Interaction of intense radiation with classically rotating atoms or molecules

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Kinetic equations, in which the orientational states of the angular momenta **J** of the levels are described classically, are obtained for the density matrix. The equations make it possible to solve the problem of the interaction between arbitrarily polarized high-intensity radiation and systems characterized by a large quantum number **J**. The level population distributions with respect to the directions of **J** are analyzed and it is noted that in an intense field these distributions acquire strongly selective angular structures. The dynamics of the polarization of intense radiation as it propagates in amplifying or absorbing media is considered.

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1. INTRODUCTION

A feature of modern lasers is high intensity of the generated radiation. At any rate, it is such that the interaction of laser radiation with matter cannot be described within the framework of the method of successive approximations in intensity, and it is necessary to seek methods that impose no restrictions on the radiation intensity. It is well known that these restrictions can be lifted in the model with two nondegenerate states. Real states of atoms and molecules, however, are degenerate in the directions of the angular momentum **J**. For these systems, the problem of interaction with radiation of arbitrary intensity can be solved if the radiation has linear or circular polarization. This possibility is due to the effective subdivision of the real multilevel system into a set of simple two-level subsystems (see, e.g., Ref. 1).

The situation becomes radically more complicated when the radiation has an arbitrary (elliptic) polarization and a high intensity. In a rigorous quantum-mechanical approach, the problem of absorption (amplification) and propagation of such radiation in a resonant medium has not lent itself to a solution to this day. As a result, no attention has been paid to a large class of interesting polarization phenomena.

It is shown in the present paper that the foregoing difficulties can be overcome in the case of transitions with large quantum numbers J, when the orientational states of the vector **J** can be described classically. The representation in which the density matrix elements are identical to the classical distribution functions of the vector **J** with respect to the direction turned out to be productive. The transition to quantum mechanics is effected in this representation with the aid of a transformation similar to that used by Wigner for translational degrees of freedom (see, e.g., Ref. 2).

In the new representation, the equations for the density matrix have assumed the form of the usual equations of the model of two nondegenerate states. All that was added was a parametric dependence on the orientation angles of **J**. The solution of the resultant equations has a lucid geometric interpretation.

The derived equations can be used to solve an entire class of new problems. We confine ourselves here, however, to an analysis of the distribution of the level populations with respect to the angular-momentum directions, to a calculation of the polarization of the medium under the influence of intense elliptically polarized radiation, and to an analysis of the dynamics of the polarization of the radiation in the course of the propagation of the latter.

2. CLASSICAL EQUATIONS FOR THE DENSITY MATRIX

In the initial (quantum) equations for the density matrix, we shall focus our attention on the dynamic part responsible for the interaction with the radiation. As for the relaxation processes, we shall simplify as much as possible and start from the model of the relaxation constants. In this model the interaction of a quantum system with radiation that is at resonance with the $m-n$ transition with angular momenta J_m and J_n is described by the following equations for the density matrix in the M representation:

$$\begin{gathered}\left(\Gamma_m+\frac{d}{dt}\right)\rho_{mm}(M|M')=Q_m\delta_{MM'}\\ +i\sum_{M_1}[\rho_{mn}(M|M_1)V_{nm}(M_1|M')-V_{mn}(M|M_1)\rho_{nm}(M_1|M')],\\ \left(\Gamma_n+\frac{d}{dt}\right)\rho_{nn}(M|M')=Q_n\delta_{MM'}\\ +i\sum_{M_1}[\rho_{nm}(M|M_1)V_{mn}(M_1|M')-V_{nm}(M|M')\rho_{mn}(M_1|M')],\\ \left(\Gamma+\frac{d}{dt}\right)\rho_{mn}(M|M')\\ =i\sum_{M_1}[\rho_{mm}(M|M_1)V_{mn}(M_1|M')-V_{mn}(M|M_1)\rho_{nn}(M_1|M')],\\ \rho_{nm}(M|M')=\rho_{mn}^*(M'|M),\quad V_{nm}(M|M_1)=V_{mn}^*(M_1|M).\end{gathered}\tag{2.1}$$

Here Γ_m and Γ_n are the decay constants of the states m and n; Γ is the impact half-width of the line; Q_m and

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Q_n characterize the rates of excitation of the levels m and n. The excitation is assumed to be isotropic.

We assume next satisfaction of the condition

$$J_m,\ J_n \gg 1, \tag{2.2}$$

which permits a classical description of the orientational states of the angular momenta J_m and J_n.

In place of the density matrix elements $\rho_{ij}(M|M')$ we introduce

$$\rho_{ij}(\bar{M},\ \mu) \equiv \rho_{ij}(\bar{M}+\mu/2|\bar{M}-\mu/2), \qquad \mu=M-M',\ \ \bar{M}={}^1/_2(M+M'). \tag{2.3}$$

The reasoning continues as follows. In classical physics there is no place for the state interference described in the quantum case by the off-diagonal elements of the density matrix. In other words, in the classical limits the correlation of states with different values of the corresponding quantum number should be weak. In particular, for a classical description of the orientational states of the angular momentum it is necessary to stipulate a weak correlation of states with different values of the angular-momentum projection.

For the quantities $\rho_{ij}(\bar{M},\mu)$ this requirement means that the effective internal μ_{eff} of the variable μ (the interval in which the values of ρ_{ij} differ substantially from zero) should be much less than the characteristic scale $\Delta\bar{M}$ of the change of ρ_{ij} relative to the variable $\bar{M}$ ($\mu_{eff} \ll \Delta\bar{M}$). We assume that this condition is satisfied.

We introduce the following representation for the density-matrix elements:

$$\rho_{ij}(\vartheta\varphi)=\sum_{\mu} e^{-i\mu\varphi}\rho_{ij}(\bar{M},\mu), \quad \cos\vartheta=\bar{M}/J,\ \ J={}^1/_2(J_m+J_n). \tag{2.4}$$

Under the conditions $\mu_{eff} \ll \Delta\bar{M}$, the summation over μ can certainly be extended to infinite limits, after which the transformation (2.4) acquires the meaning of a full-fledged Fourier transformation. The inverse transformation is of the form

$$\rho_{ij}(\bar{M},\mu)=\frac{1}{2\pi}\int_0^{2\pi} e^{i\mu\varphi}\rho_{ij}(\vartheta\varphi)\,d\varphi. \tag{2.5}$$

Relations similar to (2.4) and (2.5) hold also for the interaction matrix element $V_{mn}(M|M_1)$. We note that the transformation (2.4) is perfectly analogous to a transition to the Wigner representation in the case of translational degrees of freedom (see, e.g., Ref. 2). The angles ϑ and φ characterize the direction of the vector $\mathbf{J}$.

Since the density matrix is Hermitian, the following relation is valid

$$\rho_{ij}(\vartheta\varphi)=\rho_{ji}^*(\vartheta\varphi). \tag{2.6}$$

The density-matrix elements diagonal in the energy indices turn out to be real and describe the distribution of the populations over the angular-momentum orientations.

We change over to the $\vartheta\varphi$ representation in Eqs. (2.1). With the first equation of (2.1) is the example, we have

$$\begin{gathered}\left(\Gamma_m+\frac{d}{dt}\right)\rho_{mm}(\vartheta\varphi)=Q_m+i\sum_{\mu_1\mu_2}e^{-i(\mu_1+\mu_2)\varphi}\\ \times[\rho_{mn}(\bar{M}_1,\mu_1)V_{nm}(\bar{M}_2,\mu_2)-V_{mn}(\bar{M}_1,\mu_1)\rho_{nm}(\bar{M}_2,\mu_2)],\\ \bar{M}_1={}^1/_2(M+M_1),\quad \bar{M}_2={}^1/_2(M_1+M'),\quad \mu_1=M-M_1,\\ \mu_2=M_1-M',\quad \bar{M}={}^1/_2(M+M'),\quad \cos\vartheta=\bar{M}/J.\end{gathered} \tag{2.7}$$

The values of $\bar{M}_1$ and $\bar{M}_2$ differ from $\bar{M}$ by not more than μ_{eff} and can be replaced by $\bar{M}$, in view of the smooth dependences of ρ_{mn} and V_{mn} on $\bar{M}_1$ (or on $\bar{M}_2$) in the μ_{eff} scale. As a result, the sum over μ_1 and μ_2 is transformed into a product of sums, each of which effects a transition to the $\vartheta\varphi$ representation.

Thus, when the conditions for the classical description of the orientational states of the angular momenta are satisfied, the equations for the density matrix in the $\vartheta\varphi$ representation are diagonalized and take the form[1)]

$$\begin{aligned}&\left(\Gamma_m+\frac{d}{dt}\right)\rho_{mm}(\vartheta\varphi)=Q_m+2\,\mathrm{Re}[iV_{mn}^*(\vartheta\varphi)\rho_{mn}(\vartheta\varphi)],\\ &\left(\Gamma_n+\frac{d}{dt}\right)\rho_{nn}(\vartheta\varphi)=Q_n-2\,\mathrm{Re}[iV_{mn}^*(\vartheta\varphi)\rho_{mn}(\vartheta\varphi)],\\ &\left(\Gamma+\frac{d}{dt}\right)\rho_{mn}(\vartheta\varphi)=iV_{mn}(\vartheta\varphi)[\rho_{mm}(\vartheta\varphi)-\rho_{nn}(\vartheta\varphi)].\end{aligned} \tag{2.8}$$

These equations differ greatly from the known equations of the nondegenerate-state model in that the density-matrix elements have an additional (parametric) dependence on the angles that characterize the direction of the angular momentum. Just as in the nondegenerate-state model, they can be solved without a restriction on the radiation intensity, and what is particularly important, at an arbitrary polarization of the radiation.

3. POPULATION DISTRIBUTION IN THE ANGULAR-MOMENTUM DIRECTIONS

We consider an interaction with monochromatic radiation having a frequency ω close to the transition frequency ω_{mn}, and separate the time dependence in the interaction matrix element

$$V_{mn}(\vartheta\varphi)=\tilde{V}_{mn}(\vartheta\varphi)\exp(-i\Omega t), \qquad \Omega=\omega-\omega_{mn}, \tag{3.1}$$

where an excitation that is constant in time, Eqs. (2.8) reduce to the following algebraic equations:

$$\begin{gathered}\Gamma_m\rho_{mm}(\vartheta\varphi)=Q_m+2\mathrm{Re}[i\tilde{V}_{mn}^*(\vartheta\varphi)\tilde{\rho}_{mn}(\vartheta\varphi)],\\ \Gamma_n\rho_{nn}(\vartheta\varphi)=Q_n-2\mathrm{Re}[i\tilde{V}_{mn}^*(\vartheta\varphi)\tilde{\rho}_{mn}(\vartheta\varphi)],\\ (\Gamma-i\Omega)\tilde{\rho}_{mn}(\vartheta\varphi)=i\tilde{V}_{mn}(\vartheta\varphi)[\rho_{mm}(\vartheta\varphi)-\rho_{nn}(\vartheta\varphi)],\\ \rho_{mn}(\vartheta\varphi)=\tilde{\rho}_{mn}(\vartheta\varphi)\exp(-i\Omega t).\end{gathered} \tag{3.2}$$

The solution of these equations can be easily obtained

$$\begin{gathered}\rho_{mm}(\vartheta\varphi)=\frac{Q_m+Q_n}{\Gamma_m+\Gamma_n}-\frac{\Gamma_n}{\Gamma_m+\Gamma_n}N(\vartheta\varphi),\\ \rho_{nn}(\vartheta\varphi)=\frac{Q_m+Q_n}{\Gamma_m+\Gamma_n}+\frac{\Gamma_m}{\Gamma_m+\Gamma_n}N(\vartheta\varphi),\\ \rho_{mn}(\vartheta\varphi)=iV_{mn}(\vartheta\varphi)N(\vartheta\varphi)/(\Gamma-i\Omega),\\ N(\vartheta\varphi)\equiv\rho_{nn}(\vartheta\varphi)-\rho_{mm}(\vartheta\varphi)\\ =N_0\left[1+\frac{2\Gamma}{\Gamma^2+\Omega^2}\left(\frac{1}{\Gamma_m}+\frac{1}{\Gamma_n}\right)|\tilde{V}_{mn}(\vartheta\varphi)|^2\right]^{-1},\quad N_0=\frac{Q_n}{\Gamma_n}-\frac{Q_m}{\Gamma_m}.\end{gathered} \tag{3.3}$$

The quantity N_0 has the meaning of the difference between the level populations in the absence of radiation.

The dependence on the angles ϑ and φ in (3.3) is due to the interaction matrix element $\tilde{V}_{mn}(\vartheta\varphi)$. We shall hereafter consider electric dipole interaction, for which we have in the M representation

$$\tilde{V}_{mn}(M|M_1)=-\frac{1}{\sqrt{3}}\sum_{\sigma}(-1)^{J_n-M_1}G_\sigma\langle J_m M J_n -M_1|1\sigma\rangle, \quad G_\sigma=\frac{E_\sigma d_{mn}}{2\hbar}. \tag{3.4}$$

Here d_{mn} is the reduced matrix element of the dipole moment, E_σ are the circular components of the complex amplitude of the radiation electric vector, $\langle\cdots|\cdots\rangle$ is the vector-addition coefficient.

Using the asymptotic form ($J_m, J_n \gg 1$) of the vector-addition coefficients[3] and changing over in (3.4) to the $\vartheta\varphi$ representation, we obtain

$$\tilde{V}_{mn}(\vartheta\varphi)=-\frac{1}{(2J)^{1/2}}\sum_{\sigma}G_\sigma D^{1*}_{\sigma\Delta}(\varphi\vartheta 0), \quad \Delta=J_m-J_n, \tag{3.5}$$

where the dependence on ϑ and φ is concentrated in the Wigner D-functions $D^{1*}_{\sigma\Delta}(\varphi\vartheta 0)$.

Even simpler is the form of $\tilde{V}_{mn}(\vartheta\varphi)$ at $\Delta=0$ (J–J transition):

$$\tilde{V}_{mn}(\vartheta\varphi)=\frac{1}{(2J)^{1/2}}\mathbf{G}\mathbf{n}, \tag{3.6}$$

where the vector $\mathbf{G}$ is proportional to the complex amplitude of the electric-field vector, and $\mathbf{n}$ is a unit vector along the $\mathbf{J}$ direction.

For elliptically polarized radiation, we introduce the vectors $\mathbf{G}_\parallel$ and $\mathbf{G}_\perp$ directed along the major and minor axes of the polarization ellipse, respectively. Then

$$\mathbf{G}=\mathbf{G}_\parallel+i\mathbf{G}_\perp, \quad \mathbf{G}_\parallel\mathbf{G}_\perp=0. \tag{3.7}$$

The vectors $\mathbf{G}_\parallel$ and $\mathbf{G}_\perp$ can be regarded as real without loss of generality. We choose a coordinate system with z axis along the wave vector, and direct the x and y axis along $\mathbf{E}_\parallel$ and $\mathbf{E}_\perp$ respectively.

In the case of the J–J ($\Delta=0$) transition we obtain the following dependence on the angles for the population difference $N(\vartheta\varphi)$, in terms of which the solution (3.3) of Eqs. (3.2) is expressed:

$$N(\vartheta\varphi)=N_0[1+\varkappa_\parallel(\cos^2\varphi+\varepsilon^2\sin^2\varphi)\sin^2\vartheta]^{-1},$$

$$\varkappa_{\parallel,\perp}=\frac{2\Gamma}{\Gamma^2+\Omega^2}\left(\frac{1}{\Gamma_m}+\frac{1}{\Gamma_n}\right)\frac{1}{2J}G^2_{\parallel,\perp}, \quad \varepsilon^2=\varkappa_\perp/\varkappa_\parallel. \tag{3.8}$$

The quantity ε characterizes the degree of ellipticity. The value $\varepsilon=0$ corresponds to linear polarization and $\varepsilon=1$ to circular polarization. The quantity $\varkappa_{\parallel,\perp}$ will be called the saturation parameter, in analogy with the nondegenerate-state model. With increasing radiation intensity, the saturation parameter increases and the population difference decreases correspondingly. There exist, however, $\mathbf{J}$ directions for which there is no interaction with the field at all. For a polarization different from linear, these are the directions collinear with the wave vector. For linear polarization ($\varepsilon=0$) there is no interaction if the vector $\mathbf{J}$ lies in a plane perpendicular to the electric vector of the wave.

At a large saturation parameter ($\varkappa_{\parallel,\perp}\gg 1$) the populations become equalized for all the orientation angles of $\mathbf{J}$, with the exception of a narrow interval in the vicinity of the $\mathbf{J}$ directions indicated above. Figure 1 shows examples of the distributions of the populations of the lower state (n) in the $\mathbf{J}$ directions in the case of radiation absorption ($N_0>0$). A spherical coordinate system is used with origin at the center of the marked square (the symmetry plane). The twofold structure of the distribution can be easily traced. Highly selective angular structures stand out against the background of the isotropic part corresponding to the term $(Q_m+Q_n)/(\Gamma_m+\Gamma_n)$ in expression (3.3) for $\rho_{nn}(\vartheta\varphi)$. In the case of circular polarizations these are "peaks" at angles $\theta=0$ and π, and for linear polarizations—a narrow disk in the yz plane. It is obvious that the selective parts in the $\rho_{mm}(\vartheta\varphi)$ distribution should be "negative," i.e., have the form of "dips" for circular polarization and of an annular "valley" for linear polarization.

The characteristic angular dimension of the selective parts of the distribution at $\varkappa_\parallel\gg 1$ can be easily estimated with the aid of (3.8). At $\varepsilon=1$ we have

$$\Delta\nu\sim 1/\varkappa_\parallel^{1/2}. \tag{3.9}$$

For linear polarization, the angular dimension of the disk can be estimated from the same formula, as can be verified by putting $\varphi=0$ in (3.8).

Outwardly, the selective parts in the distributions $\rho_{mm}(\vartheta\varphi)$ and $\rho_{nn}(\vartheta\varphi)$ recall the known Bennett dips and peaks in the velocity distributions for a large dipole broadening (see, e.g., Refs. 1 and 4). They have also a farther reaching similarity: both are due to selectivity (in one case with respect to velocity, and in the

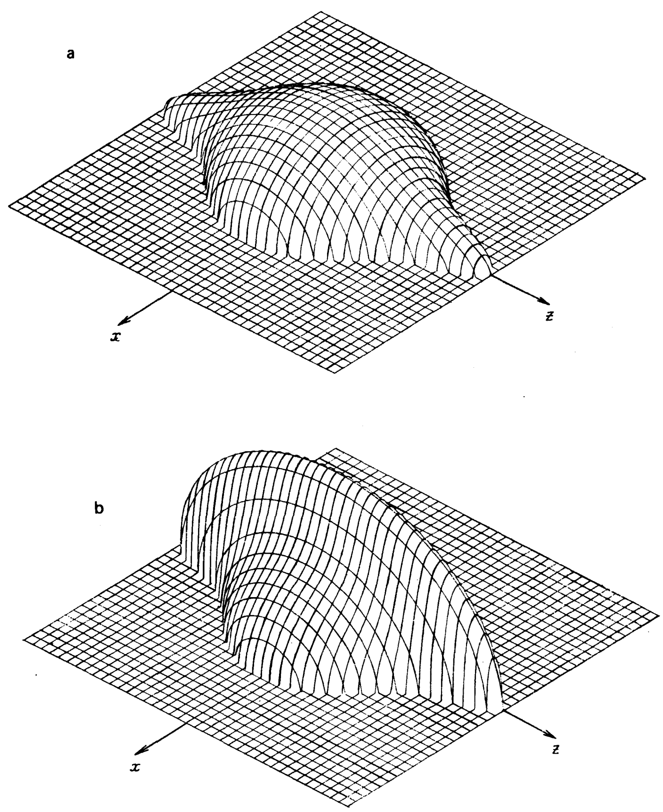

FIG. 1. Distribution of the population of the level n with respect to the directions of the angular momentum in the absorption regime (J–J transition); a—circular polarization of the radiation, b—linear polarization. $\varkappa=80$ throughout.

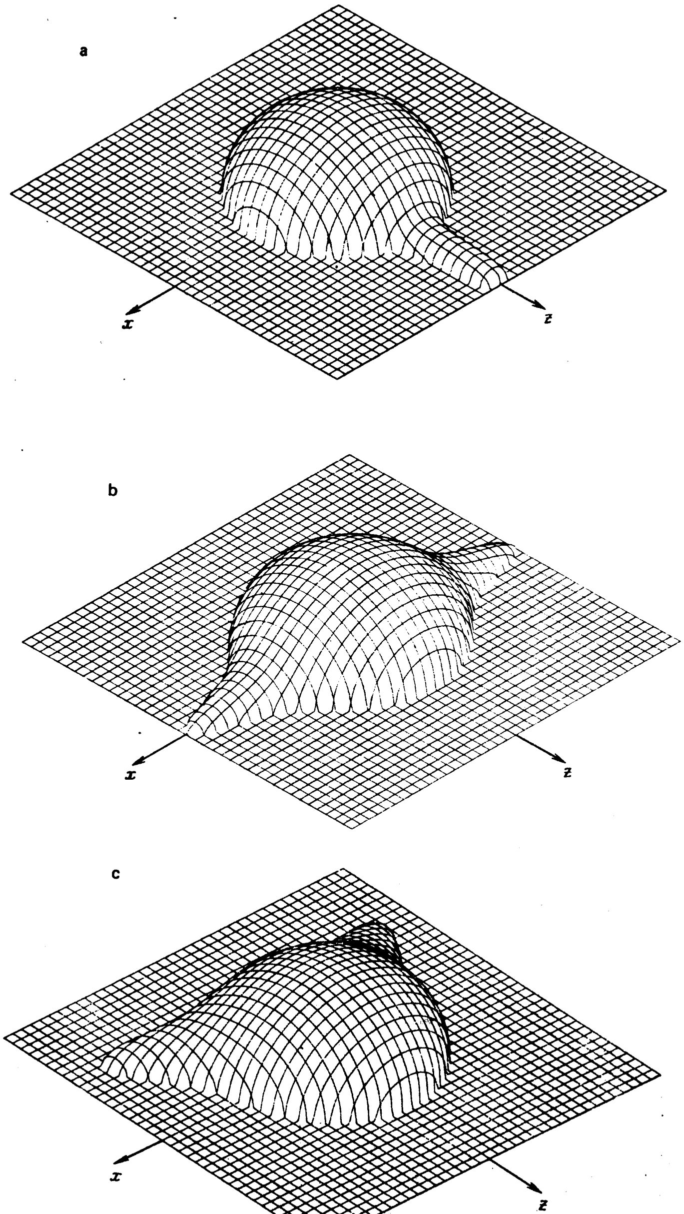

FIG. 2. The same as Fig. 1, but for the transition $J_m = J_n + 1$; a—circular polarization, $\varkappa = 8000$; b—linear polarization, $\varkappa = 80$; c—elliptic polarization, $(\varepsilon = 0.5,\ \varkappa = 80)$.

other with respect to angles) of the interaction with the radiation. There is also a radical difference. The selectivity in velocity decreases with increasing radiation intensity, whereas those parts of the distributions $\rho_{mm}(\vartheta\varphi)$ and $\rho_{nn}(\vartheta\varphi)$ that are selective in the angle become narrower with increasing intensity. The narrowing is limited only by the quantum uncertainty of the orientation angles of the angular momentum **J**.

For the $J_m = J_n + 1$ transition we obtain on the basis of expressions (3.3) and (3.5) the following dependence of the population difference on the angles of the orientation of **J** (the coordinate system is the same as before):

$$N(\vartheta\varphi) = N_0\left[1 + \frac{\varkappa_\parallel}{2}\left((\cos\vartheta - \varepsilon)^2 \sin^2\varphi + (\varepsilon\cos\vartheta - 1)^2\cos^2\varphi\right)\right]^{-1}. \tag{3.10}$$

In this case there likewise exist directions of **J** at which the particle does not interact with the field. If the radiation is circularly polarized ($\varepsilon = 1$), this direction coincides with the direction of the wave vector ($\vartheta = 0$). For linear polarization there are two such directions, $\varphi = 0$ and $\pi = \pm\pi/2$. At large saturation parameters, the population distributions in the directions of **J** also reveal strongly selective parts. Typical examples of such distributions for the population of the lower state $\rho_{nn}(\vartheta\varphi)$ are shown in Fig. 2 (case of absorption). If the polarization differs noticeably from circular the selective parts of the distribution, at the same saturation parameters, have approximately the same angular dimension as for the J–J transition see (3.9) . For circular polarization, the interaction selectivity with respect to the orientations of J in the case of the $J_m = J_n + 1$ transition decreases sharply. Indeed, putting $\varepsilon = 1$ in (3.10), we obtain for the angular dimension of the selective part at $\varkappa_\parallel \gg 1$

$$\Delta\vartheta \sim (8/\varkappa_\parallel)^{1/4}. \tag{3.11}$$

This circumstance is well illustrated in Fig. 2(a). To obtain just as narrow a selective increment as in the preceding cases, it is necessary to increase substantially the saturation parameter.

4. DYNAMICS OF POLARIZATION OF INTENSE RADIATION IN THE PROPAGATION PROCESS

For optical and spectroscopic problems, and in particular for the analysis of the effects of radiation propagation, it is necessary to know the radiation-induced polarization of the medium. The circular components P_σ of the complex polarization vector P are expressed in the following manner in terms of the off-diagonal element of the density matrix (see, e.g., Ref. 5)

$$P_\sigma = \frac{d_{mn}}{\sqrt{3}} \sum_{MM'} \tilde{\rho}_{mn}(M|M')(-1)^{J_n - M'} \langle J_m M J_n - M' | 1\sigma \rangle. \tag{4.1}$$

We change over in this equation to the $\vartheta\varphi$ representation and use the connection, given by (3.3), between $\rho_{mn}(\vartheta\varphi)$ and the population difference $N(\vartheta\varphi)$:

$$P_\sigma = \frac{1}{4\pi}\frac{id_{mn}}{\Gamma - i\Omega}\int_0^\pi \sin\vartheta\, d\vartheta \int_0^{2\pi} d\varphi \sum_{\sigma'} G_{\sigma'} D^{1*}_{\sigma'\Delta}(\varphi\vartheta 0) D^1_{\sigma\Delta}(\varphi\vartheta 0) N(\vartheta\varphi). \tag{4.2}$$

In what follows we shall need to specify the type of transition. We consider first the J–J transition ($\Delta = 0$). Using expression (3.8) for $N(\vartheta\varphi)$ and the explicit form of the D-matrices,[3] we obtain after integrating in (4.2) with respect to the angle φ:

$$P_\sigma = \frac{id_{mn} N_0 G_\sigma}{4(\Gamma - i\Omega)} \int_0^\pi \sin\vartheta\, d\vartheta \frac{1}{\varkappa_\sigma}\left[1 - \frac{1 + (\varkappa_{-\sigma} - \varkappa_\sigma)(\sin^2\vartheta)/2}{[1 + (\varkappa_\sigma + \varkappa_{-\sigma})\sin^2\vartheta + (\varkappa_\sigma - \varkappa_{-\sigma})^2(\sin^4\vartheta)/4]^{1/2}}\right],$$
$$\varkappa_\sigma = \frac{2\Gamma}{\Gamma^2 + \Omega^2}\left(\frac{1}{\Gamma_m} + \frac{1}{\Gamma_n}\right)\frac{1}{2J} G_\sigma^2. \tag{4.3}$$

where an electric field with radiation of the type (3.7) with real $G_\parallel$ and $G_\perp$, circular components of the vector **G** are also real:

$$G_1 = (G_\perp - G_\parallel)/\sqrt{2}, \quad G_{-1} = (G_\perp + G_\parallel)/\sqrt{2}. \tag{4.4}$$

It is possible to integrate in (4.3) with respect to the angle ϑ and to reduce the result to tabulated functions—elliptic integrals. However, the integral representation is also perfectly convenient both from the point of view of numerical calculations and for the analysis of limiting cases. In particular, if the radiation polarization is close to circular (e.g., $\varkappa_{-1} \ll 1$), then we obtain on the basis of (4.3)

$$P_1 = \frac{id_{mn}N_0}{\Gamma - i\Omega}\frac{1}{\varkappa_1}\left\{1 - \frac{1}{[\varkappa_1(2+\varkappa_1)]^{1/2}}\ln\left[\frac{(2+\varkappa_1)^{1/2}+\varkappa_1^{1/2}}{(2+\varkappa_1)^{1/2}-\varkappa_1^{1/2}}\right]\right\}G_1, \quad (4.5)$$

$$P_{-1} = \frac{id_{mn}N_0}{\Gamma - i\Omega}\frac{1}{\varkappa_1(2+\varkappa_1)}\left\{\frac{1+\varkappa_1}{[\varkappa_1(2+\varkappa_1)]^{1/2}}\ln\left[\frac{(2+\varkappa_1)^{1/2}+\varkappa_1^{1/2}}{(2+\varkappa_1)^{1/2}-\varkappa_1^{1/2}}\right]-1\right\}G_{-1}. \quad (4.6)$$

The imaginary and real parts of the factors preceding G_1 and G_{-1} in (4.5) and (4.6) are proportional respectively to the absorption (amplification) and refraction coefficients. It is easy to verify, by comparing (4.6) with (4.5), that the absorption (amplification) coefficient for a weak circular component of the radiation is smaller than for a strong one at all values of $\varkappa_1$. It follows therefore that if the radiation has a polarization that differs somewhat from circular and propagates in an amplifying medium ($N_0 < 0$), then with further propagation its polarization tends to become circular. In other words, circular polarization is stable for the J–J transition in the amplification regime. On the contrary, in an absorbing medium the deviation from circular polarization accumulates as the radiation propagates, i.e., the circular polarization is unstable.

Let us clarify the question of the stability of the linear polarization. To this end we calculate the Cartesian components of the polarization vector of the medium. Using the connection between the circular and Cartesian components

$$P_\parallel = -(P_1 + iP_{-1})/\sqrt{2}, \quad P_\perp = (P_1 - iP_{-1})/\sqrt{2}, \quad (4.7)$$

we obtain on the basis of (4.2)

$$P_\parallel = \frac{id_{mn}N_0}{\Gamma - i\Omega}\frac{G_\parallel}{2}\int_0^\pi \frac{(1+\varkappa_\parallel \sin^2\vartheta)^{1/2} - (1+\varkappa_\perp \sin^2\vartheta)^{1/2}}{(1+\varkappa_\perp \sin^2\vartheta)^{1/2}}\sin\vartheta\, d\vartheta. \quad (4.8)$$

The expression for $P_\perp$ is obtained from (4.8) by making the substitutions $G_\parallel \to G_\perp$ and $\varkappa_\parallel \leftrightarrow \varkappa_\perp$. We assume that $\varkappa_\perp \ll \varkappa_\parallel$, 1 and after integrating with respect to the angle ϑ we arrive at the relations

$$P_\parallel = \frac{id_{mn}N_0}{\Gamma - i\Omega}\frac{1}{\varkappa_\parallel}\left[1 - \frac{\operatorname{arctg}\varkappa_\parallel^{1/2}}{\varkappa_\parallel^{1/2}}\right]G_\parallel, \quad (4.9)$$

$$P_\perp = \frac{id_{mn}N_0}{\Gamma - i\Omega}\frac{1}{2\varkappa_\parallel}\left[(1+\varkappa_\parallel)\frac{\operatorname{arctg}\varkappa_\parallel^{1/2}}{\varkappa_\parallel^{1/2}} - 1\right]G_\perp. \quad (4.10)$$

Comparison of these equations shows that the absorption (amplification) coefficient of the weak component ($G_\perp$) is larger at all $\varkappa_\parallel$ than that for the strong one ($G_\parallel$). Consequently, the linear polarization is unstable in an amplifying medium and stable in an absorbing medium.

It follows from the foregoing analysis that in the general case when intense radiation propagates in a resonant medium its polarization state changes. For the resonant J–J transition the polarization tends to be circular in an amplifying medium and linear in an absorbing medium. At exact resonance ($\Omega = 0$) the phase relations between the components of the electric vector of the wave (both circular and Cartesian) are preserved. Consequently, the orientation of the polarization-ellipse axis remains unchanged, only the degree of ellipticity changes. On the other hand if $\Omega \neq 0$, the refractive indices for the different wave components are different and the change of the degree of ellipticity is simultaneously accompanied by a rotation of the polarization-ellipse axis.

The question of the change of the polarization of radiation propagating in a resonant medium was already discussed in the literature relatively long ago, principally in connection with an investigation of the polarization of laser radiation.[6-8] However, only the first-order linear corrections were taken into account in the analysis. Using our equations (3.2) for the density matrix, we can lift the restriction on the intensity of the radiation and trace completely the dynamics of its polarization in the course of propagation.

The compact Maxwell's equations can be transformed to the following equations for the intensities of the circular components of the radiation:

$$\frac{d}{dz}|G_\sigma|^2 = -\frac{4\pi d_{mn}\omega}{\hbar c}\operatorname{Re}[iG_\sigma^* P_\sigma], \quad \sigma = \pm 1. \quad (4.11)$$

These equations do not describe the change of the phase between the circular components, and a complete description of the state of polarization can be obtained on their basis only at $\Omega = 0$, a fact that we shall bear in mind.

Equations (4.11) were solved numerically with a computer, using Eq. (4.3) for P_σ. The amplification regime was considered. The results for the total intensity and the ellipticity parameter are shown in Fig. 3. Up to a certain distance, the intensity increases exponentially, after which the increase becomes linear. With respect to the polarization of the radiation we have the following result. Regardless of the initial state, the polarization tends in the course of propagation to become circular ($R \to 1$). The more the initial polarization differs from the unstable (linear) one, the faster its change. If the initial polarization is not too close to unstable, then the spatial scale of the variation is a distance of the order of the reciprocal gain.

We consider now a transition with a change of J, assuming for the sake of argument $J_m = J_n + 1$. After integrating in (4.2) with respect to the angle φ, we obtain ($\sigma = \pm 1$)

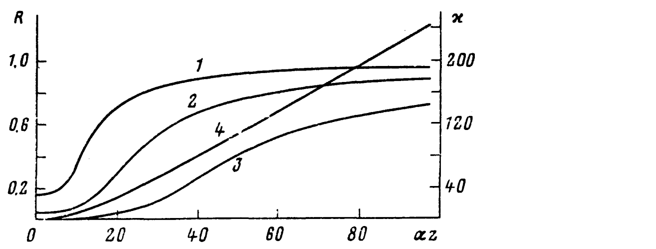

FIG. 3. Change of the polarization (curves 1–3) and of the intensity (curve 4) of the radiation in the case of propagation in an amplifying medium (J–J transition); $R = (\varkappa_{-1} - \varkappa_1)/\varkappa$; $\varkappa = \varkappa_1 + \varkappa_{-1}$; α is the linear amplification coefficient; 1) $R_0 = 0.143$; 2) 0.026; 3) 0.003; $R_0 = R_{z=0}$.

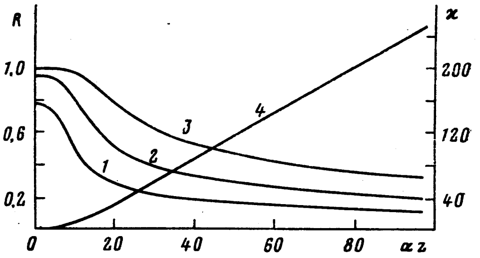

FIG. 4. The same as in Fig. 3, but for the transition $J_m = J_n + 1$; 1) $R_0 = 0.995$; 2) 0.951; 3) 0.778.

$$P_\sigma = \frac{id_{mn} N_0 G_\sigma}{2(\Gamma - i\Omega)} \int_0^\pi \sin\vartheta\, d\vartheta \frac{1}{2\varkappa_\sigma} \Big\{ 1 - \frac{1 + \varkappa_{-\sigma}\sin^4(\vartheta/2) - \varkappa_\sigma \cos^4(\vartheta/2)}{[1 + 2(\varkappa_\sigma \cos^4(\vartheta/2) + \varkappa_{-\sigma}\sin^4(\vartheta/2)) + (\varkappa_\sigma\cos^4(\vartheta/2) - \varkappa_{-\sigma}\sin^4(\vartheta/2))^2]^{1/2}} \Big\}. \quad (4.12)$$

In the limiting case $\varkappa_{-1} \ll \varkappa_1$, 1 (close to circular polarization) we obtain therefore

$$P_1 = \frac{id_{mn} N_0}{\Gamma - i\Omega} \frac{1}{\varkappa_1} \left[1 - \frac{\operatorname{arctg} \varkappa_1^{1/2}}{\varkappa_1^{1/2}} \right] G_1, \quad (4.13)$$

$$P_{-1} = \frac{id_{mn} N_0}{\Gamma - i\Omega} \frac{1}{2\varkappa_1} \left[(1 + \varkappa_1) \frac{\operatorname{arctg} \varkappa_1^{1/2}}{\varkappa_1^{1/2}} - 1 \right] G_{-1}. \quad (4.14)$$

We note the similarity of these expressions to Eqs. (4.9) and (4.10): in the approximation employed, the circular components P_σ depend in the case of a $\Delta \neq 0$ transition in exactly the same manner on the radiation intensity as the Cartesian components for the $\Delta = 0$ transition. From this it follows in particular that for the transition $\Delta = \pm 1$ the circular polarization of the radiation in an amplifying medium turns out to be unstable. Obviously, the linear polarization is stable in the amplification regime and the circular polarization in the absorption regime. Thus, with respect to stability of the radiation polarization we have for a transition in which the quantum number J changes a situation that is the converse of the situation in the case of the J–J transition.

The numerical calculation of the dynamics of the radiation polarization in an amplifying medium for the $\Delta = 1$ transition was carried out on the basis of Eqs. (4.11) and (4.12). Its results are shown in Fig. 4. The character of the change of the polarization in the course of propagation is practically the same as for the J–J transition, the only difference being that the polarization tends in the upshot not to circular but to linear. The characteristic spatial scale of the change of the polarization is, as before, the reciprocal gain.

5. LIMITS OF APPLICABILITY OF THE "CLASSICAL" EQUATIONS

We discuss in greater detail the limits of applicability of Eqs. (3.2). We consider for the sake of argument the transition J–J and change over to the M representation in expression (3.8) for the population difference:

$$N(\overline{M}, \mu) = \rho_{nn}(\overline{M}, \mu) - \rho_{mm}(\overline{M}, \mu) = \frac{N_0}{\{[1 + \varkappa_\parallel \sin^2\vartheta][1 + \varkappa_\perp \sin^2\vartheta]\}^{1/2}} \times \left\{ \frac{[(\varkappa_\parallel - \varkappa_\perp)\sin^2\vartheta]^{1/2}}{(1 + \varkappa_\parallel \sin^2\vartheta)^{1/2} + (1 + \varkappa_\perp \sin^2\vartheta)^{1/2}} \right\}^\mu. \quad (5.1)$$

$$\overline{M} = {}^1/_2(M + M'), \quad \mu = M - M', \quad \cos\vartheta = \overline{M}/J.$$

Here μ takes on only even values. From the obtained equation it is seen that the coherence of the magnetic sublevels M and M' decreases exponentially with increasing μ, since the base of the exponential function in (5.1) is always smaller than unity. The effective correlation interval is

$$\mu_{eff} \sim \ln^{-1} \left\{ \frac{(1 + \varkappa_\parallel \sin^2\vartheta)^{1/2} + (1 + \varkappa_\perp \sin^2\vartheta)^{1/2}}{[(\varkappa_\parallel - \varkappa_\perp)\sin^2\vartheta]^{1/2}} \right\}. \quad (5.2)$$

As already noted, for the transformations carried out in Sec. 2 to be valid, the condition $\mu_{eff} \ll \Delta\overline{M}$ must be satisfied. The most unfavorable situation is realized at $\vartheta \approx \pi/2$, $\varkappa_\perp \to 0$, and $\varkappa_\parallel \gg 1$, when

$$\mu_{eff} \sim \varkappa_\parallel^{1/2}. \quad (5.3)$$

Under the same conditions it is possible to obtain from (5.1)

$$\Delta\overline{M} \sim J/\varkappa_\parallel^{1/2}. \quad (5.4)$$

The requirement $\mu_{eff} \ll \Delta\overline{M}$ is satisfied under the condition

$$\varkappa_\parallel \ll J, \quad (5.5)$$

which is in fact the criterion for the validity of Eqs. (3.2). The physical meaning of this criterion can be easily understood by comparing it with relation (3.9). From (3.9), with allowance for (5.5) follows $\Delta\vartheta \gg 1/J^{1/2}$. This means that the characteristic angle scale of the population distribution in the directions of **J** should exceed the quantum uncertainty of the direction of **J**. Such a condition is perfectly natural in the classical description of the orientation of the angular momentum.

The criterion (5.5) is obtained in natural fashion also from an analysis of the angular distribution of the induced dipole moment; it remains in force also for transitions with change of the quantum number J.

We note that despite the restriction imposed by the condition (5.5), the radiation intensity can still be large enough to give rise to nonlinear effects. In particular, effective equalization of the populations takes place already at $\varkappa \sim 1$.

In experiments on nonlinear spectroscopy and nonlinear optics, the distributions of any quantity with respect to the directions is not measured directly, and certain properties averaged over the angles are registered. For example, the absorption (emission) probability w is given according to (3.2) and (3.5) by

$$w = \frac{\Gamma}{2\pi(\Gamma^2 + \Omega^2)} \int_0^{2\pi} d\varphi \int_0^\pi \sin\vartheta\, d\vartheta |V_{mn}(\vartheta\varphi)|^2 N(\vartheta\varphi). \quad (5.6)$$

As shown above, the strongest dependence on ϑ and φ in $N(\vartheta\varphi)$ is observed in the vicinity of angles that cause the quantity $V_{mn}(\vartheta\varphi)$ to vanish, and it is precisely in this vicinity that $N(\vartheta\varphi)$ is least accurately described by Eqs. (3.2). In (5.6), however, this angle region is discriminated by the factor $|V_{mn}(\vartheta\varphi)|^2$, and the errors in $N(\vartheta\varphi)$ influence the value of w to a much lesser degree. In particular, for linear or circular polarization of the radiation, the exact quantum-mechanical solution for w (see Ref. 1) goes over into the solution obtained from Eqs. (3.2) merely under the condition $J \gg 1$, without the additional requirement (5.5). For other polarizations, the condition should obviously remain the same. We note also that the population difference integrated over the angles which is connected with w by the relation

$$\frac{1}{4\pi}\int_0^{2\pi} d\varphi \int_0^{\pi} \sin\vartheta\, d\vartheta N(\vartheta\varphi) = N_0 - \left(\frac{1}{\Gamma_m} + \frac{1}{\Gamma_n}\right) w, \tag{5.7}$$

is seen to be calculated with the same accuracy as w. In other words, the integrated values of the selective angle structures discussed in Sec. 3 have a smaller error than the angular distributions themselves.

All this gives grounds for hoping that when solving nonlinear-spectroscopy or nonlinear optics problems, equations of the type (3.2) with a classical description of the orientation of the angular momentum are valid under a condition less stringent than (5.5). Namely, it sufficies to assume the only condition, which is natural for the classical approximation, $J \gg 1$.

It is easy to verify that processes of exchange of angular momentum between the radiation and the resonant particles are excluded from Eqs. (2.8) and (3.2). The action of the radiation reduces to transitions between energy levels without change in the direction of the angular momentum. Such an approximation is justified for many systems, and the condition of its applicability is a sufficiently rapid relaxation of the angular momentum with respect to the directions. It is possible, however, to include in the consideration the transfer of the angular momentum, and remain nevertheless within the framework of the classical description. To this end it is necessary to take into account in equations of the type (2.7) the small deviations of the quantities $\bar{M}_1$ and $\bar{M}_2$ from M. In the upshot we obtain in place of (2.8) the following equations for the density matrix:

$$\left(\Gamma_\varphi + \frac{d}{dt}\right)\rho_{mm}(\vartheta\varphi) = Q_m + \mathrm{Re}\left\{2iV^*_{mn}(\vartheta\varphi)\rho_{mn}(\vartheta\varphi) + \frac{1}{J}\left[\frac{\partial V^*_{mn}}{\partial\cos\vartheta}\frac{\partial\rho_{mn}}{\partial\varphi} - \frac{\partial\rho_{mn}}{\partial\cos\vartheta}\frac{\partial V^*_{mn}}{\partial\varphi}\right]\right\};$$

$$\left(\Gamma_n + \frac{d}{dt}\right)\rho_{nn}(\vartheta\varphi) = Q_n - \mathrm{Re}\left\{2iV^*_{mn}(\vartheta\varphi)\rho_{mn}(\vartheta\varphi) - \frac{1}{J}\left[\frac{\partial V^*_{mn}}{\partial\cos\vartheta}\frac{\partial\rho_{mn}}{\partial\varphi} - \frac{\partial\rho_{mn}}{\partial\cos\vartheta}\frac{\partial V^*_{mn}}{\partial\varphi}\right]\right\}, \tag{5.8}$$

$$\left(\Gamma + \frac{d}{dt}\right)\rho_{mn}(\vartheta\varphi) = iV_{mn}(\vartheta\varphi)\left[\rho_{mm}(\vartheta\varphi) - \rho_{nn}(\vartheta\varphi)\right] + \frac{1}{2J}\left[\frac{\partial V_{mn}}{\partial\cos\vartheta}\left(\frac{\partial\rho_{mm}}{\partial\varphi} + \frac{\partial\rho_{nn}}{\partial\varphi}\right) - \left(\frac{\partial\rho_{mm}}{\partial\cos\vartheta} + \frac{\partial\rho_{nn}}{\partial\cos\vartheta}\right)\frac{\partial V_{mn}}{\partial\varphi}\right].$$

Compared with (2.8), we have added here terms of differential type to the dynamic parts of the equations. They describe the change of the direction of the angular momentum under the influence of the radiation. It can be verified that when condition (5.5) is satisfied their contribution remains small.

We note that the differential terms in Eqs. (5.8) are reminiscent in form of the classical Poisson brackets. This similarity is not accidental. Assume that the interaction operator causes transitions only between magnetic sublevels of one energy state. For this purpose it is necessary to put formally $m = n$ in Eqs. (5.8). By virtue of the property in (2.6), the only dynamic terms that remain in these equations are those of the differential type. The equations themselves take the form of the known Liouville equations with classical Poisson brackets, as they should. The canonical conjugate variables are $\cos\vartheta$ and φ.

[1)]The range of validity of the equations derived is additionally discussed in Sec. 5.

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