Effect of boundary conditions on the spatial separation of a gas by spin projection in a circularly polarized optical field

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The effect of boundary conditions of two types at the cell walls—specular reflection with a partial depolarization of atoms and diffuse reflection with a partial depolarization—on the separation of a gas by spin projection in a circularly polarized optical field is analyzed. The degree of magnetization of the gas in the cell depends strongly on the nature of the collisions of atoms with the cell walls. The degree of polarization of the atomic beam leaving the cell is calculated.

1. Mechanisms for producing spatially nonuniform distributions of atoms in quantum states in electromagnetic fields have recently been the subject of active research.[1–7] The interest in these problems stems from the promising outlook for the use of spatial separation of atoms by quantum state to generate polarized beams of atoms, to separate isotopes, to produce population inversions, etc. Several schemes for a spatial separation of quantum states have been proposed: in a circularly polarized optical field,[1] in a linearly polarized optical field and a longitudinal magnetic field,[2] and in optical fields of different frequencies with a special spatial configuration.[3–7] A feature common to all these mechanisms is that the atomic density is locally at equilibrium (a Maxwellian distribution), while the velocities of the atoms in different quantum states are locally not at equilibrium. The spatial-separation effects themselves stem from the simultaneous selective effect of light on the translational and internal degrees of freedom. The condition of a local density equilibrium distinguishes these effects from the known effects of radiation pressure and photoinduced drift.

An important feature of all selective effects of light is a cancellation of the particle fluxes at the boundaries of the volume, which results in a spatial selection of quantum states. Since the interactions of the particles with the walls may differ in nature, the efficiency of the spatial selection will in general depend on the nature of the boundary conditions at the walls of the gas-filled cell. Identical boundary conditions for atoms in different quantum states, specifically, absolutely elastic collisions (specular reflection), were considered in Refs. 1–7. It is worthwhile to examine other boundary conditions from the standpoint of their effect on the efficiency of spatial selection in comparison with specular reflection.

In the present paper we use the model for the medium which was proposed in Ref. 1 to examine some new boundary conditions: absolutely elastic collisions with a partial depolarization and diffuse reflection with a partial depolarization. The boundary-value problem is formulated in accordance with a known concept (§15 in Ref. 8). The manifestations of boundary effects are analyzed in examples involving a change in the efficiency of gas separation by spin projection at the exit from a cell and in the degree of magnetization of a gas at the boundaries of a cell.

2. We begin with a brief description of the model of the medium which we will use.[1] A single-component rarefied gas of two-level atoms with spins $j_0 = j_1 = 1/2$ for the ground and excited states interact with a circularly polarized resonant optical field

$$\mathbf{E} = (\mathbf{E}_+ e^{ikz} + \mathbf{E}_- e^{-ikz}) e^{-i\omega t} + \text{c.c.}, \quad \mathbf{E}_\pm = \frac{E_\pm}{2^{1/2}} (\mp \mathbf{e}_x + i\mathbf{e}_y). \tag{1}$$

The densities of atoms in the ground state, $N_\pm(z,v,t)$, with spin projections $\pm 1/2$, respectively, are described by a system of one-dimensional balance equations in the approximation of a slight saturation:

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right) N_\pm \pm \frac{2}{3} [\Gamma_-(v) N_+ - \Gamma_+(v) N_-] = 0. \tag{2}$$

All types of collisions are ignored here, and the notation is as follows, in accordance with Ref. 1: $\Gamma_\pm(v)$ are the rates at which the states are pumped, given by

$$\Gamma_\pm = \frac{I}{2} (1 \pm \xi_2) g_\pm(v) \gamma,$$

$$g_\pm(v) = [\gamma^2 + (\delta \pm kv)^2]^{-1}, \qquad I = \frac{1}{3} \left|\frac{d}{\hbar}\right|^2 (|E_+|^2 + |E_-|^2).$$

$$\xi_2 = (|E_+|^2 - |E_-|^2)(|E_+|^2 + |E_-|^2)^{-1}, \tag{3}$$

I is the interaction energy in frequency units, ξ_2 is the degree of circular polarization, d is the reduced dipole matrix element, 2γ is the rate of radiative decay of the excited state, and $\delta = \omega - \omega_0$ is the deviation of the field frequency ω from the resonant frequency ω_0.

The behavior of the spin densities $N_\pm$ and fluxes $j_\pm$ was studied in sufficient detail in Ref. 1. As we have already mentioned, boundary conditions corresponding to specular reflection from the walls of a one-dimensional cell of length l were used in Refs. 1–7:

$$N_\pm\left(-\frac{l}{2}, v\right) = N_\pm\left(-\frac{l}{2}, -v\right),$$

$$N_\pm\left(\frac{l}{2}, v\right) = N_\pm\left(\frac{l}{2}, -v\right). \tag{4}$$

3. Following Ref. 8, we impose the following boundary conditions (at $z = \pm l/2$) in the one-dimensional problem in the general case:

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$$\left\langle w_{\parallel}\left(\frac{l}{2};\ -v',v\right)vN_\alpha\left(\frac{l}{2};\ v\right)\right\rangle_v + \left\langle w_{\perp}\left(\frac{l}{2};-v',v\right)vN_{\alpha'}\left(\frac{l}{2};v\right)\right\rangle_v = v'N_\alpha\left(\frac{l}{2};\ -v'\right), \tag{5a}$$

$$\left\langle w_{\parallel}\left(-\frac{l}{2};\ v',-v\right)vN_\alpha\left(-\frac{l}{2};\ -v\right)\right\rangle_v + \left\langle w_{\perp}\left(-\frac{l}{2};\ v',-v\right)vN_{\alpha'}\left(-\frac{l}{2},-v\right)\right\rangle_v = v'N_\alpha\left(-\frac{l}{2},\ v'\right), \tag{5b}$$

where $v,v'>0$; $\alpha=(+,-)$; and $\alpha'\neq\alpha$.

Here and below, the symbol $\langle ... \rangle_v$, which signifies an average over velocity, is intended to also imply $v>0$ and that the integration is carried out over exclusively positive velocities.

The scattering of atoms by the walls in (5a) and (5b) is described by $w_\parallel(v',v)$ and $w_\perp(v',v)$, which are the probabilities that an atom with a given velocity v and a spin projection α will, after colliding with the wall, be reflected with a velocity v' and with, respectively, the same spin projection or an altered spin projection. The normalization of these functions is[8] (at $z=-l/2$, for example)

$$\left\langle w_{\parallel}\left(-\frac{l}{2};\ v',-v\right)\right\rangle_{v'} + \left\langle w_{\perp}\left(-\frac{l}{2};\ v',-v\right)\right\rangle_{v'} = 1. \tag{6}$$

a) For specular reflection without a change in the polarization of the atoms, (4), we have

$$w_\parallel = v_0^{-1}\delta(v-v'),\quad w_\perp=0,$$

where v_0 is the thermal velocity.

There is no difficulty in incorporating the depolarization of the atoms upon elastic reflection from the walls (below we will use a subscript a to specify this case):

$$w_\parallel=[(1+\lambda)v_0]^{-1}\delta(v-v'),\quad w_\perp=\lambda w_\parallel. \tag{7}$$

The quantity λ in (7) determines the degree of depolarization ε of the spin $\Delta N=N_+-N_-$ at the walls (for simplicity, we assume that the degree of depolarization ε is the same for the two walls),

$$\varepsilon=\frac{1-\lambda}{1+\lambda}=\frac{\langle v\Delta N(l/2,-v)\rangle}{\langle v\Delta N(l/2,v)\rangle}=\frac{\langle v\Delta N(-l/2,v)\rangle}{\langle v\Delta N(-l/2,-v)\rangle}, \tag{8}$$

as the ratio of the average spin of the atoms moving away from 1 cm^2 of wall surface area per second, on the one hand, to the average spin of the incident particles, on the other.

The range of the parameter ε would be $-1\leqslant\varepsilon\leqslant 1$ in general, but a boundary at which a spin reorientation $\varepsilon<0$ occurred would be difficult to imagine physically. For this reason we will assume $0\leqslant\varepsilon\leqslant 1$. The limiting cases correspond to complete depolarization ($\varepsilon=0$) and to the preservation of the spin after the collision ($\varepsilon=1$).

b) We now consider the diffuse-reflection boundary condition (which we will specify below by the subscript b):

$$\frac{N_\pm(l/2,-v)}{C_\pm^1}=\frac{N_\pm(-l/2,v)}{C_\pm^2}=\theta(r_0-r)f_0(v),$$
$$C_+^{(1,2)}+C_-^{(1,2)}=1. \tag{9}$$

The meaning of this boundary condition is as follows: After the atoms collide with the wall ($z=\pm l/2$) they lose all information about their initial velocity; i.e., they become randomized with a Maxwellian distribution $f_0(v)$. The constants $C^{(1,2)}_\pm$ determine the average densities of the components, $N_\pm$, near the walls for the outgoing particles. Their normalization in (9) follows from the condition for a local equilibrium in terms of the density n. A cell of radius r_0 is modeled by a unit step function $\theta(x)$. In this case the scattering probabilities $w_\parallel$ and $w_\perp$ are

$$w_\parallel=2v'[(1+\lambda)v_0^2]^{-1}\exp\left(-\frac{v'^2}{v_0^2}\right),\quad w_\perp=\lambda w_\parallel. \tag{10}$$

4. The degree of magnetization of the gas is determined by the macroscopic magnetic moment $M(z)=\mu_0\langle\Delta N(z,v)+\Delta N(z,-v)\rangle$ induced in the medium. This moment is directed along the z axis. Here μ_0 is the Bohr magneton. For simplicity, we carry out a specific calculation of M in the particular case of a linearly polarized spiral [$\xi_2=0$ in (3)]. The profile $M(z)$ is thus of odd parity along z for boundaries of both types, (7) and (10), and it reaches its maximum at the boundaries. We recall[1] that in the case $\xi_2=0$ the magnetic moment originates from a frequency separation of "Bennett holes" in the different spin projections, and in the case $\delta=0$ we have $M=0$.

We can write expressions for the magnetic moment.

a) In the case of specular reflection with a partial depolarization we have

$$\frac{M_a(z)}{M_0}=2\theta(r_0-r)(1+\varepsilon)\left\langle\frac{A(z,v)}{1+\varepsilon-2\varepsilon B(l/2,v)}\right\rangle, \tag{11}$$

$$M_0=\mu_0 n,\quad A(z,v)=\Delta n_0 B(z,v),$$

$$B(z,v)=\exp\left(-\varkappa\frac{l}{2}\right)\operatorname{sh}(\varkappa z),$$

$$\Delta n_0=\frac{g_+-g_-}{g_++g_-},\quad \varkappa=\frac{1}{v}(\Gamma_++\Gamma_-). \tag{12}$$

b) In the case of diffuse reflection with a partial depolarization [$A(z)=\langle A(z,v)\rangle$, $B(z)=\langle B(z,v)\rangle$] we have

$$\frac{M_b(z)}{M_0}=2\theta(r_0-r)\left\{A(z)+\frac{2\varepsilon B(z)u\langle vA(l/2,v)\rangle}{1+\varepsilon-2\varepsilon u\langle vB(l/2,v)\rangle}\right\}$$
$$u=2\pi^{1/2}/v_0. \tag{13}$$

Let us explain the physical meaning of the notation here: $\Delta n_0(v)$ is a uniform distribution of the difference between the populations of states with spins of $\pm 1/2$ per atom which is produced by the optical pumping $\varkappa$ is the gain per unit length; M_0 is the maximum value of the magnetic moment; and the averaging over $v_0>0$ in (11) and (13) is carried out with a Maxwellian distribution $f_0(v)$.

In the case of complete depolarization ($\varepsilon = 0$), distributions (11) and (13) lead to the same results

$$M(\varepsilon=0)=2\theta(r_0-r)A(z).$$

The fact that the result is the same reflects a specific feature of the model[1]: The gas as a whole is locally at equilibrium (Maxwellian), and the depolarization of quantum states with locally nonequilibrium velocity distributions implies that these states are Maxwellized simultaneously.

To determine the dependence of the magnetization on the type of wall (7) or (10), we compare magnetic moments (11) and (13) at the cell walls locations $z = \pm l/2$. We focus on the limiting cases of long cells ($s \gg 1$) and short cells ($s \ll 1$), where the characteristic parameter of the problem is

$$s=Gkl\begin{cases}\gamma\Omega(\gamma^2+\delta^2)^{-1}, & |\delta|<\Omega,\\ \gamma\Omega^{-1} & |\delta|>\Omega,\end{cases} \tag{14}$$

$\Omega = kv_0$ is the Doppler width, and $G = \gamma^{-1}\langle\Gamma_+ + \Gamma_-\rangle < 1$ is the saturation parameter. At small deviations from resonance, $|\delta| \ll \Omega$, this saturation parameter is $G = \pi^{1/2} I(\gamma\Omega)^{-1}$, while at large deviations, $|\sigma| \gg \Omega$, we have $G = I\delta^{-2}$.

In the limiting case of long cells, $s \gg 1$, and for deviations on the order of the Doppler width, $|\delta| = \Omega$, the condition $kl \gg \Omega(\gamma G)^{-1}$ is thus satisfied for Na vapor at $T = 300$ K in cells of size $l \gtrsim (200\,G)^{-1}$ (centimeters). This condition is easily met experimentally.

a) In the limiting case of short cells, $s < 1$, the value of $|M(\pm l/2)|$ is the same for the boundaries of the two types, (7) and (10), and is independent of the degree of depolarization ε:

$$\left|M\left(\pm\frac{l}{2}\right)\right| = \frac{\pi^{1/2}}{2}\theta(r_0-r)\frac{Ikl}{\Omega^2}\operatorname{Im}\left(\frac{w(\Delta+i\sigma)}{\sigma+i\Delta}\right)M_0, \tag{15}$$

where the special function $w(x)$ is defined in Ref. 9, $\Delta = \delta/\Omega$, and $\sigma = \gamma/\Omega$.

At low frequency deviations, $|\Delta| < 1$, we have

$$\left|M\left(\pm\frac{l}{2}\right)\right| = \frac{s}{2}\theta(r_0-r)|\Delta|\exp(-\Delta^2)M_0;$$

at large deviations, $|\Delta| \gg 1$, we have

$$\left|M\left(\pm\frac{l}{2}\right)\right| = M_0\theta(r_0-r)s|\Delta|^{-1}.$$

b) In the limit of long cells, $s \gg 1$, $|M(\pm l/2)|$ does not depend on the light intensity or the dimensions of the cell. It is determined by the frequency deviation and by the nature of the collisions of the particles with the walls:

$$\left|M_a\left(\pm\frac{l}{2}\right)\right| = M_0\theta(r_0-r)\frac{(1+\varepsilon)}{\pi^{1/2}}|\Delta|\exp y\cdot E_1(y), \tag{16}$$

$$\left|M_b\left(\pm\frac{l}{2}\right)\right| = M_0\theta(r_0-r)|\Delta|\left\{\frac{1}{\pi^{1/2}}\exp y\cdot E_1(y) + \varepsilon[\pi^{1/2}-\pi y^{1/2}\exp y\operatorname{erfc} y^{1/2}]\right\},$$

$$y=\Delta^2+\sigma^2. \tag{17}$$

Here $E_1(x) = -\operatorname{Ei}(-x)$ is the integral exponential function, and $\operatorname{erfc}(x)$ is the error function.[9]

At small deviations, $|\Delta| \ll 1$, we have $|M_a(\pm l/2)| > |M_b(\pm l/2)|$, while at $|\Delta| \gg 1$ we have

$$|M_a(\pm l/2)| < |M_b(\pm l/2)|.$$

In principle, the difference between moments (16) and (17) would make it possible to experimentally study the nature of the interaction of atoms with a wall on the basis of the strength of the magnetic field, since for a given distribution $M(z)$ the magnetic field **B** is equivalent to the magnetic field of a solenoid[10] with a constant and nonuniform current in its winding:

$$j_\varphi(r,z)=-c\delta(r_0-r)M(z) \tag{18}$$

(we are using a cylindrical coordinate system here).

A current distribution with the property $j_\varphi(-z) = -j_\varphi(z)$ could be pictured as two windings with a varying density of turns, in which the currents are flowing in different directions.

5. The efficiency of the separation of a gas by spin projection can be determined from, for example, the degree of polarization of a beam of atoms emitted from a small slit at the end of the cell. Let us assume that this slit is at the boundary $z = l/2$. The degree of polarization,

$$\beta=\frac{\langle v[N_+(l/2,v)-N_-(l/2,v)]\rangle}{\langle v[N_+(l/2,v)+N_-(l/2,v)]\rangle}=u\left\langle v\Delta N\left(\frac{l}{2},v\right)\right\rangle, \tag{19}$$

is determined by the ratio of the difference between the fluxes of the particles with the different projections which are incident on the wall to the total flux of particles incident on the wall.

In turns out that in the physically interesting limit of long cells the degree of polarization in (19) does not depend on the nature of the cells or the light intensity:

$$\beta=u\langle v\Delta n_0 f_0(v)\rangle=-2\pi^{-1/2}[1-(\pi y)^{1/2}\exp y\cdot\operatorname{erfc} y^{1/2}],$$

$$y=\Delta^2+\sigma^2.$$

The degree of polarization of the beam of incident atoms reaches its maximum at frequency deviations on the order of the Doppler width, $|\Delta| \approx \Omega$, for which its value is $|\beta| \approx 0.89$.

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