf{u}|M_1'\mathbf{u}_1);$$

N_b is the density of buffer particles, $d\Omega=2\pi\sin\theta d\theta$, θ is the angle between $\mathbf{u}$ and $\mathbf{u}_1$, $W_\mu(u)$ is the Maxwellian distribution for particles of mass μ, $f_i(M\mathbf{u}|M_1\mathbf{u}_1)$ is the amplitude for the scattering of an absorbing particle in state i by a structureless buffer particle accompanied by a change in both the direction of the relative velocity $\mathbf{u}$ and the projection M of the moment of the absorbing particle, and the angle brackets specify the coefficients of a vector addition. In (A.1)–(A.3), f_i is to be understood as the scattering amplitude in a coordinate system with z axis parallel to $\mathbf{u}$ and with x axis lying in the $\mathbf{uu}_1$ plane. In this paper we have assumed that $(\nu_m-\nu_n)/\nu_n$, ν_1/ν_n, and ν_2/ν_n are all small. We have accordingly omitted the index specifying the quantum state (i) from the cross sections σ in the expressions for ν_1 and ν_2.

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