Spatial separation of mixed states of atoms and molecules in the field of a traveling radio wave

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A mechanism which effects a spatial transport of coherence in gases that interact with an rf field and which gives rise to spatial separation of an incoherent mixture of states into two coherent components is proposed.

The feasibility of spatially separating "pure" states of gas atoms (molecules) which intereact with a traveling radio wave has been pointed out in Refs. 1 and 2. In the present letter we report the observation of a new physical phenomenon in a system of this sort: a transfer of coherence in the direction of propagation of a radio wave or the spatial separation of mixed states of the quantum system.

This phenomenon can be qualitatively described in the following way. Let us consider low-density pairs of an active substance, whose spectral transitions occur in the rf region of the spectrum ($\omega \sim 10^6 - 10^{13}$ rad/s). Such transitions correspond to the Zeeman levels which are adequately resolved by a static magnetic field, to the hyperfine- and fine-structure levels, and to the transitions between the rotational-structure levels of molecules lying in the millimeter and centimeter ranges. We assume that the population of the lower-lying level is different from that of the upper level and that this difference is caused by pumping or by the Boltzmann factor [$\exp(-\hbar\omega_{21}/T)$]. Upon placement of the system in a coherent rf field, the Maxwellian velocity distribution of particles at the $|1\rangle$ and $|2\rangle$ levels (Fig. 1) becomes deformed, acquiring the Bennett structure (the dashed curve in Fig. 1).

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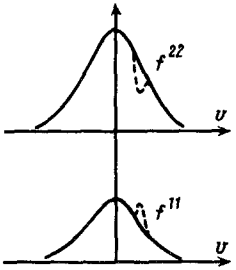

FIG. 1.

The appearance of the Bennett dips (peaks) stems from the velocity-selective (due to the Doppler effect) interaction of atoms with the rf field. The distribution asymmetry gives rise[1,2] to the appearance of oppositely directed particle fluxes in the $|1\rangle$ and $|2\rangle$ states, i.e., to the inversion transfer (freezing-in)–spatial separation of the $|1\rangle$ and $|2\rangle$ states. The basic condition that must be satisfied for separating the states a macroscopic distance from each other is that the lifetime of these states, characteristic for the part of the spectrum studied, be large enough.

In contrast with the populations, the velocity dependence of the coherence does not have the Doppler shape with the Bennett dips, but rather the shape of a dispersion curve (Fig. 2). The spatial transfer of the coherence therefore occurs even at resonance: in the absence of particle flux in each state. A graphic interpretation can easily be rendered for transitions between the levels of the Zeeman structure. In a rotating coordinate system, the preferred orientation of the transverse magnetic moments of the atoms occurs along the vector of the intensity of the magnetic field (B) of the wave. At resonance, the number of atoms moving in the direction of propagation of the wave is equal to the number of atoms moving in the opposite direction. The atoms in the first flux, however, interact coherently with the wave field for a longer time and hence transfer a larger transverse magnetic moment. The existence of this macroscopic magnetic flux means that there is a coherence transfer associated with the spatial separation of the initially incoherent mixture of states $|1\rangle$ and $|2\rangle$ into two coherent components whose phases are opposite to each other. The separation of mixed states can be observed experimentally by known methods.[3] We emphasize that the presence of an initially produced molecular beam was required in Ref. 3. In our case, however, the flux appears by itself in the course of the interaction with the rf field.

Corresponding calculations give, for the nonvanishing components of the magnet-

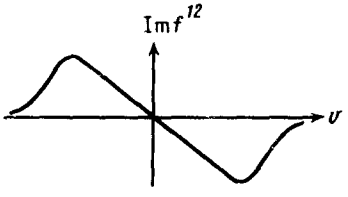

FIG. 2.

ic flux density,

$$j_{ik} = \int v_i m_k(\mathbf{v}) d^3 v$$

the expressions

$$\begin{aligned} j_{zz} &= -n\mu_B \frac{4w(\Gamma + w)}{(w+\gamma)^2} J_1 |u|^2, \\ j_{zx} &= -n\mu_B \frac{2w(\Gamma + w)}{w + \gamma} |u| J_1, \\ j_{zy} &= -n\mu_B \frac{2w|u|}{w + \gamma} (\Omega J_1 - kJ_2), \end{aligned} \tag{1}$$

where w is the pumping rate, γ and Γ are the rates of the longitudinal and transverse relaxations, $U_{21} = \hbar u$ is the matrix element of the interaction with the rf field, and μ_B is the Bohr magneton,

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

$$\Gamma_s^2 = (\Gamma + w)^2 + 4\frac{\Gamma + w}{\gamma + w} |u|^2, \qquad \Delta = \Omega - \mathbf{kv},$$

We assume that the z axis is parallel to the wave vector $\mathbf{k}$ and that the y axis is directed along the rotating component of the magnetic field of the radio wave.

Expressions (1) were obtained on the basis of the solution of kinetic quantum-mechanical equations for the density matrix.[1,4] At resonance ($\Omega = 0$), the only nonvanishing component is

$$j_{zy} = n\mu_B \frac{2kw|u|}{w + \gamma} J_2(\Gamma_s, 0). \tag{3}$$

If the cell contains a buffer gas, in the frequent-collision limit we can write the expression for this component in simple form

$$j_{zy} = v_T n\mu_B \frac{kv_T}{\nu} \frac{w|u|}{(w + \gamma)(w + \Gamma) + 4|u|^2}, \tag{4}$$

where ν is the frequency of elastic collisions.

The magnitude of the transverse magnetization flux can be characterized by the flux rate: $V = j_{zy}/n\mu_B$. For the parameter values characteristic of the experiments with optical pumping, $w \sim |u| \sim 10^4\ \mathrm{s}^{-1}$, $\gamma \sim \Gamma \sim 10^2\ \mathrm{s}^{-1}$, and $\omega_{21} \sim 10^{10}$ rad/s, we have $\Gamma_s \sim 10^4\ \mathrm{s}^{-1}$, so that $J_2(\Gamma_s, 0) \simeq 0.242\ k^{-2}$ and the flux rate is on the order of the thermal flux, $V \sim 0.4 v_T$. We are thus dealing not only with a qualitatively new phenomenon but also, from the quantitative viewpoint, with a very important effect. Magnetization transfer can play a prominent role in modern physics of atomic systems as a

tool for studying subtle features in the interaction of atoms and molecules and it can also be used in applied fields.

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