Nonlinear optical activity of a resonant gaseous medium

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The nonlinear gyration vector and the tensor of the cubic susceptibility are determined for an isotropic gaseous resonant medium that has both natural and nonlinear optical activity. The nonlinear gyrotropy and nonlinear dichroism are investigated with allowance for the dopolarizing atomic collisions and for the Doppler effect. The possibilities are examined of directly applying the obtained nonlinear relations which are governed by self-rotation and deformation of the light-wave polarization ellipse.

Nonlinear optical activity of condensed media[1–4] and of gases[5–12] is being extensively investigated of late in view of the great promise of its use in spectroscopy, so that the development of a rigorous quantum-mechanical theory of this phenomenon is relevant. Nonlinear optical activity constitutes polarization self-action of a light wave, which reduces to self-rotation of the polarization ellipse (nonlinear gyrotropy) and to deformation of its shape (nonlinear dichroism). For a gas, these phenomena were considered in Ref. 5 for resonant transitions $0 \rightleftarrows 1$ and $1 \to 1$ with small angular momenta, and in Refs. 6–11 without allowance for the thermal motion of the atoms. In the case of large angular momenta, the polarization self-action of a light wave was investigated in Ref. 12 by semiclassical theory. In practice, however, interest attaches to results for arbitrary angular momenta, with allowance for the depolarizing atomic collisions and for the Doppler effect.

A rigorous quantum-mechanical approach is used in the present paper to investigate optical phenomena in a gaseous medium that has natural and nonlinear optical activities and contains various admixtures in addition to the resonant atoms. These nonlinear phenomena set in when the light-wave frequency is close to the frequency of a transition of the atom between states with arbitrary angular momenta. They come into play under conditions of thermal motion of the atoms in the presence of depolarizing collisions and of relaxation due to spontaneous emission on the excited levels. Resonant interaction adds to the gyration vector indicative of the natural optical activity a nonlinear gyration vector that describes the nonlinear polarization-ellipse rotation that is superimposed on the usual rotation in an optically active medium. As it rotates, the polarization ellipse is simultaneously deformed—a manifcstation of nonlinear optical activity induced by the light wave. The direction of the linear rotation of the polarization ellipse depends on the signs of the detuning from resonance and of the occupancy, on the electric-vector rotation direction, and also on the type of resonant transition and of the relaxation constants of the orientation and alignment of the atom. This makes it possible to separate in the experiments the contributions of the nonlinear and usual polarization-ellipse rotations. In addition, it permits a determination of the of the collisional shift of the central line and identification of the type of resonant transition, as well as a verification of the validity of the assumed atomic-collision model, since nonlinear rotation and deformation of the polarization ellipse on the simplest atomic transitions $0 \rightleftarrows 1$ and $1 \to 1$ are results of depolarizing collisions. In contrast to natural optical activity, the direction of the nonlinear rotation of the polarization ellipse is reversed after specular reflection, but the direction of its deformation is preserved. In multiple specular reflection, the cumulation effect causes the polarization-ellipse rotation angle and shape to change in such a manner that the light-wave polarization becomes ultimately linear or circular. These stationary regimes are reached much more rapidly on homogeneously than inhomogeneously broadened transitions. The regularities derived are described by simple equations that can be readily verified by experiment.

1. NONLINEAR GYROTROPY AND DICHROISM

Consider an isotropic gas having natural optical activity and containing resonant atoms in addition to various impurities. We determine the optical-activity change induced by the electric field

$$\mathbf{E}=\mathbf{a}\exp\left[i(\mathbf{kr}-\omega t)\right]+\text{c.c.} \tag{1}$$

of a monochromatic light wave, where **a** is a slow function of the coordinates compared with $\exp(i\mathbf{k}\cdot\mathbf{r})$, and the connection between the wave vector **k** and the frequency ω will be established below. The frequency is close to the frequency $\omega_{ba} = (E_b - E_a)\hbar^{-1}$ of the atomic transition between two states of a resonant atom with zero nuclear spin; these states are characterized by energies E_a and E_b $(E_a > E_b)$ and also by total angular momenta J_a and J_b and their projections M_a and M_b on the quantization axis. This reasoning is valid also for atoms with nonzero nuclear spin, if only one hyperfine sublevel, of either the upper or lower resonance levels, participates in the interaction with the wave (1). Moreover, The results are valid for vibrational-rotational molecular transitions $vJKM \to v'J'K'M'$, where v and J are vibrational and rotational quantum numbers, and K and M are projections of the total angular momentum on the molecule and quantization axes, respectively. It is assumed here that the K-degeneracy is lifted, i.e., the distance between neighboring K-split sublevels exceeds the homogeneous and inhomogeneous widths of the spectral line.

The induction **D** takes in the presence of resonance levels the form

$$\begin{gathered}\mathbf{D}=\mathbf{D}_0+4\pi\mathbf{P},\\ \mathbf{D}_0=\left[\varepsilon(\omega)\mathbf{a}+i[\mathbf{a}\mathbf{g}_0]\right]\exp\left[i(\mathbf{kr}-\omega t)\right]+\text{c.c.},\end{gathered} \tag{2}$$

where $\mathbf{D}_0$ is the induction in the absence of resonance levels, $\varepsilon(\omega)$ is the dielectric constant that takes into account the influence of the impurities and of the nonresonant levels of the active atoms,[13] **P** is the gas dielectric-polarization vector due to the resonance levels, $\mathbf{g}_0 = b\,\mathbf{k}/k$ is the gyration vector

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that characterizes the natural optical activity, and the constant b is independent of $\mathbf{k}$.

To calculate $\mathbf{P} = \int \mathrm{Sp}(\rho \mathbf{d}) d\mathbf{v}$ it is necessary to solve an equation for the density matrix ρ in the electric field (1), viz.,

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right)\rho = \frac{i}{\hbar}[\rho(H-\mathbf{Ed}) - (H-\mathbf{Ed})\rho] + \Gamma\rho, \tag{3}$$

where H is the Hamiltonian of a free active atom moving with velocity $\mathbf{v}$, $\mathbf{d}$ is the dipole-moment operator, and $\Gamma\rho$ is the collision integral. The latter takes a simple form if the density matrix ρ is expanded in irreducible tensor operators ($3j$-symbols), thereby transforming (3) for $\mathbf{E} = 0$ into a system of independent equations:

$$\left[\frac{\partial}{\partial t} + \mathbf{v}\nabla + i(\omega_{ba} + \Delta_{ba}^{(\varkappa)}) + \gamma_{ba}^{(\varkappa)}\right] \rho_q^{(\varkappa)}(J_bJ_a) = 0,$$

$$|J_b - J_a| \leqslant \varkappa \leqslant J_b + J_a, \tag{4}$$

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \gamma_b^{(\varkappa)}\right)\rho_q^{(\varkappa)}(J_bJ_b) = \gamma_b N_b f(v)\delta_{0\varkappa}\delta_{0q}, \quad 0 \leqslant \varkappa \leqslant 2J_b;$$

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \gamma_a^{(\varkappa)}\right)\rho_q^{(\varkappa)}(J_aJ_a) = (\gamma_a N_a - \gamma N_b) f(v)\delta_{0\varkappa}\delta_{0q}$$

$$+\gamma[(2J_a+1)(2J_b+1)]^{1/2}(-1)^{\varkappa+1+J_a+J_b}\begin{Bmatrix} J_a & \varkappa & J_a \\ J_b & 1 & J_b \end{Bmatrix}\rho_q^{(\varkappa)}(J_bJ_b),$$

$$0 \leqslant \varkappa \leqslant 2J_a, \tag{5}$$

$$\gamma_{ba}^{(\varkappa)} = \gamma_{ba} + \Gamma_{ba}^{(\varkappa)}, \quad \gamma_b^{(\varkappa)} = \gamma_b + \Gamma_b^{(\varkappa)}, \quad \gamma_a^{(\varkappa)} = \gamma_a + \Gamma_a^{(\varkappa)},$$

$$f(v) = \frac{1}{(\pi^{1/2}u)^3}\exp\left(-\frac{v^2}{u^2}\right), \quad \gamma = \frac{4|d_{ba}|^2\omega_{ba}^3}{3\hbar c^3(2J_b+1)}.$$

Here $\rho_q^{(\varkappa)}(J_bJ_a)$ is a multipole moment of rank $\varkappa$, relates the states of the atom with the angular moment J_b and J_a, and is of the form

$$\rho_q^{(\varkappa)}(J_bJ_a) = (2J_b+1)^{1/2}\sum_{M_bM_a}(-1)^{J_b-M_a}\begin{pmatrix} J_b & J_a & \varkappa \\ M_b & -M_a & q \end{pmatrix}\rho_{M_bM_a}, \tag{6}$$

where $\rho_{M_bM_a}$ is a component of the density matrix and $-\varkappa \leqslant q \leqslant \varkappa$. The multipole moments $\rho_q^{(\varkappa)}(J_bJ_b)$ and $\rho_q^{(\varkappa)}(J_aJ_a)$ characterize the polarization state of the atom on levels E_b and E_a. The first is obtained from (6) by making the substitutions $J_a \to J_b$ and $M_a \to M_b$, and second by the substitutions $J_b \to J_a$ and $M_b \to M_a$. The quantities $\gamma_{ba}^{(\varkappa)}$, $\gamma_b^{(\varkappa)}$ and $\gamma_a^{(\varkappa)}$ are the relaxation constants of these multipole moments, and Δ_a^b describes the contribution from the shifts of the levels E_a and E_b by the elastic depolarization collisions. The relaxation constants γ_{ba}, γ_b, and γ_a are due to spontaneous decay and to inelastic gaskinetic collisions. Next, N_a and N_b are the stationary atom densities on the levels E_a and E_b in the absence of the field (1), $f(v)$ a Maxwell distribution, u the most probable velocity, c the speed of light in vacuum, γ the probability of spontaneous emission of a photon $\hbar\omega_{ba}$ by an isolated atom, and d_{ba} the reduced dipole moment of the atomic transition $J_a \to J_b$.[14] The quantities $\Gamma_{ba}^{(\varkappa)}$, $\Gamma_b^{(\varkappa)}$ and $\Gamma_a^{(\varkappa)}$ are due to elastic depolarizing collisions, with $\Gamma_a^{(0)} = \Gamma_b^{(0)} = 0$. They have been calculated for small angular momenta in Ref. 15. If the impurity-atom mass does not exceed that of the resonant atom, the quantities $\Gamma_{ba}^{(\varkappa)}$, $\Gamma_b^{(\varkappa)}$, and $\Gamma_a^{(\varkappa)}$ are smooth functions of the velocity v compared with $v^2 f(v)$ in the vicinity of the most probable velocity u, and can be regarded as constants. In the opposite case it is necessary to take into account the dependences of these quantities on the velocity. The terms containing $f(v)$ in Eqs. (4) and (5) are due to the Boltzmann distribution of the atoms over the levels, or to a constantly applied pump that saturates with equal probability the Zeeman levels. The last term in the right-hand side of (5) describes the influx of atoms to the lower level on account of spontaneous emission on the upper one.

We solve Eq. (3) by perturbation theory in the resonance approximation, assuming the field (1) to be weak enough:

$$|\mathbf{E}|^2|d_{ba}|^2\gamma_{ba}^{(1)}\{\hbar^2\gamma_a^{(\varkappa)}[(|\Delta|+ku)^2 + (\gamma_{ba}^{(1)})^2]\}^{-1} \ll 1,$$

$$\Delta = \omega - \omega_{ba} - \Delta_{ba}^{(1)},$$

where Δ is the detuning from resonance, $\Delta_{ba}^{(1)}$ the collisional level shift, $\gamma_{ba}^{(1)}$ the homogeneous half-width of the $J_a \to J_b$ atomic transition line, and ku the inhomogeneous Doppler width.

In the stationary regime and in the cubic approximation in the field (1), the solution obtained for Eq. (3) yields the dielectric-polarization vector

$$\mathbf{P}(\mathbf{r}, t) = [\mathbf{P}^L(\omega, \mathbf{k}) + \mathbf{P}^N(\omega, \mathbf{k})]\exp[i(\mathbf{kr} - \omega t)] + \text{c.c.},$$

where $\mathbf{P}^L(\omega,\mathbf{k})$ and $\mathbf{P}^N(\omega,\mathbf{k})$ are the linear and nonlinear parts, which are described by the equations

$$\mathbf{P}^L(\omega, \mathbf{k}) = \varkappa(\omega, \mathbf{k})\mathbf{a}, \quad P_i^N(\omega, \mathbf{k}) = \chi_{ijkl}(\omega, \mathbf{k})a_j a_k^* a_l,$$

$$P_i^N(\omega, \mathbf{k}) = \mathbf{l}_i\mathbf{P}^N(\omega, \mathbf{k}), \quad a_i = \mathbf{l}_i\mathbf{a},$$

$$\varkappa(\omega, \mathbf{k}) = -\frac{N_{ab}|d_{ba}|^2}{3\hbar\gamma_{ba}^{(1)}}[Y_1(\xi, \eta) - iY_2(\xi, \eta)], \tag{7}$$

$$Y_1(\xi, \eta) = \frac{\eta}{\pi^{1/2}}\int_{-\infty}^{\infty}\frac{(\xi - x)e^{-x^2}}{(\xi - x)^2 + \eta^2}dx,$$

$$Y_2(\xi, \eta) = \frac{\eta^2}{\pi^{1/2}}\int_{-\infty}^{\infty}\frac{e^{-x^2}}{(\xi - x)^2 + \eta^2}dx,$$

$$N_{ab} = N_a/(2J_a+1) - N_b/(2J_b+1), \quad \xi = \Delta/ku, \quad \eta = \gamma_{ba}^{(1)}/ku,$$

$$\chi_{ijkl}(\omega, \mathbf{k}) = Q[(C_1 + C_2)\delta_{ij}\delta_{kl} + (C_2 - C_1)\delta_{ik}\delta_{lj} + {}^2/_3(C_0 - C_2)\delta_{il}\delta_{jk}], \tag{8}$$

$$Q = N_{ab}|d_{ba}|^4(\gamma_{ba}^{(1)})^{-2}\hbar^{-3}[Z_1(\xi, \eta) - iZ_2(\xi, \eta)],$$

$$Z_1(\xi, \eta) = \frac{\eta^3}{\pi^{1/2}}\int_{-\infty}^{\infty}\frac{(\xi - x)e^{-x^2}}{[(\xi - x)^2 + \eta^2]^2}dx,$$

$$Z_2(\xi, \eta) = \frac{\eta^4}{\pi^{1/2}}\int_{-\infty}^{\infty}\frac{e^{-x^2}}{[(\xi - x)^2 + \eta^2]^2}dx,$$

$$C_\varkappa = \frac{1}{\gamma_a^{(\varkappa)}}\begin{Bmatrix} 1 & 1 & \varkappa \\ J_a & J_a & J_b \end{Bmatrix}^2 + \frac{1}{\gamma_b^{(\varkappa)}}\begin{Bmatrix} 1 & 1 & \varkappa \\ J_b & J_b & J_a \end{Bmatrix}^2 + \frac{\gamma(2J_b+1)}{\gamma_a^{(\varkappa)}\gamma_b^{(\varkappa)}}(-1)^{J_a+J_b}\begin{Bmatrix} 1 & 1 & \varkappa \\ J_a & J_a & J_b \end{Bmatrix} \times \begin{Bmatrix} 1 & 1 & \varkappa \\ J_b & J_b & J_a \end{Bmatrix}\begin{Bmatrix} J_a & \varkappa & J_a \\ J_b & 1 & J_b \end{Bmatrix}, \tag{9}$$

where $\varkappa = 0, 1, 2$, the indices i, j, k, and l take on values x, y, and z, with $\mathbf{l}_x$, $\mathbf{l}_y$, and $\mathbf{l}_z$ the unit vectors of the Cartesian axes. The spatial dispersion of the dielectric susceptibility (7) and of the cubic susceptibility (8) is due to the Doppler effect. The last term in (9) is due to atoms going to the lower level on account of spontaneous emission on the upper level. It makes a noticeable contribution in a rarefied gas, when γ and $\gamma_b^{(\varkappa)}$ are of the same order. In a dense gas, the collision width of the level E_b is large ($\gamma_b^{(\varkappa)} \gg \gamma$) and the indicated term can be neglected. In addition, in the expression (8) for the tensor it is assumed that $\gamma_a^{(\varkappa)}$ and $\gamma_b^{(\varkappa)}$ do not depend on the velocity v. In the opposite case the quantities C_0, C_1, and C_2 in the tensor (8) are under the integral sign in the factor Q.

Substituting the obtained dielectric polarization vector $\mathbf{P}(\mathbf{r},t)$ in expression (2) for the induction $\mathbf{D}$, we get

$$\mathbf{D}=[(\varepsilon(\omega,\ \mathbf{k})+\varepsilon^N)\mathbf{a}+[\mathbf{a}(i\mathbf{g}-\mathbf{h})]]\exp[i(\mathbf{kr}-\omega t)]+\text{c.c},$$
$$\varepsilon(\omega,\ \mathbf{k})=\varepsilon(\omega)+4\pi\varkappa(\omega,\ \mathbf{k}), \tag{10}$$
$$\varepsilon^N=8\pi(C_0+2C_2)Q|\mathbf{a}|^2/3, \quad C_0+2C_2>0, \ \mathbf{g}=\mathbf{g}_0+\mathbf{f},$$

$$\mathbf{f}=i[\mathbf{aa}^*]4\pi(C_2-C_1)Q', \quad \mathbf{h}=i[\mathbf{aa}^*]4\pi(C_2-C_1)Q'', \tag{11}$$

$$C_2-C_1=\frac{1}{30J(J+1)(2J+1)}\times\Big[(2J-1)(2J+3)\Big(\frac{1}{\gamma_a^{(2)}}+\frac{1}{\gamma_b^{(2)}}-\frac{\gamma(J^2+J-3)}{\gamma_a^{(2)}\gamma_b^{(2)}J(J+1)}\Big)$$
$$-5\Big(\frac{1}{\gamma_a^{(1)}}+\frac{1}{\gamma_b^{(1)}}+\frac{\gamma(J^2+J-1)}{\gamma_a^{(1)}\gamma_b^{(1)}J(J+1)}\Big)\Big]$$

for $J_a=J\rightarrow J_b=J \quad (J>0)$,

$$C_2-C_1=-\frac{1}{30(J+1)(2J+1)(2J+3)}\Big[\frac{5J(2J+3)}{\gamma_a^{(1)}}-\frac{J(2J-1)}{\gamma_a^{(2)}}$$
$$+\frac{5(J+2)(2J+1)}{\gamma_b^{(1)}}-\frac{(J+2)(2J+5)}{\gamma_b^{(2)}}$$
$$+\frac{\gamma J(J+2)}{(J+1)(2J+1)}\Big(\frac{5(2J+1)(2J+3)}{\gamma_a^{(1)}\gamma_b^{(1)}}$$
$$-\frac{(2J-1)(2J+5)}{\gamma_a^{(2)}\gamma_b^{(2)}}\Big)\Big] \quad \text{for} \quad J_a=J\rightarrow J_b=J+1,$$

$$C_2-C_1=-\frac{1}{30(J+1)(2J+1)(2J+3)}\Big[\frac{5J(2J+3)}{\gamma_b^{(1)}}-\frac{J(2J-1)}{\gamma_b^{(2)}}$$
$$+\frac{5(J+2)(2J+1)}{\gamma_a^{(1)}}-\frac{(J+2)(2J+5)}{\gamma_a^{(2)}}$$
$$+\frac{\gamma J(J+2)}{(J+1)(2J+3)}\Big(\frac{5(2J+1)(2J+3)}{\gamma_a^{(1)}\gamma_b^{(1)}}$$
$$-\frac{(2J-1)(2J+5)}{\gamma_a^{(2)}\gamma_b^{(2)}}\Big)\Big] \quad \text{for} \quad J_a=J+1\rightarrow J_b=J,$$

where Q' and Q'' are the real and imaginary parts of $Q = Q' + iQ''$, while $\gamma_a^{(1)}$ ($\gamma_b^{(1)}$) and $\gamma_a^{(2)}$ ($\gamma_b^{(2)}$) are the relaxation constants of the orientation and alignment of the atom on the E_a (E_b) level.

Relation (10) between the induction and the electric field contains besides the tensor $ie_{ikm}g_m$ that describes the gyrotropy, another tensor $-e_{ikm}h_m$ that takes into account the difference between the energy exchange of the resonant gas with two light waves having orthogonal circular polarizations. The ability of an isotropic substance to have absorbed differently light waves with orthogonal circular polarizations is called dichroism.[16] In contrast to linear dichroism, the energy exchange with a resonant gas is the same for right- and left-polarized circular waves if they have different propagations, and a difference occurs only when they propagate jointly. Nonlinear dichroism sets in therefore in a resonant gas when the incident light wave is elliptically polarized, while in the case of linear and circular polarizations they must be unstable. Dichroism manifests itself by a deformation of the shape of the polarization ellipse. Nonlinear dichroisim in a resonant gas is always accompanied by gyrotropy and is characterized by an antisymmetric tensor $-e_{ikm}h_m$ or by dichroism vector $\mathbf{h}$ which is its dual.

It follows from (10) and (11) that the nonlinear gyrotropy and dichroism depend on the type of the resonant transition $J_a \rightarrow J_b$, on the depolarizing collisions, on the detuning from resonance Δ, and on the Doppler width ku. At the resonance $\Delta h = 0$, the former is optimal and the latter vanishes. If the wave (1) is linearly polarized, there is no nonlinear gyrotropy or dichroism, since the vectors a and a^* are collinear and $f = h = 0$. As this wave propagates, the polarization plane rotates just as in the absence of resonance levels. The latter influence only the connection between k and ω owing to the term ε^N in the dielectric constant. If $\mathbf{g}_0 = 0$, relation (10) describes nonlinear optical activity characterized by the axial vectors (11).

2. POLARIZATION SELF-ACTION

To simplify the equations we choose the connection between $\mathbf{k}$ and ω in the form $\omega^2\varepsilon'(\omega,\mathbf{k}) = k^2c^2$, and denote the absorption coefficient by $\alpha = \omega^2\varepsilon''(\omega,\mathbf{k})/kc^2$, where the prime and double prime mark the real and imaginary parts of the dielectric constant $\varepsilon(\omega,\mathbf{k}) = \varepsilon'(\omega,\mathbf{k}) + i\varepsilon''(\omega,\mathbf{k})$. In addition, we omit the second derivatives of the slow functions. We obtain then from the Maxwell equations, with allowance for the orthogonality of $\mathbf{a}$ and $\mathbf{k}$ in an isotropic gas,

$$\frac{d}{dz}\mathbf{a}+\frac{\alpha}{2}\mathbf{a}+\frac{\omega^2}{2kc^2}[\mathbf{ag}_0]-i\frac{cA}{2\pi}|\mathbf{a}|^2\mathbf{a}-i\frac{cB}{2\pi}\mathbf{a}^2\mathbf{a}^*=0, \tag{12}$$

$$A=\frac{(2\pi\omega)^2}{kc^3}\Big(\frac{2}{3}C_0+C_1+\frac{1}{3}C_2\Big)Q, \quad B=\frac{(2\pi\omega)^2}{kc^3}(C_2-C_1)Q. \tag{13}$$

To study the polarization self-action of wave (1), we express $\mathbf{a}$ in the general form

$$\mathbf{a}=\mathbf{l}|\mathbf{a}|\exp(i\Phi), \quad \mathbf{l}=\mathbf{l}_1\cos\varphi+i\mathbf{l}_2\sin\varphi, \quad \mathbf{l}_1\mathbf{l}_2=0,$$

where the polarization vector $\mathbf{l}$, the amplitude $|\mathbf{a}|$, and the phase Φ, as well as the lengths of the polarization semiaxes, equal to $|\cos\varphi|$ and $|\sin\varphi|$, are slow functions of the coordinates compared with $\exp(i\mathbf{k}\cdot\mathbf{r})$. As the light wave (1) propagates, the mutually orthogonal unit vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ can rotate around $\mathbf{k}$, describing the rotation of the polarization-ellipse axes. For convenience, we direct the Z axis along $\mathbf{k}$, and then the angle $\Omega = \Omega(z)$ of the rotation of the polarization-ellipse axes of wave (1) is given by

$$\mathbf{l}_1=\mathbf{l}_x\cos\Omega+\mathbf{l}_y\sin\Omega, \quad \mathbf{l}_2=-\mathbf{l}_x\sin\Omega+\mathbf{l}_y\cos\Omega,$$

where the positive direction of Ω is clockwise when viewed along $\mathbf{k}$. Multiplying Eq. (12) by $\mathbf{a}$ or $\mathbf{a}'$ and separating the real and imaginary parts $A = A' + iA''$ and $B = B' + iB''$ of (13), we arrive at a closed system of equations:

$$\frac{d}{dz}I+\alpha I+2[A''+B''(1-\sin^2 2\varphi)]I^2=0, \tag{14}$$

$$\frac{d}{dz}\sin 2\varphi-2B''I\sin 2\varphi(1-\sin^2 2\varphi)=0, \tag{15}$$

$$\frac{d}{dz}\Omega=\frac{\omega^2 g_0}{2kc^2}+B'I\sin 2\varphi, \tag{16}$$

$$\frac{d}{dz}\Phi=(A'+B')I, \tag{17}$$

where $T = c|\mathbf{a}|^2/2\pi$ is the intensity of wave (1), $g_0 = \mathbf{k}\cdot\mathbf{g}_0/k$, and $\sin 2\varphi$ is expressed in terms of the respective intensities I_+ and I_- of the right- and left-polarized circular waves:

$$\begin{gathered}\sin 2\varphi=(I_+-I_-)/I, \quad I_\pm=(\cos\varphi\pm\sin\varphi)^2 I/2,\\ I=I_++I_-, \quad \mathbf{E}=\mathbf{E}_++\mathbf{E}_-,\\ \mathbf{E}_\pm=\mathbf{l}_\pm 2^{-1/2}(\cos\varphi\pm\sin\varphi)|\mathbf{a}|\exp[i(\mathbf{kr}-\omega t+\Phi_\pm)]+\text{c.c.},\\ \mathbf{l}_\pm=2^{-1/2}(\mathbf{l}_x\pm i\mathbf{l}_y), \quad \Phi_\pm=\Phi\mp\Omega=\mp\frac{\omega^2 g_0 z}{2kc^2}+A'\int_0^z I\,dz+\Phi(0).\end{gathered} \tag{18}$$

Here $\Phi(0)$ is the value of the phase at the point $z = 0$ at the entrance to the resonant gas, and the solutions of Eqs. (16) and (17) are used in the expression for $\Phi_\pm$.

We denote by $\delta\varphi(z_0)$ the small deviation from $\varphi(z_0)$ resulting from an arbitrary perturbation at the point z_0. For linearly ($\varphi(z_0) = m\pi/2$) and circularly ($\varphi(z_0) = (2m + 1)\pi/4$, $m = 0, \pm 1,\ldots$) polarized waves we obtain from (15) the course of the evolution of the perturbation along the wave propagation direction

$$\delta\varphi(z)=\delta\varphi(z_0)\exp\left(\pm 2B''\int_{z_0}^{z} I\,dz\right), \tag{19}$$

$$B''=\frac{(2\pi\omega)^2|d_{ba}|^4}{k(\hbar c)^3(\gamma_{ba}^{(1)})^2}N_{ab}(C_1-C_2)Z_2(\xi,\eta), \tag{20}$$

where the plus and minus signs pertain respectively to linear and circular polarizations. Therefore circular polarization is stable in a resonant gas if $B'' > 0$, and linear if $B'' < 0$, for arbitrary values of $\mathbf{g}_0$. According to (18), the polarization plane of a linearly polarized wave rotates when z is varied, just as in the absence of resonance levels. In the stable regime there is no polarization self-action in these waves, and their variation with intensity is given by

$$I(z)=\frac{1}{2}\left|\frac{\alpha}{\beta}\right|-\left\{\left(\frac{\alpha}{2\beta}\right)^2+\left[I^2(0)+\frac{\alpha}{\beta}I(0)\right]\exp(-\alpha z)\right\}^{1/2}.$$

where $\beta = 2A''$ for linear polarization and $\beta = 2(A'' + B'')$ circular polarization. This equation is valid for a gas population $N_{ab} < 0$ in the region $|\alpha| > 2\beta I(z)$. For $\alpha = 0$ Eq. (14) yields $I(z) = I(0)[1 + \beta I(0)z]^{-1}$.

If the wave incident on the point $z = 0$ at the entry to the resonant gas is not circular, we obtain from (15)

$$\operatorname{tg} 2\varphi(z)=\operatorname{tg} 2\varphi(0)\exp\left(2B''\int_0^z I\,dz\right), \tag{21}$$

$$(2m-1)\pi/4<\varphi(z)<(2m+1)\pi/4, \quad m=0, \pm 1,\ldots. \tag{22}$$

On the other hand, for an incident wave that is not plane-polarized, we get in place of (21) and (22)

$$\operatorname{ctg} 2\varphi(z)=\operatorname{ctg} 2\varphi(0)\exp\left(-2B''\int_0^z I\,dz\right), \tag{23}$$

$$m\pi/2<\varphi(z)<(m+1)\pi/2, \quad m=0, \pm 1,\ldots. \tag{24}$$

It can be seen that the polarization ellipse initially elongated along a certain axis approaches continuously a circle if $B'' > 0$, whereas for $B'' < 0$ it becomes even more elongated. This deformation of the polarization ellipse is much faster on a homogeneously broadened ($1 \ll \eta^2$) transition than on an inhomogeneously broadened ($\eta \ll 1$) one, since $Z_2(\xi,\eta)$ takes on in (20) essentially different values in these cases. For example, at resonance $\xi = 0$ we have

$$Z_2(0,\eta)=1 \text{ for } 1\ll\eta^2, \quad Z_2(0,\eta)=\pi^{1/2}\eta/2 \text{ for } \eta\ll 1.$$

In addition, relations (21)–(24) show that in the case of elliptic polarization the value of $\varphi(z)$ varies monotonically and does not go outside one of the sectors

$$\begin{gathered}m\pi/2<\varphi(z)<(2m+1)\pi/4,\\ (2m-1)\pi/4<\varphi(z)<m\pi/2, \quad m=0, \pm 1,\ldots,\end{gathered}$$

which is specified at the entry into the gas. The direction of rotation of the vector $\mathbf{E}$ remains therefore unchanged as the wave (1) propagates, i.e., the sign of the difference $I_+ - I_-$ is constant.

A measure the polarization-ellipse deformation per unit length of the optical path is the derivative of the ratio of the lengths of the minor and major axes:

$$\frac{d}{dz}|\operatorname{tg}\varphi|=B''|I_+-I_-|e^2, \tag{25}$$

where $e = 2(I_+I_-)^{1/4}(I_+^{1/2} + I_-^{1/2})^{-1}$ is the eccentricity of the polarization ellipse. If the major and minor semiaxes are respectively $|\sin\varphi|$ and $|\cos\varphi|$, the substitution $\tan\varphi \to \cot\varphi$ is made in (25). The sign of B'' in (25) governs the direction of the polarization-ellipse deformation, towards a circle ($e\to 0$) or towards a segment ($e \to 1$), in full accord with the condition (19) for the stability of the circular and linear polarizations. Since the derivative (25) does not reverse sign following specular reflection, the direction of the polarization-ellipse deformation is not changed by specular reflection, i.e., a cumulative effect takes place. Multiple specular reflection, in view of the cumulation, causes the elliptic polarization to turn continously into circular or linear, which are stable in a gaseous resonant medium.

Relation (25) can also be expressed in terms of the dichroism vector $\mathbf{h}$:

$$\frac{d}{dz}\operatorname{tg}\varphi=\frac{\omega^2 e^2(\mathbf{kh})}{2k^2c^2}, \quad \mathbf{h}=\mathbf{k}\frac{2c^2B''(I_+-I_-)}{\omega^2}.$$

The polarization-ellipse rotation angle $d\Omega/dz$ per unit length is expressed in terms of the summary gyration vector $g = g_0 + f$ by the simple equation

$$\frac{d}{dz}\Omega=\frac{\omega^2(\mathbf{kg})}{2k^2c^2}, \tag{26}$$

where the nonlinear gyration vector $\mathbf{f}$ is given by

$$\mathbf{f}=\mathbf{k}\frac{8\pi^2|d_{ba}|^4}{kc\hbar^3(\gamma_{ba}^{(1)})^2}N_{ab}Z_1(\xi,\eta)(C_2-C_1)(I_+-I_-). \tag{27}$$

The total rotation angle $\Omega(z) - \Omega(0)$ takes the form

$$\Omega(z)-\Omega(0)=g_0 z+\frac{(2\pi\omega)^2|d_{ba}|^4}{k(\hbar c)^3(\gamma_{ba}^{(1)})^2}N_{ab}Z_1(\xi,\eta)$$

$$(C_2-C_1)\int_0^z (I_+-I_-)\,dz. \qquad (28)$$

In the absence of resonance levels we have $d_{ba}=0$ and Eqs. (26)–(28) describe the known rotation of the polarization ellipse in an active medium without deformation of its shape. The presence of resonance levels leads to polarization self-action that alters both the rotation angle (28) and the shape of the polarization ellipse. Reversal of the sign of the nonlinear rotation angle $\Omega(z)-\Omega(0)-g_0 z$ with change of the sign of Δ or of I_+-I_- agrees with experiment.[7] Equation (28) differs from that obtained in Ref. 10 for the particular case of a homogeneously broadened transition $1\ll\eta^2$ and $g_0=0$, in view of computation errors in this reference.

In contrast to g_0, the nonlinear gyration vector (27) has opposite directions for positive and negative populations N_{ab} and also for right-hand ($I_+>I_-$) and left-hand ($I_+<I_-$) rotations of the vector **E**. It depends strongly on the type of the resonant transition $J_a\to J_b$, on the depolarizing atomic collision, and on the ratio $2\gamma_{ba}^{(1)}/ku$ of the homogeneous to inhomogeneous widths. In addition, the vector (27), as a function of Δ, reverses sign when the the detuning from resonance passes through zero. All this makes it possible in experiment to separate the contributions of the optical activity from the natural optical activity.

In a gas with natural optical activity we have $\mathbf{f}=\mathbf{h}=0$ and the rotation direction of the polarization ellipse is not changed by specular reflection. On passing forth and back along the same path in such a gas, the polarization ellipse returns to the same position. In a gas with nonlinear optical activity $\mathbf{g}=\mathbf{f}$, $\mathbf{h}\neq 0$ and $\mathbf{g}_0=0$, however, specular reflection transforms right-hand rotation of the ellipse into left-hand and vice versa, i.e., cumulation is produced in the nonlinear rotation angle upon specular reflection. In the general case of a gas with both natural and nonlinear optical activities, $\mathbf{g}=\mathbf{g}_0+\mathbf{f}$, the contributions from $\mathbf{g}_0$ cancel out after specular reflection and forth and back passage along the same path. After returning to the initial point, the polarization-ellipse axes are rotated only by the polarization self-action connected with **f**. In the case of multiple specular reflection, the nonlinear rotation angle is increased by cumulation and tens to its limiting value.

3. DISCUSSION

The peculiarities of the polarization self-action depend essentially on the sign of the difference C_2-C_1, which depends on the type of the resonance transition $J_a\to J_b$ and on the values of $\gamma_a^{(1)}(\gamma_b^{(1)})$ and $\gamma_a^{(2)}(\gamma_b^{(2)})$. For the simplest atomic transitions we have

$$C_2-C_1=-\frac{\gamma_a^{(1)}+\gamma_b^{(1)}}{9\gamma_a^{(1)}\gamma_b^{(1)}}\left(1-\frac{\gamma}{3(\gamma_a^{(1)}+\gamma_b^{(1)})}\right)$$

$$\text{for}\quad J_a={}^1/_2\to J_b={}^1/_2, \qquad (29)$$

$$C_2=C_1=(\gamma_b^{(1)}-\gamma_b^{(2)})/9\gamma_b^{(1)}\gamma_b^{(2)}\quad\text{for}\quad J_a=0\to J_b=1, \qquad (30)$$

$$C_2-C_1=(\gamma_a^{(1)}-\gamma_a^{(2)})/9\gamma_a^{(1)}\gamma_a^{(2)}\quad\text{for}\quad J_a=1\to J_b=0, \qquad (31)$$

$$C_2-C_1=\frac{1}{36}\left[\frac{\gamma_a^{(1)}-\gamma_a^{(2)}}{\gamma_a^{(1)}\gamma_a^{(2)}}+\frac{\gamma_b^{(1)}-\gamma_b^{(2)}}{\gamma_b^{(1)}\gamma_b^{(2)}}+\frac{\gamma}{2}\left(\frac{1}{\gamma_a^{(2)}\gamma_b^{(2)}}-\frac{1}{\gamma_a^{(1)}\gamma_b^{(1)}}\right)\right],\ \text{for}$$

$$J_a=1\to J_b=1. \qquad (32)$$

To be specific, we put first $\mathbf{g}_0=0$ and $N_{ab}>0$. Since the derivative $dZ_1(\xi,\eta)/d\xi$ is positive for $\xi=\Delta=0$ is positive, the signs of $Z_1(\xi,\eta)$ and Δ are the same near resonance. Consequently, in the case (29) at $\Delta>0$ polarization-ellipse axes rotate to the left if $I_+>I_-$ and to the right if $I_+<I_-$, and since (20) is positive the stable polarization is circular. The sign of the difference C_2-C_1 in (30)–(32) is the same as for $\gamma_a^{(1)}-\gamma_a^{(2)}$ and $\gamma_b^{(1)}-\gamma_b^{(2)}$. Theoretical calculations[15] for the atomic transitions $0\rightleftarrows 1$ and $1\to 1$ yield $\gamma_a^{(1)}>\gamma_a^{(2)}$ and $\gamma_b^{(1)}>\gamma_b^{(2)}$, which corresponds for $\Delta>0$ to right-hand rotation of the polarization ellipse axes at $I_+>I_-$ and left-hand if $I_+<I_-$, the stable polarization being the linear one. We emphasize in this connection that in the absence of depolarizing collisions the atomic transitions $0\rightleftarrows 1$ and $1\to 1$ are polarizationwise neutral, since the axial vectors (11) vanish. An experimental confirmation of the obtained polarization rules for the transitions $0\rightleftarrows 1$ and $1\to 1$ verifies the validity of the model of depolarizing collisions.

In the case of $J_a=J\to J_b=J$ transition (Q mode) and $J>1$ we have $C_2-C_1>0$, so that the left-hand polarization is the stable one, and the polarization-ellipse axes are rotated to the right in this transition if $\Delta>0$ and $I_+>I_-$. For the transitions $J_a=J\to J_b=J+1$ (R-mode) and $J_a=J+1\to J_b=J$ (P-mode) and for $J>0$ the opposite inequality $C_2-C_1<0$ holds. The result is left-hand rotation of the ellipse axes under the previous conditions $\Delta>0$ and $I_+>I_-$, and it is the circular polarization which is stable. In all cases, the rotation of the polarization-ellipse axes reverses direction if the changes $\Delta\to-\Delta$ or $I_+>I_-\to I_+<I_-$ is made. These rules permit experimental identification of the type of the resonant transition $J_a\to J_b$. It must be remembered here that the very simple atomic transitions (29)–(32) are different from all others with respect to the laws of polarization of the propagating wave (1).

The examples presented pertain to an absorbing gaseous medium with $N_{ab}>0$. In a gas with inverted population $N_{ab}<0$ the rotation direction of the polarization-ellipse axes is reversed, and the linear and circular polarizations in the stable regime change places.

If $\mathbf{g}_0=0$, the rotation direction of the polarization ellipse axes is reversed for any transition $J_a\to J_b$ if the frequency ω passes through the resonance $\Delta=0$ in the course of scanning. Since ω and ω_0 are known in this case, the equality $\Delta=\omega-\omega_0-\Delta_{ba}^{(1)}=0$ permits an experimental determination of the collision shift $\Delta_{ba}^{(1)}$ of the resonance levels. By varying the pressure p of the buffer gas at fixed temperature, one can also obtain the derivative $d\Delta_{ba}^{(1)}/dp$.

In the case $\mathbf{g}_0\neq 0$, the value of $\mathbf{g}_0$ is determined experimentally from the rotation of the polarization plane of a linearly polarized waves. This makes it possible to determine experimentally, for propagation of an elliptically polarized wave, the nonlinear rotation angle $\Omega(z)-\Omega(0)$. The latter can yield the same information and by the same methods as in the case $\mathbf{g}_0=0$.

The stationary regime considered above sets in after a rather long time interval $1 \ll \gamma_a^{(\varkappa)} t$ and $1 \ll \gamma_b^{(\varkappa)} t$ for $\varkappa = 0, 1, 2$. This should be kept in mind in the case of a highly rarefied gas.

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