Anomalous manifestation of static Stark effect in the ground state of an atomic gas

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The combined effects of a static electric field and the field of a linearly polarized, spatially nonuniform light beam on the Stark splitting and polarizability of the ground state of atomic gases are studied. The anisotropy which arises in the process is quite different from that in the Kerr effect in comparatively weak static fields, 10^2–10^3 V/cm. For example, the static electrification is accompanied by a magnetization of the gas. The optical anisotropy of the gas is analyzed for the case of an anomalous rotation of the polarization plane of a bounded light beam.

1. INTRODUCTION

Krasheninnikov *et al.*[1] have observed an anomalous optical anisotropy of a gas of atoms in a ground state which is degenerate with respect to projections of the angular momentum $\mathbf{j}_0$ in ultraweak magnetic fields. The physical reason for the anomaly in the anisotropy was that the average quadrupole moment induced in the gas by the linearly polarized optical field was reoriented as a result of precession of the atomic polarization vector in the weak magnetic field. The quadratic Stark effect in a static electric field gives rise to a precession quite different from that of Ref. 1. It mixes multipole moments of different parities of the atom (e.g., the quadrupole and magnetic moments). For this reason we would expect that the Stark effect of atoms in the ground state would also lead to some unusual features in the static and optical polarizabilities of gas.

The optical manifestations of this effect can be put in two categories. The first includes effects which stem from splitting of emission (or absorption) lines and which lead to birefringence, such as the Kerr effect. In the saturated vapor of atomic gases, however, the line splitting is a small effect because of the strong Doppler broadening, and can be seen only in fields of 10^5–10^6 V/cm (Ref. 2). The second category of optical manifestations of the quadratic Stark effect is directly related to splitting of atomic levels and to the formation of additional anisotropy during the joint polarization of the atoms by the static and optical external fields. This is obviously a nonlinear effect in terms of both the static and optical fields. In the absence of the optical field, the static field $\mathbf{E}_0$ does not alter the distribution of freely oriented atoms with respect to sublevels. In the absence of the static field, on the other hand, linearly polarized light $\mathbf{E}$ induces a quadrupole moment (an alignment) $\rho_{2,0}$, which is oriented along the direction of the linear polarization vector $\mathbf{e}$: $\rho_{2,0} \propto \{\mathbf{e} \otimes \mathbf{e}\}_{2,0}$. When the fields $\mathbf{E}$ and $\mathbf{E}_0$ act together, the atom acquires a magnetic moment

$$\rho_{1q} \propto (\mathbf{e}\mathbf{e}_0)\{\mathbf{e}\otimes\mathbf{e}_0\}_{1q}$$

[see expression (14) below], a quadrupole moment

$$\rho_{2q} \propto (\mathbf{e}\mathbf{e}_0)\{\mathbf{e}\otimes\mathbf{e}_0\}_{2q}$$

and other multipole moments allowed by the selection rules.

Manakov and Faĭnshteĭn[4] were the first to take up the phenomenological discussion of the appearance of a magnetic moment in absorbing media in static and optical fields. In general, the principal axes of the multipole moments will not coincide with the directions determined on the basis of the fields $\mathbf{E}$ and $\mathbf{E}_0$ separately. As a result, the anisotropy of the gas as a whole can no longer be reduced to a Kerr effect. The proportionality coefficients of the multipole moments $\rho_{\varkappa q}$ will be determined by the level splitting. The magnitude of this splitting should be compared not with the Doppler width (as in the case of the line splitting) but with the homogeneous width of the levels which are in resonance with the optical field. If one of the levels happens to be the ground state, it will be the Stark splitting of this level which plays the leading role in the formation of the anisotropy. The magnitude of the Stark splitting, Ω [see expression (4)], should (as in Ref. 1) now be compared with either the decay rate $\gamma^0_\varkappa$ of the multipole moments of order $\varkappa$ in the ground state or the reciprocal of the time scale of the interaction of the atoms with the light beam, $\bar{t}^{-1}$. For a bounded light beam, $\bar{t} = r_0/\bar{v}$ is the time taken by an atom to pass through the beam. The relations $\gamma^0_\varkappa \bar{t} \ll 1$ and $\gamma\bar{t} \gg 1$ usually hold, where γ is the rate of radiative relaxation of the excited state. Consequently, the splitting of the ground state becomes important under the condition $\Omega\bar{t} \gg 1$, which can be satisfied in comparatively weak fields, $E_0 \sim 10^2$–10^3 V/cm. It is this manifestation of the Stark effect which is the subject of the present paper.

2. FORMULATION OF THE PROBLEM; EQUATION FOR THE DENSITY MATRIX OF THE GROUND STATE, WITH ALLOWANCE FOR THE STATIC FIELD AND OPTICAL PUMPING PROCESSES

Let us consider the interaction of a linearly polarized and bounded light beam of width r_0 with a single-component gas of resonant atoms in their ground state, which has a total angular momentum j_0, in a static electric field. The optical density of the gas is small: $\varkappa_0 l < 1$, where $\varkappa_0$ is the linear absorption coefficient, and l is the length of the cell which holds the gas. In this case we can ignore collisions, assuming that the width γ of the second resonant level (with an angular momentum j_1) and the width of the transition line are purely radiative. With regard to the static electric field imposed on the cell we assume $\Omega \ll \gamma$; in other words, we take into account the Stark splitting of only the sublevels of the ground state, ignoring the splitting of the line and of the upper state.

We describe the atom by means of a density matrix in

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the Wigner representation. We ignore the ordinary saturation effects, assuming that the saturation parameter

$$G=\frac{|Ed/\hbar|^2}{(\gamma/2)^2+(\Delta\omega)^2} \tag{1}$$

satisfies $G \ll 1$. Here d is the reduced transition dipole moment, and $\Delta\omega = \omega - \omega_0 - \mathbf{kv}$ is the deviation of the frequency ω of the light from that of the atomic transition, ω_0, where we are taking the Doppler shift $\mathbf{kv}$ into account. As was shown in Ref. 1, if the lower working level is the ground state, optical pumping processes play an important role, and the condition $G \ll 1$ is not sufficient for using perturbation theory in G. Following Ref. 1, we will accordingly assume that the primary mechanism for the relaxation of the ground-state atoms is their escape from the light beam. We also assume that the number of spontaneous transitions during the time taken by an atom to pass through the beam is large: $\gamma\bar{t} \gg 1$, but $\gamma G\bar{t} < 1$. In order to legitimately ignore disorienting collisions in the ground state, we choose the optical pumping rates γG to be much larger than the typical relaxation rates of the multipole moments, $\gamma^0_\varkappa$. Summarizing all these restrictions on G,

$$\frac{\gamma_\varkappa^0}{\gamma} \ll G < \frac{1}{\gamma\bar{t}} \ll 1, \tag{2}$$

we define a region of pump rates or restrictions on the intensity of the light beam, I. Typical values for alkali metals are $\gamma^0_\varkappa \sim 10^2$–$10^{-2}\ \mathrm{s}^{-1}$, $\gamma \sim 10^7\ \mathrm{s}^{-1}$, and $\bar{t} \sim 10^{-4}$–10^{-5} s. Consequently, with tunable lasers it is easy to satisfy condition (2) over a wide range of pump light intensities, $10^{-11}\ \mathrm{W/cm}^2 < I < 10^{-4}\ \mathrm{W/cm}^2$.

In a coordinate system with an arbitrarily directed quantization axis, the equation of motion of the density matrix of the ground state, $\rho_{mm'}$ takes the following form in these approximations:

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho_{mm'}+i(\Omega_{mm_1}\rho_{m_1m'}-\rho_{mm_1}\Omega_{m_1m'})=\gamma F_{mm'}, \tag{3}$$

where a repeated index implies summation. We can find the magnitude of the Stark splitting by directing the quantization axis along $\mathbf{e}_0 = \mathbf{E}_0/E_0$; we then find

$$\Omega_{mm}-\Omega_{m'm'}=\Omega(m^2-m'^2),$$

$$\Omega=E_0^2\sum_l \frac{\alpha_{l0}}{\hbar}(-1)^{j_0+j_l+1}\frac{3[15(2j_0+1)]^{1/2}}{[2j_0(j_0+1)(2j_0-1)(2j_0+3)]^{1/2}} \times\begin{Bmatrix} j_l & 1 & j_0 \\ 2 & j_0 & 1\end{Bmatrix}. \tag{4}$$

The summation in (4) is over all of the levels l which are dipole-coupled with the ground level. It is more convenient to express the polarizabilities α_{l0} in terms of the rates of the corresponding radiative transitions, γ_{l0}, and the wavelengths λ_{l0}:

$$\alpha_{l0}=\frac{1}{2}\left(\frac{\lambda_{l0}}{2\pi}\right)^4\frac{\gamma_{l0}}{c}. \tag{5}$$

We recall that the static polarizability of an atom is

$$\alpha=\sum_l \alpha_{l0}.$$

The "collision integral" $F_{mm'} \propto G\rho$ describes redistribution of atoms among sublevels of the ground state in the optical pumping cycle. The expression for $F_{mm'}$ for the general case of an elliptically polarized pump wave is given in the Appendix.

3. ANISOTROPY OF THE GROUND STATE IN SPATIALLY NONUNIFORM FIELDS

We will formulate a perturbation theory in the parameter G which is suitable for calculating the density matrix of the ground state in spatially nonuniform fields. We direct the quantization axis along the static field $\mathbf{e}_0$. Integrating over the path of an atom, we transform (3) into an integral equation:

$$\rho_{mm'}(\mathbf{r},t)=\delta_{mm'}/(2j_0+1)+\gamma\int_0^\infty dt_1 F_{mm'}(\mathbf{r}-\mathbf{v}t_1, t-t_1) \times\exp[-i\Omega t(m^2-(m')^2)]. \tag{6}$$

In the steady state, $\rho_{mm'}(\mathbf{r},t)$ is independent of t, and $F_{mm'}(\mathbf{r},\mathbf{v}t_1, t-t_1)$ is correspondingly independent of $t-t_1$. Quantities which do depend on $\mathbf{r}$ are the saturation parameter $G(\mathbf{r})$ and the density matrix $\rho(\mathbf{r})$, which appear in $F_{mm'}$.

In $G(\mathbf{r})$ we single out a dimensionless factor $g(\mathbf{r})$ which describes the spatially nonuniform transverse field distribution in the beam: $G(\mathbf{r}) = g(\mathbf{r})G$. For definiteness, we assume that this is a Gaussian distribution:

$$g(\mathbf{r})=\exp(-r^2/r_0^2).$$

The first term in (6) describes an isotropic distribution with respect to the sublevels of the ground state before the interaction with the field $\mathbf{E}$ is turned on (adiabatically). Substituting this distribution into the second term in (6), we find a first-order perturbation approximation to which we restrict the present analysis:

$$\rho^{(1)}_{mm'}=\gamma\bar{t}_{mm'}F^{(1)}_{mm'},\qquad t_{mm'}=\int_0^\infty dt\, g(r-vt)\exp[-i\Omega t(m^2-m'^2)]. \tag{7}$$

In this approximation, the expression for $F^{(1)}_{mm'}$ simplifies considerably. Assuming

$$\rho_{mm'}=\delta_{mm'}/(2j_0+1),$$

in (A1), we find, for a linearly polarized field,

$$F_{mm'}=GA\sum_q(-1)^{j_0-m}\begin{pmatrix} j_0 & 2 & j_0\\ -m & q & m'\end{pmatrix}Y_{2q}^*(\theta,\varphi),$$

$$A=\left(\frac{8\pi}{3}\right)^{1/2}\frac{1}{(2j_0+1)}\left[(-1)^{j_0+j_1}\begin{Bmatrix} j_1 & j_0 & 1\\ 2 & 1 & j_0\end{Bmatrix}+(2j_1+1) \times\begin{Bmatrix} 1 & j_0 & j_1\\ 2 & j_1 & j_0\end{Bmatrix}\begin{Bmatrix} j_0 & 1 & j_1\\ 2 & j_1 & j_0\end{Bmatrix}\right], \tag{8}$$

where $Y_{2q}(\theta,\varphi)$ are the spherical harmonics, and θ and φ are the spherical angles of the linear polarization vector $\mathbf{e}$. As we mentioned in the Introduction, it follows from this expression that a linearly polarized field induces only a quadrupole moment in the ground state. The expansion of

$\rho^{(1)}_{mm'}$ in irreducible tensor operators (the $\varkappa q$ representation), however, contains multipole moments of other orders ($\varkappa\neq 2$) and of the other parity ($\varkappa$ is of odd parity), which result from the interaction with the static field:

$$\rho^{(1)}_{mm'}=\sum_{\varkappa}(2\varkappa+1)^{1/2}(-1)^{j_0-m}\begin{pmatrix} j_0 & \varkappa & j_0\\ -m & q & m'\end{pmatrix}\rho^{(1)}_{\varkappa q},$$

$$\rho^{(1)}_{\varkappa q}=\gamma GA(2\varkappa+1)^{1/2}\sum_{m,m'}\begin{pmatrix} j_0 & \varkappa & j_0\\ -m & q & m'\end{pmatrix}\begin{pmatrix} j_0 & 2 & j_0\\ -m & q & m'\end{pmatrix}$$

$$\times\ Y_{2q}(\theta,\varphi)\bar{t}_{mm'}. \qquad (9)$$

The entire dependence on the static field Ω, the transverse coordinates $\mathbf{r}$, and the transverse velocities $\mathbf{v}_\perp$ $[(\mathbf{kv}_\perp)=0]$ is contained in the quantities $\bar{t}_{mm'}(\Omega,\mathbf{r},\mathbf{v}_\perp)$ which determine the time scales of the interaction of the multipole moments of the atoms with the light beam. In the case $|m'|=|m|$, the quantity $\bar{t}_{mm'}$, averaged over the transverse velocities, $\langle t_{mm'}\rangle_{\mathbf{v}_\perp}=\bar{t}(\mathbf{r})$ does not depend on the static field. Its meaning is the average duration of the interaction of the atom with the field (the transit time):

$$\bar{t}(r)=\bar{t}\frac{\pi}{2}I_0\left(\frac{r^2}{2r_0^2}\right)\exp\left(-\frac{r^2}{r_0^2}\right). \qquad (10)$$

Here $I_0(x)$ is a modified Bessel function. In our case the transit relaxation has appeared in the solution in a natural way; it is usually introduced at the outset in the equations for the density matrix (3), by ignoring the spatial variation of the beam.[5,6] In the case $|m|\neq|m'|$, the interaction times $\bar{t}_{mm'}$ in the static field begin to depend on the magnetic quantum numbers. This circumstance ultimately determines the additional anisotropy which is introduced by the Stark splitting of the ground state. Since the $\mathbf{r}$ dependence of the times, $\bar{t}_{mm'}(\mathbf{r})=\langle t_{mm'}(\mathbf{r},\mathbf{v}_\perp)\rangle_{\mathbf{v}_\perp}$ is of no interest for the problem discussed below, we will analyze the asymptotic behavior $\bar{t}_{mm'}(0)$ for atoms at the center of the light beam.

In "small" static fields, with $\varepsilon=\Omega\bar{t}\ll 1$, we have

$$\frac{\bar{t}_{mm'}(0)}{\bar{t}}=\frac{\pi}{2}(1-|\varepsilon_{mm'}|)-i\varepsilon_{mm'}(1-C-\ln|\varepsilon_{mm'}|). \qquad (11)$$

Here C is Euler's constant, and $\varepsilon_{mm'}=\varepsilon(m^2-(m')^2)$. In "large" fields, with $\varepsilon\gg 1$, we have

$$\frac{\bar{t}_{mm'}(0)}{\bar{t}}=\frac{\pi}{2}\delta_{|m|,|m'|}-\frac{i}{\varepsilon_{mm'}}. \qquad (12)$$

Consequently, in the case $\varepsilon\gg 1$ the interaction time and the density matrix $\rho^{(1)}_{mm'}$ do not depend on the absolute value of the static field $\mathbf{E}_0$. We wish to stress that expression (7) for $\bar{t}_{mm'}$ has been written in a special coordinate system, aligned with the direction of $\mathbf{e}_0$, so the dependence on the relative orientation of the fields $\mathbf{e}$ and $\mathbf{e}_0$ in the density matrix $\rho^{(1)}_{mm'}$ persists even at $\varepsilon\gg 1$. Here, as in Ref. 1, we are seeing a manifestation of the anomalous nature of the saturation effects in the ground state in terms of the static field.

Let us estimate the characteristic values of the static field for which we have $\varepsilon\sim 1$. Replacing the sum over l by the static polarizability in (4), we find, in order of magnitude, $\varepsilon\sim\alpha\bar{t}E_0^2/\hbar$. Setting $\alpha\sim 10^{-22}$–10^{-24} cm^3, $\bar{v}\sim 10^4$ cm/s, and $r_0\sim 1$ cm, we find $\varepsilon\sim 1$ at $E_0\sim 10^2$–10^3 V/cm. Such fields are weak in comparison with the fields (10^5–10^6 V/cm) which are usually required for observing the Stark splitting of a line, which we discussed in the Introduction, in a saturated vapor of gases. Consequently, in comparatively weak static fields there are substantial changes in the multipole moments of the atom in its ground state, $\rho_{\varkappa q}$ [see (A1)]. We can demonstrate the point with the example of the onset of a static magnetization $\mathbf{M}$ of a gas inside a linearly polarized light beam in a static electric field. By definition, we have

$$M_{1q}=n\mu_0 g_0[j_0(j_0+1)(2j_0+1)/3]^{1/2}\langle\rho_{1q}\rangle_v, \qquad (13)$$

where μ_0 is the Bohr magnetron, and g_0 is the Landé factor of the ground state. For simplicity we consider the specific transition $j_0=1\to j_1=0$. In this case, all of the equations simplify considerably, and as a result we find

$$\mathbf{M}=\left[\frac{2j_0(2j_0+1)(j_0+1)}{3}\right]^{1/2}\langle G(v)\rangle_v(\mathbf{ee}_0)[\mathbf{ee}_0]f(\varepsilon),$$

$$f(\varepsilon)=\frac{1}{2}[\mathrm{Ei}(\varepsilon)e^{-\varepsilon}-\mathrm{Ei}(-\varepsilon)e^{\varepsilon}]. \qquad (14)$$

Here $Ei(x)$ is the integral exponential function. The asymptotic expressions for $f(\varepsilon)$ reproduce those for the imaginary part of (11), (12); i.e., we find $f(\varepsilon)\sim-\varepsilon\ln\varepsilon$ at $\varepsilon\ll 1$ and $f(\varepsilon)\sim 1/\varepsilon$ at $\varepsilon\sim 1$. The maximum of $f(\varepsilon)$ is reached at $\varepsilon\sim 1$. A magnetization of the medium of the form (13),

$$M\sim(\mathbf{ee}_0)[\mathbf{ee}_0],$$

in a linearly polarized optical field and in a static field could be found simply from general symmetry considerations,[4] aside from the explicit form of $f(\varepsilon)$. Such a magnetization obviously leads to a rotation of the polarization plane, as in the inverse Faraday effect.[7] The very nonlinear dependence on ε is characteristic not only of the magnetic moment but also of all of the multipole moments of the atom, $\rho_{\varkappa q}(\varepsilon)$. The linear dependence on ε begins in weaker fields, $\Omega\sim\bar{v}/\bar{l}\ll 1/\bar{t}$ where $\bar{l}=1/\varkappa_0$ is the range of the front in the medium ($l<\bar{l}$). In contrast with Ref. 1, where the magnetic field led to the formation of only even multipole moments, the static electric field in this case induces multipole moments of both even and odd orders $\varkappa$ in the presence of linearly polarized or unpolarized light. This is yet another anomalous manifestation of the Stark effect in the ground state of a gas.

4. ANOMALOUS ROTATION OF THE POLARIZATION PLANE IN A STATIC ELECTRIC FIELD

In addition to the static effects which were mentioned above, and which are manifested in a magnetization and an electrification of the gas, the Stark effect in the ground state is also manifested in anomalies of the optical anisotropy. We will discuss this anisotropy here in the example of the rotation of the polarization plane of a bounded light beam.

We fix the relative orientation of the vectors $\mathbf{E}=\mathbf{e}E$ and $\mathbf{E}_0=\mathbf{e}_0E_0$ in the coordinate system attached to the light beam: Axis 3 (z) runs along the e direction, axis 2 (y) runs along the $\mathbf{k}$ direction, the spherical angles θ and φ determine the vector $\mathbf{e}_0$, and axis 1 (x) is orthogonal to the vectors $\mathbf{e}$ and $\mathbf{k}$.

The problem of the rotation of the polarization plane of light after it passes through a cell reduces to a calculation of the projection of the polarization vector of the medium, $\mathbf{P}$, onto the x axis at the cell boundary $y=l$. We will not write out the expression for $\mathbf{P}$; we proceed immediately to the general expression for the dielectric susceptibility tensor $\varkappa_{ij}$ as a function of the multipole moments of the density matrix of the ground state:

$$\chi_{ij}=3in\frac{|d|^2}{\hbar}(-1)^{j_0+j_1}\sum_{\varkappa}\begin{Bmatrix}\varkappa & j_0 & j_0\\ j_1 & 1 & 1\end{Bmatrix}\left\langle\frac{(\rho_\varkappa^*\cdot\{\mathbf{e}_1^i\otimes\mathbf{e}_1^j\}_\varkappa)}{\gamma/2-i\Delta\omega}\right\rangle_v. \tag{15}$$

The notation used for the irreducible tensor product, $\{\ldots\otimes\ldots\}$; for the scalar product, $(\ldots\ldots)$; for the irreducible tensors $\rho_{\varkappa q}$ and e^i_{1q} and for the $3jN$ symbols corresponds to the notation of Ref. 3. In addition, $\langle\ldots\rangle_v$ is an average over the velocities of atoms having a Maxwellian distribution, and n is the density.

By virtue of the anisotropy created in the gas by the fields $\mathbf{E}$ and $\mathbf{E}_0$, the light leaving the cell is elliptically polarized; the ellipse is rotated through an angle ψ with respect to axis 3, and the ellipticity angle is α ($\tan\alpha$ is equal to the ratio of semiaxes, and the sign of α specifies the rotation direction). In this approximation the angles are small and are determined in terms of the component $\chi_{13}=\chi_{13'}+i\chi_{13''}$:

$$\psi=-2\pi kl\chi_{13}'',\quad \alpha=2\pi kl\chi_{13}'. \tag{16}$$

We analyze the optical characteristics of the gas in the limit $\varepsilon\gg1$, in which we can ignore the magnetic moment induced in the medium. In other words, we can ignore gyrotropy effects in χ_{ij} by virtue of the inverse Faraday effect, and we need retain only the first term in (12); in expression (15) for χ_{ij} we retain only the anisotropy which stems from the quadrupole moment of the atom, ρ_{2q}. For simplicity we assume that the light is exactly resonant with the transition frequency, $\omega=\omega_0$. We then have $\chi_{13'}=0$, and only $\chi_{13''}$ is nonzero. We are thereby singling out the rotation of the polarization plane which is caused exclusively by the dichroism, (15). We see from (15) tha the tensor $\chi_{13}=\chi_{xz}$ contains cyclic components $q=\pm1$ of the quadrupole $\{\mathbf{e}^1\otimes\mathbf{e}^3\}_{2q}=-q/2$. We thus have $(\{\mathbf{e}^1\otimes\mathbf{e}^3\}_{2q}\cdot\rho^*_{2q})=-\mathrm{Re}\,\rho_{21}=-\rho_{21}$. In the absence of a static field we would have $\rho_{21}=0$, and there would be no rotation, since the quadrupole moment created by a linearly polarized light beam would be oriented exactly along the vector $\mathbf{E}$ (the z axis): $\hat{Q}^{(0)}_{zz}=\hat{Q}^{(0)}_{20}$. A static field $\mathbf{E}_0\neq0$ rotates $\hat{Q}^{(0)}$, and the latter acquires a nonvanishing projection $\hat{Q}^{(0)}_{xz}\propto\rho_{21}$. Using (7), (9), and (12), we find an explicit expression for ρ_{21}:

$$\rho_{21}=\frac{\pi}{2\cdot5^{1/2}}\gamma G\bar{t}A\cos\varphi\sum_L P_L^1(\cos\theta)\frac{C_L(j_0)}{[L(L+1)]^{1/2}}$$

$$C_L(j_0)=(2L+1)\begin{pmatrix}2 & L & 2\\ 0 & 1 & -1\end{pmatrix}\left[\begin{pmatrix}L & 2 & 2\\ 0 & 1 & -1\end{pmatrix}+5(1+(-1)^L)\right.$$
$$\left.\times\begin{pmatrix}L & 2 & 2\\ 0 & 2 & -2\end{pmatrix}\begin{pmatrix}j_0+1 & 2 & j_0-1\\ 0 & 0 & 0\end{pmatrix}\right]. \tag{17}$$

Here $P^1_L(\cos\theta)$ are the associated Legendre polynomials. By virtue of the selection rules, only terms with $L=2$ and 4 appear in (17). These terms mean that only tensors of even rank constructed from the vectors $\mathbf{e}_0$ contribute to the quadrupole moment of the atom. This result does not depend on the approximation which we have made, $\varepsilon\gg1$. Remarkably, for $\varepsilon\gg1$ the value of ρ_{21} does not depend on the strength of the field $\mathbf{E}_0$, because of the pronounced saturation, while it depends on the direction of this field in a rather complicated way. The coefficients $C_L(j_0)$ depend on j_0 only for integer values of j_0; the dependence describes the contribution of the coherence between the sublevels of the ground state, $\rho_{m,-m}$. For half-integer values of j_0 the atomic quadrupole moment which is produced by the optical field is oriented exactly along the direction of the static field in the limit $\varepsilon\gg1$. The mechanism for this reorientation of ρ_{2q} involves the pronounced anisotropy with respect to the magnetic quantum numbers of the time scales of the interaction between the atoms and the field of the light beam. The reasons for the rotation of the polarization plane are now obvious: At $\varepsilon=0$, the field polarizing the medium is a natural wave, while in the case $\varepsilon\neq0$ the natural waves are linearly polarized, and the polarization directions do not coincide with $\mathbf{E}$. The polarization plane rotates because of the difference between the absorption coefficients for the natural waves. Here is the expression for the rotation angle corresponding to the transition $j_0=1\to j_1=0$:

$$\psi=\frac{\pi}{4}\varkappa_0 l\left|\frac{Ed}{\hbar\gamma}\right|^2\gamma\bar{t}\sin2\theta\cos\varphi. \tag{18}$$

We see that within the range of applicability of our perturbation theory the polarization plane can rotate through an angle on the order of a few degrees for $\mathbf{E}_0$ in the xz plane and for angles $2\theta=\pi(n+1/2)$.

5. CONCLUSION

The interaction of atoms with a spatially nonuniform optical field and a static electric field can cause strong effects in the ground state of the atoms which are nonlinear in the static field. These effects stem from Stark splitting of the ground state and anisotropy of the lifetimes of the multipole moments of atoms in bounded light beams. The optical anisotropy which arises in the process is quite different from the Kerr effect. It leads to an anomalously large rotation of the polarization plane of the light in the gas at static fields $E_0\sim10^2$–10^3 V/cm. The results derived here could be generalized without difficulty to the case of a ground state which is split into several hyperfine components, through the use of the results of Ref. 7. Remarkably, similar effects occur if the anisotropic distribution of particles with respect to the projection of the angular momentum is caused by factors other than optical pumping, e.g., polarization effects in an inhomogeneous molecular gas (see Ref. 8 and the bibliography there).

APPENDIX: EQUATION FOR OPTICAL PUMPING IN THE ABSENCE OF SATURATION EFFECTS ($G\ll1$)

In the general case of elliptically polarized light, the equation describing the redistribution of atoms with respect to the sublevels of the ground state in the optical pumping cycle is, for an arbitrarily oriented coordinate system,

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho_{mm'}=\gamma F_{mm'},$$

$$F_{mm'}=\sum_{(\varkappa)q}GC_{\varkappa\varkappa_1\varkappa_2}\begin{pmatrix}j_0 & \varkappa & j_0\\ -m & q & m'\end{pmatrix}(-1)^{j_0-m}\{\{\mathbf{e}^{*1}\otimes\mathbf{e}^1\}_{\varkappa_1}\otimes\rho_{\varkappa_2}\}_{\varkappa q}, \tag{A1}$$

where

$$\rho_{\varkappa_2 q_2} = (2\varkappa_2+1)^{1/2} \sum_{m,m'} (-1)^{j_0-m} \begin{pmatrix} j_0 & \varkappa_2 & j_0 \\ -m & q_2 & m' \end{pmatrix} \rho_{mm'}$$

is the density matrix of the ground state in the irreducible representation, and $\mathbf{e}^1$ are complex unit vectors of the elliptical polarization. Singling out in the coefficients $C_{\varkappa\varkappa_1\varkappa_2}$ the contributions from the spontaneous decay of the excited state, $A_{\varkappa\varkappa_1\varkappa_2}$ and from transitions out of the ground state upon the absorption of the pump light, $B_{\varkappa\varkappa_1\varkappa_2}$ we can write

$$C_{\varkappa\varkappa_1\varkappa_2} = 3[(2\varkappa_1+1)(2\varkappa_2+1)(2\varkappa+1)]^{1/2}(-1)^{\varkappa_1+\varkappa_2}(A_{\varkappa\varkappa_1\varkappa_2} - B_{\varkappa\varkappa_1\varkappa_2}), \tag{A2}$$

$$A_{\varkappa\varkappa_1\varkappa_2} = (-1)^{j_1+j_0}(2j_1+1) \begin{Bmatrix} 1 & j_1 & j_0 \\ 1 & j_1 & j_0 \\ \varkappa_1 & \varkappa_2 & \varkappa \end{Bmatrix} \begin{Bmatrix} 1 & j_0 & j_1 \\ \varkappa & j_1 & j_0 \end{Bmatrix}, \tag{A3}$$

$$B_{\varkappa\varkappa_1\varkappa_2} = (-1)^{j_1-j_0} \begin{Bmatrix} \varkappa & \varkappa_1 & \varkappa_2 \\ j_0 & j_0 & j_0 \end{Bmatrix} \begin{Bmatrix} 1 & \varkappa_1 & 1 \\ j_0 & j_1 & j_0 \end{Bmatrix} (\Delta + (-1)^{\varkappa+\varkappa_1+\varkappa_2}\Delta^*). \tag{A4}$$

In (A4) we have used the notation $\Delta = 1/2 - i\Delta\omega/\gamma$. It is not difficult to see that we have $\Sigma_m F_{mm} = 0$, i.e., that the optical pumping redistributes atoms with respect to sublevels, while the total population of the ground state is conserved.

[1]M. V. Krasheninnikov, V. S. Smirnov, M. B. Sultanov, *et al.*, Zh. Eksp. Teor. Fiz. **94**(11), 24 (1988) [Sov. Phys. JETP **67**, 2190 (1988)].

[2]A. M. Bonch-Bruevich and A. M. Khodovoĭ, Usp. Fiz. Nauk **93**, 71 (1967) [Sov. Phys. Usp. **10**, 637 (1967)].

[3]D. A. Varshalovich, A. N. Moskalev, and V. K. Khersonskiĭ, *Kvantovaya teoriya uglovogo momenta* (*Quantum Theory of Angular Momentum*, World Scientific, Singapore, 1987), Nauka, Moscow, 1975, Chs. 3, 4.

[4]N. L. Manakov and A. G. Faĭnshteĭn, Zh. Eksp. Teor. Fiz. **87**, 1552 (1984) [Sov. Phys. JETP **60**, 890 (1984)].

[5]S. G. Rautian and A. M. Shalagin, Zh. Eksp. Teor. Fiz. **58**, 962 (1970) [Sov. Phys. JETP **31**, 518 (1970)].

[6]H. Rinnebeng, T. Huhle, E. Matthias, and A. Timmerman, Z. Phys. A **295**, 17 (1979).

[7]P. S. Pershan, Phys. Rev. **130**, 919 (1963).

[8]V. D. Borman, S. Yu. Krylov, A. P. Popov, and V. I. Troyan, Zh. Eksp. Teor. Fiz. **82**, 696 (1982) [Sov. Phys. JETP **55**, 415 (1982)].

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