Spatial separation of pure quantum states of atoms and molecules by coherent electromagnetic fields

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An analysis is made of new methods for creating inverted active gas systems by producing a nonequilibrium distribution of the atomic velocities by means of a coherent electromagnetic field. The conditions for a complete spatial separation of quantum states of atoms and molecules are determined.

I. INTRODUCTION

The velocity selectivity of the interaction of coherent electromagnetic radiation with atoms and molecules in a gas is utilized in numerous practical applications (see, for example, Refs. 1–3). Both theoreticians and experimentalists have recently given much attention to light-induced gas drift,[4] and to the velocity-dependent excitation of particles.

Another important application of the selective interaction of coherent electromagnetic radiation with gases is spatial selection of quantum states.[5,6] We shall use this term to describe both spatial separation of gaseous atoms (molecules) in specific quantum states (selection of pure states or separation of populations represented by the diagonal elements of the density matrix) and spatial separation of a noncoherent mixture of states into coherent components [selection of mixed states or transport of the off-diagonal elements of the density matrix (coherences)].

Selection of quantum states is not only an interesting subject for phsyics research, but also a fairly effective method for creating inverted or frozen-out quantum systems (i.e., nonequilibrium distributions in which the population of the upper level is either substantially smaller or larger than the population of the lower level), which have various possible practical applications in quantum electronics.

Spatial selection of pure states has certain features in common with light-induced drift. In both cases the external manifestation of the effect is the appearance of a macroscopic flux of atoms. However, in constrast to light-induced drift, the methods used to achieve spatial selection of quantum states do not require the presence of a buffer gas and are related to the separation of active atoms into various quantum states[5] or separation of the initially incoherent mixture of states into two coherent components which on the average are 180° out of phase with one another.[6] The present paper will provide a description of such macroscopic manifestations of the quantum kinetics of the interaction of coherent radiation with matter.

The main condition for spatial separation of quantum states is that the lifetime τ in each of the separated quantum states be sufficiently long. This condition makes the spatial separation $l \sim \tau v_T$, comparable with the characteristic size L of the volume occupied by the gas in question. Under normal laboratory conditions we have $L \sim 1$–10 cm and the thermal velocity satisfies $v_T \sim 10^4$ cm/sec, so that significant separation of the states requires a lifetime $\tau \gtrsim 10^{-4} - 10^{-3}$ sec. These values determine the class of systems in which separation is possible.

Spatial separation of quantum states may be observed for any atoms or molecules exhibiting spectral transitions at frequencies ranging from the rf part of the spectrum to visible light. Separation of quantum states is most effective in the rf range, including transition waves. The transitions in this range occur between Zeeman levels which are well resolved by a static magnetic field, levels of the hyperfine and fine structures of atoms, and levels of the molecular rotational structure corresponding to transitions with wavelengths in the millimeter and centimeter ranges. The active media are, for example, vapors of alkali elements; the states which are separated are levels of the hyperfine structure or the Zeeman components of one of the hyperfine sublevels of the ground state. Selection of states corresponding to the fine-structure levels is possible, for example, in thallium vapor. We should mention also active media of three-level lasers in which one can separate the ground and metastable states between which a laser transition is possible, etc.

Spatial separation of quantum states is due to the interaction between atoms or molecules of an active gas and coherent electromagnetic radiation.

The velocity selectivity of this interaction (due to the Doppler effect) deforms the distribution functions of the gas atoms in the states being separated, so that this distribution function acquires an asymmetric Bennett structure. The departure of the distribution from equilibrium gives rise to oppositely directed fluxes of atoms in the separated states. Spatial selection of the particles according to the states occurs in a finite volume. Therefore, the mechanism for creation of a nonequilibrium distribution by coherent electromagnetic radiation makes it possible to establish an inverted (or frozen-out) system of quantum states. As in the case of the light-induced drift,[7] there are no special restrictions on the profile or width of the electromagnetic radiation line.

Spatial selection of atoms or molecules by states occurs in the case of finite values of the detuning and is associated with the existence of a macroscopic flux of the longitudinal component of the momentum (or a population inversion).[5] Another aspect of this effect is the appearance of transverse momentum or coherence fluxes as a result of the separation of quantum states.[6] Spatial transport of the coherence occurs also in a resonance in the absence of macroscopic fluxes of particles in each of the states. In a finite volume transport results in separation of an incoherent mixture of quantum states into two coherent components.

A quantum analysis of selection of pure quantum states is given in Sec. 2. The main equations are given and analyzed in Sec. 3.

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2. QUALITATIVE ANALYSIS

We shall now provide a qualitative description of the methods for spatial separation of quantum states and compare these methods. We shall simplify the analysis of the phenomena which occur by confining the treatment to a three-level model of an active gas and assuming that levels 1 and 2 are the states to be separated, whereas level 3 is short-lived.

The simplest method for separation of quantum states is realized[5] in rf-induced transfer of states when a coherent electromagnetic field is applied directly to a transition between states to be separated. Spatial separation of the states $|1\rangle$ and $|2\rangle$ requires then a significant initial inversion of these states (or a significant degree of freezing of these states). In some cases (for example, when the active medium is thallium vapor at room temperature) this condition is satisfied because of the Boltzmann factor $\exp(\hbar\omega_{21}/T)$. If the active medium is a vapor of an alkali element (in which case the levels 1 and 2 are the hyperfine sublevels of the ground state), a room-temperature population inversion is usually established. by optical pumping. We shall consider the specific case when pumping is provided by incoherent radiation from level 1 via level 3. Since the 2–1 transition is strongly forbidden, the population of level 2 is higher than that of level 1, but the atomic velocity distribution functions in these states remain Maxwellian (curve 2 in Fig. 1).

We shall now assume that a coherent electromagnetic field is applied to the 1–2 transition in the form of a plane monochromatic traveling wave with a frequency which is detuned from the transition frequency. The selectivity of the interaction of atoms with the electromagnetic field associated with the Doppler effect deforms the Maxwellian distributions of particles in levels 1 and 2, so that these distributions acquire a Bennett structure (curve 3 in Fig. 1). The asymmetry of the distributions creates[5,6] opposite fluxes of particles in the states $|1\rangle$ amd $|2\rangle$, i.e., it transports the population inversion (freezing-out of the state) and separates spatially the states $|1\rangle$ and $|2\rangle$.

Coherent electromagnetic radiation plays a dual role in this method for separating states. On the one hand, it creates nonequilibrium distributions of particles in levels 1 and 2 and in this sense it is a direct cause of the separation. On the other hand, it mixes the states $|1\rangle$ and $|2\rangle$, equalizes their populations, and reduces the degree of spatial separation. Therefore, this method cannot create a significant separation of states although it is suitable for the formation of an excess inversion at one of the ends of a cell. There is also an inversion at the other end of the cell, but it is weaker. The states are not frozen-out at any point inside the cell.

Another method for spatial separation of quantum states is based on the interaction of an atomic sytem with coherent electromagnetic radiation applied to a transition connected to 1–2 (via a rapidly decaying excited state $|3\rangle$). This mechanism ensures a more effective spatial separation of quantum states and requires in all cases just one field. There is no need to create a preliminary population inversion of the states to be separated. Selective interaction of atoms with a coherent electromagnetic field in the case of nonzero detuning results, as before, in an anisotropic deformation of the Maxwellian distribution of the particle velocities at levels 1 and 2, so that a Bennett structure is established. Reversal of the sign of the detuning causes the Bennett dips or peaks to travel from one wing of Maxwellian distribution to the other and reverses the directions of the opposite fluxes of particles in the states being separated.

This method makes it possible to achieve a much more complete separation of states than the preceding one. However, even this method cannot be used to form frozen-out states (if the field is applied to the 1–3 transition and if there are initially no frozen-out states); throughout the cell there is some degree of inversion, which at one end of the cell can reach values close to unity but disappears at the opposite end. If a preliminary inversion (freezing-out) is established in this method and then a coherent electromagnetic field is applied to the level with the larger number of atoms, it is possible to establish a population inversion at one end of the cell and freezing-out of the states at the other (see Fig. 5 below).

Complete spatial separation of quantum states is possible when two coherent electromagnetic fields are used and they are applied to the contiguous transitions 1–3 and 2–3. (A very special case of such separation is considered in Ref. 8.) A suitable selection of the values of the detuning $\Omega_1 = \omega_1 - \omega_{31}$ and $\Omega_2 = \omega_2 - \omega_{32}$ can ensure the necessary deformation of the distribution of particle velocities in the states being separated (Fig. 2), which gives rise to the maximum oppositely directed fluxes of the states. This method can ensure a complete separation of the states so that at one

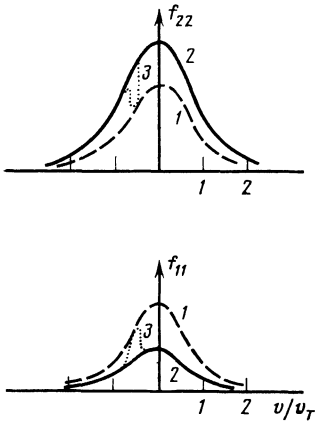

FIG. 1.

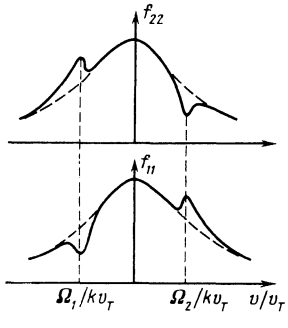

FIG. 2. Deformation of the particle distributions at the levels 1 and 2 due to the velocity-selective interaction of the atomic system with two coherent fields: the dashed curve represents an equilibrium Maxwellian distribution, whereas the continuous curve is a Bennett distribution; $|\Omega_1| = |\Omega_2| = 0.5kv_T$; $|\mathbf{k}_2| = |\mathbf{k}_2| = k$.

end of a cell the atoms are in the state $|2\rangle$ and at the opposite end they are in the state $|1\rangle$.

In all the separation methods described above the presence of a buffer gas reduces the degree of spatial separation of the states because collisions of active and buffer atoms create equilibrium distributions in the states $|1\rangle$ and $|2\rangle$.

The methods for separating quantum states utilizing a coherent field which mixes one of the states being separated with an exited state are much more effective than the method utilizing an electromagnetic field applied to the transition between the states being separated. However, the operational range of these methods is limited by the smallness of the Doppler widths of the 3–1 and 3–2 lines compared with the transition frequency ω_{21} between the states being separated: $kv_T < \omega_{21}$. For example, when the level 3 is optical, this condition is satisfied for $\omega_{21} \gtrsim 10^{10}$ c^{-1}. If this condition does not hold, an asymmetry of the velocity distributions in the separated states is extremely small, because the Bennett dips and peaks (Figs. 1 and 2) are close to the center of the distribution. This reduces strongly the fluxes of the states being separated and reduces to zero the degree of separation.

We shall also consider a hybrid method for spatial separaton of quantum states utilizing two coherent electromagnetic fields, one of which is applied to the transition between the states being separated and the other to one of the neighboring transitions. This method can compete in terms of the degree of separation of the states with the last of the above methods, but its use requires a sufficiently high initial difference between the populations of the states. It is effective in the case of active atoms for which the energy transition scheme is similar to that in thallium. An interesting feature of this hybrid method is the use of electromagnetic fields with frequencies in different parts of the spectrum. The complexity of the method is compensated by its efficiency.

One can also separate quantum states by using a standard electromagnetic wave as one of the coherent fields. This ensures the required initial difference between the populations of the states to be separated and then the distribution functions depart from equilibrium under the influence of a traveling wave. This method can alter the initial difference between the populations over a fairly wide range, and if that difference is optimal the separation efficiency can be increased.

3. QUANTITATIVE ANALYSIS OF SPATIAL SEPARATION OF QUANTUM STATES

In this section we shall provide a quantitative description of some of the methods for spatial separation of quantum states.

We shall consider a cell filled with the vapor of an active element (describing the atoms of this element in the three-level approximation) and of a buffer gas (this gas is not essential). We shall assume that coherent electromagnetic radiation of frequency differing from the transition frequency ω_{21} by $\Omega = \omega - \omega_{21}$ acts on the cell. The initial difference between the populations may be due to the Boltzmann factor (as in the case of thallium) or due to the pumping from level 1 by incoherent optical radiation (as in the case of vapors of alkali elements). We shall assume that the population difference is due to the latter factor (pumping with incoherent optical radiation) and write down a system of quantum kinetic equations for the density matrix of the active atoms allowing for all the main interactions of these atoms[5]:

$$\begin{aligned} \partial f/\partial t + \mathbf{v}\nabla f &= -(w+\gamma)f + \hat{S}f - 4\,\mathrm{Re}\,(iuf_{12}) + wM(\mathbf{v}), \\ \partial f_{12}/\partial t + \mathbf{v}\nabla f_{12} &= -(w+\Gamma+i\Delta)f_{12} + \hat{S}f_{12} - iu^*f, \end{aligned} \tag{1}$$

where $f = f_{22} - f_{11}$ is the inversion of the populations of the states being separated, f_{12} is the corresponding coherence, $U_{21} = \hbar u$ is the matrix element of the operator representing the interaction of an atom with the coherent electromagnetic field of a traveling wave, $2w$ is the optical pumping rate, $\gamma = T_1^{-1}$ and $\Gamma = T_2^{-1}$ are the rates of longitudinal and transverse relaxation, $\Delta = \Omega - \mathbf{kv}$; $M(\mathbf{v})$ is the initial Maxwellian distribution of the active atoms and $\hat{\mathbf{S}}$ is the collisional operator.

The steady-state spatially homogeneous solution of the system (1) considered in the model of strong collisions[9] is of the form[5]

$$f(\mathbf{v}) = \frac{\varepsilon\rho_0}{\varepsilon+\nu R}\left\{1 + \frac{\alpha+\nu R}{\Gamma_s^2+\Delta^2}\left[\frac{\nu\Delta\,(\Omega J_0 - kJ_1)}{1-\nu\,(E_1+\alpha)J_0} - E_1\right]\right\} M(\mathbf{v}), \tag{2}$$

where $\rho_0 = w/\varepsilon$ is the population inversion due to incoherent optical pumping;

$$R = \frac{\alpha}{\nu}\left\{\frac{1-\nu\,(E_1+\alpha)J_0}{\nu^2(\Omega J_0 - kJ_1)^2 + (1-\nu E_1 J_0)\,[1-\nu\,(E_1+\alpha)J_0]} - 1\right\}, \tag{3}$$

$$E = w+\Gamma, \quad \varepsilon = w+\gamma, \quad E_1 = E+\nu,$$
$$\alpha = 4|u|^2/(\varepsilon+\nu), \quad \Delta = \Omega - \mathbf{kv};$$

ν is the gaskinetic frequency of collisions of active and buffer atoms; and

$$J_n = J_n(\Gamma_s, \Omega) = \int_{-\infty}^{+\infty} \frac{v^n M(v)}{\Gamma_s^2+\Delta^2}\,dv, \quad \Gamma_s^2 = E_1(E_1+\alpha). \tag{4}$$

In the case when the initial population difference ρ_0 results from the Boltzmann factor, the solution is still given by Eqs. (2)–(4), but now the expressions for ε and E are taken from $w = 0$.

The inversion flux is

$$\begin{aligned} \mathbf{j} &= \int \mathbf{v} f(\mathbf{v})\,d^3v \\ &= -\rho_0 \frac{\mathbf{k}}{k}\varepsilon\frac{\alpha+\nu R}{\varepsilon+\nu R}\left[E_1 J_1 - \nu\frac{(\Omega J_0 - kJ_1)(\Omega J_1 - kJ_2)}{1-\nu\,(E_1+\alpha)J_0}\right]. \end{aligned} \tag{5}$$

The expression (5) for the flux simplifies in the limits of low and high collision rates. If $\nu = 0$, then

$$j = -4\rho_0|u|^2 J_1 E/\varepsilon, \tag{6}$$

whereas in the limit $\nu \to \infty$, we have

$$j = -4\rho_0\frac{|u|^2}{\nu^2}v_T\frac{kv_T\Omega}{\Omega^2+E^2+4|u|^2E/\varepsilon}, \tag{7}$$

where Eq. (7) is valid irrespective of the actual collision model. A numerical treatment shows that an increase in ν reduces the inversion flux monotonically, i.e., collisions with buffer atoms simply reduce the degree of spatial separation of quantum states.

The velocity of the inversion flux

$$\mathbf{v}_n = \mathbf{j} \Big/ \int f(\mathbf{v})\,d^3v$$

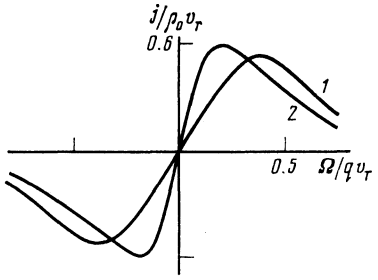

FIG. 3. Dependence of the inversion flux on the detuning Ω: 1) $u = w = 10^3\ \text{sec}^{-1}$; 2) $u = 10^3\ \text{sec}^{-1}$, $w = 5\times 10^2\ \text{sec}^{-1}$.

corresponding to specific values of the parameters u, w, and Ω become of order v_T. Figure 3 shows graphically the dependence of j on Ω. It should be pointed out that the values of Ω corresponding to the maximum flux ($\Omega \approx 0.3kv_T$) are considerably smaller than the values of Ω which maximize v_n ($\Omega \approx 1.1kv_T$).

The presence of an inversion flux in a spatially homogeneous medium results in spatial separation of quantum states in cells of finite size. Obviously, a more effective separation of pure states requires the use of cells free of buffer gas, but with a coating preventing population inversion as a result of relaxation due to collisions with the walls. In the case of coherence fluxes the separation of mixed states occur irrespective of whether the coherence is conserved or destroyed completely as a result of collisions with the walls.

We shall consider a cell of length $2L$. We shall assume that a traveling wave is propagating parallel to the z axis, so that f and f_{12} depend only on z. We shall assume that the boundary conditions are

$$f(v, \pm L) = f(-v, \pm L), \quad f_{12}(-v, L) = f_{12}(v, -L) = 0, \qquad (8)$$

which corresponds to conservation of the longitudinal momentum in collisions with the walls and complete relaxation of the transverse momentum. Consequently, under steady-state conditions the integrated population inversion

$$\rho(z) = \int_{-\infty}^{+\infty} f(v, z)\, dv \qquad (9)$$

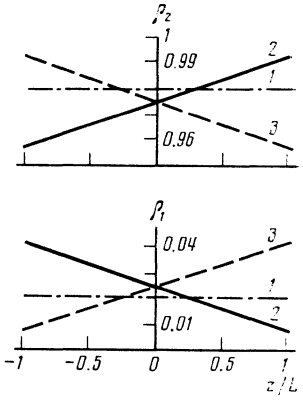

FIG. 4. Spatial dependences of the populations of the states for $L = 16$ cm, $\gamma = 10\ \text{sec}^{-1}$, $u = 10^3\ \text{sec}^{-1}$, $w = 5\times 10^2\ \text{sec}^{-1}$ and various values of Ω: 1) $\Omega = 0$; 2) $\Omega = -0.2kv_T$; 3) $\Omega = 0.2kv_T$.

is given by

$$\rho(z) = \rho_0[1 + P(z)], \qquad (10)$$

$$P(z) = -|u|^2 \int_{-\infty}^{+\infty} dvM(v)\,(\varepsilon^2+\delta^2)^{-1} + \int_0^{\infty} dvM(v)\,\{[f_1^+ + f_2^+ c_+(z) + f_3^+ s_+(z)]e^{-\varepsilon z/v} + [f_1^- + f_2^- c_-(z) - f_3^- s_-(z)]e^{\varepsilon z/v}\}. \qquad (11)$$

Here,

$$\begin{aligned}
&c_\pm(z) = \cos(\delta_\pm z/v), \quad s_\pm(z) = \sin(\delta_\pm z/v),\\
&f_1^\pm = \Phi^\pm + (\Delta_\pm^2/|u|^2)[f_2^\pm c_\pm(L) - f_3^\pm s_\pm(L)],\\
&f_2^\pm = [F^\pm - f_3^\pm c_\pm(L)]/s_\pm(L),\\
&f_3^\pm = d^{-1}[g_1^\mp g_2^\pm - s_+(L)s_-(L)g_2^\mp/\operatorname{ch}(\varepsilon L/v)],\\
&d = \{[c_-^2(L) + \Delta_-^2/|u|^2][c_+^2(L) + \Delta_+^2/|u|^2] + s_-^2(L)s_+^2(L)\}\operatorname{th}(\varepsilon L/v)\\
&\qquad + [c_-^2(L) + \Delta_-^2/|u|^2]s_+^2(L) + [c_+^2(L) + \Delta_+^2/|u|^2]s_-^2(L),\\
&g_1^\pm = s_\pm^2(L) + [c_\pm^2(L) + \Delta_\pm^2/|u|^2]\operatorname{th}(\varepsilon L/v),\\
&g_2^\pm = c_\pm(L)\delta_\pm^2 F^\pm/|u|^2 + s_\pm(L)(\Phi^\pm \mp F),\\
&F = 2|u|^2 kv\Delta/(\varepsilon^2+\delta_+^2)(\varepsilon^2+\delta_-^2)\operatorname{sh}(\varepsilon L/v),\\
&F^\pm = [|u|^2\varepsilon/\delta_\pm(\varepsilon^2+\delta_\pm^2)]e^{-\varepsilon L/v}, \quad \Phi^\pm = -[\Delta_\pm^2/(\varepsilon^2+\delta_\pm^2)]e^{-\varepsilon L/v},\\
&\Delta_\pm = \Omega \mp |\mathbf{kv}|, \quad \delta = (\Delta^2+|u|^2)^{1/2}, \quad \delta_\pm = (\Delta_\pm^2+|u|^2)^{1/2}.
\end{aligned} \qquad (12)$$

A numerical calculation carried out using Eqs. (11) and (12) shows that the spatial separation is maximized by values of the detuning Ω which maximize the inversion flux in a spatially homogeneous medium (Fig. 3).

Figure 4 plots the dependence $\rho_i(z)$, where

$$\rho_i(z) = \int d^3v f_{ii}(\mathbf{v}, z).$$

Figure 4 demonstrates clearly the spatial separation of the states along the cell. However, it also follows from Fig. 4 that the separation of the states by this method is fairly weak. In the case of an exact resonance ($\Omega = 0$) there is no separation, just as expected.

We shall now turn to the separation methods in which coherent radiation interacts with the transitions between an excited state and one of the states to be separated. We shall discuss the specific case in which the field is applied to the 1–3 transition and the levels 1 and 2 are linked by a transition whose frequency is in the rf part of the spectrum.

The system of quantum kinetic equations for the density matrix of active atoms in this case is as follows (this holds in the absence of a buffer gas):

$$\begin{aligned}
&\partial f_{11}/\partial t + \mathbf{v}\nabla f_{11} = 2\operatorname{Re}(iVf_{13}) + {}^1/_2\gamma(f_{22}-f_{11}) + {}^1/_2 Af_{33},\\
&\partial f_{22}/\partial t + \mathbf{v}\nabla f_{22} = -{}^1/_2\gamma(f_{22}-f_{11}) + {}^1/_2 Af_{33},\\
&\partial f_{33}/\partial t + \mathbf{v}\nabla f_{33} = -2\operatorname{Re}(iVf_{13}) - Af_{33},\\
&\partial f_{13}/\partial t + \mathbf{v}\nabla f_{13} = -\left(\frac{A}{2} + i\Delta\right)f_{13} + iV^*(f_{11}-f_{33}).
\end{aligned} \qquad (13)$$

Here, $\Delta = \Omega - \mathbf{kv}$; $\Omega = \omega - \omega_{31}$; ω is the frequency of the traveling wave field; $\mathbf{k}$ is the wave vector of the traveling wave; ω_{31} is the frequency of the 3–1 transition; γ is the rate of the longitudinal relaxation of the ground state; $V_{31} = \hbar V$ is the matrix element of the dipole interaction; A^{-1} is the natural lifetime of level 3.

A steady-state spatially homogeneous solution of the system (13) yields the following expressions for the rf inversion ρ and the inversion flux $\mathbf{j}$:

$$\rho(\Omega)=\frac{A}{2\gamma}|V|^2J_0(\Gamma_s,\Omega),\quad \mathbf{j}(\Omega)=\frac{\mathbf{k}}{k}\frac{A}{2\gamma}|V|^2J_1(\Gamma_s,\Omega), \tag{14}$$

where

$$\Gamma_s^2=A(A+2|V|^2/\gamma)/4.$$

The flux velocity is $v_n=J_1/J_0$ and, in contrast to the preceding method, the maxima of j and v_n occur at similar values of the detuning: $\Omega\approx 0.5kv_T$ for j and $\Omega\approx 0.6kv_T$ for v_n.

In the case of cells of finite size with the boundary conditions of the type given by Eq. (8) the solution of the system (13) is

$$\rho(z)=\int_{-\infty}^{+\infty}dvM(v)\frac{\Delta W(v)}{W(v)}+\int_0^{\infty}dvM(v)\left(\frac{\Delta W^-}{W^-}-\frac{\Delta W^+}{W^+}\right)$$
$$\times\left[\exp\left(-\frac{W^+}{v}z\right)\mathrm{sh}\left(\frac{W^-}{v}L\right)-\exp\left(\frac{W^-}{v}z\right)\mathrm{sh}\left(\frac{W^+}{v}L\right)\right]\Big/$$
$$\mathrm{sh}\left[\frac{L}{v}(W^++W^-)\right], \tag{15}$$

where

$$W(v)=\gamma+{}^1/_2W_1,\quad \Delta W(v)=\gamma\rho_0+{}^1/_2W_1,$$
$$W^\pm=W(\pm v),\quad \Delta W^\pm=\Delta W(\pm v),$$
$$W_1=4A|V|^2/[A^2+4(\Omega-\mathbf{kv})^2],$$

and ρ_0 is the initial difference between the populations.

Figure 5 shows plots of the dependence $\rho_i(z)$ illustrating the spatial separation. In the absence of an initial population difference ($\rho_0=0$) this method makes it possible to produce atoms practically only in state $|2\rangle$ at one end of a cell (curve 1); if the field is applied to the 2–3 transition, then atoms collect in state $|1\rangle$. However, if $\rho_0\neq 0$, then separation of the states in the cell is even more effective and we can have a situation in which a population inversion occurs at one end of the cell and the states are frozen-out at the other end (curves denoted by 2).

We shall now consider a method which provides complete separation of the quantum states $|1\rangle$ and $|2\rangle$.

We shall assume that coherent electromagnetic fields in the form of traveling monochromatic waves interact with the transitions 1–3 and 2–3. The interaction of atoms with these fields deforms the Maxwellian distributions of the particle velocities in levels 1 and 2, which acquire a Bennett structure (Fig. 2). The asymmetry of the distributions gives rise to opposite fluxes of the particles in states $|1\rangle$ and $|2\rangle$, i.e., it causes spatial separation of these states. In this method the separation is most effective subject to two conditions. The first condition specifies equality of the amplitudes of the fields applied to the transitions 1–3 and 2–3. The second requires collinearity of the wave vectors of these fields, but here we can have two cases: 1) the wave vector $\mathbf{k}_1$ of the first field is parallel to the wave vector $\mathbf{k}_2$ of the second field; 2) $\mathbf{k}_1$ is antiparallel to $\mathbf{k}_2$.

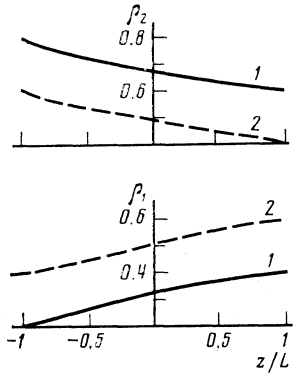

FIG. 5. Spatial dependences of the populations of the states: 1) $\rho_0=0$, $\Omega=-0.5kv_T$; 2) $\rho_0=-0.8$, $\Omega=-0.7kv_T$.

However, the conditions which must be satisfied by the detuning of the fields are different in these two cases. In the first case we must have $\Omega_1=-\Omega_2\equiv\Omega$, where $\Omega_1=\omega_1-\omega_{31}$; $\Omega^2=\omega_2\omega_{32}$; ω_1 is the frequency of the first field; ω_2 is the frequency of the second field. In the second case we have to satisfy $\Omega^1=\Omega^2\equiv\Omega$, so that in this case the detuning between ω_1 and ω_2 is exactly equal to the frequency ω_{21} of the transition between the states being separated. Reversal of the sign of the detuning Ω reverses the direction of fluxes of the atoms in the states $|1\rangle$ and $|2\rangle$, which transposes the regions in the cell where the atoms in these states collect.

The system of equations for the density matrix of active atoms in terms of the convenient notation of Ref. 10 is as follows ($\mu,\nu=1,2;\mu\neq\nu$)

$$\hat{L}_{\mu\mu}f_{\mu\mu}={}^1/_2Af_{33}+{}^1/_2\gamma f_{\nu\nu}+2\mathrm{Re}\{iV_{3\mu}f_{\mu\nu}\exp[i(\mathbf{k}_\mu\mathbf{r}-\omega_\mu t)]\},$$
$$\hat{L}_{33}f_{33}=-2\sum_{\mu=1}^{2}\mathrm{Re}\{iV_{3\mu}f_{\mu3}\exp[i(\mathbf{k}_\mu\mathbf{r}-\omega_\mu t)]\},$$
$$\hat{L}_{\mu3}f_{\mu3}=iV_{3\mu}^*(f_{\mu\mu}-f_{33})\exp[i(\omega_\mu t-\mathbf{k}_\mu\mathbf{r})]$$
$$+iV_{3\nu}f_{\mu\nu}\exp[i(\omega_\mu t-\mathbf{k}_\mu\mathbf{r})], \tag{16}$$
$$\hat{L}_{12}f_{12}=iV_{32}f_{13}\exp[i(\mathbf{k}_2\mathbf{r}-\omega_2 t)]-iV_{13}f_{32}\exp[i(\mathbf{k}_1\mathbf{r}-\omega_1 t)],$$
$$f_{11}+f_{22}+f_{33}=M(\mathbf{v}),$$

where

$$\hat{L}_{ik}=\mathbf{v}\nabla+\Gamma_{ik}+\partial/\partial t,\quad \Gamma_{33}=A,$$
$$\Gamma_{\mu\mu}=\gamma/2,\ \Gamma_{\mu3}=A/2-i\omega_{3\mu},\quad \Gamma_{12}=\Gamma-i\omega_{21},$$

and the rest of the notation is the same as before.

The steady-state solution of the system (16) subject to the boundary conditions of the type (8) for cells of length $2L$ gives Eq. (15) for the velocity-integrated inversion $\rho(z)$, but now we have

$$\Delta W={}^1/_2(W_1-W_2);\quad W=\gamma+{}^1/_2(W_1+W_2),$$
$$W_\mu=4A|V_{3\mu}|^2/[A^2+4(\Omega_\mu-k_\mu v)^2],$$

where it is assumed that $|V_{31}|,\ |V_{32}|\ll kv_T\sim 10^9\ \mathrm{sec}^{-1}$. When the conditions for the most effective separation mentioned above are satisfied, Eq. (15) simplifies greatly to

$$\rho(z)=2\int_0^{\infty}dvM(v)\frac{\Delta W^+}{W^+}\mathrm{sh}\left(\frac{W^+}{v}z\right)\Big/\mathrm{ch}\left(\frac{W^+}{v}L\right). \tag{17}$$

Analysis of Eq. (17) shows that there is an optimal value of the field amplitude for spatial separation of order $V_{opt}\sim kv_T(\gamma/A)^{1/2}$, and the optimal detuning is $\Omega_{opt}\sim kv_T$.

Figure 6 shows calculated curves illustrating the change in the degree of inversion $\rho(z)=\rho_2(z)-\rho_1(z)$ along a cell for different values of the parameters. It is clear that the atoms in the states $|1\rangle$ and $|2\rangle$ collect at the opposite

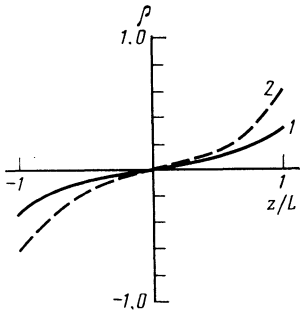

FIG. 6. Variation of the degree of inversion along a cell in the case when $V = 6\times10^6$ sec^{-1}, $\gamma = 10$ sec^{-1}, and $A = 3\times10^7$ sec^{-1} for different values of the parameters: 1) $\Omega_1 = -\Omega_2 = 0.5kv_T$, $L = 1$ cm; 2) $\Omega_1 = -\Omega_2 = 0.6kv_T$, $L = 16$ cm.

ends of a cell most effectively when the parameters have the optimal values.

A quantitative analysis of the hybrid method for spatial separation of quantum states is analogous to that given above, but the solution is fairly cumbersome and will not be given here.

We shall finally note that we investigated also the problem of spatial separation for other boundary conditions. For example, separation occurs quite efficiently when Eq. (8) is replaced with boundary conditions of the diffusion type when collisions with a wall do not alter the number of atoms in a given quantum state, but merely give rise to a Maxwellian distribution. In the case of uncoated cells, when collisions with the walls destroy the inversion, the separation efficiency decreases by approximately a factor of two compared with cells that have a protective coating.

4. CONCLUSIONS

The present paper describes new methods for the creation of inverted (frozen-out) systems based on formation of a nonequilibrium distribution by coherent electromagnetic fields. The proposed methods for spatial separation of quantum states are distinguished by their simplicity and can be applied to a wide class of quantum systems. An increase in the degree of spatial selection of quantum states by these methods should extend greatly the range of technical means available for physical experiments. Their use will make it possible to construct new quantum electronic devices operating in a wide spectral range. An example of a physical task in which an allowance is necessary for the resultant macroscopic fluxes of separated states is the problem of the nonlinear intra-Doppler structure of hyperfine transitions in alkali atoms.

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[1]V. S. Letokhov and V. P. Chebotayev, *Nonlinear Laser Spectroscopy*, Springer Verlag, Berlin (1977) [Springer Series in Optical Sciences, Vol. 4].
[2]V. S. Letokhov, Nonlinear Selective Photoprocesses in Atoms and Molecules [in Russian], Nauka, Moscow (1983).
[3]A. P. Kazantsev, Usp. Fiz. Nauk **124**, 113 (1978) [Sov. Phys. Usp. **21**, 58 (1978)].
[4]F. Kh. Gel'mukhanov and A. M. Shalagin, Zh. Eksp. Teor. Fiz. **78**, 1672 (1980) [Sov. Phys. JETP **51**, 839 (1980)].
[5]B. D. Agap'ev and B. G. Matisov, Pis'ma Zh. Tekh. Fiz. **12**, 123 (1986) [Sov. Tech. Phys. Lett. **12**, 51 (1986)].
[6]B. D. Agap'ev and B. G. Matisov, Pis'ma Zh. Eskp. Teor. Fiz. **44**, 66 (1986) [JETP Lett. **44**, 81 (1986)].
[7]A. K. Popov, A. M. Shalagin, V. M. Shalaev, and V. Z. Yakhnin, Zh. Eksp. Teor. **80**, 2175 (1981) [Sov. Phys. JETP **53**, 1134 (1981)].
[8]V. S. Smirnov and A. M. Tumaĭkin, Zh. Eksp. Teor. Fiz. **90**, 866 (1986) [Sov. Phys. JETP **63**, 505 (1986)].
[9]S. G. Rautian, Zh. Eksp. Teor. Fiz. **51**, 1176 (1966) [Sov. Phys. JETP **24**, 788 (1967)].
[10]A. K. Popov, Introduction to Nonlinear Spectroscopy [in Russian], Nauka, Novosibirsk (1983), p. 35.

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