

# Redistribution of the velocities of atoms under the influence of light

S. N. Luzgin

*Institute of Terrestrial Magnetism, Ionosphere, and Radiowave Propagation, Siberian Branch of the Academy of Sciences of the USSR*

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The model of weak collisions is used to consider the redistribution of atomic velocities in a low-pressure gas under the influence of light. A consistent derivation is given of a Fokker-Planck type equation for atoms in resonant optical field. It is shown that in the case of a traveling wave the diffusion coefficients of atoms in the momentum space are limited by the rate of spontaneous decay, whereas for a standing wave they are proportional to the intensity of light. A calculation is made of the minimum temperature which can be reached by cooling atoms in a strong optical field. It is shown that, in principle, narrow dips can appear in the momentum distribution and the width of these dips expressed in terms of frequency units can be of the order of the homogeneous width.

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## 1. INTRODUCTION

When an atom absorbs or emits a quantum of light, its momentum changes by an amount equal to the photon momentum  $\hbar k$ , i.e., such an atom experiences recoil. The influence of a recoil on the spatial motion of atoms can be divided into redistributions of the atomic velocities and redistributions of the atoms themselves in space under the influence of light. The latter effects include deflection,<sup>1</sup> scattering,<sup>2</sup> and focusing<sup>3</sup> of an atomic beam by light, capture of atoms in electromagnetic microtraps<sup>4,5</sup> and macrotraps,<sup>6,7</sup> appearance of density gradients in atomic vapors under the influence of light,<sup>8</sup> and several other effects. Many (but not all) of these effects are characterized by a strong spatial inhomogeneity of the optical field which can either be microinhomogeneous, as is the case for a standing wave, or macroinhomogeneous in the case of a spatially inhomogeneous distribution of the intensity of light. However, inhomogeneity of the optical field is unimportant in the redistribution of the velocities of atoms under the influence of light, which is the subject of the present paper. The existence of an optical field inhomogeneity simply makes it difficult to analyze theoretically the effects but makes no basic contribution. For this reason we shall use only the rate equations, even when the configuration of the optical field is assumed to be a standing wave (one- or three-dimensional) in order to make the problem symmetric and when the influence of coherent effects is no longer weak. One of the most interesting effects in the redistribution of the atomic velocities in a resonant optical field is the cooling of gases which occurs on excitation of the low-frequency half of a Doppler-broadened absorption line. This effect was first predicted by Hänsch and Schawlow<sup>9</sup> in 1975 and has already been observed in experiments involving trapped ions.<sup>10,11</sup> Many papers have since been published on the optical cooling of atoms and gases. The kinetic theory of the cooling effect was developed in an earlier paper by Klimontovich and the present author,<sup>12</sup> where a simple formula was obtained for the rate of optical cooling of a gas under weak saturation conditions. It is important to stress that this formula is valid only when light does not deform the Maxwellian distribution function of the atomic

velocities. It is thus assumed that the whole cooling process reduces to an adiabatically slow reduction in the gas temperature. This assumption is justified only when collisions can restore the Maxwellian distribution function which is continuously disturbed by the incident light. A more detailed analysis of the gas cooling kinetics requires rigorous criteria for the validity of adiabatic approximation. Allowance for the collisions of atoms is essential also in another interesting effect predicted earlier.<sup>13</sup> If the collisions of atoms are few and they do not manage to restore the Maxwellian distribution, the incident light transfers atoms from the resonant to the nonresonant range and produces a narrow dip, whose width can be close to the resonance width. Formation of such a dip in the momentum distribution thus occurs exactly when the adiabatic approximation is invalid. If this effect occurs during the optical cooling of atoms, it can reduce considerably the rate of cooling because of the shortage of resonant atoms, which are displaced to the range of lower velocities. The method of a narrow dip produced by the incident light (hole burning method) may become one of the more promising methods in the optical and  $\gamma$  spectroscopy inside the Doppler profile and may compete successfully with, for example, the ultrahigh-resolution method of three-level laser spectroscopy.<sup>14</sup> The kinetics of filling the dip after the end of illumination, as well as the kinetics of its formation and the steady-state profile all contain information on the velocity-changing atomic collisions.

The present paper represents the first attempt (apart from the communication mentioned earlier<sup>13</sup>) to develop a theory of the redistribution of the atomic velocities under the influence of resonant light, subject to an allowance for the collisions of atoms. The problem can be solved analytically if the collisions are assumed to be weak.

## 2. "RATE EQUATIONS" WITH ALLOWANCE FOR RECOIL

In the description of the recoil effects applicable to the dipole interaction of an atom with an electromagnetic field Hamiltonian  $\hat{H}$  can assume to be<sup>15</sup>

$$\dot{H} = \dot{H}_A - dE(\mathbf{R}, t), \quad (1)$$

where the atomic Hamiltonian  $\hat{H}_A$  contains also the operator of the kinetic energy of the center of mass of the atom  $\hat{P}^2/2M$ . In general, the state of an atom is described by the density operator  $\hat{f}$  satisfying the Neumann equation of motion. We shall use the density operator representation, which is of the Wigner type in the case of the spatial variables of an atom and of the energy type in the case of the variables of its internal motion. Then, the Neumann equation becomes the following system of equations for the matrix elements  $f_{mn}(\mathbf{R}, \mathbf{P}, t)$ :

$$\begin{aligned} & \left( \frac{\partial}{\partial t} + \mathbf{v} \frac{\partial}{\partial \mathbf{R}} + i\omega_{mn} \right) f_{mn}(\mathbf{R}, \mathbf{P}, t) \\ &= \frac{1}{(2\pi)^3} \frac{i}{\hbar} \sum_{\mathbf{l}} \int \left\{ d_{ml} \mathbf{E} \left( \mathbf{R} + \frac{1}{2} \hbar \mathbf{\Pi}, t \right) f_{ln}(\mathbf{R}, \mathbf{P}', t) \right. \\ & \left. - d_{ln} \mathbf{E} \left( \mathbf{R} - \frac{1}{2} \hbar \mathbf{\Pi}, t \right) f_{ml}(\mathbf{R}, \mathbf{P}', t) \right\} e^{i\mathbf{\Pi}(\mathbf{P}' - \mathbf{P})} d\mathbf{\Pi} d\mathbf{P}' + I_{mn}(\mathbf{R}, \mathbf{P}, t) \\ & \quad + J_{mn}(\mathbf{R}, \mathbf{P}, t). \end{aligned} \quad (2)$$

Equation (2) includes also the relaxation matrices  $I_{mn}$  and  $J_{mn}$  linked directly to the electromagnetic and collisional relaxational processes. If we ignore the terms  $\pm \frac{1}{2} \hbar \mathbf{\Pi}$  in the arguments of the field, the integrals with respect to  $\mathbf{\Pi}$  and  $\mathbf{P}'$  are calculated readily and we obtain the well-known system of equations without recoil. The system (2) applies to an arbitrary atomic spectrum. It simplifies greatly if the field is resonant with just one electron transition in an atom. In the case of the problems considered here it is essential that the lower active level 1 is the ground or metastable state, whereas the upper level 2 decays most likely to the ground state. In this case the relaxation matrix  $I_{mn}$  has the form<sup>16</sup>

$$I_{mn}(\mathbf{R}, \mathbf{P}, t) = \begin{pmatrix} 2\Gamma_{f_{22}}(\sim) & -\Gamma_{f_{12}}(0) \\ -\Gamma_{f_{12}}(0) & -2\Gamma_{f_{22}}(0) \end{pmatrix}, \quad (3)$$

where  $\Gamma = 2d_0^2\omega^3/\hbar c^3$  and in the optical part of the spectrum it is of the order of  $10^7 - 10^8 \text{ sec}^{-1}$ . If the recoil effect is ignored, the arguments of all the functions  $f_{mn}$  are not shifted [i.e., they are identical with the corresponding arguments on the left-hand sides of the equations in the system (2)]. In the case of spontaneous emission of light by an atom excited to the upper active level, its momentum changes in accordance with the angular distribution of the scattered light. If the light is linearly polarized, the probability of resonant scattering into a solid angle  $d\Omega$  is governed by the angular distribution of the electric dipole excited along the electric vector of the wave. Consequently, the arguments of the functions in the relaxation matrix are shifted by an amount of the order of the photon momentum. The following notation is used in Eq. (3) and below:

$$\begin{aligned} f_{mn}(\mathbf{R}, P_i \pm (N/2)\hbar k \delta_{3i}, t) &= f_{mn}(\pm N), \quad N=0, 1, 2; \\ \frac{3}{8\pi} \int d\mathbf{n} (1-n_i^2) f_{22}(\mathbf{R}, \mathbf{P} + \hbar k \mathbf{n}, t) &= f_{22}(\sim). \end{aligned}$$

1. Let us assume that the field has the configuration of a linearly polarized traveling wave

$$\mathbf{E}(\mathbf{R}, t) = E_0 \mathbf{e}_1 \cos(\omega t - kR_3).$$

Then, the integrals in Eq. (2) can be calculated. Next, expressing the nondiagonal matrix elements in terms of the diagonal elements, and substituting there the equations for the diagonal elements, we obtain the equations

$$\begin{aligned} \frac{\partial f_{mn}(0)}{\partial t} &= \Gamma G \mathcal{L}(\Omega + kV_3 \pm \delta) [f_{nn}(\pm 2) - f_{mn}(0)] \\ & \pm 2\Gamma f_{22} \left( \begin{smallmatrix} \sim \\ 0 \end{smallmatrix} \right) + J_{mn}(\mathbf{R}, \mathbf{P}, t) + \left[ V_{nm}^{(\pm)} \frac{\partial f_{mn}(\pm 1)/\partial t'}{\pm(\Omega + kV_3 \pm \delta) + i\Gamma} + \text{c.c.} \right] \end{aligned} \quad (4)$$

$$V_{nm}^{(+)} = -V_0 \exp(\pm i\Omega t + ikR_3), \quad V_{nm}^{-} = -V_0 \exp(\pm i\Omega t - ikR_3), \quad V_0 = d_0 E_0 / 2\hbar.$$

In the system (4) we have again used the notation in which  $\Omega = \omega_0 - \omega$  is the detuning,  $G = 2V_0^2/\Gamma^2$  is the saturation parameter,  $\mathcal{L}(\Omega) = \Gamma^2/(\Omega^2 + \Gamma^2)$  is the Lorentz factor. The frequency  $\delta$  is equal to the recoil energy  $R_r = \hbar^2 k^2/2M$ , divided by  $\hbar$ . In all cases the upper sign should be taken as corresponding to  $m=1, n=2$ , where as the lower sign corresponds to  $m=2, n=1$ . In the derivation of Eq. (4) it is assumed that the relaxation of the nondiagonal matrix elements is mainly due to electromagnetic processes, which is valid at pressures discussed here.

2. In the case of a standing wave, we have

$$\mathbf{E}(\mathbf{R}, t) = 2E_0 \mathbf{e}_1 \cos \omega t \cos kR_3$$

and in Eq. (4) we have to add terms differing from those already present by the replacement of  $k$  with  $-k$  (Ref. 17):

$$\begin{aligned} \frac{\partial f_{mn}(0)}{\partial t} &= \Gamma G \mathcal{L}(\mp \Omega - kV_3 - \delta) [f_{nn}(2) - f_{mn}(0)] \\ & + \Gamma G \mathcal{L}(\mp \Omega + kV_3 - \delta) [f_{nn}(-2) - f_{mn}(0)] \pm 2\Gamma f_{22} \left( \begin{smallmatrix} \sim \\ 0 \end{smallmatrix} \right) \\ & + J_{mn}(\mathbf{R}, \mathbf{P}, t) + \left[ V_{nm}^{(+)} \frac{\partial f_{mn}^{(-)}(1)/\partial t'}{\pm \Omega + kV_3 + \delta + i\Gamma} + V_{nm}^{(-)} \frac{\partial f_{mn}^{(+)}(-1)/\partial t'}{\pm \Omega - kV_3 + \delta + i\Gamma} + \text{c.c.} \right]. \end{aligned} \quad (5)$$

The nondiagonal matrix elements  $\tilde{f}_{mn} = f_{mn} \exp(\mp i\omega_0 t)$  depend on time in two ways: they oscillate rapidly as  $\exp(\mp i\Omega t)$  under the action of the field and they also vary slowly with the phase density. In Eqs. (4) and (5) the symbol  $\partial/\partial t'$  denotes differentiation with respect to the "slow time."

Equations (4) and (5) could have been called the rate equations had they not contained additional terms associated with the delay processes. Transient processes contribute to the diffusion coefficients and to the optical pressure (Sec. 4).

### 3. APPROXIMATION OF WEAK COLLISIONS FOR $J(\mathbf{R}, \mathbf{P}, t)$

The relaxation of the Wigner distribution function  $f(\mathbf{R}, \mathbf{P}, t)$  is due to the term  $J(\mathbf{R}, \mathbf{P}, t) = J_{11} + J_{22}$  known as the collision integral. In the model of weak collisions, it has the Fokker-Planck form<sup>18</sup>

$$J(\mathbf{R}, \mathbf{P}, t) = \gamma \frac{\partial}{\partial \mathbf{P}} (\mathbf{P} f) + D \frac{\partial^2 f}{\partial \mathbf{P}^2}, \quad (6)$$

where  $\gamma$  is the effective collision frequency and  $D$  is the diffusion coefficient. If  $J(\mathbf{R}, \mathbf{P}, t)$  in the form of Eq. (6) is related to the collisions of atoms with one another in the investigated gas, we must also ensure that the distribution function is close to equilibrium. Otherwise,  $J(\mathbf{R}, \mathbf{P}, t)$  depends nonlinearly on this function. The condition of closeness of the distribution function to the equilibrium form can be lifted if we assume that Eq. (6) is governed mainly by the interaction of atoms with the thermostat. This thermostat can be equilibrium ther-

mal radiation, but its intensity is far too low to compete with the incident laser radiation. Therefore, we shall assume that the collision integral (6) can be accounted for by the scattering of light atoms of thermostatting gas from the heavy atoms of the investigated gas, when the latter are just an impurity in the thermostatting gas. The thermostatting gas is in an equilibrium state if the laser radiation produces no transitions between the levels of its atoms.

#### 4. FIRST APPROXIMATION WITH RESPECT TO $\delta/\Delta\omega_D$ AND $\delta/\Gamma_G$

Adding both equations in (4) or (5), we obtain an equation for the Wigner distribution function for the cases of traveling and standing waves, respectively. The momentum functions occurring in these equations can be expanded as the Taylor series at the point  $P_3$ . We can easily see that this corresponds to the expansion of the right-hand sides of the equations generally in terms of two small parameters:  $\delta/\Delta\omega_D$  and  $\delta/\Gamma_G$  [ $\Gamma_G = \Gamma(1+G)^{1/2}$ , where  $\Delta\omega_D$  is the width of the momentum distribution measured in frequency units]. Since the quasiclassical parameter  $\delta/\Delta\omega_D$  is inversely proportional to the width of the momentum distribution and the parameter  $\delta/\Gamma_G$  is proportional to the width of a Bennett hole which is burnt in this distribution, the former is usually smaller than the latter.

1. In the first approximation with respect to the recoil parameters, we obtain from Eq. (4)

$$\frac{\partial f^{(1)}}{\partial t} = -\frac{\partial}{\partial P_3} [\hbar k \Gamma \mathcal{L}(\Omega + kV_3)(f_{11}^{(0)} - f_{22}^{(0)})] + J^{(1)}(\mathbf{R}, \mathbf{P}, t). \quad (7)$$

Here and later, an index in parentheses denotes the order of the approximation in which a given function is selected. The solution, in the zeroth approximation with respect to the recoil parameters, giving the population difference is well known:

$$f_{11}^{(0)} - f_{22}^{(0)} = \frac{f^{(0)}}{1 + G \mathcal{L}(\Omega + kV_3)}. \quad (8)$$

If we use  $\mathcal{L}_G(\Omega) = \Gamma_G^2 / (\Omega^2 + \Gamma_G^2)$ , we obtain the kinetic equation for  $f^{(1)}(\mathbf{R}, \mathbf{P}, t)$ :

$$\frac{\partial f^{(1)}}{\partial t} = -\frac{\partial}{\partial P_3} \left[ \frac{\hbar k \Gamma G}{1+G} \mathcal{L}_G(\Omega + kV_3) f^{(1)} \right] + J^{(1)}(\mathbf{R}, \mathbf{P}, t), \quad (9)$$

from which it follows directly that the "spontaneous optical pressure" is given by:

$$F_{sp}^{(1)} = \frac{\hbar k \Gamma G}{1+G} \mathcal{L}_G(\Omega + kV_3). \quad (10)$$

We shall find the steady-state solution of Eq. (9) using the collision integral (6). The distribution with respect to  $P_1$  and  $P_2$  retains its Maxwellian form. However, the

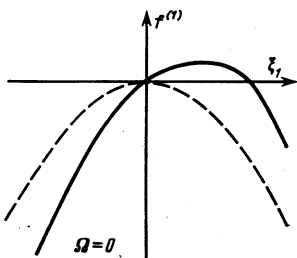


FIG. 1.

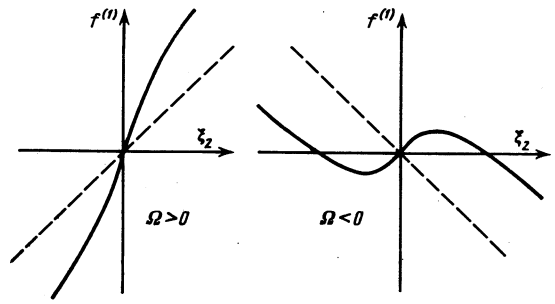


FIG. 2.

dependence of  $f^{(1)}$  on  $u = kV_3$  is now

$$f^{(1)}(u) \sim \exp \left[ -\frac{\gamma M^2}{2k^2 D} u^2 + \frac{\hbar \Gamma^2 G M}{(1+G)^{1/2} D} \arctg \left( \frac{\Omega + u}{\Gamma_G} \right) \right]. \quad (11)$$

a) Let us assume that  $\Omega = 0$ . Then,

$$f^{(1)}(\xi_1) \sim \exp \left[ -\frac{\Gamma_G^2}{(\Delta\omega_D)^2} (\xi_1^2 + \alpha_1 \arctg \xi_1) \right], \quad (12)$$

where  $\alpha_1 = 2\delta G / (1+G)\gamma$ ,  $\xi_1 = u/\Gamma_G$ . If  $|\xi_1| \leq 1$ , then the function  $f^{(1)}(\xi_1)$  has the form shown in Fig. 1.

b) We shall now consider an alternative to the case a) which represents large detuning  $|\Omega| \gg \Gamma_G$ . If we introduce

$$\alpha_2 = 2\delta \Gamma G / (1+G)\gamma \Omega, \quad \xi_2 = (\Omega + u)/\Gamma_G,$$

we find that if  $|\xi_2| \ll \Omega/\Gamma_G$ , then from Eq. (11)

$$f^{(1)}(\xi_2) \sim \exp \left( -\frac{\Omega^2}{(\Delta\omega_D)^2} \right) \exp \left[ \frac{2\Omega \Gamma_G}{(\Delta\omega_D)^2} (\xi_2 + \alpha_2 \arctg \xi_2) \right]. \quad (13)$$

The graph of  $f^{(1)}(\xi_2)$  is shown in Fig. 2 for two signs of detuning. We can see from this that if  $\Omega < 0$  and  $|\alpha_2| > 1$ , a dip (hole) forms in the distribution. Its depth is usually less than its width.

2. In the case of a field configuration in the form of a standing wave in Eq. (9), the spontaneous optical pressure contains a contribution from both traveling waves<sup>5</sup>:

$$F_{sp}^{(0)} = \hbar k \Gamma G \frac{\mathcal{L}(\Omega + kV_3) - \mathcal{L}(\Omega - kV_3)}{1 + G[\mathcal{L}(\Omega + kV_3) + \mathcal{L}(\Omega - kV_3)]}. \quad (14)$$

If  $\Omega = 0$ , this pressure vanishes and, therefore, the distribution function is not deformed. However, if  $|\Omega| \gg \Gamma_G$ , the distribution function in the vicinity of a resonance still has the form given by Eq. (13).

#### 5. SECOND APPROXIMATION WITH RESPECT TO $\delta/\Delta\omega_D$ AND $\delta/\Gamma_G$

In the first approximation with respect to the recoil parameters, the kinetic equation acquires a term with the optical pressure. The second approximation allows us also to include fluctuations of this pressure.

1. In the case of a traveling wave, we obtain from Eq. (4)

$$\begin{aligned} \frac{\partial f^{(2)}}{\partial t} = & -\frac{\partial}{\partial P_3} [\hbar k \Gamma G \mathcal{L}_G(\Omega + kV_3)(f_{11}^{(1)} - f_{22}^{(1)})] + \frac{1}{2} \hbar^2 k^2 \Gamma G \frac{\partial}{\partial P_3} \left( \mathcal{L}_+ \frac{\partial f^{(0)}}{\partial P_3} \right) \\ & + \frac{1}{2} \hbar^2 k^2 \Gamma G \sum_{i=1}^2 c_i \frac{\partial^2}{\partial P_i^2} \left( \frac{\mathcal{L}_+}{1+G \mathcal{L}_+} f^{(0)} \right) \\ & + 2\hbar k \frac{\partial}{\partial P_3} \text{Re} \left[ \frac{V_{21}^{(+)}}{\Omega + kV_3 + i\Gamma} \frac{\partial f_{12}^{(1)}}{\partial t'} \right] + J^{(2)}(\mathbf{R}, \mathbf{P}, t), \end{aligned} \quad (15)$$

where  $c_1 = 1/5$  and  $c_2 = c_3 = 2/5$  (Refs. 16 and 19). Here and later,  $\mathcal{L}(\Omega \pm kV_3)$  is reduced to  $\mathcal{L}_\pm$ .

The difference between the populations in the first approximation  $f_{11}^{(1)} - f_{22}^{(1)}$  occurring in Eq. (15) and the term with  $\partial f_{12}^{(1)}/\partial t'$  can be expressed in terms of the phase density. For example,

$$f_{11}^{(1)} - f_{22}^{(1)} = \frac{f^{(1)} + \frac{1}{2} \hbar k G \mathcal{L}_+ \partial f^{(0)}/\partial P_3}{1 + G \mathcal{L}_+} + \frac{1}{2} \hbar k \frac{1 + 2G \mathcal{L}_+ (1 - 2\mathcal{L}_+)}{(1 + G \mathcal{L}_+)^2} \frac{\partial}{\partial P_3} \left( \frac{G \mathcal{L}_+}{1 + G \mathcal{L}_+} f^{(0)} \right). \quad (16)$$

Substituting Eq. (16) into Eq. (15), we obtain the kinetic equation in the second approximation

$$\begin{aligned} \frac{\partial f^{(2)}}{\partial t} = & -\hbar k \Gamma \frac{\partial}{\partial P_3} \left\{ \frac{G \mathcal{L}_+}{1 + G \mathcal{L}_+} + \frac{1}{2} \hbar k \frac{\partial}{\partial P_3} \left( \frac{G \mathcal{L}_+}{1 + G \mathcal{L}_+} \right) \right. \\ & \left. + \frac{1}{2} \hbar k \frac{G \mathcal{L}_+}{1 + G \mathcal{L}_+} \frac{\partial}{\partial P_3} \left[ \frac{G \mathcal{L}_+ (1 - 4\mathcal{L}_+)}{(1 + G \mathcal{L}_+)^2} \right] \right\} \\ & + \frac{1}{2} \hbar^2 k^2 \Gamma \sum_{i=1}^3 \frac{\partial^2}{\partial P_i^2} \left\{ (\delta_{3i} + c_i) \frac{G \mathcal{L}_+}{1 + G \mathcal{L}_+} + \delta_{3i} \frac{G^2 \mathcal{L}_+^2 (1 - 4\mathcal{L}_+)}{(1 + G \mathcal{L}_+)^3} \right\}. \quad (17) \end{aligned}$$

Fluctuations of the optical pressure are described by the second term on the right-hand side of Eq. (17). In its turn, this term consists of three components, the first of which is associated with the recoil occurring in the induced absorption of a photon by an atom which alters only  $P_3$ , whereas the second component is due to the recoil in the case of spontaneous deexcitation of the excited upper state contributing to all the projections of the momentum in accordance with the angular distribution of the scattered light. The two processes (absorption and emission) are aspects of the same two-photon process of resonant fluorescence and, therefore, the separation into absorption and emission is arbitrary. Each photon absorption event is necessarily accompanied by a photon emission event, so that the sum of the coefficients  $c_i$  in Eq. (17) is unity irrespective of the polarization of light. It follows from Eq. (17) that in the momentum space the diffusion coefficients are limited only by the rate of spontaneous decay. This is to be expected because the induced processes of the reemission of photons in a traveling wave cannot broaden the momentum distribution. In the second approximation the spontaneous pressure contains a correction which is  $\sim \delta/\Gamma_G$  times less than the main part of the pressure.<sup>11</sup> Its origin can be explained by a shift of the absorption resonance if allowance is made for the recoil by  $\delta$ . The last terms in the expressions for the pressure and diffusion coefficients are related to the delay processes and, therefore, cannot be explained in terms of the photon language.

2. In the case of a standing wave we have, by complete analogy with Sec. 5.1,

$$\begin{aligned} F_3 = & \hbar k \Gamma G \frac{\mathcal{L}_+ - \mathcal{L}_-}{1 + G(\mathcal{L}_+ + \mathcal{L}_-)} + \frac{1}{2} \hbar^2 k^2 \Gamma G \frac{\partial}{\partial P_3} \left[ \frac{\mathcal{L}_+ + \mathcal{L}_- + 4G \mathcal{L}_+ \mathcal{L}_-}{1 + G(\mathcal{L}_+ + \mathcal{L}_-)} \right] \\ & + \frac{1}{2} \hbar^2 k^2 \Gamma G^2 \frac{\mathcal{L}_+ - \mathcal{L}_-}{1 + G(\mathcal{L}_+ + \mathcal{L}_-)} \\ & \times \frac{\partial}{\partial P_3} \left\{ \frac{(\mathcal{L}_+ - \mathcal{L}_-)[1 - 4(\mathcal{L}_+ + \mathcal{L}_-) - 8G \mathcal{L}_+ \mathcal{L}_-]}{[1 + G(\mathcal{L}_+ + \mathcal{L}_-)]^2} \right\}, \\ D_i = & \frac{1}{2} \hbar^2 k^2 \Gamma G \left\{ \frac{(\delta_{3i} + c_i)(\mathcal{L}_+ + \mathcal{L}_-) + 4\delta_{3i} G \mathcal{L}_+ \mathcal{L}_-}{1 + G(\mathcal{L}_+ + \mathcal{L}_-)} \right. \\ & \left. + G \frac{(\mathcal{L}_+ - \mathcal{L}_-)^2 [1 - 4(\mathcal{L}_+ + \mathcal{L}_-) - 8G \mathcal{L}_+ \mathcal{L}_-]}{[1 + G(\mathcal{L}_+ + \mathcal{L}_-)]^2} \right\}. \quad (18) \end{aligned}$$

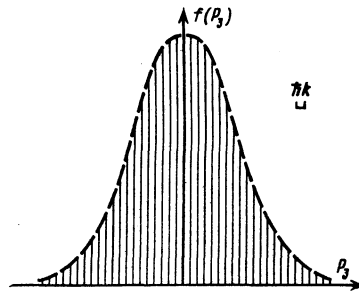


FIG. 3.

It is clear from the system (18) that the diffusion spreading of the momentum distribution function in a standing light wave occurs not only because of the spontaneous scattering but also because of the induced re-emission of photons by an atom and these photons are transferred from one traveling wave to another.<sup>20</sup>

a) If the detuning is  $\Omega = 0$ , we find from Eq. (18) that

$$\begin{aligned} \frac{\partial f^{(2)}}{\partial t} = & \hbar^2 k_0^2 \Gamma G \frac{\partial}{\partial P_3} \left[ \mathcal{L}(k_0 V_3) \frac{\partial f^{(2)}}{\partial P_3} \right] \\ & + \hbar^2 k_0^2 \Gamma G \sum_{i=1}^3 c_i \frac{\partial^2}{\partial P_i^2} \left[ \frac{\mathcal{L}(k_0 V_3)}{1 + 2G \mathcal{L}(k_0 V_3)} f^{(2)} \right] + J^{(2)}(\mathbf{R}, \mathbf{P}, t). \quad (19) \end{aligned}$$

We can see that the first term on the right-hand side of Eq. (19) increases proportionally to the intensity of light, whereas the corresponding term in Eq. (17) is limited by the rate of spontaneous decay and is saturated in strong fields. To illustrate the role of induced re-emission of photons in the broadening of the momentum distribution, we show in Fig. 3 its form at times  $t \gg (\Gamma_G)^{-1}$ . Spontaneous processes are ignored here and it is also assumed that initially the velocities of all the atoms vanish.

b) When the detuning is large so that  $|\Omega| \gg \Gamma_G$ , traveling waves interact with different groups of atoms, so that the induced re-emission of photons is a rare event. It then follows from Eq. (18) that the diffusion coefficients are governed mainly by the spontaneous scattering.

## 6. FORMATION OF A NARROW DIP IN THE MOMENTUM DISTRIBUTION

It is clear from Figs. 1 and 2 that the action of the optical pressure results in a fairly smooth deformation of the momentum distribution. Only fluctuations of this force can, under certain conditions, result in the formation of narrow dips in the distribution. The dips appear at points corresponding to resonant atoms which are optically displaced to the nonresonant region. We shall consider here only the case of a standing light wave tuned to the transition frequency. With this in mind, we shall obtain the steady-state solution of the kinetic equation (19) with the collision integral in the form of Eq. (6). Consequently, the distribution function  $f^{(2)}(u)$  is found from the differential equation

$$\frac{df^{(2)}}{f^{(2)}} = -\frac{\gamma M^2}{2k_0^2 D} \frac{1 - \beta_1 \mathcal{L}_{20}^2(u)}{1 + \beta_2 \mathcal{L}(u) + \beta_3 \mathcal{L}_{20}(u)} d(u^2), \quad (20)$$

where

$$\beta_1 = \frac{4\hbar^2 k_0^4 G}{5(1+2G)^2 M^2 \Gamma}, \quad \beta_2 = \frac{\hbar^2 k_0^2 \Gamma G}{D}, \quad \beta_3 = \frac{2\hbar^2 k_0^2 \Gamma G}{5D(1+2G)}.$$

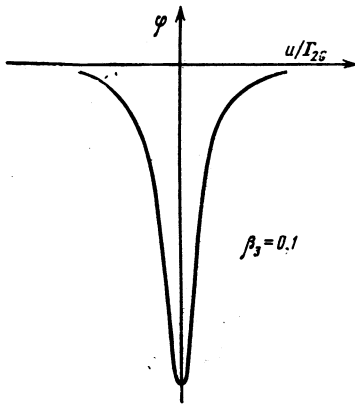


FIG. 4.

The condition  $df^{(2)}/du = 0$  gives the positions of the maxima in the distribution:

$$u_1^2 + \Gamma_{2G}^2 = \Gamma_{2G}^2 \beta_1^{1/2}. \quad (21)$$

Thus, a dip forms at  $\beta_1 > 1$ . At lower values of  $\beta_1$  the top of the distribution is flattened. In Sec. 4 we find that, in the first approximation with respect to the recoil parameters, the dip in the distribution appears for  $|\alpha_2| > 1$ . As expected,  $\beta_1 \propto \delta |\alpha_2| / \Gamma_{2G}$ . We shall show that the width of the dip measured in terms of frequency units has a lower limit set by the resonance width  $\Gamma_{2G}$ . Broadening of the dip by strong light can be avoided if  $G \leq 1$ . Then,  $\beta_2 \propto \beta_3 \propto \beta_1 (\Gamma / \Delta \omega_D)^2$ . Consequently, if  $\beta_2, \beta_3 \ll 1$  and  $|u| \leq \Gamma_{2G}$ , the denominator of Eq. (20) is close to unity, whereas the numerator depends strongly on  $u$ . The solution of Eq. (20) can then be easily obtained in this approximation:

$$f^{(2)}(u) \propto \exp\left(-\frac{\gamma M^2}{2k_0^2 D} u^2\right) \varphi(u), \quad (22)$$

where the dip function  $\varphi(u)$  is

$$\varphi(u) = \exp[-\beta_3 \mathcal{L}_{2G}(u)]. \quad (23)$$

The half-width of this function at half-maximum is given by

$$u_{1/2}^2 = \Gamma_{2G}^2 \left( \frac{\beta_3}{-\ln[1/2(1+e^{-\beta_3})]} - 1 \right). \quad (24)$$

If  $\beta_3 \ll 1$ , then  $u_{1/2} \approx \Gamma_{2G}$ . The graph of  $\varphi(u)$  is shown in Fig. 4. If  $G \ll 1$ , the solution of this problem is the same as that obtained earlier.<sup>13</sup>

## 7. MINIMUM TEMPERATURE OBTAINABLE IN OPTICAL COOLING OF ATOMS

We shall not analyze in detail the problem of optical cooling of atoms. We shall simply refer to earlier investigations<sup>5,12</sup> of this problem. In particular, the minimum cooling temperature, governed by the competition between the processes of cooling of atoms by the average optical pressure and their heating because of fluctuations of this force, is found in Ref. 5. Our aim is to determine how fluctuations of the optical pressure in the case of remission of photons, ignored in Refs. 5 and 12, can affect the minimum temperature attainable in such cooling. We shall ignore the collisions of atoms assuming that  $J^{(2)} = 0$  and we shall consider only that part of the distribution where the following inequality is

satisfied:

$$kV_s \ll \Gamma(\Omega/\Gamma + \Gamma/\Omega).$$

Under these conditions, Eq. (17), which is generalized to the case of a three-dimensional standing wave in order to symmetrize the distribution,<sup>5</sup> can be reduced directly to the Fokker-Planck equation

$$\frac{\partial f^{(2)}}{\partial t} = -\gamma' \frac{\partial}{\partial \mathbf{P}} (\mathbf{P} f^{(2)}) + D' \frac{\partial^2 f^{(2)}}{\partial \mathbf{P}^2} \quad (25)$$

with the friction coefficient

$$\gamma' = \frac{8\delta \Omega G \mathcal{L}^2(\Omega)}{\Gamma[1+6G\mathcal{L}(\Omega)]} \quad (26)$$

and the diffusion coefficient

$$D' = 4\delta M G \hbar \Gamma \mathcal{L}(\Omega) \frac{1+G\mathcal{L}(\Omega)}{1+6G\mathcal{L}(\Omega)}. \quad (27)$$

It is well known that the steady-state solution of Eq. (25) is the Maxwellian distribution with the following temperature

$$k_B T = \frac{D'}{\gamma' M} = \frac{1}{2} \hbar \Gamma \left( \frac{\Omega}{\Gamma} + \frac{\Gamma}{\Omega} \right) [1+G\mathcal{L}(\Omega)]. \quad (28)$$

At this temperature the quasiclassical parameter  $\delta/\Delta\omega_D$  is still small. If  $G\mathcal{L}(\Omega) \ll 1$ , we obtain the result of Ref. 5.

## 8. ESTIMATES OF PRINCIPAL QUANTITIES

The collision frequency  $\gamma$  in Eq. (6) is proportional to the gas pressure  $p$ . We shall use the estimate  $\gamma[\text{sec}^{-1}] \sim 10^6 p [\text{Torr}]$ . In the optical part of the spectrum the frequency is  $\delta \sim 10^5 \text{ sec}^{-1}$ . If the intensity of light is  $100 \text{ mW/cm}^2$ , then  $G \sim 1$ . Light deforms strongly the Maxwellian distribution if  $\alpha_1 \sim 1$ , i.e., when the pressure is  $p \sim 10^{-1} \text{ Torr}$ . A narrow dip appears in the distribution at much lower pressures of  $p \sim 10^{-5} \text{ Torr}$ , when  $\gamma \sim 10 \text{ sec}^{-1}$ ,  $\beta_3 \sim 10^{-1}$ , and  $\beta_1 \sim 10^2$ . If the gas is in a conventional ampoule, we have to allow also for the contribution made to  $\gamma$  by the collisions of atoms with the ampoule walls. Even if the dimensions of the ampoule are  $\sim 10 \text{ cm}$ , this contribution is considerable:  $\sim 10^3 \text{ sec}^{-1}$ . Thus, only a "smooth" deformation of the distribution function under the action of light can be observed in an ampoule. A narrow dip is obtained only if atoms are confined in a special electromagnetic trap where light acts as the walls.<sup>21</sup> Such traps are being developed and will be realized in the near future. Optical traps provide ideal conditions not only for spectroscopic measurements but also for the investigation of atomic collisions.

## 9. CONCLUSIONS

The optical burning of a narrow dip (hole) in the distribution of the atomic velocities can be observed in the following proposed experiment. A gas of atoms which is confined to an optical trap where the pressure is  $p \leq 10^{-5} \text{ Torr}$  is illuminated with resonant light of intensity  $w \sim 100 \text{ mW/cm}^2$  for a time  $\tau > \gamma^{-1}$  sufficient to produce a dip; next, illumination is terminated abruptly (so that the radiation used to confine the atoms itself would not disturb the distribution function; a trap should be hollow, i.e., it should be a field-free region in space surrounded by light characterized by  $\Omega < 0$ ). Then, after a

time  $\sim \Gamma^{-1} \sim 10^{-7}$  sec all the atoms are again in the ground state and, therefore, the distribution of the atomic velocities at the lower active level no longer has a Bennett dip (hole) associated with the resonant nature of the excitation of atoms. In spite of this, for an additional time of  $\sim \gamma^{-1} \sim 10^{-1}$  sec the momentum distribution still has a dip of half-width (24), which is explained by a redistribution of the velocities of atoms under the influence of light. This can be discovered from the resonant reduction in the absorption by a test light wave in the case of exact tuning to one of the allowed transitions from the ground state. An experiment of this kind can also be used to resolve closely spaced spectral lines masked by the Doppler broadening. Moreover, there is a possibility of observing directly the process of relaxation of the nonequilibrium distribution of the atomic velocities to its equilibrium form, i.e., in the final analysis this makes it possible to study collisions of atoms in a gas (for example, the reciprocal of the time needed for the disappearance of a dip is equal to the frequency of the velocity-changing atomic collisions). Information on atomic collisions is contained also in the steady-state dip profile.

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<sup>1)</sup>The author's attention to this fact was drawn by V. G. Minogin.

<sup>1)</sup>A. Ashkin, Phys. Rev. Lett. 25, 1321 (1970).

<sup>2)</sup>E. Arimondo, H. Lew, and T. Oka, Phys. Rev. Lett. 43, 753 (1979).

<sup>3)</sup>Yu. L. Klimontovich and S. N. Luzgin, Pis'ma Zh. Eksp. Teor. Fiz. 30, 645 (1979) [JETP Lett. 30, 610 (1979)].

<sup>4)</sup>A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 63, 1628 (1972) [Sov. Phys. JETP 36, 861 (1973)].

<sup>5)</sup>V. S. Letokhov, V. G. Minogin, and B. D. Pavlik, Zh. Eksp. Teor. Fiz. 72, 1328 (1977) [Sov. Phys. JETP 45, 698 (1977)].

<sup>6)</sup>A. Ashkin, Phys. Rev. Lett. 40, 729 (1978).

<sup>7)</sup>V. S. Letokhov and V. G. Minogin, Appl. Phys. 17, 99 (1978).

<sup>8)</sup>J. E. Bjorkholm, A. Ashkin, and D. B. Pearson, Appl. Phys. Lett. 27, 534 (1975).

<sup>9)</sup>T. W. Hansch and A. L. Schawlow, Opt. Commun. 13, 68 (1975).

<sup>10)</sup>W. Neuhauser, M. Hohenstatt, P. E. Toschek, and H. G. Dehmelt, Appl. Phys. 17, 123 (1978).

<sup>11)</sup>D. J. Wineland, R. E. Drullinger, and F. L. Walls, Phys. Rev. Lett. 40, 1639 (1978).

<sup>12)</sup>Yu. L. Klimontovich and S. N. Luzgin, Zh. Tekh. Fiz. 48, 2217 (1978) [Sov. Phys. Tech. Phys. 23, 1269 (1978)].

<sup>13)</sup>S. N. Luzgin, Pis'ma Zh. Tekh. Fiz. 5, 777 (1979) [Sov. Tech. Phys. Lett. 5, 319 (1979)].

<sup>14)</sup>V. S. Letokhov and V. S. Chebotayev, Printsipy nelineinoi lazernoj spektroskopii (Nonlinear Laser Spectroscopy, Springer Verlag, Berlin, 1977).

<sup>15)</sup>S. Stenholm, Phys. Rep. C 43, 151 (1978).

<sup>16)</sup>E. V. Baklanov and B. Ya. Bubetskii, Opt. Spektrosk. 41, 3 (1976) [Opt. Spectrosc. (USSR) 41, 1 (1976)].

<sup>17)</sup>S. Stenholm and J. Javanainen, Appl. Phys. 16, 159 (1978).

<sup>18)</sup>S. G. Rautian and I. I. Sobel'man, Usp. Fiz. Nauk 90, 209 (1966) [Sov. Phys. Usp. 9, 701 (1967)].

<sup>19)</sup>A. Yu. Pusep, Zh. Eksp. Teor. Fiz. 70, 851 (1976) [Sov. Phys. JETP 43, 441 (1976)].

<sup>20)</sup>A. P. Botin and A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 68, 2075 (1975) [Sov. Phys. JETP 41, 1038 (1975)].

<sup>21)</sup>A. Ashkin and J. P. Gordon, Opt. Lett. 4, 161 (1979).

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## Hyperfine shift of x-ray lines excited in internal conversion

A. I. Grushko, K. E. Kir'yanov, N. M. Miftakhov, A. S. Ryl'nikov, Yu. P. Smirnov, and V. V. Fedorov

*B. P. Konstantinov Leningrad Institute of Nuclear Physics, Academy of Sciences of the USSR*

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The energy shift of x-ray lines following internal conversion, which is due to the nonstatistical population of the hyperfine structure sublevels, is discussed. The nonstatistical population arises from the interaction of the magnetic moment of the nucleus in the final state with the electron current in the atomic shell ( $K$  or  $L$ ) that has a single vacancy. Expressions are obtained for the corresponding populations and shifts for the case of conversion transitions of arbitrary multipolarity. It is pointed out that from the experimental values of the shift one can derive the magnetic moment of the nucleus (provided the conversion-transition parameters are known) or the characteristics of the conversion transition itself (provided the nuclear magnetic moment is known). The shift of the barium  $K_{\alpha 1}$  line has been measured on isotopically enriched specimens, and the previously unknown magnetic moment of the excited state of the  $^{133}\text{Ba}$  nucleus has been determined from the results as  $+0.51 \pm 0.07$  nuclear magneton.

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The effect of the nonstatistical population of hyperfine-structure (HFS) sublevels of x-ray terms of atoms excited as a result of internal conversion was first noted in Ref. 1, where it was used to determine the magnetic

moment of a nucleus.

In the first part of the present paper we present a theoretical description of the effect and discuss its influ-

ence on the energy shifts of the x-ray emission lines<sup>1)</sup> that follow the internal conversion process. In the second part of the paper we describe an experiment to determine the magnetic moment of an excited state of the <sup>133</sup>Ba nucleus, using known crystal-diffraction techniques<sup>2</sup> for measuring small energy shifts of x-ray lines.

## THEORY

We shall calculate the probability  $W_e^F$  for excitation of a definite HFS sublevel ( $F$ ) of the final atom resulting from internal conversion. Let us consider the following process: the atom undergoes a transition from an initial state with total angular momentum  $I_0$  (the nucleus is in an excited state with spin  $I_0$  and the electron shell (the  $K$  and  $L$  electrons) is closed with zero angular momentum) to a final state (in which the nucleus has spin  $I$  and the corresponding shell has one vacancy and angular momentum  $j_0$ , the total angular momentum of the atom being<sup>2)</sup>  $F=I+j_0$ ) with emission of an electron into the continuum with angular momentum  $j$ . The matrix element for such a transition can be written in the form

$$H_e^F = \sum_{M, \mu, m_0, m} C_{j_0 m_0 M}^{I_0 0} C_{j m \mu}^{F M} \langle \Psi_{I_0 \mu} \Psi_{j_0 m_0} | H_e | \Psi_{I \mu} \Psi_{j m} \rangle (-1)^{j_0 + m_0} \\ = \sum_{M, \mu, m_0, m} C_{j_0 m_0 M}^{I_0 0} C_{j m \mu}^{F M} \langle \Psi_{I_0 \mu} \Psi_{j_0 m_0} | H_e | \Psi_{I \mu} \Psi_{j m} \rangle (-1)^{j_0 - m_0}, \quad (1)$$

where  $m$ ,  $m_0$ ,  $\mu$ ,  $\mu_0$ , and  $M$  are the projections of the angular momenta  $j$ ,  $j_0$ ,  $I$ ,  $I_0$ , and  $F$ , respectively;  $C_{j_1 m_1 j_2 m_2}^{j_3 m_3}$  is a Clebsch-Gordan coefficient;  $\Psi_{I_0 \mu_0}$  and  $\Psi_{I \mu}$  are the initial- and final-state wave functions of the nucleus while  $\Psi_{j_0 m_0}$  and  $\Psi_{j m}$  are those of the electron; and  $H_e$  is the operator for the conversion transition. The matrix element on the right in Eq. (1) is well known from the theory of internal conversion (see, e.g., Ref. 3, pp. 25 and 34) and has the form

$$\langle \Psi_{I_0 \mu_0} \Psi_{j_0 m_0} | H_e | \Psi_{I \mu} \Psi_{j m} \rangle = \sum_{L \Lambda} C_{I_0 \mu_0 L \Lambda}^{I_0 0} C_{j m \Lambda}^{I \mu} b_{\kappa}(\tau L). \quad (2)$$

Here  $L$  and  $\Lambda$  are the transition angular momentum (multipolarity) and its projection, respectively ( $|I_0 - I| \leq L \leq I_0 + I$ );  $\tau$  specifies the type of the transition ( $\tau = M$  for a magnetic transition and  $\tau = E$  for an electric transition); and  $\kappa$  characterizes the final state of the ejected (continuum) electron in the central field:  $\kappa = (l + j)(2j + 1)$  and  $j = |\kappa| - (\frac{1}{2})$ , where  $l$  and  $j$  are the orbital and total angular momenta of the electron, respectively.

On substituting (2) into (1) and performing the summation over the magnetic quantum numbers, we obtain (except for an unimportant phase factor) the matrix element for the conversion transition to a definite HFS state ( $F$ ) of the final atom:

$$H_e^F = [(2F+1)(2j+1)]^{1/2} \sum_L (-1)^L \left\{ \begin{matrix} I_0 & I & L \\ j_0 & F & L \end{matrix} \right\} b_{\kappa}(\tau L), \quad (3)$$

where the curly brackets denote the Wigner  $6j$  symbol.<sup>4</sup> The desired probability  $W_e^F$  itself is given by

$$W_e^F = \frac{2\pi}{\hbar} \sum_{\kappa} |H_e^F|^2. \quad (4)$$

The shift of the center of gravity of an x-ray line accompanying conversion with respect to that of a line emitted in the absence of hyperfine interaction is

$$\Delta E = \sum_F \Delta^F W_e^F / \sum_F W_e^F. \quad (5)$$

where  $\Delta^F$  is the hyperfine shift of the sublevel  $F$ . Formula (5) is valid if the hyperfine splitting of the final term of the x-ray transition can be neglected. We note that  $\Delta E$  vanishes in the case of a statistical population (when the probability  $W_e^F$  is proportional to the statistical weight  $2F+1$  of the final state of the atom). Such a situation arises, for example, in the photoexcitation of an atom,<sup>5</sup> and this makes it possible to use fluorescence lines as reference standards in experiments. In this case  $\Delta E = E^{\text{conv}} - E^{\text{phot}}$ , where  $E^{\text{conv}}$  and  $E^{\text{phot}}$  are the energies of the x-ray lines excited in internal conversion and by photoexcitation, respectively.

As a specific example we shall derive the expression for the shift  $\Delta E^{K\alpha_1}$  of the  $K\alpha_1$  line (arising from the transition between the  $K$  and  $L_{III}$  x-ray terms) excited as a result of an internal conversion transition of specified multipolarity  $\tau L$ . Then  $j_0 = \frac{1}{2}$ ,  $\Delta^{F=I+1/2} = -\Delta_K(I+1)/(2I+1)$ , and  $\Delta^{F=I-1/2} = \Delta_K I/(2I+1)$ ,<sup>5,6</sup> where  $\Delta_K$  is the hyperfine splitting of the  $K$  level<sup>3)</sup>:

$$\Delta_K = \alpha E_0 \left( \frac{m_p}{m_e} \right) \mu_I \frac{2I+1}{I} \frac{2(\alpha Z)^3}{3\gamma(2\gamma-1)} (1 - e_e - e_m), \quad (6)$$

in which  $\alpha$  is the fine structure constant,  $Z$  is the charge number of the nucleus,  $[\gamma = 1 - (\alpha Z)^2]^{1/2}$ ,  $m_e/m_p$  is the electron: proton mass ratio, and  $E_0 = m_e c^2$  is the electron rest energy,  $\mu_I$  is the magnetic moment of the nucleus in nuclear magnetons, and  $\omega_e$  and  $\omega_m$  are corrections for the distributions of charge and magnetization within the nucleus. Equation (6) is accurate to within 1%. Using Eqs. (3)–(5) in this case, we obtain

$$\Delta E^{K\alpha_1} = \Delta_K \frac{(I_0 - I)(I_0 + I + 1) - L(L + 1)}{2L(2I + 1)} \frac{1 - \rho}{1 + (L + 1)\rho/L}, \quad (7)$$

where  $\rho = |b_{\kappa_2}(\tau L)|^2 / |b_{\kappa_1}(\tau L)|^2$ ,  $|\kappa_1| < |\kappa_2|$ , and  $L > 0$  ( $\kappa_1$  and  $\kappa_2$  specify the two final continuum states possible for the electron in  $K$  conversion).

In the case of allowed nuclear transitions,  $b_{\kappa}$  is actually determined by matrix elements that are proportional to the amplitude for  $\gamma$ -ray emission and can be calculated fairly well numerically; use can be made of this circumstance to derive the magnetic moment from the hyperfine shift of the x-ray line.

We note that the above method of determining nuclear magnetic moments  $\mu$  differs from the known methods that make use of external magnetic fields in that the magnetic field acting on the nucleus can be calculated accurately enough since it is due to the current of the inner atomic electrons. Thus, the proposed method makes it possible to measure  $\mu$  directly. This is also of interest in connection with the experimental study of the physics of atomic effects that may, for example, strengthen or weaken external magnetic fields in the vicinity of the nucleus. In the case of strongly hindered transitions for which the amplitude for  $\gamma$ -ray emission is small (the case of anomalous conversion) the main contribution to  $b_{\kappa}$  comes from intranuclear conversion matrix elements, which are calculated on the basis of definite nuclear-structure models. In this case, if the magnetic moment of the final state of the nucleus is

known the line shift provides information on the adequacy of the model used in the calculation, which supplements the information that can be obtained by known methods from conversion-coefficient measurements and correlation experiments.

We also note that in the case of a mixed transition the expression for  $W_0^{\alpha}$  contains an interference term due to interference between transitions of different multiplicities (there is no such term in the total conversion coefficients). The expression for the shift  $\Delta E$  therefore contains a term that is linear in the multipole mixing parameter  $\delta_{\gamma}$ .<sup>3</sup> This may prove to be useful in studying mixed transitions.<sup>4)</sup>

For ordinary (not anomalous) conversion, expression (7) for the shift reduces (when using the explicit form of the matrix elements  $b_{\kappa}$ , see p. 25 of Ref. 3) to the form

$$\Delta E^{K_{\alpha_1}=\Delta_{\kappa}} = \frac{(I_0-I)(I_0+I+1)-L(L+1)}{2L(2I+1)} \frac{1-Lr/(L+1)}{1+r}, \quad (8)$$

where  $r = |M_{\kappa_2}(\tau L)|^2 / |M_{\kappa_1}(\tau L)|^2$ ,  $M_{\kappa}(\tau L)$  being the partial conversion matrix element [ $\alpha(\tau L) = \sum_{\kappa} |M_{\kappa}(\tau L)|^2$ , where  $\alpha(\tau L)$  is the total coefficient for conversion on a definite atomic shell or subshell<sup>3,8)</sup>]. The  $M_{\kappa}(\tau L)$  have been partially tabulated,<sup>8</sup> or they may be calculated with special computer programs (see, e.g., Ref. 9). Equation (8), together with Eq. (6), yields an equation for the magnetic moment of the nucleus in the final state. In the case of mixed transitions, the expressions for the shifts are more cumbersome, but they can always be obtained from Eqs. (3) and (4) when the multipole mixing parameter  $\delta_{\gamma}$  is known. We note that the shifts given in Ref. 1 for M1 transitions correspond to Eq. (8) with  $r=0$ . This approximation is accurate within 10% for all ML transitions with energies up to 0.5 MeV in nuclei with  $Z \geq 55$ .

## EXPERIMENT

To determine the unknown magnetic moment of the 12.3 keV  $3/2^+$  excited state  $^{133}\text{Ba}^*$  of the  $^{133}\text{Ba}$  nucleus, whose lifetime<sup>10</sup> is  $\tau = 6.8 \pm 0.4$  nsec, we measured the energy shift of the barium  $K_{\alpha_1}$  line accompanying internal conversion of the 276 keV M4 nuclear transition  $^{133}\text{Ba}^m(11/2^- \rightarrow 3/2^+)^{133}\text{Ba}^*$ . The  $^{133}\text{Ba}^m$  isomer was obtained by exposing a  $\text{BaCO}_3$  specimen enriched in  $^{132}\text{Ba}$  (the isotopic compositions of the specimens used in the study are given in Table I) to a  $\sim 10^{14}$   $\text{cm}^{-2}\text{sec}^{-1}$  neutron flux at the reactor. The barium  $K_{\alpha_1}$  fluorescence line excited by radioactive  $^{170}\text{Tm}$  in a  $\text{BaCO}_3$  specimen enriched in  $^{135}\text{Ba}$  was used as a reference standard. The measurements were made with a Cauchois crystal-diffraction spectrometer according to the scheme that we ordinarily employ (see, e.g., Ref. 2).

A new setup was constructed, which differed from

earlier setups in that the source exchanger was suitable for work with activities up to 100 Ci. To increase the luminosity of the spectrometer, the height of the entrance slit to the detector was doubled and two FEU-93 photomultipliers with NaI(Tl) crystals 45 mm in diameter were mounted behind the slit, one above the other and symmetrically disposed with respect to the plane of the focal circle. For the same purpose (i.e., to minimize the time required to measure the shifts of the barium  $K_{\alpha_1}$  line) we optimized the cut parameters of the single-crystal quartz plate, so that reflection was from the (203) planes, which were perpendicular to the large faces and parallel to the small faces of the plate. The bending coefficient  $k$  of the reflecting planes<sup>2</sup> turned out to be  $k = 2 \times 10^{-4} \text{ cm}^{-1}$ ; the plate was 1.2 mm thick, the entrance slit to the detector was 0.4 mm wide, and the focal circle was 2 m in diameter. As a result of all this, the spectrometer luminosity increased by a factor of three [as compared with the use of a single detector and a plate cut in the standard way with reflection from the (130) planes<sup>2</sup>].

The specimens to be compared consisted of wafers 20 mm in diameter and 2–3 mm thick and were introduced alternately into the field of view of the spectrometer. Statistics were accumulated for a total of  $\sim 120$  hr in separate runs of 1–2 hr each. Because of the short half-life of the  $^{133}\text{Ba}^m$  isomer ( $T_{1/2} = 38.9$  hr) the shape of the barium  $K_{\alpha_1}$  line changed during a single run, so a correction for the radioactive decay of  $^{133}\text{Ba}^m$  was included in the data processing procedure. The experimental shift turned out to be  $+57.8 \pm 7.4$  (6.8) meV (two errors are shown, the external and internal rms deviations, respectively).

The principal difficulty inherent in the present method is that of distinguishing the  $K_{\alpha_1}$  radiation corresponding to the investigated conversion transition against the background of such other barium  $K_{\alpha_1}$  radiation as may present. In the case of  $^{133}\text{Ba}^m$ , all the  $K_{\alpha_1}$  radiation emitted following its decay corresponds to the investigated 276M4 transition. The only source of extraneous barium  $K_{\alpha_1}$  radiation is the  $^{134}\text{Ba}$  present as an impurity in the  $^{132}\text{Ba}$  specimen. The isomer  $^{135}\text{Ba}^m$  resulting from the reaction  $^{134}\text{Ba}(n, \gamma)^{135}\text{Ba}^m$  decays to the ground state by the well converted 268M4 ( $11/2^- \rightarrow 3/2^+$ ) transition with the half life  $T_{1/2} = 28.7$  hr. The resulting extraneous barium  $K_{\alpha_1}$  line is also shifted with respect to the fluorescence line as a result of nonstatistical population. This shift can be calculated with formula (8), using the known<sup>11</sup> magnetic moment of the  $^{135}\text{Ba}$  ground state. The admixture of the barium  $K_{\alpha_1}$  line from the  $^{135}\text{Ba}^m$  isomer can also be calculated quantitatively provided the reaction cross section ratio

$$\sigma(^{134}\text{Ba}(n, \gamma)^{135}\text{Ba}^m) / \sigma(^{132}\text{Ba}(n, \gamma)^{133}\text{Ba}^m)$$

and the yields of  $K_{\alpha_1}$  radiation on conversion are known. Unfortunately, the data in the literature on these reaction cross sections are very contradictory,<sup>12,13</sup> so we determined the relative admixture of the barium  $K_{\alpha_1}$  line from the  $^{135}\text{Ba}^m$  isomer experimentally. For this purpose we irradiated  $\text{BaCO}_3$  specimens, enriched in  $^{132}\text{Ba}$  and  $^{134}\text{Ba}$  but otherwise identical, in the reactor under identical conditions and then measured the inten-

TABLE I.

Principal isotope	Composition, %						
	130	132	134	135	136	137	138
132	0.1	28	7.58	8.85	7.28	8.12	40.07
134	<0.06	0.06	85.5	5.98	1.42	1.36	5.47
135	<0.05	<0.05	0.26	92.7	3.62	0.80	2.62



sity ratio of the barium  $K_{\alpha_1}$  lines of the two specimens at the crystal-diffraction spectrometer. Knowing the isotopic compositions of the compared specimens and the half lives of  $^{133}\text{Ba}^m$  and  $^{135}\text{Ba}^m$ , we can calculate the relative contribution to the barium  $K_{\alpha_1}$  line from the  $^{135}\text{Ba}$  isomer in the working specimen; it varied from 4.5 to 2.0% during the entire time in which statistics were being collected. The experimental shift was corrected for the shift of this impurity line; the corrected shift turned out to be  $+57.2 \pm 7.6$  (6.8) meV.

The experimental ratio of the intensities of the barium  $K_{\alpha_1}$  line from the  $\text{BaCO}_3$  specimens enriched in  $^{134}\text{Ba}$  and  $^{132}\text{Ba}$  and irradiated in the reactor was also used to determine the reaction cross section ratio  $\sigma[^{134}\text{Ba}(n, \gamma)^{135}\text{Ba}^m] / \sigma[^{132}\text{Ba}(n, \gamma)^{133}\text{Ba}^m]$ , using the known yields of  $K_{\alpha_1}$  radiation for the 276M4 and 268M4 conversion transitions.<sup>14</sup> This ratio was found to be  $0.18 \pm 0.01$ , which agrees with the value  $0.23 \pm 0.04$  given in Ref. 13, but is in conflict with the value given in Ref. 12.

The measured shift depends on the isotopic and chemical compositions of the specimens (the "isotopic" and "chemical" shifts), as well as on the difference of the volume isotopic shifts between the excited and ground states of the investigated isotope (the "isomeric" shift), and corrections for these effects should be made.

Experimental data from Ref. 15 were used to correct for the isotopic shift; the corrected experimental shift turned out to be  $+55.9 \pm 7.6$  (6.8) meV. The chemical shift was assumed to be zero, since all the specimens had the same chemical form:  $\text{BaCO}_3$ . The isomeric shift can be estimated from the known<sup>16</sup> experimental values of  $\delta\langle r^2 \rangle$  (the increment of the rms charge radius of the nucleus) for the  $3/2^+$  excited states of the neighboring even-odd nuclei  $^{119}\text{Sn}$ ,  $^{125}\text{Te}$ , and  $^{123}\text{Xe}$ . The isomeric shifts of the  $K_{\alpha_1}$  lines of these nuclei corresponding to the experimental  $\delta\langle r^2 \rangle$  values do not exceed 0.3 meV, so no correction was made for the isomeric shift.

On substituting the experimental value of the shift into Eq. (8) we obtain the value  $+0.51 \pm 0.07$  (0.06)  $\mu N$  for the magnetic moment of the  $3/2^+$  state of  $^{133}\text{Ba}^*$ . This value agrees well with the calculated value  $\mu^{\text{calc}} = +0.54$  given by Kisslinger and Sorensen.<sup>17</sup>

In concluding, we note that the possibility of using coincidence techniques or of investigating the shifts of the conversion-electron lines may be of interest in connection with the difficulties discussed above in separating out the x-radiation corresponding to the conversion transition under investigation.

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- <sup>1</sup>We recall<sup>1</sup> that the conversion-electron lines themselves, as well as the corresponding Auger-electron lines, suffer analogous shifts.
- <sup>2</sup>It is not necessary to take the outer shells of the atom into account when  $K$  or  $L$  terms are excited.
- <sup>3</sup>In Ref. 5 the authors made an error in editing: in the expression for the constant  $G$  from formulas (2)–(4) of Ref. 5 the quantity  $\mu_N$  should be replaced by  $m_e/m_p$  (the electron:proton mass ratio). In addition, the small correction for shielding  $\eta$  from Eq. (6) vanishes when it is correctly defined.
- <sup>4</sup>In Ref. 7 it was suggested that the analogous interference between Fermi and Gamow-Teller transitions in the case of  $K$  capture with  $\Delta I = 0$  be used to determine the ratio of the corresponding matrix elements.
- <sup>5</sup>A. I. Egorov, A. A. Rodionov, A. S. Ryl'nikov, A. E. Sovestnov, O. I. Sumbaev, and V. Z. Shaburov, *Pis'ma Zh. Eksp. Teor. Fiz.* **27**, 514 (1978) [*JETP Lett.* **27**, 483 (1978)].
- <sup>6</sup>O. I. Sumbaev, *Usp. Fiz. Nauk* **124**, 281 (1978) [*Sov. Phys. Usp.* **21**, 141 (1978)].
- <sup>7</sup>I. M. Band, M. A. Listengarten, and A. P. Feresin, *Anomalii v koeffitsientakh vnutrennei konversii gamma-lucheĭ* (Anomalies in gamma-ray internal conversion coefficients), Nauka, L., 1976.
- <sup>8</sup>L. A. Sliv (editor), *Deformatsiya atomnykh yader* (Nuclear deformation), M., 1958.
- <sup>9</sup>A. S. Ryl'nikov, A. I. Egorov, V. I. Marushenko, A. F. Mezentsev, A. I. Smirnov, O. I. Sumbaev, and V. V. Fedorov, *Zh. Eksp. Teor. Fiz.* **63**, 53 (1972) [*Sov. Phys. JETP* **36**, 27 (1973)].
- <sup>10</sup>G. Breit, *Phys. Rev.* **35**, 1447 (1930).
- <sup>11</sup>V. V. Fedorov and A. S. Ryl'nikov, *Zh. Eksp. Teor. Fiz.* **76**, 1986 (1979) [*Sov. Phys. JETP* **49**, 1007 (1979)].
- <sup>12</sup>Kai Siegbahn (editor), *Alpha-, beta-, and gamma-ray spectroscopy*, North-Holland, Amsterdam, 1965.
- <sup>13</sup>I. M. Band and M. B. Trzhaskovskaya, Preprint LIYaF-300, 1977.
- <sup>14</sup>W. Andrejtscheff, Zh. T. Zhelev, M. Enikova, K. M. Muminov, T. M. Muminov, and U. S. Salikhbaev, *Yad. Fiz.* **29**, 849 (1979) [*Sov. J. Nucl. Phys.* **29**, 849 (1979) [*Sov. J. Nucl. Phys.* **29**, 437 (1979)]].
- <sup>15</sup>M. P. Avotina and A. V. Zolotavin, *Momenty osnovnykh i vzbuzhdennykh sostoyaniĭ yader* (Spins of nuclear ground and excited states), Atomizdat, 1979, Part 1.
- <sup>16</sup>I. V. Mednis, *Spravochnye tablitsy dlya neitrono-aktivatsionnogo analiza* (Handbook tables for neutron activation analysis), "Zinatne", Riga, 1974.
- <sup>17</sup>Neutron cross sections, Vol. 1, B. N. L. Upton, New York, 1973.
- <sup>18</sup>N. G. Gusev and P. P. Dmitriev, *Kvantovoe izluchenie radioaktivnykh nuklidov, Spravochnik* (Quantum radiations of radioactive nuclides, a Handbook). Atomizdat, 1977.
- <sup>19</sup>K. Bekk, A. Andl, S. Göring, A. Hanser, G. Nowicki, H. Rebel, and G. Shatz, *Z. Phys.* **A291**, 219 (1979).
- <sup>20</sup>G. M. Kalvius and G. K. Shenoy, *At. Data and Nucl. Data Tables* **14**, 639 (1974).
- <sup>21</sup>Leonard S. Kisslinger and Raymond A. Sorensen, *Rev. Mod. Phys.* **35**, 853 (1963).

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