EFFECT OF ATOMIC COLLISIONS ON POLARIZATION OF LASER RADIATION

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The effect of van der Waals, short-range, and resonance atom-atom interactions on the polarization of a monochromatic wave passing through an active gaseous medium and also on the polarization of the radiation from a laser with an isotropic resonator is investigated. The rotation and shape variation of the polarization ellipse that occur during passage of the wave are expressed as functions of the parameters of the medium. It is shown that, in accordance with experiment, the polarization of the laser radiation is circular in the case of atomic transitions involving changes in the total momentum $1 \rightleftarrows 0$ and $1 \rightarrow 1$.

THE polarization of the radiation of a gas laser with an isotropic resonator in the absence of an external magnetic field has been measured for various atomic transitions in a number of experiments.[1-4] 1) Satisfactory agreement between theory and experiment has been established[4,5] for atomic transitions with changes in total momentum

$$J+1 \rightleftarrows J(J>0), \qquad J \rightarrow J(J>1). \tag{1}$$

In the case of atomic transitions $1 \rightleftarrows 0$ and $1 \rightarrow 1$, the theory proposed in[4,5] did not give a preference to any specific polarization. On the other hand, circular polarization was obtained in an experiment with helium-neon and helium-xenon lasers with an isotropic resonator (atomic transition $1 \rightarrow 0$). It was indicated in[6] that the source of circular polarization is the differing relaxation rates of the quadrupole and magnetic momenta of the excited level. However, a concrete calculation of the relaxation time was not performed.

The present paper is devoted to a description of the mechanism of atomic relaxation and to a concrete calculation of the necessary parameters, as well as to an establishment of the connection between the nature of the atomic relaxation and the character of the polarization of the electromagnetic wave. Since the polarization of an electromagnetic wave is characterized by a vector, it is necessary to describe the state of the active atoms by a density matrix that includes level degeneracy. This means that degeneracy must be taken into account also in the relaxation collision term of the density matrix equation. Different atomic collisions lead to a different relaxation of the individual elements of the density matrix, which is reflected in the interaction of the wave with the active medium and in the polarization.

In[7-9], where the Lamb theory of the gas laser is developed, as well as in[10], the collision term is introduced without taking account of level degeneracy. Hence the attempt to use the Lamb theory to explain the polarization of radiation from the atomic transitions $1 \rightleftarrows 0$ and $1 \rightarrow 1$ in an isotropic resonator[4,5] did not yield the desired result.

Below, the collision term is obtained on the basis of the quantum-mechanical theory of atomic collisions, with level degeneracy taken into account. In doing this, the van der Waals and short-range interactions of the active atoms with each other and with impurity atoms are included. In addition, an important role can be played by the resonant (dipole-dipole) interaction between identical atoms, one of which with total momentum J = 1 is in the excited, and the other, in the ground state.[11-15] These interactions lead to a rather cumbersome collision term, which includes a characteristic component with a transposed density matrix. The latter makes the controlling contribution to the establishment the particular polarization of the electromagnetic wave resonant with the atomic transitions $1 \rightleftarrows 0$ and $1 \rightarrow 1$. In the absence of degeneracy, the component with the transposed density matrix disappears.

The part of the collision term due to resonant collisions is calculated completely and represented in a form convenient for application. The tensor and matrix structure of the collision term associated with the van der Waals and short-range interactions, as well as the signs of two of its components, are established on the basis of the general theorem on the unitarity of the S-matrix. In order to determine the sign of the coefficient G_b (20) irrespective of the nature of the atomic collisions, we examine the different regions where perturbation theory of the adiabatic approximation is valid. The result is confirmed by an exact calculation in the case of van der Waals collision of neon with helium.

Introduction of this collision term into the usual laser equations[9,10] leads to circular polarization for all three atomic transitions $1 \rightarrow 0$, $0 \rightarrow 1$, and $1 \rightarrow 1$, if the van der Waals and short-range interactions overwhelm the resonance. In the opposite situation, the character of the polarization of the laser radiation can vary as a function of the pressure and temperature of the gas. Thus, an investigation of the polarization effects of laser radiation assists the establishment of the nature of the atomic collisions.

1. POLARIZATION OF THE WAVE IN AN ACTIVE MEDIUM

First we consider the passage of the monochromatic wave

1) There are references to previous work in the cited papers.

through an active medium in which the polarization current

$$\mathbf{j} = (c/4\pi)(a\mathbf{A} + b\mathbf{A}^2\mathbf{A}^*) \tag{4}$$

is induced. Here the coefficients a and b depend on field and in an isotropic medium are defined uniquely by the expansion (4). In the first nonzero approximation of perturbation theory, the coefficient b is independent of field. The coefficient a, on the other hand, in the same approximation contains a component that is proportional to $|\mathbf{A}|^2$.

The function $\mathbf{R}$, which varies slowly compared to $\exp(i\omega z/c)$, is the solution of the equation

$$i\frac{2}{\lambda}\frac{d\mathbf{R}}{dz} + a\mathbf{R} + b\mathbf{R}^2\mathbf{R}^* = 0, \tag{5}$$

which is obtained from d'Alambert's equation after neglecting the second derivative of the slow function $\mathbf{R}$ with respect to z. As the wave propagates, the orthogonal unit vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ can rotate about the wave vector $\mathbf{k}$. Clockwise rotation with respect to the direction of $\mathbf{k}$ is symbolized by $\Omega(z)$. From (5) it is easy to obtain

$$\Phi(z) = \lambda \int_0 (a' + b'|\mathbf{R}|^2)\,dz + \Phi(0), \tag{6}$$

$$\Omega(z) = (\lambda/2)\sin[2\varphi(z)]\int_0^z b'|\mathbf{R}|^2 dz + \Omega(0), \tag{7}$$

where $\lambda = c/\omega$, and the angle $\varphi(z)$ is defined as follows:

$$\operatorname{tg} 2\varphi(z) = \operatorname{tg}[2\varphi(0)]\cdot\exp\left(\lambda\int_0^z b''|\mathbf{R}|^2 dz\right), \tag{8}$$

$$-\pi/4 \leqslant \varphi(z) \leqslant \pi/4.$$

Here $\Phi(0)$, $\Omega(0)$, and $\varphi(0)$ are the values of the corresponding functions at the point z = 0 on the boundary of the medium.

The function $|\mathbf{R}|^2$ is found from the simultaneous solution of the equations

$$\frac{1}{\lambda}\frac{d|\mathbf{R}|^2}{dz} + \left[a'' + \frac{1}{2}b''(1+\cos 4\varphi)|\mathbf{R}|^2\right]|\mathbf{R}|^2 = 0,$$

$$\frac{1}{\lambda}\frac{d(\cos 4\varphi)}{dz} + b''|\mathbf{R}|^2(1-\cos^2 4\varphi) = 0$$

with given boundary conditions. The single and double primes represent the real and imaginary parts of the coefficients, respectively. By means of (8), the last two equations may be brought together into one integro-differential equation for the function $|\mathbf{R}|^2$; this, however, is less convenient.

According to (8), linear polarization is stable in the case $b'' < 0$ and circular polarization is stable for $b'' > 0$. If an elliptically polarized wave is initially incident on the active medium, then as it propagates the axes of the polarization ellipse are rotated through the angle $\Omega(z)$, and the ellipse itself becomes a circle when $b'' > 0$ and flattens out when $b'' < 0$. The direction of rotation of the axes as a function of z depends on the sign of the real part of b, which is proportional to the difference $\omega - \omega_0$, where ω_0 is the frequency of the atomic transition with collisional level shift taken into account. Hence, as the field frequency changes, the direction of rotation reverses when ω passes through resonance. Observation of this rotation permits determination of the collisional shift.[2)]

[2)] An experimental investigation of this rotation in a non-absorbing liquid with induced anisotropy has been carried out in [16,17].

The rotation of the polarization ellipse in a resonant medium is different from the usual optical activity and the Faraday effect. For example, in the medium considered, linear polarization is stable when $b'' < 0$, and the plane of polarization of the wave is not rotated. If $b'' > 0$, the linearly polarized wave is unstable, and as it propagates it is transformed into elliptically polarized light with a rotating polarization ellipse. The elliptically polarized wave, in turn, becomes circular, which is then stable.

Inside an operating laser the picture is complicated by the fact that besides the forward wave

$$\mathbf{A}_+ = \mathbf{R}_+ \exp[i(kz - \omega t)]$$

there is a backward wave

$$\mathbf{A}_- = \mathbf{R}_- \exp[-i(kz + \omega t)].$$

Each of the functions $\mathbf{A}_+$ and $\mathbf{A}_-$ satisfies the D'alambert equation with its own polarization current, $\mathbf{j}_+$ and $\mathbf{j}_-$, respectively; for example,

$$\mathbf{j}_+ = (c/4\pi)[a_+\mathbf{A}_+ + b_+\mathbf{A}_+^2\mathbf{A}_+^* + c_+(\mathbf{A}_+\mathbf{A}_-^*)\mathbf{A}_- + d_+(\mathbf{A}_+\mathbf{A}_-)\mathbf{A}_-^*], \tag{9}$$

where the field-dependent coefficients have different values for the forward and backward waves. The current $\mathbf{j}_-$ is the same form as (9) with the replacement $+ \to -$ and $- \to +$.

In order to find the frequency, polarization, and intensity of the field in the steady state, we need to impose boundary conditions on the mirror reflections. If the latter are anisotropic, then the solution of the problem depends on the structure of the specific resonator. Hence, in what follows we shall limit ourselves to an isotropic resonator with plane-parallel mirrors. In this case the polarization is completely determined by the physical processes taking place inside the laser. The boundary conditions of an isotropic resonator are most simple:

$$\mathbf{R}_+(0) = \sqrt{r_1}\mathbf{R}_-(0)e^{i\Phi_1}, \qquad \mathbf{R}_-(L) = \sqrt{r_2}\mathbf{R}_+(L)e^{i\Phi_2}, \tag{10}$$

where r_1, r_2, Φ_1, and Φ_2 are the reflection coefficients and phase losses at the boundaries of a resonator of length L. The amplitude of the forward and backward waves has the form (3) with indexes + and −, respectively.

For simplicity, we shall set the frequency ω of the standing wave equal to the frequency ω_0 of the atomic transition. As will be seen below, all the coefficients of the polarization current (9) become purely imaginary and identical for the forward and backward waves:

$$a_+ = a_- \equiv ia'', \tag{11}$$

$$b_+ = c_+ = d_+ = b_- = c_- = d_- \equiv ib'', \tag{12}$$

where a'' and b'' in the first nonzero approximation are the same as in the case of a single traveling wave (2). The assumption we have made $\omega = \omega_0$ is not basic, since the imaginary parts of the coefficients of (9) are smooth functions of $\omega - \omega_0$ with constant sign.

As is seen from (6) and (7) rotation of the axes of the polarization ellipse is absent, and the phases of the forward (Φ_+) and backward (Φ_-) waves are constant. The polarization of the waves is described by the equations

$$\frac{2}{\lambda}\frac{d\varphi_+}{dz} = b''\sin 2\varphi_+(|\mathbf{R}_+|^2\cos 2\varphi_+ + |\mathbf{R}_-|^2\cos 2\varphi_-), \tag{13}$$

$$-\frac{2}{\lambda}\frac{d\varphi_-}{dz} = b''\sin 2\varphi_-(|\mathbf{R}_-|^2\cos 2\varphi_- + |\mathbf{R}_+|^2\cos 2\varphi_+), \quad (14)$$
$$-\pi/4 \leqslant \varphi_+ \leqslant \pi/4, \qquad -\pi/4 \leqslant \varphi_- \leqslant \pi/4.$$

The boundary conditions (10) and the solution to Eqs. (13) and (14) are consistent only in two cases

$$\varphi_+(z) \equiv \varphi_-(L - z) \equiv 0, \qquad \varphi_+(z) \equiv \varphi_-(L - z) \equiv \pm\pi/4.$$

Consequently, at resonance $\omega = \omega_0$ steady state is established with linear or circular polarization. Investigation of the solution of Eqs. (13) and (14) for stability shows that when $b'' < 0$ linear, and when $b'' > 0$ circular, polarization is stable. When $\omega \neq \omega_0$, the situation is more complex, but the conclusion is the same.

2. EQUATIONS FOR A LASER ON THE ATOMIC TRANSITION 1 → 0

The polarization current (9)

$$\mathbf{j}_+ + \mathbf{j}_- \equiv \int d\mathbf{v}(\mathbf{I}_+ + \mathbf{I}_-), \qquad \mathbf{I}_+ + \mathbf{I}_- = \mathbf{I}$$

will be sought for single-mode operation, in which the vector potential $\mathbf{A} = \mathbf{A}_+ + \mathbf{A}_-$ and the current $\mathbf{I}$ are proportional to the factor

$$(e^{ikz} + e^{-ikz})e^{-i\omega t}.$$

The basic equations for the laser may be written in the form[3)]

$$\left(i\frac{\partial}{\partial t} + i\mathbf{v}\nabla - \omega_0 + i\frac{\Gamma}{2}\right) I_\alpha = \frac{3}{4}\gamma c \lambda (\rho_{\alpha\beta} - \rho_1\delta_{\alpha\beta}) A_\beta, \quad (15)$$

$$\frac{\partial}{\partial t}\rho_{\alpha\beta} = \frac{1}{3}W_2 n f \delta_{\alpha\beta} - \gamma_2\rho_{\alpha\beta} - \delta S_{\alpha\beta} - \delta_b S^b_{\alpha\beta} - i\frac{1}{c}(I_\alpha A_\beta^* - A_\alpha I_\beta^*), \quad (16)$$

$$\frac{\partial}{\partial t}\rho_1 = W_1 n f - \gamma_1\rho_1 + \gamma\rho_{\sigma\sigma} + i\frac{1}{c}(I_\sigma A_\sigma^* - A_\sigma I_\sigma^*). \quad (17)$$

The quantities I_α and $\rho_{\alpha\beta}$ appearing in these equations are connected with the density matrix $R_{\mu 0}$ which describes the transitions between levels and the density matrix in the upper degenerate level $\rho_{\mu\mu'}$ in the following way:

$$I_\alpha = -i\omega_0 d^\alpha_{0\mu} R_{\mu 0}, \quad \rho_{\alpha\beta} = 3 d^\alpha_{0\mu}\rho_{\mu\mu'} d^\beta_{\mu' 0}/|d^1_0|^2, \quad (18)$$

where $d^\alpha_{\mu 0}$ and d^1_0 are the dipole and reduced dipole moments of the transition. The transformation (18) facilitates the transition from the matrix to the tensor representation, which is more convenient.

The other quantities appearing in (15)—(17) have the following significance: W_1 and W_2 are the probabilities of excitation per unit time of the lower and upper levels as a result of pumping, n is the density of active atoms in the ground state, γ is the probability of a radiative transition between the upper and lower levels, connected with the reduced dipole moment by the relation $\gamma = 4|d^1_0|^2/9\lambda^3$. The term $i\mathbf{v}\nabla$ takes account of the Doppler change in frequency due to motion of an atom with velocity v. The quantity Γ is

$$\Gamma = \gamma_1 + \gamma_2 + \delta + \delta_b,$$

where γ_1 and γ_2 are the widths of the lower and upper levels due to both radiative transitions and gas-kinetic inelastic collisions, δ is the broadening of the upper level due to resonant collisions, and δ_b is the broadening of the lower level due to van der Waals and short-range forces.

The quantities $\delta S_{\alpha\beta}$ and $\delta_b S^b_{\alpha\beta}$ represent collision integrals. The expressions for them take into account the collision of excited atoms only with unexcited ones, since the concentration of excited atoms is low. We also neglect the change of velocity of the atoms upon collision. This change leads to a relaxation of the atomic distribution in velocity space, which, evidently, significantly influences the shape of the Lamb dip.[10] However, in the polarization problem, the deciding circumstance is the distribution of the atoms over the sublevels, i.e., the tensor structure of the density matrix. The change in the tensor structure of the density matrix as a result of collisions is determined by the atomic transitions between sublevels, for the calculation of which small changes in velocity due to collision are not important. With these assumptions the collision integrals may be written in the following general form:

$$\delta S_{\alpha\beta} = \delta\left[P\rho_{\sigma\sigma}\delta_{\alpha\beta} + Q\rho_{\alpha\beta} + G\rho_{\beta\alpha} + f\int d\mathbf{v}(p\rho_{\sigma\sigma}\delta_{\alpha\beta} + q\rho_{\alpha\beta} + g\rho_{\beta\alpha})\right], \quad (19)$$

$$\delta_b S^b_{\alpha\beta} = \delta_b(P_b\rho_{\sigma\sigma}\delta_{\alpha\beta} + Q_b\rho_{\alpha\beta} + G_b\rho_{\beta\alpha}). \quad (20)$$

Here f is a function of the Maxwell distribution of active atoms in the ground state. The dependence on density and temperature is contained in the parameters δ and δ_b, and the coefficients P, P_b, Q, etc., are dimensionless numerical factors.

For resonant neon-neon collisions the parameter

$$\delta = 10.97 n \lambda_{02}^3 \gamma_{02} \quad (21)$$

contains the probability γ_{02} of spontaneous emission with transition of a neon atom from an excited state with momentum J = 1 to the ground state, ω_{02} the frequency of this atomic transition, and $\lambda_{02} = c/\omega_{02}$. The coefficients of the resonant collision term were calculated on an electronic computer and are

$$P = -0.064, \quad Q = 0.660, \quad G = 0.013,$$
$$p = -0.151, \quad q = 0.051, \quad g = -0.080. \quad (22)$$

The operator $V_{\alpha\beta}$ of the energy of the van der Waals interaction of excited neon (energy E_{a_1} and total momentum J = 1) with helium in the ground state with energy E_{b_0} has the form

$$V_{\alpha\beta} = \sum_{n,n'}\frac{|d^{n'1}_{b0}|^2}{9r^8}\left[\frac{|d^{n0}_{a1}|^2(r^2\delta_{\alpha\beta} + 3r_\alpha r_\beta)}{E_{a1} + E_{b0} - E_{n0} - E_{n'1}} \times \frac{1}{3}\frac{|d^{n1}_{a1}|^2(5r^2\delta_{\alpha\beta} - 3r_\alpha r_\beta)}{E_{a1} + E_{b0} - E_{n1} - E_{n'1}} + \frac{1}{10}\frac{|d^{n2}_{a1}|^2(19r^2\delta_{\alpha\beta} + 3r_\alpha r_\beta)}{E_{a1} + E_{b0} - E_{n2} - E_{n'1}}\right]$$

where E_{nJ} and $d^{nJ}_{a_1}$ are the energy and reduced dipole moment of neon, with the index n′ marking the analogous quantities for helium, and r is the distance between the neon and helium. The summation is carried over all intermediate states of neon and helium, including the continuous spectrum, with the term with zero denominator left out. If among the atomic transitions of neon $E_{nJ} - E_{a_1}$ and helium $E_{n'1} - E_{b_0}$ there are any which are quasi-resonant, then in the first nonzero approximation in $V_{\alpha\beta}$ it is possible to retain only one quasi-resonant term. Then in the case of van der Waals neon-helium collisions we have

[3)] Repeated vector and tensor subscripts imply summation everywhere. The system of units in which $\hbar = 1$ and c is the velocity of light in vacuum is used.

$$\delta_b = 3.99 n_0 \left(\frac{2T}{M_0}\right)^{3/10} \left[\frac{9}{16} \frac{\gamma_{01}\gamma_{02}}{|\omega_{01}-\omega_{02}|} \left(\frac{c^2}{\omega_{01}\omega_{02}}\right)^3\right]^{2/5}, \quad (23)$$

$$P_b = -0.406, \quad Q_b = 1.318, \quad G_b = -0.099. \quad (24)$$

Here γ_{01} is the probability of spontaneous emission of helium which is found in a state with atomic transition frequency ω_{01} closest to ω_{02}; T is the temperature; n_0 and M_0 are the density and mass of the helium.

The coefficients of the collision term (20) were not calculated for the general case of van der Waals and short-range interactions. However, they satisfy certain general relations coming out of the conservation of number of particles and Boltzmann's H-theorem:

$$3P_b + Q_b + G_b = 0,$$
$$\delta_b > 0, \quad P_b < 0, \quad Q_b > 0, \quad |G_b| < Q_b. \quad (25)$$

3. POLARIZATION OF THE RADIATION OF A LASER ON THE ATOMIC TRANSITION 1 → 0

We solve Eqs. (15)–(17) by the method of successive approximations, expanding in series with respect to the parameter

$$c|A|^2/\hbar\omega\Gamma \ll 1. \quad (26)$$

In the first nonzero approximation for the traveling monochromatic wave (2) we obtain the following expression for the coefficient b of the polarization current (4):

$$b = \frac{9\pi}{4} n \lambda^2 \gamma^2 \left[\left(\frac{1}{3\gamma} - \frac{1}{\gamma_1}\right)\frac{\gamma W_2}{\gamma_2} - \frac{W_1}{\gamma_1}\right] \left\{ \frac{(\delta G + \delta_b G_b)(F_1 - F_2)}{(\delta Q + \delta_b Q_b + \gamma_2)^2 - (\delta G + \delta_b G_b)^2} \right.$$
$$\left. \times \frac{[\delta(G+g) + \delta_b G_b] F_2}{[\delta(Q+q) + \delta_b Q_b + \gamma_2]^2 - [\delta(G+g) + \delta_b G_b]^2} \right\}, \quad (27)$$

$$F_1 = \int \frac{\Gamma f(v)\, dv}{(\Delta - kv + i\Gamma/2)[(\Delta - kv)^2 + \Gamma^2/4]},$$
$$F_2 = \int \frac{f(v) dv}{\Delta - kv + i\Gamma/2} \int \frac{\Gamma f(v')\, dv'}{(\Delta - kv')^2 + \Gamma^2/4},$$
$$f(v) = (1/\sqrt{\pi} u) \exp(-v^2/u^2),$$
$$\Delta = \omega - \omega_0, \quad u^2 = 2T/M,$$

where M is the mass of an active atom.

We see that the real part of the coefficient b is proportional to $\omega - \omega_0$, while its imaginary part is of constant sign in the resonance region $\omega - \omega_0$. The coefficients of (9) have analogous properties.

In order to determine the polarization of the laser radiation, it is sufficient to find the coefficients of (9) at resonance. In the considered approximation (26) we obtain the equalities (11) and (12), in which b'' is the imaginary part of the coefficient (27) for $\omega = \omega_0$. To finally establish the sign of b'' we clarify the signs of the separate parts of (27). The expression in the extreme left-hand brackets of (27) is positive, which is a condition for laser action. The denominators in the curly brackets of (27) are also positive, which follows from (22) and (25).

In an experiment[4] on a helium-neon laser with wavelength $\lambda = 1.523\ \mu$, the parameters appearing in (21) and (23) had the following values:

$$\hbar\omega_{01} = 19.82\ \text{eV}, \ \hbar\omega_{02} = 19.69\ \text{eV},$$
$$n = 7.1 \cdot 10^{15}\ \text{cm}^{-3}, \ ku \approx 5 \cdot 10^8\ \text{sec}^{-1}, \ \gamma_{01}/\gamma_{02} \approx 160.$$

For these values we obtain $\delta/\delta_b \approx 1/30$. The smallness of this ratio follows also from a comparison of the theoretical value of δ with the experimental value of $\delta + \delta_b$ obtained from the dependence of the Lamb dip on gas density.

Thus, under the experimental conditions of[4] resonant collisions were unimportant. In this case Eq. (27) simplifies:

$$b = \frac{9\pi}{4} n \lambda^2 \gamma^2 \left[\left(\frac{1}{3\gamma} - \frac{1}{\gamma_1}\right)\frac{\gamma W_2}{\gamma_2} - \frac{W_1}{\gamma_1}\right] \frac{G_b F_1/\delta_b}{(Q_b + \gamma_2/\delta_b)^2 - G_b^2}. \quad (28)$$

From Eq. (28) it follows that the sign of the imaginary part of b is determined by the sign of G_b.

Experimental results show[1-4] that $G_b < 0$. The following three arguments support this assignment.

1. The coefficient G_b determines the difference in the rate of relaxation of symmetric and antisymmetric parts of the density matrix $\rho_{\alpha\beta}$. It is a natural assumption that the nondiagonal elements of the density matrix do not decay more slowly than the diagonal elements, which means that $G_b < 0$.

2. The coefficient G_b is easily calculated independently of the form of the interaction between atoms in those regions of the range parameters of collisions where the perturbation theory or the adiabatic approximation is valid. In this way we obtain the very same result $G_b < 0$.

3. A negative sign for G_b is confirmed by direct calculations of (24) in the special case of quasi-resonant collisions of neon and helium carried out without the use of perturbation theory or the adiabatic approximation.

These arguments lead us to expect that negativity of G_b is a general property of non-resonant atom-atom collisions. Consequently for a gas laser on the atomic transition $J_2 = 1 \to J_1 = 0$, circular polarization ($b'' > 0$) is stable.

Another limiting case is of interest: when the van der Waals and short-range interactions are small in comparison with resonance. Setting $\delta_b = 0$ in (27), we find that there is a region of change of pressure and temperature where either circular ($b'' > 0$) or linear ($b'' < 0$) polarization is stable. We can indicate those values of the parameters for which $b'' = 0$ and there occurs a transition from one polarization to the other. These are found from the solution of the algebraic equation

$$\frac{ku}{\Gamma} = \sqrt{\pi}\left(1 - \frac{1}{\pi} - \frac{(1 + g/G)[(Q + \gamma_2/\delta)^2 - G^2]}{(Q + q + \gamma_2/\delta)^2 - (G + g)^2}\right), \quad (29)$$

where ku is the Doppler width of the level. In particular, for $\gamma_2/\delta \ll 1$, we obtain $ku/\Gamma = (6\pi - 1)/\pi^{1/2}$.

If the magnitude of ku/Γ is greater than the numerical value of (29), then $b'' < 0$. In the opposite case, we have $b'' > 0$.

4. LASERS ON THE ATOMIC TRANSITIONS 0 → 1 AND 1 → 1

In the case of a laser on the atomic transition $J_2 = 0 \to J_1 = 1$, the discussion is analogous, and the conclusion about the polarization of the electromagnetic wave is the same as before. The coefficient b in a weak field (26) is obtained from (27) by changing γ_1 to $9\gamma_1$ in the brackets on the extreme left.

The transition $J_2 = 1 \to J_1 = 1$ is noteworthy because the matrix structure of the collision term is important for both the upper and lower working levels. The collision term for the upper level is written

$$\delta_1 S_{\mu\mu'} + \delta_{1b} S^b_{\mu\mu'},$$

$$S_{\mu\mu'} = P\rho_{\mu''\mu''}\delta_{\mu\mu'} + Q\rho_{\mu\mu'} + GU^{\alpha\beta}_{\mu\mu''}\rho_{\mu''\mu'''}U^{\alpha\beta}_{\mu'''\mu'} \times f\int dv(p\rho_{\mu''\mu''}\delta_{\mu\mu'} + q\rho_{\mu\mu'} + gU^{\alpha\beta}_{\mu\mu''}\rho_{\mu''\mu'''}U^{\alpha\beta}_{\mu'''\mu'}), \quad (30)$$

$$S^b_{\mu\mu'} = P_b\rho_{\mu''\mu''}\delta_{\mu\mu'} + Q_b\rho_{\mu\mu'} + G_bU^{\alpha\beta}_{\mu\mu''}\rho_{\mu''\mu'''}U^{\alpha\beta}_{\mu'''\mu'}, \quad U^{\alpha\beta}_{\mu\mu'} = \delta_{\alpha\beta}\delta_{\mu\mu'} - (\hat{J}_\beta\hat{J}_\alpha)_{\mu\mu'}, \quad (31)$$

where $\hat{J}_\alpha$ is the total momentum operator, and all the coefficients have been defined above. The parameters δ_1 and δ_{1b} have the form (21) and (23) with account taken of the difference in the radiation widths of the levels.

The matrices $S_{mm'}$ and $S^b_{mm'}$ of the collision term in the lower level

$$\delta_2 S_{mm'} + \delta_{2b} S^b_{mm'}$$

differ from (30) and (31) by the index transformation $m \to \mu$, $m' \to \mu'$.

The desired coefficient b is obtained from (27) by the replacement in the brackets on the extreme left $\gamma_1 \to 3\gamma_1$, with simultaneous division of the entire expression by 4. In the curly brackets we must then set

$$\delta = \delta_i, \quad \delta_b = \delta_{ib}, \quad \Gamma = \gamma_1 + \gamma_2 + \delta_1 + \delta_2 + \delta_{1b} + \delta_{2b},$$

and the expression obtained is summed over i, with $i = 1, 2$.

Since b has the identical structure, the polarization of the radiation of lasers on all three atomic transitions

$$J_2 = 1 \to J_1 = 0, \quad J_2 = 0 \to J_1 = 1, \quad J_2 = 1 \to J_1 = 1$$

depends in the same way on the experimental conditions.

In conclusion, we remark that in the cited papers[4,5] the polarization of laser radiation on the transitions (1) was explained without taking the matrix structure of the collision term into account. Hence, application of the general formula of[5] to the atomic transitions $1 \rightleftarrows 0$ and $1 \to 1$ gave exactly zero, leaving the question of polarization unresolved. Taking level degeneracy into account in writing down the collision term adds to the general formula of[5] an additional term which, for typical values of the experimental parameters, coincides in order of magnitude with the principal term. This means that in the case of atomic transitions with large momenta (1), the polarization of the laser radiation can become sensitive to atomic collisions.

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[1] W. Culshaw and J. Kannelaud, Phys. Rev. **156**, 308 (1967).

[2] H. de Lang and G. Bouwhuis, Phys. Lett. **A25**, 406 (1967).

[3] H. de Lang, Physica **33**, 163 (1967).

[4] W. J. Tomlinson and R. L. Fork, Phys. Rev. **164**, 466 (1967).

[5] W. Van Haeringen, Phys. Rev. **158**, 256 (1967).

[6] D. Polder and W. Van Haeringen, Phys. Lett. **A25**, 337 (1967).

[7] W. E. Lamb, Jr., Phys. Rev. **134**, A1429 (1964).

[8] M. Sargent, W. E. Lamb, and R. L. Fork, Phys. Rev. **164**, 436, 450 (1967).

[9] B. L. Gyorffy, M. Borenstein, and W. E. Lamb, Phys. Rev. **169**, 340 (1968).

[10] S. G. Rautian, Zh. Eksp. Teor. Fiz. **51**, 1176 (1966) [Sov. Phys.-JETP **24**, 788 (1967)].

[11] A. A. Blasov and V. S. Fursov, Zh. Eksp. Teor. Fiz. **6**, 750 (1936).

[12] A. I. Vaĭnshteĭn and V. M. Galitskiĭ, Preprint IYaF, 1965.

[13] M. I. D'yakonov and V. I. Perel', Zh. Eksp. Teor. Fiz. **48**, 345 (1965) [Sov. Phys.-JETP **21**, 227 (1965)].

[14] A. P. Kazantsev, Zh. Eksp. Teor. Fiz. **51**, 1751 (1966) [Sov. Phys.-JETP **24**, 1183 (1967)].

[15] Yu. A. Vdovin and V. M. Galitskiĭ, Zh. Eksp. Teor. Fiz. **52**, 1345 (1967) [Sov. Phys.-JETP **25**, 894 (1967)].

[16] R. Terhune, P. Maker, and C. Savage, Phys. Rev. Lett. **12**, 507 (1964).

[17] P. McWane and D. Sealer, Appl. Phys. Lett. **8**, 278 (1966).

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