Separation of a gas according to atomic spin projections in a helically polarized external field

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A solution is found for the steady interaction between a helically polarized field and atoms in a ground state with angular momentum $J_0 = 1/2$. It is shown that as a result of optical selfpumping, the populations of the sublevels of the ground state are independent of the total intensity of the light (they depend only on the intensity ratio of the circular components!) and depend selectively on the velocities of the atoms. It is found in the collisionless model that the velocity distribution of ground-state atoms with a given spin projection departs from equilibrium, while the velocity distribution of all the atoms without regard to spin remains thermal. A boundary-value problem is solved for the drift of polarized spins relative to one another in a closed tube. This drift causes a spatial separation of atoms into components with different spin projections $\pm 1/2$ at opposite ends of the tube. The analogy between this effect and photoinduced drift is discussed.

1. The selective influence of light on the translational motion of atoms, which can give rise to macroscopic effects in gases, is currently attracting a great deal of interest.¹⁻³ These studies, however, have not considered the possibility of a simultaneous selective influence on the translational and internal degrees of freedom of the atoms, e.g., the spin (angular momentum) in a one-component gas.

In this paper it is shown that a selective (with respect to velocity and spin projection) interaction of atoms with resonance radiation is possible even in the absence of the buffer gas if the optical orientation of the ground state by a field of special configuration, such as a helically polarized wave, is correctly taken into account. Such a polarization of the pump field can be obtained by the superposition of two counterpropagating orthogonal circularly polarized waves⁴

$$\mathbf{E} = e^{-i\omega t} \left(\mathbf{E}_{-} e^{+ikz} + \mathbf{E}_{+} e^{-ikz} \right) + \text{ c.c.}$$
$$\mathbf{E}_{\pm} = \pm 2^{-\frac{1}{2}} E_{\pm} \left(\mathbf{e}_{\pm} \pm i \mathbf{e}_{y} \right). \tag{1}$$

Here \mathbf{E}_{\pm} specifies the σ_{\pm} polarizations of the counterpropagating waves. We show below that a one-component gas in such a field, while remaining in equilibrium as a whole, acquires a latent velocity disequilibrium in the populations of the magnetic sublevels. This effect is manifested in an extra optical orientation and a relative drift of the oriented atoms with different spin projections.

2. Let us consider the interaction of the field (1) with two-level atoms in an unpolarized ground state with angular momentum $J_0 = 1/2$. The first excited state, also having angular momentum $J_1 = 1/2$, is separated from the ground state by a frequency ω_0 which is in resonance with the field frequency: $\omega = \omega_0 + \delta(|\delta| \ll \omega_0)$. A typical example of transitions of this sort are electronic transitions in alkali metals and transitions between hyperfine components in certain isotopes of rare earth elements with $F_0 = F_1 = 1/2$. As we see from Fig. 1, for this type of transition there is no coherent interaction of the counterpropagating σ_+ polarized components. Then the equations of motion for the density matrix elements for the interaction of the atoms with field (1) can be reduced to balance equations for the sublevel populations

$$\langle \pm 1/2 | \hat{\rho}^1 | \pm 1/2 \rangle \equiv N_{\pm}$$

and

$$\langle \pm 1/_2 | \hat{\rho}^0 | \pm 1/_2 \rangle \equiv N_{\pm}^0$$

of the excited and ground states, respectively⁵:

$$\left(\frac{\partial}{\partial t} + \gamma + v\nabla\right) N_{\pm}^{i} = \Gamma^{\pm h} (N_{\pm}^{i} - N_{\mp}^{0}), \qquad (2)$$

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right)N_{\pm}^{0} - \left[\frac{1}{3}\gamma N_{\pm}^{1} + \frac{2}{3}\gamma N_{\mp}^{1}\right] = -\Gamma^{\mp k}(N_{\mp}^{1} - N_{\pm}^{0}).$$
(3)

In these equations γ is the rate of radiative decay of the excited state to the ground state, and $\Gamma^{\pm k}(v)$ is the pumping rate, which is proportional to the intensities of the σ_{\pm} components with allowances for the Doppler shift $\mathbf{k} \cdot \mathbf{v}$:

$$\Gamma^{\pm k}(v) = \frac{I}{2} \cdot (1 + \xi_2) g_{\pm}(v),$$

$$g_{\pm}(v) = [(\gamma/2)^2 + (\delta \pm \mathbf{k} \mathbf{v})^2]^{-1},$$
(4)



FIG. 1. Interaction scheme of an atom at the transition $1/2 \rightarrow 1/2$ with field (1) in a cycle of the optical pump. The heavy lines with the two arrows correspond to the counterpropagating σ_{\pm} components of the wave. The wavy lines represent spontaneous decay.

$$I = \frac{1}{3} \left| \frac{d^{10}}{\hbar} \right|^2 (|E_+|^2 + |E_-|^2),$$

$$\xi_2 = (|E_+|^2 - |E_-|^2)/(|E_+|^2 + |E_-|^2).$$

Here *I* is the total intensity, ξ_2 is the degree of circular polarization of the external field, and d^{10} is the reduced matrix element for the dipole moment. It is the dependence of the pump on the velocity *v* of the atoms that makes possible velocity-selective optical orientation of the atoms in the ground state. Velocity selectivity usually involves the presence of collisions and the Doppler effect. To isolate the effect under discussion in pure form we are disregarding collisions altogether. As a result, the Maxwellian velocity distribution N(v) is an integral of the motion for the system (2), (3):

$$N_{+}^{i} + N_{-}^{i} + N_{+}^{o} + N_{-}^{o} = N(v).$$
⁽⁵⁾

The function N(v) is normalized to the total number of particles N. Here N(v) is conserved because we have allowed for the spontaneous decay of the excited state in (3) [the second term on the left-hand side of (3)].

3. Let us consider the solution of system (2), (3) in the approximation of low saturation $\Gamma^{\pm k} \ll \gamma$. In the steady state the populations of the excited levels can be neglected, and the populations of the sublevels in the ground state, as follows from (2) and (3), depend selectively on the velocity v and are independent of the total intensity I of the external field:

$$N_{+}^{0}(v) = \frac{(1+\xi_{2})g_{+}(v)N(v)}{(1+\xi_{2})g_{+}+(1-\xi_{2})g_{-}},$$

$$N_{-}^{0}(v) = \frac{(1-\xi_{2})g_{-}(v)N(v)}{(1+\xi_{2})g_{+}+(1-\xi_{2})g_{-}}.$$
(6)

The characteristic dependence of $N^0_{\ \pm}$ on the degree of circular polarization ξ_2 reflects the specifics of the interaction of an elliptically polarized field with a ground state in the absence of an intensity saturation effect. Interestingly, the velocity-selective orientation of the spins remains present even when $|E_+| = |E_-|$ ($\xi_2 = 0$). In this case the opticalorientation effect in its pure form is due solely to the velocity disequilibrium in the populations of the magnetic sublevels of the ground state. The physical nature of the velocity-selectivity of the spins is due to the separation (for $\delta \neq 0$) of the Bennett dips created by the counterpropagating circularly polarized waves in a cycle of the optical pump and to the transfer of these dips from the excited state to the ground state on account of spontaneous emission. In the collisionless model considered here, the nonequilibrium-velocity state with a given spin projection lives for an infinitely long time in the ground state, although the velocity distributions on the whole remains thermal, since the normalization $N^{0}_{+} + N^{0}_{-} = N(v)$ is preserved. Here the external field plays the role of a Maxwell's demon, sorting the particles with a given spin projection with respect to velocity directions. Since in our approximation $(\Gamma^{\pm k}/\gamma \leq 1)$ Eq. (6) exhibits no dependence on the total energy of the external field, the effect is purely entropic in origin and is explained from a thermodynamic standpoint by the transfer of order from the

field to the atoms. We see that nonzero spin currents arise with opposite directions (along and counter to k):

$$j_{\pm} = \int N_{\pm}^{0}(v) v \, dv \neq 0.$$

Of course, since the normalization (5) is preserved, the total current in an unbounded medium is zero $(j = j_+ + j_- = 0)$, and the polarized components drift relative to each other with velocities $v_{\pm}^{dr} = j_{\pm} \langle N_{\pm}^0 \rangle$. We note that the velocity-dependent selection of atomic states under continuous optical pumping between levels of the hyperfine structure in a beam of sodium atoms was studied experimentally in Ref. 6.

4. The presence of boundaries will cause cancellation of the currents but will give rise to gradients in the spin density. It follows from (2) and (3) that in the steady state

$$\mathbf{v}\nabla N_{+} - \Gamma^{-k}N_{+} + \Gamma^{+k}N_{-} = 0,$$

$$\mathbf{v}\nabla N_{-} - \Gamma^{+k}N_{-} + \Gamma^{-k}N_{+} = 0.$$
(7)

Let us consider a bounded medium in the form of a tube of length L, closed on both ends, and assume that there is no loss of angular momentum in collisions with the wall and that the impact is perfectly elastic. Then the boundary conditions for Eqs. (7) become

$$N_{\pm}(v, 0) = N_{\pm}(-v, 0), \quad N_{\pm}(v, L) = N_{\pm}(-v, L).$$
(8)

It is easy to find the general solution of the system (7), (8):

$$N_{\pm}(v, z) = N_{\pm}^{0}(v) \mp C(v) \exp [\varkappa(v)z], \qquad (9)$$

$$C(v) = (N_{+}^{0} - N_{-}^{0}) \frac{\exp[\varkappa(v)L] - 1}{\exp[L\varkappa(v)] - \exp[L\varkappa(-v)]}, \qquad (9)$$

$$\varkappa(v) = -\frac{\gamma}{3v} \frac{I}{2} [g_{+}(v) (1 + \xi_{2}) + g_{-}(v) (1 - \xi_{2})].$$

In an unbounded medium the nonequilibrium velocity distribution in the populations of the sublevels of the ground state thus leads to a spatial separation of the oriented spins. In this respect the present phenomenon is similar in effect to photoinduced drift.^{1,7}

However, unlike photoinduced drift, the spatial separation of spins occurs in a one-component gas even in the absence of collisions with a buffer gas. Let us consider some limiting cases. As in the case of photoinduced drift, the present effect depends substantially on the detunings δ of the field frequency. For $\delta = 0$ the Bennett dips which stem from the counterpropagating polarized waves merge together, and so the velocity-selective influence on the different spin projections vanishes. In this case there is a homogeneous orientation of the atoms by the elliptically polarized light, giving rise to a macroscopic magnetic moment in the gas:

$$M = \mu_0 (N_+ - N_-),$$

where μ_0 is the Bohr magneton. When the counterpropagating waves have equal intensities, $\xi_2 = 0$ (the field is a linearly polarized helix), the magnetic moments on opposite ends of the tube are oriented in opposite directions. After averaging over velocities, we have in the limit $|x|L \ge 1$

$$M(0) = -M(L) = -\frac{\delta \mu_0 N}{\pi^{1/4} k \overline{v}} e^{-\theta} \operatorname{Ei}(-\theta);$$

$$\theta = [\delta^2 + (\gamma/2)^2] / (k \overline{v})^2, \qquad (10)$$

where $\operatorname{Ei}(\theta)$ is the exponential integral function. Thus, in a long tube the magnetization of the gas ceases to depend on the intensity of the light. In the opposite limiting case, $|\kappa|L < 1$, the magnetic moment is directly proportional to the intensity and to the length of the tube:

$$M(0) = -M(L) = \frac{2}{3} \pi^{\frac{1}{2}} I \frac{L}{\bar{v}} \frac{\delta}{\delta^2 + (\gamma/2)^2} \mu_0 N, \qquad (11)$$

and is similar in its frequency dependence to photoinduced drift. If the counterpropagating waves have different intensities, $\xi_2 \neq 0$ (the helix is elliptical), and if $|\kappa| L < 1$, we have

$$M(L) = \xi_2 \mu_0 N,$$

$$M(0) = M(L) + M_0 N \cdot 2\pi^{\frac{1}{5}} \xi_2 (1 - \xi_2^2) \delta^2 / k \overline{v} [(\gamma/2)^2 + \delta^2 (1 - \xi_2^2)]^{\frac{1}{5}}. (12)$$

The magnetic moments at the ends of the tube are different in absolute magnitude and, as in (10), are independent of the intensity and of the length of the tube. For $\delta = 0$ there remains the usual homogeneous magnetization

 $M(0) = M(L) = \mu_0 \xi_2 N.$

5. We have thus demonstrated for an extremely simple example that it is possible in principle to selectively separate a gas in space according to the spin projections of its atoms in a one-component system. It is easy to see that the effect remains present for arbitrary values of the angular momenta of the ground and excited states. Moreover, it is not necessary to have the specific helical field configuration (1), since the effect stems from the frequency separation of the Bennett holes for the different spin projections. For this reason one may use, for example, a single linearly polarized traveling plane wave in place of (1) and employ a longitudinal magnetic field to separate the Bennett holes. Then, in particular, at exact resonance (for $\delta = \omega - \omega_0 = 0$) one can use all the results of this paper if in place of δ one substitutes $\delta = \mu_0 gH$, where g is the Landé factor and H is the strength of the longitudinal magnetic field.

In conclusion we note that for states with angular momentum $J_0 > 1/2$ a latent velocity disequilibrium can arise in the ground state for multipole moments of higher rank \varkappa as well $(1 < \varkappa \leq 2J_0)$. In the present paper we have specifically considered the case in which collisions are absent in the ground state. However, the effect persists in the presence of collisions if $\gamma^1 \ll \Gamma^{\pm k} \ll \gamma$, where γ^1 is the relaxation constant for the orientation of the ground state; this condition is easily established in view of the large difference between γ^1 and γ .

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