

# Effect of the polarization of atoms on the propagation of quasi-resonance emission in a gaseous medium

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We consider the propagation of incoherent, arbitrarily polarized light in an optically dense polarized gaseous medium. We use Keldysh's diagram technique to derive the equations describing the transfer of radiation. We obtain and study, for the case when the polarization of the medium changes little in direction over distances of the order of the photon mean free path, expressions for the density matrix of the transmitted and the fluorescent light. We show that taking the finite optical depth into account leads both to significant quantitative differences from the thin-layer case and to the appearance of qualitatively new effects. We discover, for instance, that in a dense medium with atoms which are polarized in the ground state the Hanle effect can occur even when the excited state is not polarized. The effects analyzed in this paper may be used to set up experiments for studying atoms by optical alignment methods.

## 1. INTRODUCTION

The method of optical detection consisting of the analysis of the changes in the properties of light passing through a gaseous medium has recently been applied widely in connection with problems of optical pumping, the physics of a weakly ionized plasma, astrophysics, and so on. The main source of information about the state of the system in this method are the magnitude of the reduction in the light intensity and the change of its polarization. For a quantitative interpretation of the experiments it is necessary to be able to describe the evolution of the density matrix of the light in a medium with an arbitrary polarization. Cohen-Tanoudji, Laloë, and coworkers<sup>1-4</sup> (see also Ref. 5) have shown in the optically thin layer approximation that with respect to the absorption and dispersion of quasi-resonant light a gas polarized in accord with the internal moment behaves like an optically anisotropic medium. They studied in this approximation the quantitative connection between the polarizations of the light and the gas. In actual experiments with optically aligned atoms the conditions of a small optical thickness are often not fulfilled. Nonetheless, there is in the literature no generalization of the results of Refs. 1-5 to the case of an optically thick medium.

D'yakonov and Perel' took the finite optical thickness into account when studying (in Ref. 6 and in later papers<sup>7,8</sup>) the capture of resonance radiation. However, they assumed that the gas in the ground state has an equilibrium distribution over the Zeeman sublevels and a Maxwellian velocity distribution. In Ref. 9 a subsequent generalization of the results of Ref. 6 was made when the effects of a strong field were taken into account. The effect of the polarization of either the light or of the atoms was here neglected. Deviation from equilibrium of the absorbing gas was studied in Ref. 10 where, however, the model case of a four-level system which is rarely realized in experiments was considered.

The aim of the present paper is a study of the propagation of incoherent, arbitrarily polarized radiation in an optically thick gaseous medium with atoms which have arbitrary polarization moments (PM).

## 2. CORRELATION FUNCTION OF THE ELECTROMAGNETIC FIELD

The polarization and spectral characteristics of the electromagnetic field are determined from the first-order correlation function:<sup>11</sup>

$$D_{\mu\nu}(x_1, x_2) = \langle E_{\mu}^{(+)}(x_1) E_{\nu}^{(-)}(x_2) \rangle. \quad (1)$$

Here  $E_{\mu}^{(+)}(x_1)$  and  $E_{\nu}^{(-)}(x_2)$  are the positive- and negative-frequency parts of the electrical field operator in the Heisenberg representation,  $x_1, x_2$  are the space-time coordinates, and  $\mu$  and  $\nu$  are polarization indexes. The angle brackets indicate averaging over the non-equilibrium density operator of the system. For a free electromagnetic field, such as it is when entering and leaving the cell, the correlation function (1) is directly connected with the spectral density of the components  $I_{\mu\nu}(\omega, \mathbf{r}t)$  of the polarization tensor of the radiation propagating in the  $z$ -direction at the point  $\mathbf{r}$  and time  $t$ :

$$\begin{aligned} I_{\mu\nu}(\omega, \mathbf{r}t) & \delta\left(\frac{\omega}{c} - k_z\right) \delta(k_x) \delta(k_y) \\ & = \frac{c}{2\pi} \int_{-\infty}^{\infty} d^3\mathbf{p} \int_{-\infty}^{\infty} d\tau \exp(-i\omega\tau + i\mathbf{k}\mathbf{p}) \\ & \times D_{\mu\nu}\left(t + \frac{\tau}{2}, \mathbf{r} + \frac{\mathbf{p}}{2}; t - \frac{\tau}{2}, \mathbf{r} - \frac{\mathbf{p}}{2}\right), \\ & \mathbf{p} = \mathbf{r}_1 - \mathbf{r}_2, \quad \tau = t_1 - t_2, \quad \mathbf{r} = (\mathbf{r}_1 + \mathbf{r}_2)/2, \quad t = (t_1 + t_2)/2. \end{aligned} \quad (2)$$

We have assumed in (2) that the geometric-optics approximation  $\lambda = 1/k \rightarrow 0$  is satisfied, so that the correlator (1) varies rapidly in  $\mathbf{p}$  and  $\tau$  and slowly in  $\mathbf{r}$  and  $t$ . The quantity  $I_{\mu\nu}(\omega, \mathbf{r}t)$  determines the polarization density matrix:

$$\Phi_{\mu\nu}(\omega, \mathbf{r}t) = \left[ \sum_{\mu} I_{\mu\mu}(\omega, \mathbf{r}t) \right]^{-1} I_{\mu\nu}(\omega, \mathbf{r}t). \quad (3)$$

It is convenient to use for calculations the notation and definitions of Keldysh's diagram technique,<sup>12</sup> and to introduce the Green function  $D_{\mu\nu}^{-+}(x_1, x_2)$ :

$$iD_{\mu\nu}^{-+}(x_1, x_2) = \langle A_{\nu}(x_2) A_{\mu}(x_1) \rangle, \quad (4)$$

where  $A_\mu(x_1)$  and  $A_\nu(x_2)$  are Heisenberg operators of the vector potential. In the case of a quasi-monochromatic field of frequency  $\omega_0$  the correlator (1) and the Green function (4) are connected through the relation

$$D_{\mu\nu}(x_1, x_2) = i \frac{\omega_0^2}{c^2} \iint_0^\infty \frac{d\omega_1}{2\pi} \frac{d\omega_2}{2\pi} \iint_{-\infty}^\infty dt_1' dt_2' \times \exp[i\omega_1(t_1 - t_1') - i\omega_2(t_2 - t_2')] D_{\nu\mu}^{-+}(t_2' \mathbf{r}_2, t_1' \mathbf{r}_1). \quad (5)$$

To distinguish the function (4) from other Green functions, we shall for simplicity call it in what follows the density matrix.

### 3. EQUATION FOR THE DENSITY MATRIX

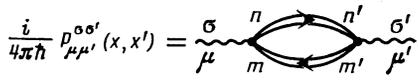
We can use Keldysh's diagram technique<sup>12,13</sup> to obtain an equation for the density matrix of radiation with a Hamiltonian corresponding to dipole interaction with the gas atoms. In abbreviated notation the corresponding Dyson equation has the form

$$\begin{aligned} \square_1 D^{-+} &= \Pi_\perp^{-+} D^{-+} + \Pi_\perp^{-+} D^{++}, \\ \square_2 D^{-+} &= -D^{-+} \Pi_\perp^{++} - D^{--} \Pi_\perp^{-+}. \end{aligned} \quad (6)$$

In the first (second) Eq. (6) the polarization operators  $\Pi_\perp^{\sigma\sigma'}$  act upon the first (second) arguments and indexes of the Green functions;  $\square_1, \square_2$  are D'Alembert operators in  $x_1$  and  $x_2$ , respectively. The kernels of the polarization operators  $\Pi_\perp^{\sigma\sigma'}$  have the form

$$P_\perp^{\sigma\sigma'} = \delta_\perp * P^{\sigma\sigma'} * \delta_\perp, \quad \sigma, \sigma' = \pm, \quad (7)$$

$$(\delta_\perp)_{\mu\nu}(x-x') = \delta_{\mu\nu} \delta(x-x') + \frac{1}{4\pi} \frac{\partial^2}{\partial r_\mu \partial r_\nu} \left( \frac{1}{|\mathbf{r}-\mathbf{r}'|} \right) \delta(t-t'), \quad (8)$$



$$\frac{i}{4\pi\hbar} P_{\mu\mu'}^{\sigma\sigma'}(x, x') = \left( \frac{\omega_0}{\hbar c} \right)^2 \sum_{mm'} \sum_{nn'} (-1)^\eta \varepsilon_{mn} \varepsilon_{n'm'} (d_\mu)_{nm} (d_{\mu'})_{m'n'} \times G_{n'n}^{\sigma'\sigma}(x', x) G_{mm'}^{\sigma\sigma'}(x, x'). \quad (9)$$

The \* sign in (7) and below in (11) and (13) indicates integration and summation over the internal space-time and polarization arguments and indexes. The presence of the  $\delta_\perp$  function is connected with the use of the Coulomb gauge of the vector potential ( $\text{div } \mathbf{A} = 0, \varphi = 0$ ). In Eq. (9)  $G^{\sigma\sigma'}$  is the Green function of the atoms;  $d_\mu, d_{\mu'}$  are the operators of the components of the dipole moments;  $\varepsilon_{mn} = +1$ , if  $m$  is an excited state, and  $n$  the ground state, and  $\varepsilon_{mn} = -1$  in the opposite case;  $\eta = 0$  for bosons and  $\eta = 1$  for fermions.

We introduce, apart from the density matrix (4), the retarded Green function of a photon in the medium:

$$D^R = D^{--} - D^{-+} = D^{+-} - D^{++},$$

which satisfies the equation

$$\square_1 D^R = -4\pi\hbar \delta_\perp + \Pi_\perp^R D^R, \quad \Pi_\perp^R = \Pi_\perp^{--} + \Pi_\perp^{-+}. \quad (10)$$

We perform in Eqs. (6) the substitution

$$D^{-+} = \bar{D}^{-+} - (4\pi\hbar)^{-1} D^R * P^{-+} * D^A. \quad (11)$$

Here  $D^A$  is the advanced Green function:

$$D_{\mu\nu}^A(x_1, x_2) = D_{\nu\mu}^{R*}(x_2, x_1).$$

Using (10) we then get the following equation for  $\bar{D}^{-+}$ :

$$\begin{aligned} \square_1 \bar{D}^{-+} &= \Pi_\perp^R \bar{D}^{-+}, \quad \square_2 \bar{D}^{-+} = \bar{D}^{-+} \Pi_\perp^A, \\ P_{\nu'}^A(x', x) &= P_{\nu\nu'}^{R*}(x, x'). \end{aligned} \quad (12)$$

We restrict ourselves to the stationary case, which is of practical importance. It is then convenient to change to the Fourier representation in the time argument. In all differential equations given above one must replace the D'Alembert operator by the Helmholtz operator:

$$\square_i \rightarrow k_i^2 + \Delta_i, \quad k_i = \omega_i/c, \quad i=1, 2,$$

$\omega_i$  are the frequencies corresponding to the variables  $t_i$ .

To solve Eq. (12) we specify the following boundary conditions. There is incident upon the cell from infinity a plane wave which is characterized by specifying the correlation function  $D_s$  on some surface  $S$  which is perpendicular to the direction of propagation. We shall assume that the boundaries of the cell are transparent so that at infinity the intensity of the scattered light decreases in proportion to  $1/R^2$ . In the geometric optics limit the solution of Eqs. (6) and (12) has the form

$$D^{-+} = \frac{4k^2}{(4\pi\hbar)^2} D^R * D_s * D^A + \frac{1}{4\pi\hbar} D^R * P^{-+} * D^A, \quad k = \frac{\omega_0}{c}. \quad (13)$$

The first term in (13) determines the correlator of the transmitted light, the second that of the fluorescent light (for the regions inside the cell—the trapped radiation). For a practical use of Eq. (13) one needs solve Eq. (10) for the retarded Green function  $D^R$ .

### 4. RETARDED GREEN FUNCTION

Using the connection between the atomic Green functions and the components of the single-particle density matrix  $\rho_{n'n}(\mathbf{p}, \mathbf{r}, t)$  in the Wigner representation (see Appendix I) we can express the kernel of the polarization operators in terms of  $\rho_{n'n}(\mathbf{p}, \mathbf{r}, t)$ . In particular, we have for the kernel of the operator  $\Pi_\perp^R$  (without the index  $\perp$ ) in the Fourier representation with respect to the time argument

$$\begin{aligned} \frac{i}{4\pi\hbar} P_{\mu\mu'}^R(\omega, \omega') &= \left( \frac{\omega_0}{\hbar c} \right)^2 \sum_{mm'} \sum_{nn'} (d_\mu)_{nm} (d_{\mu'})_{m'n'} \\ &\times \iint \frac{d^3 p}{(2\pi\hbar)^3} \frac{d^3 p'}{(2\pi\hbar)^3} \exp \left[ -\frac{i}{\hbar} (\mathbf{p}-\mathbf{p}') \cdot (\mathbf{r}-\mathbf{r}') \right] \\ &\times \left\{ \delta_{n'n} \rho_{mm'} \left( \mathbf{p}', \frac{\mathbf{r}+\mathbf{r}'}{2} \right) \right. \\ &\times 2\pi\delta(\omega-\omega') \left[ i(\omega-\omega_{mn}) - \frac{i}{\hbar} [\varepsilon(p') - \varepsilon(p)] + \gamma_m + \gamma_n \right]^{-1} \\ &\left. - \delta_{m'm} \rho_{n'n} \left( \mathbf{p}, \frac{\mathbf{r}+\mathbf{r}'}{2} \right) 2\pi\delta(\omega-\omega'-\Omega_{n'n}) \right. \\ &\times \left[ \frac{i}{2} (\omega+\omega') - \frac{i}{\hbar} \left( E_m - \frac{1}{2} (E_n + E_{n'}) \right) \right. \\ &\left. \left. - \frac{i}{\hbar} (\varepsilon(p') - \varepsilon(p)) + \gamma_m + \gamma_n \right]^{-1} \right\}. \end{aligned} \quad (14)$$

Formula (14) corresponds to the stationary case. The indexes  $m, m'$  indicate the quantum numbers of the excited state,  $n, n'$  the quantum numbers of the ground state,  $E_m, E_n, E_{m'}, E_{n'}$  are the energies of the states,  $\omega_{mn}$  is the frequency of the transition,  $\gamma_m, \gamma_n$  are the reciprocals of the lifetimes of the states, and  $\varepsilon(p) = p^2/2M$  is the kinetic energy of the atom.

We have taken into account in Eq. (14) the possibility of quantum oscillations in the ground state, i.e., the existence of an oscillating coherence

$$\rho_{n'n}(\mathbf{p}, \mathbf{r}, t) = \rho_{n'n}(\mathbf{p}, \mathbf{r}) \exp(-i\Omega_{n'n}t),$$

which can be excited in both Zeeman and hyperfine transitions with frequency  $\Omega_{n'n}$ . A detailed study of quantum oscillations in the case of Zeeman coherences was made in Refs. 1-4 for optically thin cells. The specific feature of the hyperfine coherences is that a situation is possible when  $\Omega_{n'n}$  is much larger than the Doppler ( $\gamma_D$ ) and homogeneous ( $\gamma$ ) line widths. When the cell is illuminated by light with a spectrum centered on one of the resonance frequencies  $\omega_1$  of an atomic transition there appear in the spectral intensity, on leaving the cell, combination components at frequencies  $\omega_2 = \omega_1 \pm \Omega_{n'n}$  and an interference component at the frequency  $(\omega_1 + \omega_2)/2$ , oscillating with frequency  $\Omega_{n'n}$ . We shall not analyze in the present paper the role of quantum beats.

We solve Eq. (10) for the case when there are no quantum beats. The polarization operator, and as a consequence the Green function  $D^R$ , will then be diagonal in the frequency arguments. We look for the function  $D^R$  in the form

$$D_{\mu\nu}^R(\omega_1\mathbf{r}_1, \omega_2\mathbf{r}_2) = -2\pi\hbar\delta(\omega_1 - \omega_2) X_{\mu\nu}(\omega, \mathbf{r}_1\mathbf{r}_2) \exp(ik|\mathbf{r}_1 - \mathbf{r}_2|)/|\mathbf{r}_1 - \mathbf{r}_2|. \quad (15)$$

Thanks to the presence of the  $\delta_1$  function in Eq. (10), in a coordinate frame with the  $z$ -axis along  $\mathbf{r}_1 - \mathbf{r}_2$ , the indexes  $\mu, \nu$  in (15) take on two values corresponding to the two possible polarization directions along the  $x$ - and  $y$ -axes. Considering  $X_{\mu\nu}$  to be a slowly varying function of  $\mathbf{r}_1$  as compared to  $\exp(ik|\mathbf{r}_1 - \mathbf{r}_2|)$ , we get for it in the  $xyz$  system of coordinates

$$\frac{d}{dz_1} X_{\mu\nu}(\omega, \mathbf{r}_1\mathbf{r}_2) = -\frac{1}{2} \sum_{\lambda} \alpha_{\mu\lambda}(\omega, \mathbf{r}_1) X_{\lambda\nu}(\omega, \mathbf{r}_1\mathbf{r}_2) \quad (16)$$

with boundary conditions  $X_{\mu\nu} \rightarrow \delta_{\mu\nu}$  as  $z_1 \rightarrow z_2$ . The quantity  $\alpha_{\mu\lambda}(\omega, \mathbf{r}_1)$  has the form

$$\alpha_{\mu\lambda} = -\frac{4\pi k}{\hbar} \sum_{m m'} \sum_{n n'} \langle j_0 n | d_{\mu} | j m \rangle \langle j m' | d_{\lambda} | j n' \rangle \times \int \frac{d^3 p}{(2\pi\hbar)^3} \left[ i \left( \omega - \omega_{j_0} - \frac{\mathbf{k}\mathbf{p}}{M} \right) + \gamma_j + \gamma_{j_0} \right]^{-1} \times \{ \delta_{n'n} \rho_{j m, j m'}(\mathbf{p}, \mathbf{r}_1) - \delta_{m'm} \rho_{j_0 n', j_0 n}(\mathbf{p}, \mathbf{r}_1) \}. \quad (17)$$

Here  $j_0, j$  are the total angular momenta of the ground and excited states,  $m, m', n, n'$  are the  $z$ -components of the angular momenta.

We change in Eq. (16) to cyclic components which are covariant in the first polarization index and contravariant in

the second.<sup>14</sup> We expand the two-dimensional co-contravariant matrix  $\alpha_q^p(\omega, \mathbf{r}_1)$  in terms of Pauli matrices  $\sigma = (\sigma_1, \sigma_2, \sigma_3)$ :

$$\alpha_q^p(\omega, \mathbf{r}_1) = \sigma_{kij}(\omega) \{ \alpha(\mathbf{r}_1) \delta_q^p + \beta(\mathbf{r}_1) \sigma_q^p \}. \quad (18)$$

We express the expansion coefficients in terms of the PM of the atomic density matrix  $\rho_{j_0(j)}$  and the reduced complex cross section  $\sigma_{j_0(j)}(\omega)$ :

$$\sigma_{j_0(j)}(\omega) = \frac{4\pi k}{\hbar} |d_{j_0(j)}|^2 \int \frac{d^3 p}{(2\pi\hbar)^3} \frac{f_0(p)}{i(\omega - \omega_{j_0} - \mathbf{k}\mathbf{p}/M) + \gamma_{j_0} + \gamma_j}, \quad (19)$$

$d_{j_0(j)} \equiv \langle j_0 || d || j \rangle$ . The quantity  $\alpha(\mathbf{r}_1)$  and the components of the real vector  $\beta(\mathbf{r}_1)$  have the form

$$\alpha(\mathbf{r}_1) = \frac{\rho_{j_0^{00}}(\mathbf{r}_1)}{3(2j_0+1)} - \frac{\rho_j^{00}(\mathbf{r}_1)}{3(2j+1)} + (-1)^{j+j_0+1} \left[ \frac{5}{6(2j_0+1)} \right]^{1/2} \left\{ \begin{matrix} 1 & 1 & 2 \\ j_0 & j_0 & j \end{matrix} \right\} \rho_{j_0^{20}}(\mathbf{r}_1) - (-1)^{j+j_0+1} \left[ \frac{5}{6(2j+1)} \right]^{1/2} \left\{ \begin{matrix} 1 & 1 & 2 \\ j & j & j_0 \end{matrix} \right\} \rho_j^{20}(\mathbf{r}_1), \quad (20)$$

$$\beta_z(\mathbf{r}_1) = (-1)^{j+j_0} \left[ \frac{3}{2(2j_0+1)} \right]^{1/2} \left\{ \begin{matrix} 1 & 1 & 1 \\ j_0 & j_0 & j \end{matrix} \right\} \rho_{j_0^{10}}(\mathbf{r}_1) + (-1)^{j+j_0} \left[ \frac{3}{2(2j+1)} \right]^{1/2} \left\{ \begin{matrix} 1 & 1 & 1 \\ j & j & j_0 \end{matrix} \right\} \rho_j^{10}(\mathbf{r}_1), \quad (21)$$

$$\beta_x - i\beta_y = (-1)^{j+j_0+1} \left( \frac{5}{2j_0+1} \right)^{1/2} \left\{ \begin{matrix} 1 & 1 & 2 \\ j_0 & j_0 & j \end{matrix} \right\} \rho_{j_0^{22}}(\mathbf{r}_1) - (-1)^{j+j_0+1} \left( \frac{5}{2j+1} \right)^{1/2} \left\{ \begin{matrix} 1 & 1 & 2 \\ j & j & j_0 \end{matrix} \right\} \rho_j^{22}(\mathbf{r}_1). \quad (22)$$

For the sake of simplicity we assume a Maxwellian momentum dependence of the density matrix, which is typical for optical alignment experiments.

$$\rho_{j_0 n', j_0 n}(\mathbf{r}, \mathbf{p}) = f_0(\mathbf{p}) \rho_{j_0 n', j_0 n}(\mathbf{r}).$$

The PM are defined in the standard way:

$$\rho_{j_0^{2q}}(\mathbf{r}) = \sum_{n' n} (-1)^{j_0-n'} (2j_0+1)^{1/2} \begin{pmatrix} j_0 & \times & j_0 \\ -n' & -q & n \end{pmatrix} \rho_{j_0 n', j_0 n}(\mathbf{r}), \quad (23)$$

and similarly for the excited state.

If we restrict ourselves in the solution of Eq. (16) to the first-order approximation in the matrix  $\alpha_q^p$  we arrive, after substituting (15) into (13), at the case of an optically thin layer which was considered in detail in Refs. 1-5. The constant parameters  $\rho^{10}$  and  $\rho^{22}$  determine in that case the anisotropic optical properties of the medium:  $\rho^{10}$  the optical activity and the magneto-circular dichroism,  $\rho^{22}$  the birefringence in the optical transparency region. It will be clear from what follows that taking the finite optical thickness into account leads both to significant quantitative changes, and to the appearance of qualitatively new effects.

In the general case of arbitrary polarization and arbitrary dependence of the matrix  $\alpha_q^p(\omega, \mathbf{r}_1)$  on the coordinate  $\mathbf{r}_1$ , one cannot solve Eq. (16) analytically, since the matrices  $\beta(\mathbf{r})\sigma$  in different points  $\mathbf{r}$  do not commute. As a consequence this leads to the impossibility of obtaining for the atomic density matrix a closed kinetic equation (such as the D'yakonov-Perel' equation<sup>6</sup>) which takes into account the

trapping of radiation for arbitrary spatially distributed atomic PM. Using the fact that

$$[\beta(\mathbf{r})\sigma, \beta(\mathbf{r}')\sigma] = 2i[\beta(\mathbf{r}) \times \beta(\mathbf{r}')]\sigma, \quad (24)$$

we can neglect the non-commutativity if the vector  $\beta$  changes little in direction over distances of the order of the photon mean free path at a given frequency  $l_{ph}(\omega)$ . Such a case is rather often realized in experiments and we shall therefore consider it first in what follows. We show that besides the trivial experimental situations when the PM are constant in the cell or the momenta  $j_0$  and  $j$  do not exceed  $\frac{1}{2}$ , this assumption is satisfied in the case of a magnetic field which is strong relative to the relaxation of the PM when there is no alignment. If we analyze radiation trapping in a dense layer the polarization of the medium with respect to the internal moment changes also here weakly over distances of the order of  $l_{ph}(\omega)$ , since the atoms of the gas which are separated by distances of the order of  $l_{ph}(\omega)$  from one another are approximately under identical optical pumping conditions. We consider in the limiting problem in section 7 possible effects connected with the non-commutativity (24).

If we neglect the non-commutativity (24) the solution of Eq. (16) rewritten for the co-contravariant cyclic components has the form

$$X_{p'q}(\omega, \mathbf{r}_1, \mathbf{r}_2) = \exp(-\frac{1}{2}\sigma_{j_0}\langle\alpha\rangle|\mathbf{r}_1 - \mathbf{r}_2|) [\text{ch}\{\frac{1}{2}\sigma_{j_0}\beta|\mathbf{r}_1 - \mathbf{r}_2|\}\delta_{p'q} - \text{sh}\{\frac{1}{2}\sigma_{j_0}\beta|\mathbf{r}_1 - \mathbf{r}_2|\}\langle\beta\rangle\sigma_{p'q}/\beta]. \quad (25)$$

The angle brackets in (25) indicate averaging along the ray:

$$\langle\beta\rangle = \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \int_{\mathbf{r}_2}^{\mathbf{r}_1} \beta(\mathbf{r}) ds, \\ \langle\alpha\rangle = \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \int_{\mathbf{r}_2}^{\mathbf{r}_1} \alpha(\mathbf{r}) ds, \quad \beta = |\langle\beta\rangle|. \quad (26)$$

Using Eq. (25) we analyze the polarization of the transmitted light and of the fluorescent light.

## 5. POLARIZATION OF THE TRANSMITTED LIGHT

Using the connection (2) between the correlation function and the spectral density of the radiation polarization tensor, we get from the first term of (13)

$$I_{S p'q}(\omega, \mathbf{r}) = \sum_{p'q'} X_{p'p''}(\omega, \mathbf{r}\mathbf{r}') X_{q''q'}(\omega, \mathbf{r}\mathbf{r}') I_{S p'q'}(\omega, \mathbf{r}'). \quad (27)$$

Here  $I_{S p'q'}(\omega, \mathbf{r}')$  and  $I_{I p'q}(\omega, \mathbf{r})$  are the covariant cyclic components of the polarization tensor in the incident and transmitted waves, and the coordinates  $\mathbf{r}$  and  $\mathbf{r}'$  in (27) refer to the same ray. It follows from Eqs. (25) and (26) that the coordinate dependence in (27) is in fact determined by the length of the path transversed by the ray in the medium.

We introduce the PM for the polarization density matrix (3) of the radiation:

$$\Phi^{KQ}(\omega, \mathbf{r}) = \sum_{pq} C_{1p1-q}^{KQ} \Phi_{p'q'}(\omega, \mathbf{r}). \quad (28)$$

Using (27) we find that the PM of the transmitted and incident light are connected through the relation

$$\Phi_I^{KQ}(\omega, \mathbf{r}) I_I(\omega, \mathbf{r}) = \sum_{K'Q'} T_{K'Q'}^{KQ}(\omega, \mathbf{r}\mathbf{r}') \Phi_S^{K'Q'}(\omega, \mathbf{r}') I_S(\omega, \mathbf{r}'), \quad (29)$$

where  $I_S(\omega, \mathbf{r}')$  and  $I_I(\omega, \mathbf{r})$  are the spectral intensities of the incident and the transmitted light. The matrix  $T_{K'Q'}^{KQ}$  has the form

$$T_{K'Q'}^{KQ}(\omega, \mathbf{r}\mathbf{r}') = \exp(-\text{Re} \sigma_{j_0}\langle\alpha\rangle|\mathbf{r} - \mathbf{r}'|) \sum_{pq} \sum_{p'q'} C_{1p1-q}^{KQ} \\ \times C_{1p'1-q'}^{K'Q'} \left( a\delta_{p'p} - b \frac{\langle\beta\rangle\sigma_{p'p}}{\beta} \right) \left( a^*\delta_{q'q} - b^* \frac{\langle\beta\rangle\sigma_{q'q}}{\beta} \right). \quad (30)$$

We have used in (30) the notation

$$a = \text{ch}\{\frac{1}{2}\sigma_{j_0}\beta|\mathbf{r} - \mathbf{r}'|\}, \quad b = \text{sh}\{\frac{1}{2}\sigma_{j_0}\beta|\mathbf{r} - \mathbf{r}'|\}. \quad (31)$$

The matrix  $T_{K'Q'}^{KQ}$  has the dimensions  $5 \times 5$ , since  $KQ(K'Q') = 00, 10, 2-2, 20$ , and  $22$ .

The use of a detecting ray in a medium which is optically thick for resonance radiation is particularly effective in the region of optical transparency. We consider as an example the polarization of quasi-resonance light in the case when the medium is doubly refractive (i.e., when  $\rho_{j_0(j)}^{1q} = 0, \rho_{j_0(j)}^{2q} \neq 0$ ). Let there be linearly polarized light incident upon the cell:  $\Phi_S^{KQ} \neq 0$  for  $KQ = 00, 20, 2 \pm 2$ . The real part,  $\text{Re} \Phi_S^{22}$ , determines the degree of linear polarization along the  $x$ - or  $y$ -axes and the imaginary part,  $\text{Im} \Phi_S^{22}$ , that along an axis making an angle of  $\pi/4$  with the  $x$ -axis. We consider the component  $\Phi_I^{10}$  which determines the circular polarization of the radiation when it leaves the cell:

$$\Phi_I^{10}(\omega) = -\{\text{ch}(\text{Re} \sigma\beta L) - 2 \text{sh}(\text{Re} \sigma\beta L) [\langle\beta_x\rangle \text{Re} \Phi_S^{22}(\omega) + \langle\beta_y\rangle \text{Im} \Phi_S^{22}(\omega)]/\beta\}^{-1} \\ \times 2^{1/2} \sin(\text{Im} \sigma\beta L) [\langle\beta_x\rangle \text{Im} \Phi_S^{22}(\omega) + \langle\beta_y\rangle \text{Re} \Phi_S^{22}(\omega)]/\beta. \quad (32)$$

Here  $L$  is the length traversed by the ray in the cell,  $\sigma \equiv \sigma_{j_0}$ . We used in (32) the fact that  $\Phi^{KQ}$  is independent of  $\mathbf{r}$  when entering and leaving the cell. Necessary conditions for the occurrence of alignment are the following requirements: the frequency  $\omega$  must not be the same as the frequency of the atomic transition  $\omega_0 = \omega_{j_0}$ , so that  $\text{Im} \sigma \neq 0$ ; the angle between the vector  $\beta$  and the polarization vector of the incident radiation must be different from 0 and  $\pi/2$ .

The effect of the occurrence of circular polarization in initially linearly polarized light when it passes through a gas of aligned atoms was predicted in Refs. 1-4. Also given there is an optical interpretation of that effect as the consequence of birefringence when the radiation propagates in a direction at right angles to the optical axis, a direction determined in our case by the vector  $\beta$ . The results of Refs. 1-4 referred to the case of an optically thin layer and the alignment  $\Phi_I^{10}$  was a small quantity. Formula (32) shows that the effect is in the general case not small, since for large mismatches  $\text{Re} \sigma \sim (\omega - \omega_0)^{-2}$  and tends to zero faster than  $\text{Im} \sigma \sim (\omega - \omega_0)^{-1}$ . When  $L \sim \pi/2(\text{Im} \sigma\beta)$  it is possible to have  $\Phi_I^{10} \approx 2^{-1/2}$ , if  $\Phi_S^{22} = 1/2$  which corresponds to light which is linearly polarized along the  $x$  axis. Also characteristic is the oscillating dependence of the degree of alignment on the length  $L$  with a half-period  $\sim \pi(\beta \text{Im} \sigma)^{-1}$ . This dependence is, however, not strictly periodic. We bear in mind that in deriving (32) the vector  $\beta$  is assumed to be constant only in direction, but not in magnitude.

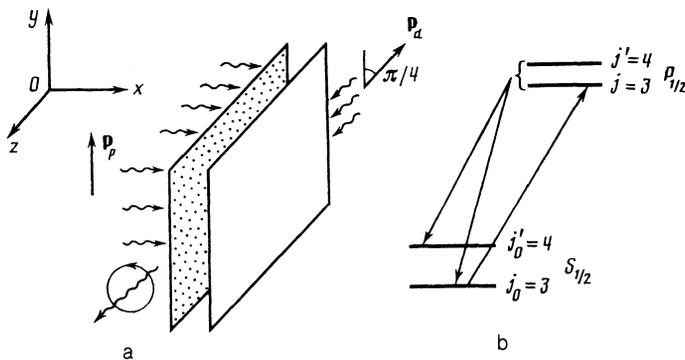


FIG. 1. Experimental scheme for the circular polarization of light by vapors of aligned atoms (a), system of driver levels and pumping scheme for Cs<sup>133</sup> (b);  $\mathbf{P}_p$ ,  $\mathbf{P}_d$  are the polarization vectors of the pumping and of the detecting radiation,  $j_0, j'_0, j, j'$  are the hfs sublevels of the ground,  $S_{1/2}$ , and of the excited,  $P_{1/2}$ , states of Cs<sup>133</sup>.

To give a clear quantitative illustration of the effect of circular polarization of the transmitted radiation we performed a numerical calculation for transitions between the hyperfine structure (hfs) components of the  $D_1$ -line of Cs<sup>133</sup>. As the driver we chose the transition between the lower sublevels of the hfs of the ground,  $^2S_{1/2}$ , and of the excited,  $^2P_{1/2}$ , states ( $j_0 = j = I - 1/2 = 3$ ). We consider the following experimental situation. Optical-pumping light, linearly polarized along the  $y$ -axis, aligns a layer of gas which is optically thin in the direction of the  $x$ -axis; see Fig. 1. A detector ray of frequency  $\omega$ , which is initially linearly polarized at an angle  $\pi/4$  to the  $y$ -axis, propagates in the  $z$ -direction. If the intensity of the detecting radiation is sufficiently weak, we must take into account only the pumping light in the optical-pumping equations. We have also used the assumption of strong collisional mixing on the excited sublevels. We give the form of the equation in Appendix 2. We show in Fig. 2 the way  $\Phi_t^{10}$  depends on the dimensionless optical length of the cell  $\tau = Ln_0\sigma_0$  ( $n_0$  is the density of the driver atoms and  $\sigma_0$  the absorption cross section at the center of the line) and on the relative mismatch  $\Delta/\gamma_D$ ,  $\Delta = \omega - \omega_0$ ,  $\gamma_D$  is the Doppler width, for a ratio  $W/\Gamma = 0.5$  of the rates of the optical pumping  $W$  and of the collisional relaxation in the ground state  $\Gamma$ . In dimensional units, the length  $L_{\max}$  corresponding to maximum alignment is large in the present case, of the order of  $10-10^2$  cm when  $n_0 \sim 10^{11}$  cm<sup>-3</sup>, since the degree of alignment of the ground state in the example considered is small:  $\sim 10\%$ .

In concluding this section we indicate the possibility of a practical use of the effect discussed as a means of registering radio-optical double resonance in alkali-atom vapors. Indeed, when switching on a radio-field on a 0-0 transition of the ground-state hfs of an optically aligned alkali atom, there occurs in the hyperfine sublevels an alignment in the direction of the static magnetic field  $\mathbf{H}_0$ . This alignment is re-

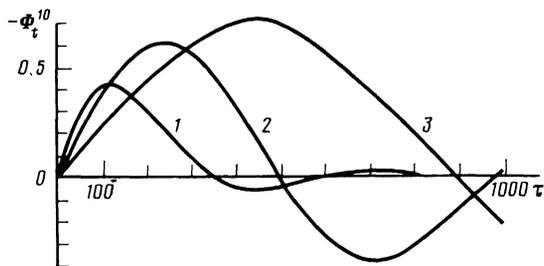


FIG. 2. The degree of circular polarization of the detecting radiation  $\Phi_t^{10}$  as function of the optical thickness  $\tau$  in Cs<sup>133</sup> vapors for a pumping parameter  $A = W/\Gamma = 0.5$  and a relative mismatch  $\Delta/\gamma_D = 1, 2, 3$ .

vealed by the appearance of circular polarization in an initially linearly polarized detector ray propagating at right angles to  $\mathbf{H}_0$ .

## 6. POLARIZATION OF THE FLUORESCENCE LIGHT

Substituting the equation for the retarded (advanced) photon Green function (15), (25) into the second term in (13) and performing the transformations (2) and (5), we get the spectral density of the fluorescence polarization tensor:

$$\Phi_f^{KQ}(\omega, \mathbf{nr}) I_j(\omega, \mathbf{nr}) = \frac{ck^4}{2\pi} \int d^3r' \frac{1}{|\mathbf{r}-\mathbf{r}'|^2} \delta\left(\mathbf{n} - \frac{\mathbf{r}-\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}\right) \times \sum_{K'Q'} T_{K'Q'}^{KQ}(\omega, \mathbf{r}\mathbf{r}') (-1)^{j+j_0+1} \left(\frac{2k'+1}{2j+1}\right)^{1/2} \left\{ \begin{matrix} 1 & 1 & K' \\ j & j & j_0 \end{matrix} \right\} \times |\hat{a}_{j_0}|^2 \int \frac{d^3p}{(2\pi\hbar)^3} f_0(\mathbf{p}) \frac{2\gamma_j}{(\omega-\omega_0-\mathbf{k}\mathbf{p}/M)^2 + \gamma_j^2} \rho_j^{K'Q'}(\mathbf{r}'). \quad (33)$$

Here  $\Phi_f^{KQ}(\omega, \mathbf{nr})$  and  $I_j(\omega, \mathbf{nr})$  are the PM and the spectral intensity of the fluorescent radiation in the direction  $\mathbf{n}$  at the point  $\mathbf{r}$ . The PM of the photon and atomic density matrices are defined in (33) in a system of coordinates with the  $z$ -axis along the direction of  $\mathbf{n}$ . The transition to the laboratory frame is accomplished through the standard transformation:

$$\rho_j^{KQ} = \sum_{\bar{Q}} D_{Q\bar{Q}}^K(\vartheta, \varphi) \rho_j^{K\bar{Q}}, \quad (34)$$

where  $\rho_j^{K\bar{Q}}$  is the PM of the atomic density matrix in the laboratory frame of reference, and  $\vartheta, \varphi$  are the polar and azimuthal angles of the direction of  $\mathbf{n}$ . The matrix  $\Phi_f^{KQ}$  is transformed similarly.

In contrast to the traditionally used expressions for the fluorescence polarization density matrices,<sup>15,16</sup> we have taken consistently into account in Eq. (33) the effect of the non-equilibrium state of the radiating gas with respect to its internal state. The PM of the fluorescent light  $\Phi_f^{KQ}$  depend on the PM of the atomic density matrix of not only the excited but also of the ground state through the matrix  $T_{K'Q'}^{KQ}(\omega, \mathbf{r}\mathbf{r}')$  given by Eq. (30). In particular, in an optically thick layer a situation is possible when the fluorescent polarization is completely determined by the PM of the ground state, i.e., when  $\rho_j^{KQ} = 0$ , but  $\rho_{j_0}^{KQ} \neq 0$ .<sup>1)</sup>

We consider this effect in more detail using the Hanle effect as an example. We assume an experimental situation similar to the one shown in Fig. 1, with the difference that the fluorescent light is registered in the direction of the  $z$ -axis (there is no test ray) and there is a static magnetic field  $\mathbf{H}_0$  applied. The degree of linear polarization and its direction are determined by the moment  $\Phi_f^{22}(\omega, n_z)$ :

$$\Phi_j^{22} = - \left[ 2 \int_0^L dz \exp(-\text{Re } \sigma \langle \alpha \rangle_z z) \text{ch}(\text{Re } \sigma \beta(z) z) \right]^{-1} \times \int_0^L dz \exp(-\text{Re } \sigma \langle \alpha \rangle_z z) \text{sh}(\text{Re } \sigma \beta(z) z) \frac{\langle \beta_x \rangle_z + i \langle \beta_y \rangle_z}{\beta(z)}. \quad (35)$$

$$\Phi_j^{22} = - \frac{\beta_x + i \beta_y}{2\beta} \frac{\beta - \exp(-\text{Re } \sigma \langle \alpha \rangle L) \{ \langle \alpha \rangle \text{sh}(\text{Re } \sigma \beta L) + \beta \text{ch}(\text{Re } \sigma \beta L) \}}{\langle \alpha \rangle - \exp(-\text{Re } \sigma \langle \alpha \rangle L) \{ \langle \alpha \rangle \text{ch}(\text{Re } \sigma \beta L) + \beta \text{sh}(\text{Re } \sigma \beta L) \}}. \quad (36)$$

Equation (36) shows that maximum polarization in a zero magnetic field is possible in a cell of infinite optical thickness when  $\beta \sim \langle \alpha \rangle$ . In the case of the optical pumping of Cs<sup>133</sup> atoms using the scheme shown in Fig. 1, maximum fluorescent polarization is reached when the parameter  $A = W/\Gamma \rightarrow \infty$ . We show in Fig. 3 the degree of linear polarization  $p$  and the angle  $\psi$  of the rotation of the direction of the polarization relative to the  $y$ -axis as functions of  $\omega_{j_0}/\Gamma$  ( $\omega_{j_0}$  is the Zeeman splitting frequency) for different values of the parameter  $A$  and as  $L \rightarrow \infty$  for fluorescent radiation at the transition  $j = 3 \rightarrow j_0 = 3$ . The parameters  $p$  and  $\psi$  are connected with  $\Phi_j^{22}$  through the relations

$$p = 2[(\text{Re } \Phi_j^{22})^2 + (\text{Im } \Phi_j^{22})^2]^{1/2}, \quad \cos 2\psi = -2 \text{Re } (\Phi_j^{22})/p. \quad (37)$$

In concluding this section we note that a direct quantitative account of the effect of a finite optical thickness on the Hanle effect is, for example, important for problems of magnetometry of solar protuberances (see Ref. 18 and the literature given there). This problem has not been widely studied at the present time.

## 7. EFFECT OF SPATIAL DIRECTIONAL INHOMOGENEITY OF THE POLARIZATION OF THE MEDIUM

We take the spatial inhomogeneity in direction of the vectors  $\beta(\mathbf{r})$  into account in a model problem. We consider a cell consisting of two regions, in each of which the direction of  $\beta(\mathbf{r})$  is constant. We assume that a detecting ray passes through the cell and has a spectrum which lies in the optical transparency region where one can neglect fluorescence. The polarization density matrix of the radiation incident on the photodetector is given by the expression

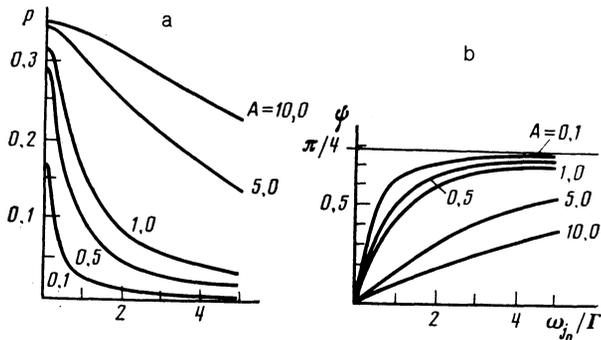


FIG. 3. The degree of fluorescent polarization  $p$  and the angle of rotation of the polarization direction  $\psi$  as functions of the ratio of the Zeeman splitting  $\omega_{j_0}$  to the rate of relaxation  $\Gamma$  for the  $P_{1/2}(j=3) \rightarrow S_{1/2}(j_0=3)$  transition in Cs<sup>133</sup>;  $A = W/\Gamma$  is the pumping parameter.

The quantities  $\langle \alpha \rangle_z$ ,  $\langle \beta \rangle_z$ , and  $\beta(z)$  are given by Eqs. (26), in which we take zero as the lower integration limit and  $z$  as the upper one. If the pumping is uniform, the parameters  $\langle \alpha \rangle$  and  $\langle \beta \rangle$  in (35) are independent of  $z$  and one can perform the integration:

$$\Phi_i^{KQ}(\omega) I_i(\omega) = \sum_{K''Q''} \sum_{K'Q'} T_{K''Q''}^{KQ}(\omega, 2) T_{K'Q'}^{K''Q''}(\omega, 1) \Phi_s^{K'Q'}(\omega) I_s(\omega), \quad (38)$$

which is an obvious generalization of Eq. (29). The matrices  $T_{K'Q'}^{K''Q''}(\omega, 1)$  and  $T_{K''Q''}^{KQ}(\omega, 2)$  determine the change in the PM of the light in the first and the second parts of the cell, respectively. One shows easily that the matrix

$$T_{K'Q'}^{KQ}(\omega, 21) = \sum_{K''Q''} T_{K''Q''}^{KQ}(\omega, 2) T_{K'Q'}^{K''Q''}(\omega, 1) \quad (39)$$

will be given by Eq. (30) in which we must perform the substitution

$$a \rightarrow a_1 a_2 + b_1 b_2 (\beta_1 \beta_2) / \beta_1 \beta_2, \quad b\beta/\beta \rightarrow a_1 b_2 \beta_2 / \beta_2 + a_2 b_1 \beta_1 / \beta_1 - i b_2 b_1 [\beta_2 \beta_1] / \beta_2 \beta_1, \quad (40)$$

where the indexes 1 and 2 indicate, respectively, the parameters for the first and second parts of the cell.

One can distinguish explicitly in experiments the effects connected with the inhomogeneity of the directions of the  $\beta(\mathbf{r})$  if we reverse the direction of propagation of the test ray. The polarization density matrices of the transmitted light for different directions of propagation will differ in the model considered by the quantity

$$\Delta \Phi_i^{KQ}(\omega) = \sum_{K'Q'} A_{K'Q'}^{KQ}(\omega, 21) \Phi_s^{K'Q'}(\omega), \quad (41)$$

where the transformation matrix has the form

$$A_{K'Q'}^{KQ}(\omega, 21) = T_{K'Q'}^{KQ}(\omega, 21) - T_{K'Q'}^{KQ}(\omega, 12) = 2i \frac{[\beta_1 \beta_2]}{\beta_1 \beta_2} \sum_{pq} \sum_{p'q'} C_{1p1-q}^{KQ} C_{1p'1q'}^{K'Q'} \times \{ b_1^* b_2^* \sigma_q^q [(a_1 a_2 + b_1 b_2 (\beta_1 \beta_2) / \beta_1 \beta_2) \delta_p^{p'} - (a_1 b_2 \beta_2 / \beta_2 + a_2 b_1 \beta_1 / \beta_1) \sigma_p^{p'}] - b_1 b_2 \sigma_p^{p'} [(a_1^* a_2^* + b_1^* b_2^* (\beta_1 \beta_2) / \beta_1 \beta_2) \delta_q^{q'} - (a_1^* b_2^* \beta_2 / \beta_2 + a_2^* b_1^* \beta_1 / \beta_1) \sigma_q^{q'}] \}. \quad (42)$$

We have neglected for the sake of simplicity absorption in Eqs. (41) and (42) putting  $\text{Im } \sigma_{j,j} \gg \text{Re } \sigma_{j,j}$ . The quantity inside the braces in (42) is, after summation over the indexes  $p, q, p',$  and  $q'$ , purely imaginary so that the whole expression (42) is real.

Equations (38)–(40) show that the effects of the transformation of the PM of the density matrix of the transmitted ray in the case of inhomogeneity in the directions of the  $\beta(\mathbf{r})$

are qualitatively the same as in the homogeneous case. Indeed, Eqs. (38)–(40) which lead to the transformation matrix in the form (30) can be generalized to the case of an arbitrary number of spatially homogeneous layers in which a real cell can be split up. In order to reveal experimentally the inhomogeneity of the  $\beta(\mathbf{r})$  one can compare the polarization of the radiation passing in opposite directions through the cell.

## 8. CONCLUSION

In the present paper we have analyzed examples of experiments in which the interaction of quasi-resonance radiation with gas atoms that are polarized by external pumping sources leads to a significant change of its polarization characteristics. The arrangement of such experiments expands the possibilities of studying atoms and molecules by optical alignment methods. In this connection we note that the example of hyperfine transitions in the  $D_1$  line of  $\text{Cs}^{133}$  considered in the paper was chosen because of methodological considerations. The  $\text{Cs}^{133}$  atom is a convenient study object because of the high saturated-vapor pressure and the well-resolved hfs of the ground and excited states. The experiments proposed are suitable also for atoms which have not often been studied by optical alignment methods. Most effective are such experiments for elements which have a rather large ground state angular momentum  $j_0 \geq 1$ . The rare earth atoms may serve as an example. In particular, the proposed realization of the Hanle effect for ground state PM enables one to determine the cross section for the collisional depolarization of the ground state of these atoms. The appearance of circular polarization for initially linearly polarized light when it passes through a gas of aligned atoms may be used to determine the magnitude and direction of the alignment in the cell. We note also that the expressions for the spectral density of the polarization tensor of the transmitted light, (29), (30), and of the fluorescent light, (33), may be used to derive self-consistent equations for optical pumping in an optically dense gaseous medium. To do this it is necessary to substitute the spectral density of the polarization tensor of the transmitted and the fluorescent light in the usual optical pumping equations for a thin layer (see, e.g., Refs. 18, 19) and regard in them the atomic density matrix as a function of the coordinates. The equations obtained in that way are a generalization of the D'yakonov-Perel' equations<sup>6</sup> to the case of non-vanishing ground-state PM when the direction of the polarization of the medium changes little over distances of the order of the photon mean free path.

## APPENDIX 1

The atomic Green functions which occur in the expressions for the kernels of the polarization operators (9) are connected with the single-particle density matrix through the relations

$$iG_{n'n}^{+-}(t\mathbf{r}, t'\mathbf{r}') = \int \frac{d^3p}{(2\pi\hbar)^3} \exp\left\{\frac{i}{\hbar} \mathbf{p}(\mathbf{r}-\mathbf{r}') - \frac{i}{\hbar} \varepsilon(p)(t-t') - \frac{i}{\hbar} \frac{E_n+E_{n'}}{2}(t-t') - (\gamma_n+\gamma_{n'})|t-t'|\right\} \left\{ \delta_{n'n} \theta(t-t') \pm \rho_{n'n} \left( \mathbf{p}, \frac{\mathbf{r}+\mathbf{r}'}{2}, \frac{t+t'}{2} \right) \right\}, \quad (\text{A1.1})$$

$$iG_{n'n}^{++}(t\mathbf{r}, t'\mathbf{r}') = [iG_{n'n}^{--}(t'\mathbf{r}', t\mathbf{r})]^*, \quad (\text{A1.2})$$

$$iG_{n'n}^{+-}(t\mathbf{r}, t'\mathbf{r}') = \int \frac{d^3p}{(2\pi\hbar)^3} \exp\left\{\frac{i}{\hbar} \mathbf{p}(\mathbf{r}-\mathbf{r}') - \frac{i}{\hbar} \varepsilon(p)(t-t') - \frac{i}{\hbar} \frac{E_n+E_{n'}}{2}(t-t') - (\gamma_n+\gamma_{n'})|t-t'|\right\} \times \left\{ \delta_{n'n} \pm \rho_{n'n} \left( \mathbf{p}, \frac{\mathbf{r}+\mathbf{r}'}{2}, \frac{t+t'}{2} \right) \right\}, \quad (\text{A1.3})$$

$$iG_{n'n}^{+-}(t\mathbf{r}, t'\mathbf{r}') = \pm \int \frac{d^3p}{(2\pi\hbar)^3} \exp\left\{\frac{i}{\hbar} \mathbf{p}(\mathbf{r}-\mathbf{r}') - \frac{i}{\hbar} \varepsilon(p)(t-t') - \frac{i}{\hbar} \frac{E_n+E_{n'}}{2}(t-t') - (\gamma_n+\gamma_{n'})|t-t'|\right\} \rho_{n'n} \times \left( \mathbf{p}, \frac{\mathbf{r}+\mathbf{r}'}{2}, \frac{t+t'}{2} \right). \quad (\text{A1.4})$$

The upper sign in these formulae corresponds to bosons and the lower one to fermions.

## APPENDIX 2

The set of optical pumping equations of an alkali atom the solution of which was used in sections 5 and 6 has the form

$$(i\omega_{j_0} q + \Gamma_{j_0}^{\kappa}) \rho_{j_0}^{\kappa q} = W \delta_{\kappa 0} \delta_{q_0} \sum_{\kappa' q'} \frac{2j_0+1}{(2S+1)(2I+1)} \cdot 3\bar{\Pi}_{j_0} \times (-1)^{\bar{j}_0+j+1} \left\{ \begin{matrix} \bar{j}_0 & \bar{j}_0 & \kappa' \\ 1 & 1 & j \end{matrix} \right\} \cdot (-1)^{\kappa'-q'} \rho_{j_0}^{\kappa' q'} \Phi_p^{\kappa'-q'} - W \sum_{\kappa' q'} \sum_{KQ} \delta_{j_0 \bar{j}_0} \bar{\Pi}_{j_0}^3 \Pi_{\kappa' \kappa K} (-1)^{\kappa-q} \left( \begin{matrix} K & \kappa & \kappa' \\ Q & -q & q' \end{matrix} \right) \times (-1)^{1+j-j_0+\kappa'} \left\{ \begin{matrix} j_0 & j_0 & K \\ 1 & 1 & j \end{matrix} \right\} \left\{ \begin{matrix} \kappa & \kappa' & K \\ j_0 & j_0 & j_0 \end{matrix} \right\} \rho_{j_0}^{\kappa' q'} \Phi_p^{KQ} + \Gamma_{j_0}^{\kappa} \rho_{j_0}^{\kappa q}. \quad (\text{A2.1})$$

Here  $j_0$  and  $j'_0$  are two sublevels of the ground state hfs of an alkali atom,  $S = \frac{1}{2}$  is the electron spin,  $I$  the nuclear spin,  $\bar{j}_0$  the sublevel involved in the pumping,  $\omega_{j_0}$  the Zeeman splitting frequency,  $\bar{\Pi}_{XY\dots} = [(2X+1)(2Y+1)\dots]^{1/2}$ ,  $W$  and  $\Phi_p^{KQ}$  the rate and the polarization density matrix of the optical pumping light. If we assume that the nuclear spin is conserved in depolarizing collisions, the relaxation matrix has the form

$$\Gamma_{j_0}^{\kappa} = \frac{1}{3} \Gamma \frac{\kappa(\kappa+1)}{(2I+1)^2} + \frac{1}{3} \Gamma \frac{2j'_0+1}{2I+1}, \quad \Gamma_{j_0' j_0}^{\kappa} = (-1)^{\kappa} \cdot \frac{2}{3} \Gamma (2j_0+1) \frac{[I(I+1)]^{1/2}}{2I+1} \left\{ \begin{matrix} \kappa & j_0 & j_0 \\ 1 & j_0' & j_0' \end{matrix} \right\}. \quad (\text{A2.2})$$

The quantity  $\Gamma$  is proportional to the probability of electron spin flipping in the alkali atom per collision. One must add to the homogeneous set of Eqs. (A2.2) the normalization condition

$$\rho_{j_0}^{00} + \rho_{j_0'}^{00} = 1. \quad (\text{A2.3})$$

<sup>1)</sup>In the case of the  $D_1$  line of  $\text{Cs}^{133}$  considered below the effective loss of alignment of the excited state always occurs for sufficiently high buffer gas pressures,  $p > 10$  Torr, because of the high values of the depolarization cross sections in the excited  $^2P_{1/2}$  states.<sup>17</sup>

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