The interaction of arbitrarily polarized oppositely propagating waves in a resonant medium

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We study the interaction of two arbitrarily polarized oppositely propagating single-frequency plane waves in a resonant medium of randomly moving two-level systems. We find that in a medium of isotropically oriented molecules the oppositely directed waves always write a spatial lattice of the dielectric constant ε, on which they rescatter into each other. Collinearly polarized waves interact in such a way that the stronger wave has the greater local growth rate. On the other hand, as a result of interaction of orthogonally polarized waves the weaker wave has the greater local growth rate. Interaction alters the eccentricities and the orientation of the polarization ellipses of the oppositely propagating waves. We study the polarization states as functions of the intensities and polarization characteristics of the oppositely propagating waves, of the detuning δ of the field frequency from the resonant transition frequency, and of the medium parameters. When $\delta=0$ holds, the fields of the oppositely propagating waves are found to transform in such a way that far from the cross section under examination their polarizations tend to be orthogonal. When the detuning is finite, the axes of the polarization ellipse of each wave undergo an additional rotation in a direction determined by the sign of δ. When particle diffusion in the medium is at its maximum, the polarization state of each wave does not change and the equations for the intensities are transformed into the equations of radiation transfer.

1. INTRODUCTION

The principles of operation of lasers, bleachable filters reversing the wavefront at a mirror, and many other devices of quantum electronics are based on the interaction of two oppositely propagating waves of optical radiation with a resonant medium. Generally the fields of the waves can be polarized arbitrarily and their intensities can even be similar to the saturation field intensity. The effectiveness of the nonlinear interaction of waves in a resonant medium depends on the wave polarizations, as a result of which the polarizations of the oppositely propagating waves change.[1]

In describing the interaction of field and matter semiclassically it is customary to think of the resonant medium as a collection of two-level quantum systems whose vector $\tilde{\mathbf{d}}_{21}\equiv\mathbf{d}$ of the matrix elements of the signal-transition dipole-moment operator is collinear to the field.[2] Such a model medium enables using a single scalar quantity to describe the electric field. The model is believed to be proper only for a linearly or circularly polarized field $\mathbf{E}$ and only for substances whose particles can form regular patterns (say, crystals). In other cases the "scalar" description does not describe the real situation properly for at least two reasons. First, all resonant media except crystals consist of molecules randomly oriented and moving (or transferring excitation).[3–5] Second, the vector $\mathbf{d}$ can be polarized either linearly if in the transition of a particle from one energy level to another the total angular momentum $|\mathbf{J}|$ of the molecule is conserved, or circularly if $|\mathbf{J}|$ changes by one unit.[4,6] Thus, polarization of $\mathbf{d}$ does not generally coincide with the polarization of $\mathbf{E}$, and it is unclear whether the polarization of the field and the polarization of the dipole moment of a unit volume of the medium, $\mathbf{P}$, are identical. If not, the wave polarizations change and nonlinear polarization effects become possible. There is practically no way in which the "scalar" description scheme can be employed for interpreting such phenomena. In such cases it becomes necessary to take into account the random orientations and movements (or excitation transfer) of the resonant particles. In spite of its importance, no semiclassical theory that describes the interaction of high-power radiation with matter and allows for the motion of randomly oriented molecules has been very well developed. One of the goals of this paper is to partially eliminate this deficiency by using the example of solution of the specific physical problem of interaction of arbitrarily polarized oppositely propagating waves in an active resonant medium.

We know of several treatments that use the semiclassical approach in conjunction with a description of certain aspects of the interaction between radiation and an isotropic medium of randomly oriented particles. Several important results of these works contradict each other, either partially or completely .

Pantell and Puthoff[3] gave an equation of motion for the macroscopic polarizability $\mathbf{P}$ of an isotropic resonant medium obtained by a quantum mechanical method with an approximation in which the active levels of all the molecules have the same populations irrespective of the molecules' orientations and position in space. They found that $\mathbf{P}$ is collinear to the active field vector $\mathbf{E}$. This result correlates with the implications of Refs. 1 and 5, in which the general laws governing the interaction of single-frequency laser radiation (oppositely propagating waves, for one thing) with a resonant medium are examined. There it was postulated that the nonlinear susceptibility of a resonant medium depends on the square of the absolute value of the field strength, and it was thereby assumed that in such a medium a strong elliptically polarized field **E** does not induce anisotropy. From the results of Refs. 1 and 5 it follows that the polarization state of a single travelling wave never changes and that in the case of two oppositely propagating noninterfering waves the population difference of the resonant medium remains spatially homogeneous.

On the other hand, Lempiski[7] and Casperson and Reyser,[8] who studied the energy and polarization characteristics of dye lasers with coherent polarized pumping, showed that a polarized strong laser field applied to an isotropic resonant medium induces anisotropy. A similar result follows from the approximate expression, given in Ref. 4 and obtained by an iterative method, for the off-diagonal element of the density matrix of a two-level molecule placed in a weak resonant field. The conclusions agree with those arrived at in Refs. 9–14, which describe research involving the propagation of an elliptically polarized wave in a homogeneously broadened resonant medium of randomly oriented immobile molecules (in a solid amorphous substance). For one thing, in Refs. 9–13 it was found that owing to saturation of the resonant transition[12,13] the polarization vector of the nonlinear response of the medium, **P**, does not coincide with the elliptical polarization of the active field **E**, and that because of such induced anisotropy the polarization ellipse of the field changes its parameters. Moreover, as established in Ref. 15, molecular motion weakens this anisotropy.

This paper studies the laws governing transformation of the polarization and energy characteristics of oppositely propagating waves in an inverted resonant medium of randomly oriented two-level particles with a signal transition in which $|\Delta J|=1$, a transformation that occurs because polarized laser radiation induces anisotropy in the nonlinear dielectric constant.

2. THE EQUATIONS FOR ARBITRARILY POLARIZED MONOCHROMATIC RADIATION PROPAGATING IN A DENSE RESONANT MEDIUM

Allowance for the motion of the resonant objects, which is one of the most important sections of the theory developed here, is based on three well-known and now often used ideas about the properties of objects in the microworld. First, the magnitude and polarization state of the vector **d** of the matrix elements of the signal-transition dipole-moment operator are assumed to be determined exclusively by the position of the atoms and electrons inside the molecule, with the result that the direction of **d** is determined entirely by the orientation of the molecule's symmetry axes in space.[16] Second, particles in fluids, while rotating and moving according to the laws of thermal motion, retain their shape. Finally, in glasses and, for high dye concentrations, in liquids the exchange interaction can generate excitation transfer between neighboring resonant molecules oriented arbitrarily in relation to each other. These ideas make it possible to write equations that allow for the changes in similarly oriented excited and unexcited molecules brought on not only by the interaction with the resonant field but also by the rotation and movement in space or by excitation transfer.

Let us examine molecules whose vectors **d** are collinear, i.e., are oriented in space in the same way. The energy state of each such particle is described by the equation of motion of the density matrix $\hat{\rho}$ of a two-level system with an operator $\hat{W}=-(\hat{\mathbf{d}}\mathbf{E})$ of the energy of interaction with an external field. Assuming that the concentration N of the resonant objects is constant, we introduce the following variable quantities related to the density-matrix elements $\tilde{\rho}_{ij}$:

$$N_2=N\rho_{22},\quad N_1=N\rho_{11},\quad n=N_2-N_1, \tag{1}$$

which have the meaning of partial number densities of excited (N_2) and unexcited (N_1) molecules that are identical in their parameters and have the same orientation in space. These quantities depend on the interaction energy (**dE**) and are therefore functions of the coordinates of the observation point and of the angles determining the directions of **d** and **E** at this point.

According to the laws of diffusion, the amount by which Brownian motion in fluids and excitation transfer in solids and liquids reduces the spatial inhomogeneities in the number densities N_j and their differences over a time interval δt is

$$\delta(N_j,n)=D\Delta(N_j,n)(\delta t). \tag{2}$$

In gases, where these laws are well known, the diffusion coefficient $D=\bar{u}^2/3\nu$ is determined[17] by the average molecular thermal velocity $\bar{u}=\sqrt{k_BT/m}$ and the collision frequency

$$\nu=\frac{16a^2P\sqrt{\pi}}{\sqrt{m/k_BT}},$$

which depend on the pressure P and temperature T of the gas (k_B is Boltzmann's constant) and the particle mass m and radius a.

By analogy with translational motion, it can easily be established that the random rotation of molecules levels out the angular dependence of $N_{2,1}$. Actually this process is the diffusion of the orientations of molecules and is similar to the process, studied by Debye,[18] of variation of the angular number density of particles whose constant dipole moments form an angle with a certain preferred direction in space. In addition to Ref. 18, other rigorous methods[19,20] can be used to calculate the variations in the angular number density, δN_j, in a time interval δt. The result is equations of the form

$$\delta((N_j),n)=(1/\tau)\Delta_\Omega((N_j),n)(\delta t), \tag{3}$$

where Δ_Ω is the angular part of the Laplacian operator in a spherical system of coordinates. Note that using such a rotational diffusion equation, Phillion *et al.*[21] described the variation in the number density in a dye solution of molecules with the same spatial orientations. For a gas the characteristic time of orientational (rotational) diffusion can be expressed[18] in terms of the collision frequency ν, the molecular volume $V=4\pi a^3/3$, and the number density N_ν of the colliding particles as $\tau=6VN_\nu/\nu$.

The motion of resonant objects with the same physical properties resembles self-diffusion of excited and unexcited molecules. Allowing for such self-diffusion of molecules in a resonant medium, as shown in Ref. 15, amounts to changing a single material equation for the partial density of the population difference, n, in the complete semiclassical system of equations. The equation acquires two terms, $D\Delta n$ and $(1/\tau)\Delta_\Omega n$, which describe the variations in n caused, respectively, by the random translational and rotational movements of the particles (or migration of the particle energy states). The result is an equation of the Einstein–Fokker–Planck (EFP) type in which the coefficients of diffusion D and of orientational diffusion $1/\tau$ depend on the energy-level structure, the aggregate state of matter, the particle number density, and other properties of the medium.

The above ideas and the results of Ref. 15 allow us to write a system of equations for the interaction of the field E and a dense resonant medium:

$$\left(\Delta-\frac{1}{v_f^2}\frac{\partial^2}{\partial t^2}-\frac{4\pi\sigma}{\varepsilon v_f^2}\frac{\partial}{\partial t}\right)\mathbf{E}=\frac{4\pi}{\varepsilon v_f^2}\frac{\partial^2}{\partial t^2}<\mathbf{d}\tilde{R}_{12}>+\text{c.c.},$$

$$\left(\frac{\partial}{\partial t}-i\omega_{21}+\frac{1}{T_2}\right)\tilde{R}_{12}=+\frac{i}{\hbar}\mathbf{d}^*\mathbf{E}n, \tag{4}$$

$$\left(\frac{\partial}{\partial t}+\frac{1}{T_1}-D\Delta-\frac{1}{\tau}\Delta_\Omega\right)n-\frac{n_0}{T_1}=+\frac{2i}{\hbar}\overline{(\mathbf{Ed})\tilde{R}_{12}}+\text{c.c.},$$

where the (partial) population difference $n=N(\rho_{22}-\rho_{11})$ and the (partial) polarizability of a unit volume of the medium, $<\mathbf{d}\tilde{R}_{12}>\equiv<\mathbf{d}N\tilde{\rho}_{12}>$, depend on the direction of $\mathbf{d}$ and are proportional to the number density N of resonant particles and to the corresponding elements ρ_{ij} of the density matrix $\hat{\rho}$ of a two-level object with a fixed orientation in space; n_0 is the steady-state value of the partial population inversion in a zero field; $T_{1,2}$ are, respectively, the longitudinal and transverse relaxation times; v_f is the speed of light in the matrix of the medium; σ is the conductivity of the medium, which determines linear losses; and $\hbar$ is Planck's constant. All other notation is practically standard.

The angular brackets $\langle\cdots\rangle$ stand for averaging over the possible orientations of the vectors $\mathbf{d}\equiv\tilde{\mathbf{d}}_0|\mathbf{d}|$ and, generally, over other statistical parameters, and the bar designates averaging over the high-frequency oscillation period. Hence the system of equations (4) must be augmented by an equation for the distribution function W over the orientations of $\mathbf{d}$. For liquids and solids W must be assumed to depend neither on time nor on the acting field. For gases the variations in W due to the resonant field can be estimated by the approach developed by Gaponov and Miller.[22] Estimates, whose justification is given in Ref. 15, show that at room temperature and in the optical range the molecular distribution function W over the orientations of $\mathbf{d}$ can be assumed isotropic with an accuracy of 10^{-7}–10^{-9}. In what follows we assume, for the sake of simplicity, that W is always isotropic, so that the system of equations (4) is supplemented by an additional relationship:

$$W(\theta,\psi)=1/4\pi. \tag{5}$$

The two constitutive equations in (4) are valid for amorphous solids, liquids, and dense gases with a homogeneously broadened luminescence line and a particle collision frequency $\nu\simeq T_2^{-1}$ exceeding the Doppler linewidth $(\omega/c)\bar{u}$, which means that for such systems the approximation $|(\mathbf{u}\nabla)\tilde{R}_{12}|\ll|\tilde{R}_{12}/T_2|$ is valid. For solids, liquids, and dense gases the transverse relaxation time T_2 can be much shorter than the time T_1 of longitudinal relaxation.[1)] The steady-state solution

$$\mathbf{E}=\tfrac{1}{2}\tilde{\mathbf{E}}\exp(i\omega t)+\text{c.c.},\quad \tilde{R}_{12}=\tilde{R}\exp(i\omega t),\quad n=n(z) \tag{6}$$

of the system of equations (4) satisfy the following equations:

$$\Delta\tilde{\mathbf{E}}+k(k-2i\Gamma)\tilde{\mathbf{E}}=-2ik\frac{3\kappa}{2}(1-i\delta)<\tilde{\mathbf{d}}_0(\tilde{\mathbf{d}}_0^*\tilde{\mathbf{E}})\frac{n}{n_0}>, \tag{7}$$

$$\left(-DT_1\Delta-\frac{1}{\xi}\Delta_\Omega+1+3|\tilde{\mathbf{d}}_0^*\tilde{\mathbf{E}}|^2\right)\frac{n}{n_0}=1, \tag{8}$$

where $\kappa=4\pi kn_0|\mathbf{d}|^2T_2/[3\hbar\varepsilon(1+\delta^2)]$ is the linear gain and $\Gamma=2\pi\sigma(\mu/\varepsilon)^{1/2}/c$ the linear absorption coefficient in the medium, $\delta=(\omega-\omega_{12})T_2$ is the dimensionless detuning of the frequency of the field from that of the luminescence line center, $k=\omega/v_f$ is the wave number, and $\xi=\tau/T_1$ is a small parameter characterizing the diffusion of orientations of the dipole moments $\mathbf{d}$ of the resonant transition.[2)] In Eqs. (7) and (8) the field $\tilde{\mathbf{E}}$ is normalized to a field

$$\tilde{E}_s=\frac{\hbar}{|\mathbf{d}|}\sqrt{\frac{3(1+\delta^2)}{T_1T_2}}=\sqrt{3}E_s,$$

whose strength exceeds that of the commonly accepted saturation field strength by a factor of three.

Before we begin a detailed study of the problem of propagation of two oppositely propagating waves, let us establish the equivalence of the two theoretical approaches to describing the interaction of arbitrarily polarized high-intensity radiation and resonant media developed in Refs. 1 and 9. In one approach[1,3–5,12–15] the resonant medium is a collection of two-level objects (atoms or molecules) whose polarizations are the same in both magnitude and state but whose vectors $\mathbf{d}$ of the matrix element of the operator of the signal-transition dipole moment are randomly oriented in space.

The other approach was developed by Nasyrov and Shalagin[9–11] for a resonant medium where the resonant objects (atoms or molecules) are in a state of quasiclassical rotational motion with large angular momenta $\mathbf{J}$. There it was shown that the usual equations for the density matrix that allow for degeneracy can be transformed into equations for two nondegenerate states that take into account the orientation of $\mathbf{J}$ via two angles, θ and ψ.

To compare the two approaches we use the steady-state solution of Eq. (8) for an inverted medium of isotropically oriented molecules ($n_0=\text{const}>0$) in the simple limit of the absence of the diffusion of particles and their orientations ($D=0$ and $\tau^{-1}=\xi^{-1}=0$) to examine the angular dependence of the partial population difference

$$n=n_0(1+3|\tilde{\mathbf{d}}_0^*\tilde{\mathbf{E}}|^2)^{-1} \tag{9}$$

on the assumption that the distribution of n is formed as a result of the optically polarized field

$$\tilde{\mathbf{E}}=\mathscr{E}_x\mathbf{x}_0+i\mathscr{E}_y\mathbf{y}_0 \tag{10}$$

of a plane wave propagating along the z axis acting on the two-level molecules.[3)] For a medium of molecules characterized by a signal transition with conservation of angular momentum (the emission Q-branch) and by a linearly polarized unit vector $\mathbf{d}_0$ of each particle arbitrarily oriented in space, i.e.,

$$\mathbf{d}_0=\mathbf{z}\cos\theta+\sin\theta(\mathbf{x}_0\cos\psi+\mathbf{y}_0\sin\psi), \tag{11}$$

the expression for the partial population difference

$$n=n_0\{i+3[(\mathscr{E}_x\cos\psi)^2+(\mathscr{E}_y\sin\psi)^2]\sin^2\theta\}^{-1} \tag{12}$$

coincides, except for minor differences in notation and normalization, with the corresponding Eq. (3.8) in Ref. 9 for the population difference of molecules with the same direction of the angular momentum $\mathbf{J}$. For media of the second type consisting of molecules characterized by a signal transition with the angular momentum changing by a unit, $|\Delta\mathbf{J}|=1$, and by a circularly polarized unit vector $\mathbf{d}_0$ of each particle arbitrarily oriented in space, i.e.,

$$\mathbf{d}_0=[i\mathbf{z}_0\sin\theta+\mathbf{x}_0(\sin\psi-i\cos\psi\cos\theta)-\mathbf{y}(\cos\psi \\ +i\sin\psi\cos\theta)]/\sqrt{2}, \tag{13}$$

the expression for the partial population difference

$$n=n_0(1+\tfrac{3}{4}[(\mathscr{E}_x^2+\mathscr{E}_y^2)(1+\cos^2\theta)+(\mathscr{E}_x^2 \\ -\mathscr{E}_y^2)\sin^2\theta\cos^2\psi-4\mathscr{E}_x\mathscr{E}_y\cos\theta])^{-1} \tag{14}$$

coincides, except for the differences in notation and normalization mentioned, with Eq. (3.10) in Ref. 9 (transformed to the same compact form) for the population difference in the molecules characterized by transitions with $J_m\rightarrow J_n+1$. The fact that the compared expressions coincide suggests that there is total equivalence of the two semiclassical approaches to describing the interaction of polarized radiation of infinitely high intensity and a resonant medium of randomly oriented molecules.

Note that when there is orientational diffusion, Eqs. (12) and (14) may change considerably, and so will the result of propagation of elliptically polarized radiation in such a medium. As shown in Ref. 14, where the polarization instability of a traveling-wave laser was studied, the critical excess of pumping above the threshold value at which the steady-state lasing mode breaks down may decrease by tenfold or more owing to a decrease in orientational diffusion.

3. STATEMENT OF THE PROBLEM AND THE EQUATIONS FOR THE VECTOR AMPLITUDES OF THE OPPOSITELY PROPAGATING WAVES

Let us examine the propagation in an active medium of two oppositely propagating plane waves,

$$\tilde{\mathbf{E}}=\tilde{\mathbf{e}}_+(z)\exp\{-ikz\}+\tilde{\mathbf{e}}_-(z)\exp\{ikz\}, \tag{15}$$

whose complex-valued amplitudes $\tilde{\mathbf{e}}_\pm(z)$ vary little over one wavelength λ. We assume that the waves are polarized elliptically, with the result that each can be represented in the form of two circularly polarized waves rotating in opposite directions,

$$\begin{aligned}\tilde{\mathbf{e}}_+(z)&=\tilde{A}(z)(\mathbf{x}_0-i\mathbf{y}_0)/\sqrt{2}+\tilde{B}(z)(\mathbf{x}_0+i\mathbf{y}_0)/\sqrt{2},\\ \tilde{\mathbf{e}}_-(z)&=\tilde{a}(z)(\mathbf{x}_0-i\mathbf{y}_0)/\sqrt{2}+\tilde{b}(z)(\mathbf{x}_0+i\mathbf{y}_0)/\sqrt{2},\end{aligned} \tag{16}$$

with the complex-valued amplitudes $\tilde{U}=U\exp(i\varphi_U)$. The polarization ellipse of each travelling wave (say the wave $\tilde{\mathbf{e}}_+$) is determined by the three parameters: the major axis $2(A+B)$, the ratio $s_+=(A-B)/(A+B)$ of the axes, and the angle $\varphi_+=(\varphi_A-\varphi_B)/2$ between the major axis and the x axis. An important role in describing the polarization effects that emerge in the interaction of two oppositely propagating waves is also played by the relative position of the field vectors, which is specified by the angle

$$\frac{\Phi}{2}=\tfrac{1}{2}[(\varphi_A-\varphi_B)-(\varphi_a-\varphi_b)]=\varphi_+-\varphi_- \tag{17}$$

between the major axes of their polarization ellipses.

Equation n/n_0 in (4) is real and contains the variable coefficient

$$3|\mathbf{d}_0^*\tilde{\mathbf{E}}|^2=w_0(\theta,\psi,z)+\tilde{w}(\theta,\psi,z)\exp(-2ikz) \\ +\tilde{w}^*(\theta,\psi,z)\exp(2ikz), \tag{18}$$

whose components w_0 and $\tilde{w}$ depend on the angular coordinates and the complex-valued amplitudes of the interacting waves. We write the solution of this equation in the form of series in spatial and spherical harmonics,

$$\begin{aligned}\frac{n}{n_0}&=Q_0(\theta,\psi,z)+\left[\sum_{f=1}^{\infty}\tilde{Q}_f(\theta,\psi,z)\exp(2ifkz)+\text{c.c.}\right],\\ Q_f&=\sum_{s=0}^{\infty}\sum_{m=0}^{s}[(\tilde{C}_s^m)_f\exp(im\psi)\\ &\quad+(\tilde{D}_s^m)_f\exp(-im\psi)]P_s^m(\cos\theta),\end{aligned} \tag{19}$$

in which the coefficients $(\tilde{C}_s^m)_f$ and $(\tilde{D}_s^m)_f$ depend weakly on z. Substituting (19) into (7) and employing the Van der Pol method, we arrive at the truncated equations

$$\pm\frac{\partial\tilde{\mathbf{e}}_\pm}{\partial z}+\Gamma\tilde{\mathbf{e}}_\pm=\frac{3\kappa}{2}(1-i\delta)(<\tilde{\mathbf{d}}_0(\tilde{\mathbf{d}}_0^*\tilde{\mathbf{e}}_\pm)Q_0>+ \\ <\tilde{\mathbf{d}}_0(\tilde{\mathbf{d}}_0^*\tilde{\mathbf{e}}_\mp)\tilde{Q}_{\mp 1}>) \tag{20}$$

for the complex-valued amplitudes of the oppositely propagating waves, with $\tilde{Q}_{-1}=\tilde{Q}_1^*$. From the right-hand sides of Eqs. (20) we see that to describe the propagation of oppositely propagating waves in a resonant medium we only need to find the mean value and the first spatial harmonic of n/n_0. The first term on the right-hand side of each equation in (20) gives the contribution to the gain of a wave provided by the mean (saturated) population difference, and the second term gives the contribution to the gain of a wave provided by the rescattering of the other (oppositely propagating) wave on the lattice of the nonlinear dielectric constant.

The spatial harmonic amplitudes $\tilde{Q}_f(\theta,\psi,z)$ can be found by solving the following infinite system of coupled equations:

$$\left(\frac{1}{\beta}f^2-\frac{1}{\xi}\Delta_\Omega+1+w_0\right)\tilde{Q}_f+\tilde{w}\tilde{Q}_{f+1}+\tilde{w}^*\tilde{Q}_{f-1}=\delta_{f0}. \qquad (21)$$

In deriving (21) from Eq. (8) we used the approximation

$$\nabla^2\{\tilde{Q}_f\exp(-2ifkz)\simeq-4f^2k^2\tilde{Q}_f\exp(-2ifkz), \qquad (22)$$

which is less accurate than the method used in deriving Eqs. (20). Besides ξ, Eq. (21) contains another small parameter, $\beta=1/4k^2DT_1$, which characterizes the magnitude of the spatial diffusion of the particles.[4)] Because of this diffusion the ratio $|\tilde{Q}_f|/|\tilde{Q}_{f-1}|$ of the amplitudes of the spatial harmonics of the population difference proves to be small (equal to β by order of magnitude). Hence if the calculation accuracy is fixed, there could be a reduction in the number of equations in (21).

Using the properties of spherical harmonics, we can obtain a system of coupled linear equations of the band type in the coefficients $\tilde{C}^m_{sf}(\xi,\beta)$ and $\tilde{D}^m_{sf}(\xi,\beta)$. The system consists of one inhomogeneous equation and an infinite number of homogeneous equations. The fact that each equation has two small parameters makes it possible to seek the unknown coefficients in the form of power series in ξ and β. Employing this method, we can calculate all the necessary coefficients $(\tilde{C}^m_s)_{0,1}$ and $(\tilde{D}^m_s)_{0,1}$ and hence find, with any preassigned accuracy, the angular distributions of the mean value $Q_0(\theta,\psi)$ and the amplitude of the first spatial harmonic, $\tilde{Q}_1(\theta,\psi)$, of the partial population difference. These functions are solutions of the equation for n/n_0 and depend on the quantity $|\mathbf{d}_0^*\mathbf{E}|$ in (8), which is proportional to the energy of the interaction between an arbitrarily polarized molecule and the field and differs for the resonant media of the two different types.

In order to be specific, let us consider the active medium of molecules whose signal-transition dipole moments are circularly polarized. For such a medium the unit vector $\mathbf{d}_0$ of a particle arbitrarily oriented in space has the form (13), and the angular dependence of $3|\mathbf{d}_0^*\mathbf{E}|^2$ is determined by the functions

$$\begin{aligned}4w_0/3&=(1+\alpha^2)J+2\alpha(J_R-J_L)-(1-\alpha^2)\\&\quad\times[\tilde{R}\exp(2i\psi)+\tilde{R}^*\exp(-2i\psi)],\\4\tilde{w}/3&=(1+\alpha^2)(\tilde{\rho}_R+\tilde{\rho}_L)+2\alpha(\tilde{\rho}_R-\tilde{\rho}_L)-(1-\alpha^2)\\&\quad\times[\tilde{A}^*\tilde{b}\exp(2i\psi)+\tilde{a}\tilde{B}^*\exp(-2i\psi)],\end{aligned} \qquad (23)$$

where the following notation has been employed:

$$\begin{aligned}&\alpha=\cos\theta,\quad J_R=|\tilde{A}|^2+|\tilde{a}|^2,\quad J_L=|\tilde{B}|^2+|\tilde{b}|^2,\\&J=J_R+J_L,\quad \tilde{\rho}_R=\tilde{A}^*\tilde{a},\quad \tilde{\rho}_L=\tilde{B}^*\tilde{b},\quad \tilde{R}=\tilde{A}^*\tilde{B}+\tilde{a}^*\tilde{b}.\end{aligned} \qquad (24)$$

In this case fourteen orthogonal components of the expansion of n/n_0 in spatial and spherical harmonics contribute to the right-hand sides of the equations for the complex-valued amplitudes of the circularly polarized oppositely propagating waves,

$$\begin{aligned}\left(\frac{d}{dz}+\Gamma\right)\tilde{A}&=\frac{\kappa}{2}(1-i\delta)[(\bar{Q}+\tilde{Q}_R)\tilde{A}-\tfrac{6}{5}(\tilde{D}^2_2)_0\tilde{B}+\tilde{q}^*_R\tilde{a}\\&\quad-\tfrac{6}{5}(\tilde{C}^2_2)^*_1\tilde{b}],\\\left(-\frac{d}{dz}+\Gamma\right)\tilde{a}&=\frac{\kappa}{2}(1-i\delta)[(\bar{Q}+\tilde{Q}_R)\tilde{a}-\tfrac{6}{5}(\tilde{D}^2_2)_0\tilde{b}+\tilde{q}_R\tilde{A}\\&\quad-\tfrac{6}{5}(\tilde{D}^2_2)^*_1\tilde{B}],\\\left(\frac{d}{dz}+\Gamma\right)\tilde{B}&=\frac{\kappa}{2}(1-i\delta)[(\bar{Q}+\tilde{Q}_L)\tilde{B}-\tfrac{6}{5}(\tilde{C}^2_2)_0\tilde{A}+\tilde{q}^*_L\tilde{b}\\&\quad-\tfrac{6}{5}(\tilde{D}^2_2)^*_1\tilde{a}],\\\left(-\frac{d}{dz}+\Gamma\right)\tilde{b}&=\frac{\kappa}{2}(1-i\delta)[(\bar{Q}+\tilde{Q}_L)\tilde{b}-\tfrac{6}{5}(\tilde{C}^2_2)_0\tilde{a}+\tilde{q}_L\tilde{B}\\&\quad-\tfrac{6}{5}(\tilde{C}^2_2)_1\tilde{A}].\end{aligned} \qquad (25)$$

These components form five combinations:

$$\begin{aligned}\bar{Q}&=[(\tilde{C}^0_0)_0+(\tilde{D}^0_0)_0+\text{c.c.}],\\\tilde{Q}_{R,L}&=\pm\tfrac{1}{2}[(\tilde{C}^0_1)_0+(\tilde{D}^0_1)_0]+\tfrac{1}{10}[(\tilde{C}^0_2)_0+(\tilde{D}^0_2)_0],\\\tilde{q}_{R,L}&=[(\tilde{C}^0_0)_1+(\tilde{D}^0_0)_1]\pm\tfrac{1}{2}[(\tilde{C}^0_1)_1+(\tilde{D}^0_1)_1]+\tfrac{1}{10}[(\tilde{C}^0_2)_1\\&\quad+(\tilde{D}^0_2)_1].\end{aligned} \qquad (26)$$

The main laws governing the interaction of oppositely propagating waves in the resonant medium considered here can be established by calculating $(\tilde{C}^m_s)_{0,1}$ and $(\tilde{D}^m_s)_{0,1}$ to first order in ξ and β and then finding the coefficients

$$\begin{aligned}\tilde{Q}_{R,L}&=-\left[\frac{\xi}{60}(23J_{R,L}-22J_{L,R})\right]\frac{1}{1+J},\\(\tilde{C}^2_2)_0&=(\tilde{D}^2_2)^*_0=\frac{\xi}{24}\frac{\tilde{R}}{1+J},\\(\tilde{C}^2_2)_1&=\frac{\xi}{24}\frac{\tilde{A}^*\tilde{b}}{1+J},\quad(\tilde{D}^2_2)_1=\frac{\xi}{24}\frac{\tilde{B}^*\tilde{a}}{1+J},\\\bar{Q}&=\frac{1}{1+J}+2\beta\frac{|\tilde{\rho}_R+\tilde{\rho}_L|^2}{(1+J)^2}+\xi\Lambda,\\\Lambda&=\frac{\chi(1+J)-[\tilde{\chi}(\tilde{\rho}^*_R+\tilde{\rho}^*_L)+\tilde{\chi}^*(\tilde{\rho}_R+\tilde{\rho}_L)]}{(1+J)^2-|2\tilde{\rho}_R+\tilde{\rho}_L|^2},\\\chi&=\frac{23(J_L-J_R)^2+38|\tilde{\rho}_R-\tilde{\rho}_L|^2+8(J_RJ_L+\rho^2_R+\rho^2_L)}{60(1+J)},\\\tilde{\chi}&=\frac{42(J_L-J_R)(\tilde{\rho}_R-\tilde{\rho}_L)+4J(\tilde{\rho}_R+\tilde{\rho}_L)}{60(1+J)},\\\tilde{q}_{R,L}&=\frac{\xi\tilde{q}-\beta(\tilde{\rho}_R-\tilde{\rho}_L)+(\xi/60)(23\tilde{\rho}_{R,L}-22\tilde{\rho}_{L,R})}{1+J},\\\tilde{q}&=\tilde{\chi}-(\tilde{\rho}_R+\tilde{\rho}_L)\Lambda,\end{aligned} \qquad (27)$$

which give the contribution of each physical mechanism to the gain of a separate circularly polarized wave.

4. THE INTERACTION OF COLLINEARLY AND ORTHOGONALLY POLARIZED WAVES

It is assumed that interfering waves in a nonlinear medium always write a dielectric-constant lattice. In this terminology interfering waves are those oppositely propagating waves whose interaction with the resonant medium forms the first spatial harmonic of the population difference.

4.1. Let us examine the interaction of oppositely propagating waves that are circularly polarized and are characterized by the same (right) rotation of the electric field vector, i.e., $B=b=0$. In this case from Eqs. (24)–(27) we first find $\tilde{q}_R^*$ and then from Eqs. (25) we obtain the equations

$$\pm\frac{dJ_\pm}{dz}+2\Gamma J_\pm=\frac{\kappa J_\pm}{1+J_++J_-}\left[1+\beta\frac{J_\mp(J_\pm-J_\mp-1)}{1+J_++J_-}\right.$$
$$\left.-\frac{23}{60}\xi\frac{J_+^2+2J_\mp+J_\pm+J_-^2}{(1+J_++J_-)^2-2J_+J_-}\right] \tag{28}$$

for the intensities $J_+=A^2$ and $J_-=a^2$. Equations (28) show that the stronger wave has a greater local gain. Qualitatively this conclusion coincides with the results of the "scalar" theory,[23–25] according to which the mechanism of redistribution at the first spatial harmonic of n/n_0 ensures that the growth of the stronger wave dominates.

From Eqs. (25) we can find the growth rates

$$\frac{d}{dz}(\ln\tilde{B})=-\frac{d}{dz}(\ln\tilde{b})=-\Gamma$$
$$+\frac{\kappa}{2(1+J_++J_-)}\left[1+2\beta\frac{J_+J_-}{1+J_++J_-}\right.$$
$$\left.+\frac{\xi}{60}\frac{19J+61J^2+46J_+J_-+42J(J_+^2+J_-^2)}{(1+J_++J_-)^2-2J_+J_-}\right] \tag{29}$$

of the field perturbations in the form of oppositely rotating weak waves $(B,b\ll 1)$ and establish that the gains are positive if the linear absorption Γ is fairly small. This means that the solution of system (28) is convectively unstable.

4.2. The results obtained in Refs. 1,3 and 5 have given rise to the idea that orthogonally polarized oppositely propagating waves, which do not interfere in linear media, do not write a spatial dielectric-constant lattice in an isotropic resonant media. This, however, has not proved to be quite the case.

Let us study the interaction of rotating, circularly polarized oppositely propagating waves $(B=a=0)$. Equations (27) suggest that the first spatial harmonic of the population difference in this case is finite and that the lattice amplitude $|(\tilde{C}_2^2)_1|\neq 0$ is independent of spatial diffusion (the coefficient β). The amplitude increases with ξ (i.e., as angular diffusion decreases) and, obviously, is largest for a medium without angular diffusion. The interaction of these orthogonally polarized waves is described by the equations

$$\pm\frac{dJ_\pm}{dz}+2\Gamma J_\pm=\frac{\kappa J_\pm}{1+J_++J_-}\left[1\right.$$
$$\left.+\frac{1}{60}\xi\frac{42J_\mp(J_\mp-J_\pm)-23J_\pm+19J_\mp}{1+J_++J_-}\right] \tag{30}$$

for the intensities $J_+=A^2$ and $J_-=b^2$. In contrast to Eqs. (28), Eqs. (30) imply that due to rescattering on such a lattice the weaker wave has the greater growth rate. The solution of Eqs. (30) proves to be convectively unstable, too.

4.3. Using the angle $\varphi_+-\varphi_-=\Phi/2$, we examine the interaction of oppositely propagating waves with the same linear polarizations $(A=B,\ a=b,\ \varphi_a=\varphi_b,\ \text{and}\ \varphi_A=\varphi_B)$. Combining (25) and (27), we arrive at the equations

$$\frac{d}{dz}\left(\frac{A-B}{A+B}\right)\equiv\frac{ds_+}{dz}=0,$$
$$\frac{d}{dz}\left(\frac{a-b}{a+b}\right)\equiv\frac{ds_-}{dz}=0, \tag{31}$$
$$\frac{d\Phi}{dz}=0,$$

which can be interpreted as the law of conservation of polarizations of the interacting waves. Here the equations for the wave intensities, $J_+=2A^2$ and $J_-=2a^2$, coincide with (28) except that the coefficient $\xi/30$ replaces $23\xi/60$. This means that, all other things being equal, for the resonant medium considered here waves with the same linear polarization of their fields are amplified more than collinear circularly polarized waves.

4.4. Assuming that in a certain cross section the oppositely propagating waves are linearly polarized and orthogonal to each other $(A=B,\ a=b,\ \varphi_A-\varphi_B=0,\ \text{and}\ \varphi_a-\varphi_b=\pi)$, we can obtain the equations

$$\pm\frac{dJ_\pm}{dz}+2\Gamma J_\pm=\frac{\kappa J_\pm}{1+J_++J_-}\left[1\right.$$
$$\left.-\frac{1}{30}\xi\frac{J_++J_-+(21/2)J_\mp(1+J_\mp-J_\pm)}{1+J_++J_-}\right] \tag{32}$$

for the intensities $J_\pm$ and the equations for the polarization characteristic $s_\pm$ and Φ, which coincide with (31). Equations (31) and (32) suggest that linearly polarized orthogonal waves propagate without changing their polarization states and that the stronger wave has the greater growth rate.

4.5. In the case of maximum diffusion ($\beta\to 0$ and $\xi\to 0$) Eqs. (28), (30), and (32) transform into equations of radiation transfer.

In the other limiting case of no diffusion ($D=0$ and $\tau^{-1}=0$), the amplitude of the first spatial harmonic of population inversion,

$$\tilde{Q}_1=\overline{(n/n_0)\exp(+2ikz)},$$

where the bar designates averaging over one wavelength, has the maximum value (other things being equal) and hence the mechanism of rescattering of the two distinct oppositely

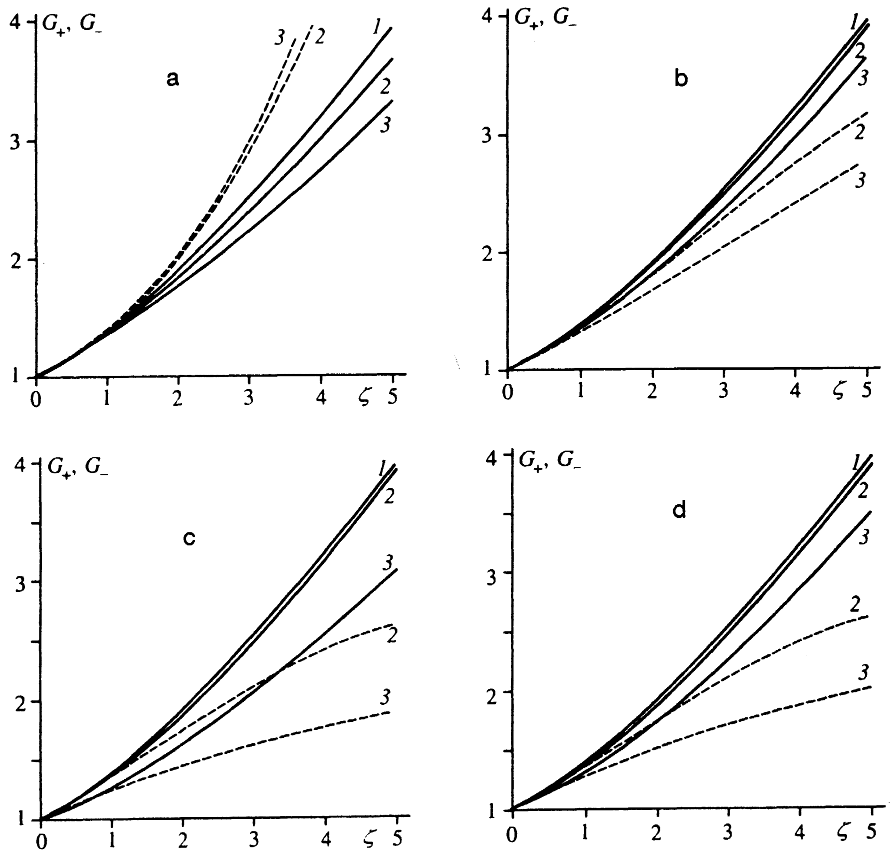

FIG. 1. $G_+ = J_+(\zeta)/J_+(0)$ and $G_- = J_-(0)/J_-(\zeta)$ as functions of the integral linear growth rate $\zeta = \kappa(z - z_0)$ of the layer $z - z_0$ of the active medium (solid curves depict the G_+ vs ζ dependence, and the dashed curves the G_- vs ζ dependence) for four cases: (a) circularly polarized orthogonal waves, (b) linearly polarized orthogonal waves, (c) circularly polarized collinear waves, and (d) linearly polarized collinear waves. At the boundary $z = 0$ of the active medium the intensities of the interacting waves are equal: $J_+(0) = J_-(0) = 1$. Curves *1* in all diagrams are constructed for media with maximum diffusion of the molecules ($\beta = \xi = 0$), curves *3* are constructed for media without diffusion, and curves 2 are constructed for media with the following parameters: $\xi = 0.30$ [(a) and (b)], and $\xi = 10^{-5}$ and $\beta = 0.20$ [(c) and (d)].

propagating waves into each other plays the most important role here. In the Van der Pol approximation, $\tilde{Q}_1$ and the mean value of the partial population inversion, $Q_0 = n/n_0$, are the coefficients in the Fourier-series expansion of the almost-periodic (in the coordinate z) function n/n_0, i.e., oscillating with a period of $\lambda/2$ and at the same time slowly varying, in the form of the steady-state solution (9) of Eq. (8).

These analytical consequences of Eqs. (28), (30), and (32) are illustrated in Fig. 1 by curves representing the dependence of the gains of the oppositely propagating waves, $G_+ = J_+(\zeta)/J_+(0)$ and $G_- = J_-(0)/J_-(\zeta)$, on the integral linear growth rate $\zeta = \kappa(z - z_0)$ of the layer $z - z_0$ of the active medium.

5. NONLINEAR POLARIZATION EFFECTS IN THE INTERACTION OF OPPOSITELY PROPAGATING WAVES

With the exception of the two particular cases considered above, the polarizations of oppositely propagating waves always undergo transformations: the major axis of the polarization ellipse rotates and the ellipticity of each wave changes. Variations in polarization are caused by the anisotropic nonlinear dielectric constant $\tilde{\varepsilon}^{NL}$ induced by the field in the medium. As a result the local propagation constants $\tilde{h}_\pm = h'_\pm + ih''_\pm$ of the oppositely rotating circularly polarized components of each of the oppositely propagating waves differ. The real parts $h'_\pm$ differ, which causes the polarizations of the oppositely propagating waves to rotate, while the difference in the imaginary parts $h''_\pm$ leads to changes in the ellipticities of the waves. Generally, $\tilde{\varepsilon}^{NL} = \varepsilon' + i\varepsilon''$ has both an imaginary part ε'' and a real part ε', whose value is proportional to the detuning δ of the field frequency from the frequency of the center of the luminescence line. Each contributes to the transformation of polarizations of the propagating waves, and each contribution can be considered a manifestation of an independent physical mechanism.

The inhomogeneous anisotropic distribution of $\tilde{\varepsilon}^{NL}$ is determined entirely by the spatial distribution of the population inversion n/n_0 formed as a result of the interaction of the two-level medium and the polarized radiation. Such a distribution acts as a lattice on which the waves rescatter into each other and as a result their polarizations change. This physical mechanism of changing the polarizations of oppositely propagating waves operates independently of δ and, therefore, can be defined as a mechanism related to the presence of ε''. Yakubovich[11] was the first to point out that the rescattering of waves on the $\tilde{\varepsilon}^{NL}$ lattice is a physical mechanism for changing the mutual orientation of the wave polarizations, and later experiments were carried out [26] in which the value of rotation of the polarization planes of two linearly polarized oppositely propagating waves was measured for the active medium of a gas laser.

The second physical reason for the rotation of the polarization planes is the presence of $\varepsilon' \propto \delta$ and is related to the additional dispersion introduced into the medium by resonant molecules.

5.1. The changes in the polarizations of the oppositely propagating waves in the interaction space can be described by equations for $\varphi_\pm$ and $s_\pm$. These equations are obtained directly from the system of equations (25) and can be represented in the form of two symmetric pairs:

$$\pm\frac{d\varphi_\pm}{dz}=\frac{\kappa}{4(1+J)}\{\pm e^{\pm\gamma}\sin\Phi(\eta_-J_\pm-\eta F_\pm)+\delta[\eta_+F_\mp+\eta(1+J_\pm)+e^{\pm\gamma}\cos\Phi\times(-\eta_-F_\pm+\eta J_\pm)]\}, \tag{33}$$

$$\pm\frac{ds_\pm}{dz}=-\frac{-\kappa/\mathrm{ch}^2\theta_\pm}{4(1+J)}\{\delta e^{\pm\gamma}\sin\Phi(\eta_-J_\pm-\eta F_\pm)+[\eta_+F_\mp+\eta(1+J_\pm)+e^{\pm\gamma}\cos\Phi(-\eta_-F_\pm+\eta J_\pm)]\}, \tag{34}$$

where we have introduced the following notation:

$$\begin{gathered}\eta_\pm=\beta+\xi\Lambda+\frac{\xi}{30}\frac{1-J}{1+J}\pm0.35\xi,\\ \eta=0.7\xi\frac{J_R-J_L}{1+J},\quad \gamma=\ln\frac{ab}{AB},\\ J_+=A^2+B^2,\quad J_-=a^2+b^2,\\ \theta_+=\frac{1}{2}\ln\frac{A}{B},\quad \theta_-=\frac{1}{2}\ln\frac{a}{b},\\ F_+=s_+(A+B)^2\equiv A^2-B^2,\\ F_-=s_-(a+b)^2\equiv a^2-b^2.\end{gathered} \tag{35}$$

Equations (33) and (34) can be transformed into each other pairwise by the following substitutions:

$$z\leftrightarrow-z,\quad s_+\leftrightarrow s_-,\quad a\leftrightarrow A,\quad b\leftrightarrow B. \tag{36}$$

Equations (33)–(35) clearly show that the effectiveness of changes in the polarizations of the interacting waves depends on the wave intensities $J_\pm$, the wave ellipticities ($F_\pm \propto s_\pm$), and the diffusion in the medium. The right-hand sides in Eqs. (33) and (34) strongly depend on Φ, since the angle $\varphi_+-\varphi_-=\Phi/2$ between the major axes of the polarization ellipses of the oppositely propagating waves is one of the main parameters determining the local anisotropy of the $\tilde{\varepsilon}^{NL}$ lattice. All other things being equal, their absolute values increase monotonically with β and ξ and, obviously, reach their maximum value when there is no diffusion. The contributions of ε' to the changes in the polarization states of the waves are taken into account by terms whose value is proportional to the detuning δ.

5.2. First we analyze Eqs. (33) for zero detuning ($\delta=0$), when $\varepsilon'=0$, and establish the main laws governing the variations in $\varphi_\pm$ caused by the rescattering of the waves on the anisotropic ε'' lattice. When the condition $\beta\gg\xi$ (i.e., $\eta_-\gg\eta$) is met (e.g., for dense gases), from Eqs. (33) we can find $d\varphi_\pm/d(\kappa z)\propto\sin\Phi$ and that these polarizations can rotate in two opposite directions, depending on Φ. For $0<\Phi<\pi$ the axes rotate clockwise and the angle between the axes, $\varphi_+-\varphi_-$, monotonically increases with ζ, asymptotically approaching the value $\pi/2$. For $-\pi<\Phi<0$ the axes rotate counterclockwise, and the angle between these axes changes, with ζ increasing toward the limit of $-\frac{1}{2}\pi$. The variation of $\varphi_\pm$ is fastest when $|\varphi_+-\varphi_-|\simeq\frac{1}{4}\pi$. If the major axes of the ellipses are orthogonal to each other, $\varphi_+-\varphi_-\simeq\frac{1}{4}\pi$, or are oriented in the same direction, the $\varphi_\pm$ do not change.

The anisotropic properties of ε'' and hence the right-hand sides of Eqs. (33) also depend on the ellipticities of the interacting waves. The corresponding contributions to the variation of $\varphi_\pm$ are provided by the terms proportional to ξ, $s_\pm$, and the difference J_R-J_L of the dextrorotatory and levorotatory parts of the intensity, J_R and J_L, of the radiation interacting with the medium. They reach their maximum value when there is no diffusion, at $\Phi=\pi/2$, and as $|s_\pm|\to1$.

5.3. The ellipticities of the oppositely propagating waves transform because of the interaction on the ε'' lattice. At $\delta=0$ the right-hand sides of Eqs. (34) have three terms, which depend differently on the medium parameters and the intensities and polarization states of the interacting waves and which, therefore, can be interpreted as four independent components of the physical mechanism of transformation of the polarization ellipse of each of the two oppositely propagating waves on the ε'' lattice.

The sign of the first term in the four terms on the right-hand side of each equation in (34) coincides with the sign of the corresponding quantity $-s_\mp$. If the $s_\pm$ have the same sign, this component of the physical mechanism "tries" to lower $|s_\pm|$ and to asymptotically transform elliptically polarized waves into linearly polarized waves.

The second term on the right-hand side of each equation in (34) is proportional to $J_L-J_R=-(F_+-F_-)$. If the $s_\pm \propto F_\pm$ have the same sign, this term also lowers $|s_\pm|$ in each wave in the direction of the wave's propagation and, therefore, evens out the fractions J_L and J_R. When there is no second wave ($J_-=0$ and $s_-=0$), this term on the right-hand side of (34) does not vanish, in contrast to the other terms, and ensures that $|s_+|$ decreases, so that asymptotically, on a large path, an elliptically polarized wave is transformed into a linearly polarized wave [9,13].

The signs of the remaining two terms on the right-hand sides of (34) coincide with those of $s_\pm\cos\Phi$ and $(J_L-J_R)\cos\Phi$, respectively. Hence, depending on $s_\pm$ and Φ, the corresponding components of the physical mechanism of ellipticity variations can lower or raise the value of $|s|$ of each wave. These terms provide the greatest contributions to the variation of $s_\pm$ when the major axes of the polarization ellipses are collinear ($\Phi=0$) or orthogonal to each other ($\Phi=\pi$). For $\beta\gg\xi$, i.e., $|\eta_-F_\pm\cos\Phi|\gg|\eta J_\pm\cos\Phi|$, these components of the physical mechanism are responsible for the decrease in $|s_\pm|$ in the direction of propagation of the wave, provided that $\cos\Phi<0$.

5.4. When $\varepsilon'\propto\delta$, the propagation constants $\tilde{h}_\pm$ of circularly polarized waves change, and Eqs. (33) and (34) acquire the appropriate terms proportional to δ. This second physical mechanism for polarization transformation operates even in the absence of one of the oppositely propagating waves [9,10,12,13]. For one thing, as a traveling wave passes a section of the active medium, the axes of the polarization ellipse of the wave rotate clockwise if the field frequency is shifted from the frequency of the gain-line center to the violet part of the spectrum ($\delta > 0$) and of the field vector is dextrorotatory ($A > B$ and $s > 0$).

The contribution of ε' to the polarization variation in the oppositely propagating waves becomes similar to that of ε'' in general for $|\delta| \sim 1$, i.e., when ε' and ε'' are of the same order of magnitude. However, its dependence on $J_\pm$, $\varphi_+ - \varphi_-$, and $s_\pm$ is quite different. For instance, ε' has a small effect on the angles $\varphi_\pm$ if the waves are polarized almost linearly ($|s_\pm| \to 0$) and the angle between the major axes of the polarization ellipses, $\varphi_+ - \varphi_-$, is $\Phi/2 \simeq \pm\pi/4$. On the other hand, ε' may have a strong effect on the ellipticities of the waves in this case.

5.5. When diffusion is strong ($\beta \to 0$ and $\xi \to 0$), anisotropy disappears and the amplitude $\tilde{Q}_1$ of the first spatial harmonic of n/n_0 vanishes. Then Eqs. (31) are valid, so that the shape of the polarization ellipse of each wave remains the same in all cross sections of a layer of the resonant medium. The equations for the intensities of the oppositely propagating waves and, for one thing, Eqs. (28), (30), and (32), acquire the form of a radiation transfer equation (the nonlinear Bouger law). They have repeatedly been used in the past (see, e.g., Refs. 27 and 28) to describe noninterfering waves in resonant media within the "scalar" theory of interaction of radiation and matter. Physically, the reason for such transformations is that strong diffusion transforms a nonlinear medium of randomly oriented two-level systems into a medium that for all practical purposes is homogeneous and isotropic. Every molecule of the medium takes part on an equal basis (to one-third of its capacity) in the interaction with the strong field, irrespective of the polarization state of the field and that of the dipole moment $\tilde{\mathbf{d}}_{21}$ of themolecule. Under these conditions the waves interact as a if they were in a medium where the $\tilde{\mathbf{d}}_{21}$ of each molecule is collinear with the field and the resonant absorption cross section is lower by a factor of three.$^{5)}$

5.6. The results of calculations of the dependence of the polarization characteristics $\varphi_\pm$ and $s_\pm$ on the total linear growth rate $\zeta = \kappa(z - z_0)$, which reflect the main behavior of the variations in the polarizations of the oppositely propagating waves, are depicted in Figs. 2 and 3. The curves are constructed for three different values of the detuning of the field frequency from the frequency of the luminescence-line center ($\delta = 0$ and $\delta = 0.5$) for oppositely propagating waves that in the cross section at $z_0 = 0$ are linearly polarized, $\tilde{\mathbf{e}}_+ = (\mathbf{x}_0 - \mathbf{y}_0)/\sqrt{2}$ and $\tilde{\mathbf{e}}_- = \mathbf{x}_0$, at an angle of 45° to each other, so that their intensities and the ratios of the polarization ellipse axes are the same ($J_+(0) = J_-(0) = 1$ and $s_+(0) = s_-(0) = 0$) and the angles $\varphi_\pm$ between the major axes of the polarization ellipses of the strong and weak waves and the x axis are, respectively, $\varphi_+(0) = -45°$ and $\varphi_-(0) = 0$. The solid curves in Figs. 2 and 3 depict the characteristics φ_+ and s_+ of the stronger wave, as a function of ζ while the dashed curves depict the angle $\Phi/2 = \varphi_+ - \varphi_-$ and the quantity s_-as a function of ζ. Curves *1* are constructed for media with maximum diffusion ($\beta = \xi = 0$), curves *3* are constructed for media without diffusion, and curves *2* are constructed for a medium with arbitrarily chose parameters: $\beta = 0.10$ and $\xi = 10^{-5}$.

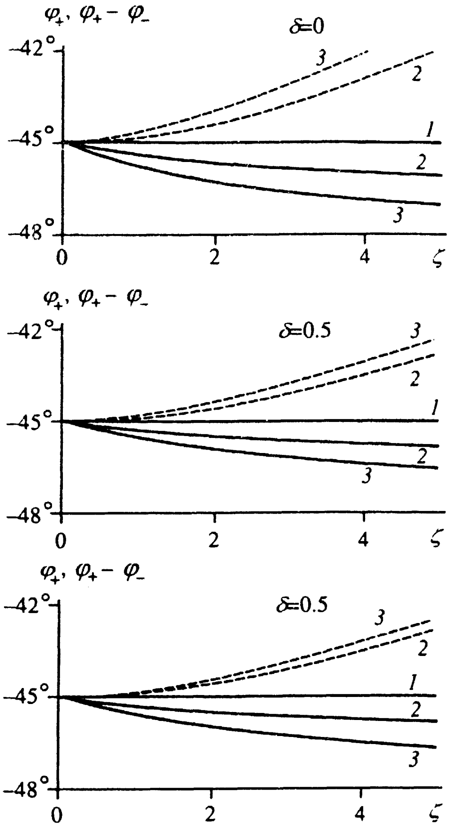

FIG. 2. $\varphi_\pm$ (solid curves) and $\varphi_+ - \varphi_-$ (dashed curves) as functions of the integral linear growth rate ζ of a layer of the medium: curves *1*, $\beta = \xi = 0$; curves *2*, $\beta = 0.10$ and $\xi = 10^{-5}$; and curves *3*, medium without diffusion.

Figure 2 shows that at zero detuning ($\delta = 0$) the angles $\varphi_\pm$ vary monotonically because of the rescattering of waves on the ε'' lattice. As each oppositely propagating wave travels through a layer of the active resonant medium, its polarization ellipse rotates counterclockwise through an angle $\Delta\varphi_\pm$ (if the wave is observed along the direction of its propagation). The polarization ellipse of the weaker field experiences the greater rotation.

The main information as to how the detuning δ influences the nonlinear polarization effects is contained in the curves *1* and *3*. The parameters β and ξ of real media are determined by the diffusion processes taking place in the media, with the result that an arbitrarily chosen combination of the values of β and ξ does not give a proper picture of real physical conditions. A proper choice of these parameters should place the curves *2* inside the area bounded by the curves *1* and *3* for the two limiting cases.

6. CONCLUSION

Equations (25) and their particular cases (28)–(34) can be used to describe the interaction of arbitrarily polarized oppositely propagating waves in a passive two-level medium. For this one need only substitute $-\kappa$ for κ in these equations.

In resonant media where the molecules are characterized by a linearly polarized vector $\mathbf{d}$ of the signal-transition matrix element, the transformation of the polarizations of the

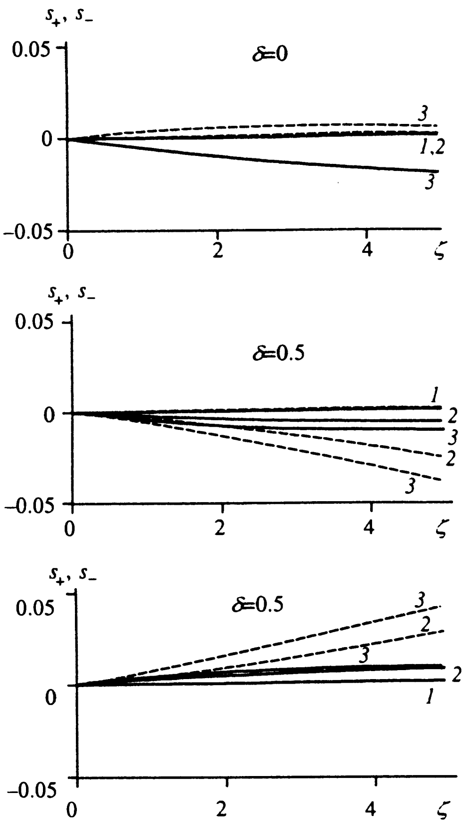

FIG. 3. The ratios s_+ (solid curves) and s_- (dashed curves) of the axes of the polarization ellipses of the oppositely propagating waves as functions of the integral linear growth rate ζ of a layer of the medium: curves *1*, $\beta=\xi=0$; curves *2*, $\beta=0.10$ and $\xi=10^{-5}$; and curves *3*, medium without diffusion.

oppositely propagating waves is also described by equations of the form (25), (33), and (34). However, their right-hand sides depend differently on the intensities and the polarization characteristics of the interacting waves. Hence the results of our study cannot be employed without additional modifications for interpreting the nonlinear polarization effects occurring in such media.

Our results can be used to estimate the potentials of such adaptive devices as a two-way amplifier with a wavefront-reversal mirror and high-power pulsed lasers with a nonlinear shutter, to design wavefront-conjugation mirrors on the basis of quarter-wave mixing in a resonant medium, and to study the properties of matter by methods of nonlinear polarization spectroscopy.

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[1)] Strictly speaking, for a dense gas, in addition to the diffusion terms the last equation in (4) must contain the terms $(\mathbf{u}\nabla n)$ and $(\mathbf{w}\nabla_{\theta\psi}n)$ where **u** and **w** are, respectively, the linear and angular velocities of the moving particles. In calculating the macroscopic polarizability the necessary statistical averaging must also be done over the random parameters **u** and **w** using the same particle energy distribution function as in calculating the diffusion coefficients D and τ^{-1}. The results of these calculations can be shown to coincide with those that can be obtained by solving Eqs. (4) in which the last EFP equation for n/n_0 contains diffusion coefficients increased by a factor of 3 to 10, depending on the thermodynamic parameters of the gas. Hence we can assume without loss of generality in the statement of the problem that the system of equations (4) provides a proper description of the interaction of radiation and a dense resonant medium of randomly oriented moving particles.

[2)] Estimates show that for carbon dioxide at atmospheric pressure and room temperature $\xi\simeq10^{-6}$.

[3)] Without loss of generality we can assume $\mathscr{E}_x$ and $\mathscr{E}_y$ to be real.

[4)] For carbon dioxide at atmospheric pressure and room temperature β is less than 0.01 for radiation with a wavelength of approximately 10^{-3}.

[5)] In Refs. 27 and 28 the intensities are normalized to a quantity three times smaller than the one we used.

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