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Determination of the relaxation characteristics by a polarization method in nonlinear spectroscopy

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Various aspects are considered of a new spectral method, free of Doppler broadening and proposed by Wieman, Hansch, *et al.* (Phys. Rev. Lett. **36**, 1170, 1976; Opt. Comm. **18**, 227, 1976). The method is based on registration of the signal of a test field passing through a crossed polaroid, with a definite choice of the polarization of the strong laser field that produces the nonlinear phenomena. It is noted in the cited papers that this "polarization" method is much more sensitive than earlier nonlinear spectroscopy methods. In the present paper it is shown theoretically that, in addition to sensitivity, the polarization method affords unique possibilities for the investigation of relaxation processes. It can be used to investigate the relaxation of the "polarization moment" of a quantum system (the total population of the levels, the orientation, the alignment) separately, something impossible with the earlier methods. An optimal experimental sequence is recommended for the investigation of the relaxation characteristics. In the analysis of concrete cases it is noted that in practice the homogeneous saturation characteristic of longlived systems (molecules, atomic metastable formations) should not appear in experiments with crossed polaroids. This conclusion is most important for the problem of developing lasers with very high frequency stability.

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1. INTRODUCTION

A new method of nonlinear spectroscopy has been recently proposed,[1,2] based on the following known phenomenon. Polarizing laser radiation produces anisotropy when it interacts nonlinearly with the medium: a gas medium becomes uniaxial (if the radiation is linearly polarized) or gyrotropic (in the case of circular polarization), and is characterized by phenomena typical of anisotropic media, such as dichroism and birefringence. In particular, a linearly polarized trial wave passing through a medium becomes elliptically polarized and at the same time the axes of the ellipse rotate. The gist of the method of[1,2] consists of recording the signal of the trial wave passing first through the medium and then through a polaroid crossed relative to the initial polarization of the wave. The resultant signal is due only to the anisotropy induced by the strong field, and it is therefore patently of nonlinear origin. It was proposed in[1,2] to use this "polarization" method for high-resolution spectroscopy in systems with Doppler-broadened lines. By registering only the nonlinear part of the signal, the influence of the Doppler broadening is eliminated. It was shown at the same time[1,2] that the polarization method is much more sensitive than the

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earlier methods of nonlinear spectroscopy. With respect to the sensitivity, this method is apparently capable of competing with another method whose experimental development has been initiated recently, and in which it is proposed to eliminate the influence of the Doppler broadening by using two-phonon processes with participation of photons that are close in frequency[3] (in a particular case—with participation of two photons with equal frequency[4]).

In this paper we wish to call attention to the fact that the polarization is highly promising not only (and not so much) because of its sensitivity but because of new unusual possibilities of investigating on its basis the relaxation processes in a medium. The point is the following. The levels of real systems (atoms or molecules) are degenerate with respect to the direction of the angular momentum, and in an analysis of relaxation processes it is important to take into account the relaxation produced in the sublevel system by the nonuniform population of the sublevels and the coherence between them. It is customary to characterize the distribution over the sublevels by means of the "polarization moments," such as the total population of the level, the orientation, the alignment, etc.[5,6] This representation is convenient from the point of view that in many cases polarization moments relax independently of one another. In ordinary methods of registration, relaxation characteristics of several polarization moments appear in the nonlinear spectra, and this raises substantial difficulties in the reduction of the experimental data.

It will be shown below that with the aid of the polarization method it is possible to investigate the relaxation of the polarization moments separately. On this basis, the problem of developing the complete relaxation picture is also simplified.

The anisotropy effects referred to above become stronger if the strong-field frequency is close to one of the natural frequencies of the system, as will be assumed from now on. We consider here two characteristic situations: the trial field is resonant with the same transition as the strong field (two-level system, Fig. 1a) or is resonant with an adjacent transition (three-level system, Fig. 1b).

We make a minimum number of assumptions with respect to the character of the relaxation, in order to show that the possibilities of the method with respect to the separation of the individual relaxation characteristics are not limited to particular relaxation models. Under these conditions, we calculate the induced dipole moment at the frequency of the trial field for a two-level level (Sec. 3) and three-level (Sec. 4) system. Section 5 consists of an analysis of the results, a discussion of the organization of polarization experiments in which individual relaxation characteristics are separated, and particular relaxation models are analyzed.

The theoretical description of systems with degenerate levels usually entails cumbersome forms not only as a result but of the equations themselves (see, e.g., [5-7]). In Sec. 2 is proposed a compact matrix

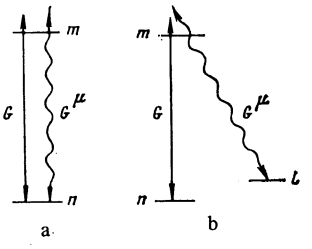


FIG. 1. Transitions between energy levels: a—two-level system; b—three-level system.

form of the equations for the elements of the density matrix and for the representation of the polarization moments. The matrix method of description is subsequently used extensively since it is the most convenient from the methodological point of view.

2. GENERAL EQUATIONS FOR DEGENERATE SYSTEMS

The equations for the elements of the density matrix, for systems with level degeneracy with respect to the projection of the angular momentum, can be represented in the form

$$\left(\frac{d}{dt}+\mathbf{v}\nabla+\Gamma_{kj}+i\omega_{kj}\right)\rho_{kj}=Q_{kj}+A_{kj}\rho_{kj}+i\sum_{l}\left(U_{lj}^{k}\rho_{kl}-U_{kl}^{j}\rho_{lj}\right). \quad (2.1)$$

The indices k, j, and l label here the energy levels. Each element ρ_{kj} constitutes a vector whose components are characterized by angle variables and a velocity; A_{kj} is a statistical operator that describes the result the collision; U_{lj}^{k} is a dynamic operator responsible for the interaction with the external field. Γ_{kj} and ω_{kj} are the constants of the radiative relaxation and of the frequency of the transition $k\rightarrow j$; Q_{kj} is the excitation vector.

In the irreducible tensor operator representation[5,6] (the polarization-moment representation or the $\varkappa q$ representation), the vectors and operators contained in (2.1) have the following structure and symmetry relations[1)]:

$$\begin{aligned}
\rho_{kj}(\varkappa q\mathbf{v})&=(-1)^{J_j-J_k+q}\rho_{jk}^{*}(\varkappa -q\mathbf{v}),\\
Q_{kj}(\varkappa q\mathbf{v})&=(-1)^{J_j-J_k+q}Q_{jk}^{*}(\varkappa -q\mathbf{v}),\\
A_{kj}(\varkappa q\mathbf{v}|\varkappa_1 q_1\mathbf{v}_1)&=(-1)^{q-q_1}A_{jk}^{*}(\varkappa -q\mathbf{v}|\varkappa_1 -q_1\mathbf{v}_1),\\
U_{lj}^{k}(\varkappa q\mathbf{v}|\varkappa_1 q_1\mathbf{v}_1)&=(-1)^{J_j-J_l+q-q_1}U_{jl}^{kk}(\varkappa -q\mathbf{v}|\varkappa_1 -q_1\mathbf{v}_1).
\end{aligned} \quad (2.2)$$

The quantity $\rho_{kj}(\varkappa q\mathbf{v})$ is connected with the elements of the density matrix in the JM representation by the following relation:

$$\rho_{kj}(\varkappa q\mathbf{v})=\sum_{M_kM_j}(-1)^{J_j-M_j}c(J_kJ_j\varkappa|M_k-M_jq)\rho_{J_kM_k;J_jM_j}(\mathbf{v}). \quad (2.3)$$

Here J_k and J_j are the total angular momenta of the levels k and j; M_k and M_j are their projections; $c(\ldots|\ldots)$ is a Clebsch-Gordan coefficient. The matrices U_{jl}^{k} are given explicitly by

$$\begin{aligned}
U_{jl}^{k}(\varkappa q\mathbf{v}|\varkappa_1 q_1\mathbf{v}_1)&=\delta(\mathbf{v}-\mathbf{v}_1)\sum_{\lambda\alpha}(-1)^{\varkappa_1+\lambda-J_k-J_l}\\
&\times[(2\lambda+1)(2\varkappa_1+1)]^{1/2}\begin{Bmatrix}\lambda & \varkappa_1 & \varkappa\\ J_k & J_l & J_j\end{Bmatrix}c(\lambda\varkappa_1\varkappa|\alpha q_1q)V_{jl}(\lambda\alpha),
\end{aligned} \quad (2.4)$$

where

$$V_{jl}(\lambda\alpha)=\sum_{M_jM_l}(-1)^{J_l-M_l}c(J_jJ_l\lambda|M_j-M_l\alpha)V_{J_jM_j;J_lM_l}.$$

Here $V_{J_jM_j;J_lM_l}$ is the matrix element of the interaction, $V_{jl}(\varkappa q)$ are the expansion coefficients in the irreducible tensor operators, and is the multipolarity of the interaction (we confine ourselves henceforth to the dipole approximation, i.e., $\lambda=1$).

Equations (2.1) will serve as the basis for the subsequent analysis.

3. TWO-LEVEL SYSTEM

We consider the solution of Eqs. (2.1) in the case when the trial field is resonant to the same transition as the strong field (two-level system, transition $m-n$, Fig. 1a). The matrix element of the interaction is given by

$$V_{mn}(1\alpha)=G_\alpha e^{-i(\omega t-\mathbf{kr})}+G_\alpha^\mu e^{-i(\omega_\mu t-\mathbf{k}_\mu \mathbf{r})}, \tag{3.1}$$

where

$$G_\alpha=E_\alpha d_{mn}/2\hbar,\ G_\alpha^\mu=E^\mu d_{mn}/2\hbar,$$

and E_α and E_α^μ are the cyclic components of the strong and trial waves; ω, ω_μ and $\mathbf{k}$, $\mathbf{k}_\mu$ are the frequencies and the wave vectors; d_{mn} is the reduced matrix element of the dipole moment. The density-matrix elements that play an important role in the problem have the following structure:

$$\begin{aligned}&\rho_{jj}=\rho_{jj}^0+R_{jj}+r_{jj}e^{-i\varphi}+\tilde r_{jj}e^{i\varphi},\ (j=m,n),\\ &\rho_{mn}=e^{-i(\omega t-\mathbf{kr})}(R_{mn}+r_{mn}e^{-i\varphi}+\tilde r_{mn}e^{i\varphi}),\\ &\varphi\equiv\varepsilon t-\mathbf{pr},\ \varepsilon=\omega_\mu-\omega,\ \mathbf{p}=\mathbf{k}_\mu-\mathbf{k}.\end{aligned} \tag{3.2}$$

Here ρ_{jj}^0 is the value of ρ_{jj} in the absence of external fields. On the other hand, the elements R and r satisfy under stationary and spatially-homogeneous conditions the following equations in matrix form (in perfect analogy to the case of nondegenerate systems[8]):

$$\hat\Lambda\hat R=i\hat U\hat\rho_0+i\hat U\hat R,\quad \hat\Lambda(\varepsilon')\hat r=i\hat u\hat\rho_0+\hat U\hat r, \tag{3.3}$$

$$\hat\Lambda\equiv\hat\Lambda(\varepsilon')_{\varepsilon'=0},\quad \varepsilon'=\varepsilon-\mathbf{pv}.$$

$$\hat U=\begin{pmatrix}0&0&U_{nm}^m&-U_{mn}^m\\0&0&-U_{nm}^n&U_{mn}^n\\U_{mn}^m&-U_{mn}^n&0&0\\-U_{nm}^m&U_{nm}^n&0&0\end{pmatrix},$$

$$\hat R=\begin{pmatrix}R_{mm}\\R_{nn}\\R_{mn}\\R_{nm}\end{pmatrix},\quad \hat r=\begin{pmatrix}r_{mm}\\r_{nn}\\r_{mn}\\\tilde r_{nm}\end{pmatrix},\quad \hat\rho_0=\begin{pmatrix}\rho_{mm}^0\\\rho_{nn}^0\\0\\0\end{pmatrix},$$

$$\hat u=\begin{pmatrix}0&0&0&-\tilde u_{mn}^m\\0&0&0&u_{mn}^n\\u_{mn}^m&-\tilde u_{mn}^n&0&0\\0&0&0&0\end{pmatrix},\quad \hat\Lambda=\begin{pmatrix}\Lambda_{mm}&0&0&0\\0&\Lambda_{nn}&0&0\\0&0&\Lambda_{mn}&0\\0&0&0&\Lambda_{nm}\end{pmatrix}, \tag{3.4}$$

$$\Lambda_{mm}=(\Gamma_{mm}-i\varepsilon')I-A_{mm},\ \Lambda_{nn}=(\Gamma_{nn}-i\varepsilon')I-A_{nn},$$
$$\Lambda_{mn}=(\Gamma_{mn}-i\Omega'-i\varepsilon')I-A_{mn},\ \Lambda_{nm}=(\Gamma_{nm}+i\Omega'-i\varepsilon')I-A_{nm},$$
$$\Omega'=\Omega-\mathbf{kv},\ \Omega=\omega-\omega_{mn},\ I(\varkappa qv|\varkappa_1q_1v_1)=\delta(\mathbf{v}-\mathbf{v}_1)\delta_{\varkappa\varkappa_1}\delta_{qq_1}.$$

Each matrix element or vector element in (3.3) and (3.4) is in turn a matrix or a vector, respectively. Thus, we are dealing with the so called supermatrices and supervectors. The matrices U_{jl}^k and u_{jl}^k are given by formula (2.4) in which $V_{jl}(\lambda\alpha)$ is replaced respectively by the quantities G_α and $G_\alpha^\mu(\lambda=1)$.

The next task is the solution of Eqs. (3.3) by iteration with respect to the strong field. Bearing in mind the general character of the analysis, it is convenient to introduce for the operators Λ_{jk} the Green's functions defined by the equation

$$\Lambda_{jk}(\varkappa qv|\varkappa_1q_1v_1)f_{jk}(\varkappa_1q_1v_1|\varkappa_0q_0v_0)=\delta(\mathbf{v}-\mathbf{v}_0)\delta_{\varkappa\varkappa_0}\delta_{qq_0}. \tag{3.5}$$

The solution of (3.3) by iteration is obtained with the aid of the Green's function in elementary fashion. Thus, for the matrix element r_{mn}, which describes the polarization induced at the frequency of the trial field, we obtain the following result:

$$\begin{aligned}r_{mn}=if_{mn}\{&(u_{mn}{}^m\rho_{mm}{}^0-\tilde u_{mn}{}^n\rho_{nn}{}^0)\\&-[(U_{mn}{}^mf_{mm}U_{nm}{}^m+\tilde U_{mn}{}^nf_{nn}\tilde U_{nm}{}^n)f_{mn}(u_{mn}{}^m\rho_{mm}{}^0-\tilde u_{mn}{}^n\rho_{nn}{}^0)\\&+(U_{mn}{}^mf_{mm}\tilde u_{mn}{}^m+\tilde U_{mn}{}^nf_{nn}u_{mn}{}^n)F_{nm}(\tilde U_{nm}{}^m\rho_{mm}{}^0-U_{nm}{}^n\rho_{nn}{}^0)\\&+(u_{mn}{}^mF_{mm}U_{nm}{}^m+\tilde u_{mn}{}^nF_{nn}\tilde U_{nm}{}^n)F_{mn}(U_{mn}{}^m\rho_{mm}{}^0-\tilde U_{mn}{}^n\rho_{nn}{}^0)\\&+(u_{mn}{}^mF_{mm}\tilde U_{mn}{}^m+\tilde u_{mn}{}^nF_{nn}U_{mn}{}^n)F_{nm}\cdot(\tilde U_{nm}{}^m\rho_{mm}{}^0-U_{nm}{}^n\rho_{nn}{}^0)]\}\end{aligned} \tag{3.6}$$

Here F_{jk} is the Green's function for the operators Λ_{jk} and $\varepsilon'=0$. Unfortunately, without specifying the form of the Green's function it is impossible to simplify the result (3.6) significantly. We therefore make several simplifying assumptions concerning the character of the relaxation. We start from the model of the isotropic collisional perturbation.[5] On the basis of the ideas of of[5,9], we can assume that the Green's functions have the following structure[2)]:

$$f_{jk}(\varkappa qv|\varkappa_1q_1v_1)=\delta_{\varkappa\varkappa_1}\delta_{qq_1}f_{jk}^{(\varkappa)}(\mathbf{v}|\mathbf{v}_1) \tag{3.7}$$

and analogously for F_{jk}. It is assumed, as usual, that in the absence of fields the population distribution over the levels is Maxwellian in the velocities and uniform in the magnetic sublevels, i.e.,

$$\rho_{jj}^0(\varkappa qv)=N_j(2J_j+1)^{1/2}\delta_{\varkappa0}\delta_{q0}W(\mathbf{v}). \tag{3.8}$$

In optical experiments, a quantity $r_{mn}(\varkappa qv)$ appears with a value $\varkappa=1$ (the induced dipole moment). We obtain for this quantity the following final expression, averaged over the velocities:

$$\begin{aligned}r_{mn}(1\alpha)\equiv\int r_{mn}(1\alpha v)\,dv=iN\Big\{&G_\alpha{}^\mu a_{mn}-9\sum_{\varkappa q\alpha_1}(-1)^{1-\alpha_1}c(11\varkappa|\alpha-\alpha_1q)\\&\times[G_{\alpha_1}J(\varkappa q)\beta_{mn}(\varkappa)+G_{\alpha_1}{}^\mu I(\varkappa q)B_{mn}(\varkappa)]\Big\}.\end{aligned} \tag{3.9}$$

We have introduced here the notation

$$\begin{aligned}\beta_{mn}(\varkappa)=&\langle f_{mn}(\mathbf{v}|\mathbf{v}_1)[c_m^{(\varkappa)}f_{mm}^{(\varkappa)}(\mathbf{v}_1|\mathbf{v}_2)+c_n^{(\varkappa)}f_{nn}^{(\varkappa)}(\mathbf{v}_1|\mathbf{v}_2)]\\&\times[f_{mn}(\mathbf{v}_2|\mathbf{v}_3)+F_{mn}{}^*(\mathbf{v}_2|\mathbf{v}_3)]W(\mathbf{v}_3)\rangle,\\ B_{mn}(\varkappa)=&\langle f_{mn}(\mathbf{v}|\mathbf{v}_1)[c_m^{(\varkappa)}F_{mm}^{(\varkappa)}(\mathbf{v}_1|\mathbf{v}_2)\\&+c_n^{(\varkappa)}F_{nn}^{(\varkappa)}(\mathbf{v}_1|\mathbf{v}_2)][F_{mn}(\mathbf{v}_2|\mathbf{v}_3)+F_{mn}{}^*(\mathbf{v}_2|\mathbf{v}_3)]W(\mathbf{v}_3)\rangle,\\ a_{mn}=&\langle f_{mn}(\mathbf{v}|\mathbf{v}_1)W(\mathbf{v}_1)\rangle,\ f_{mn}=f_{mn}^{(1)},\ F_{mn}=F_{mn}^{(1)},\\ I(\varkappa q)=&\sum_{\alpha'\alpha_1'}(-1)^{1-\alpha_1'}c(11\varkappa|\alpha'-\alpha_1'q)G_{\alpha'}G_{\alpha_1'}^*,\\ J(\varkappa q)=&\sum_{\alpha'\alpha_1'}(-1)^{1-\alpha_1'}c(11\varkappa|\alpha'-\alpha_1'q)G_{\alpha'}{}^\mu G_{\alpha_1'}^*,\\ c_m^{(\varkappa)}=&\begin{Bmatrix}1&\varkappa&1\\J_m&J_n&J_m\end{Bmatrix}^2,\quad c_n^{(\varkappa)}=\begin{Bmatrix}1&\varkappa&1\\J_n&J_m&J_n\end{Bmatrix}^2,\quad N=N_m-N_n.\end{aligned} \tag{3.10}$$

The angle brackets denote integration over all the velocities. $I(\varkappa q)$ is the strong-field polarization tensor in

the $\varkappa q$ representation[6,10]; $J(\varkappa q)$ is the crossing polarization tensor. $J(\varkappa q)$ is constructed in analogy with the usual polarization tensor, but in contrast to the latter, $J(\varkappa q)$ contains bilinear combinations of cyclic components of different origin.

Thus, formula (3.9) describes the circular component ($\alpha = 0, \pm 1$) of the macroscopic dipole moment of the medium at the trial-field frequency. Before we proceed to analyze it, we obtain an analogous result for the case when the trial field is resonant to a neighboring transition (three-level system, Fig. 1b).

4. THREE-LEVEL SYSTEMS

The matrix elements of the interaction take the form

$$V_{mn}(1\alpha)=G_\alpha e^{-i(\omega t-\mathbf{kr})}, \quad V_{ml}(1\alpha)=G_\alpha^\mu e^{-i(\omega_\mu t-\mathbf{k}_\mu \mathbf{r})},$$
$$G_\alpha=E_\alpha d_{mn}/2\hbar, \quad G_\alpha^\mu=E_\alpha^\mu d_{ml}/2\hbar. \tag{4.1}$$

The density matrix elements on the transition $m-n$ are described as before by the first equation of (3.3). The additional significant elements satisfy the following relations:

$$\begin{aligned}(\Gamma_{ml}-i\Omega'-i\varepsilon')r_{ml}&=A_{ml}r_{ml}-i\bar{U}_{mn}{}^{l}r_{nl}\\&+iu_{ml}{}^{m}R_{mm}+i[u_{ml}{}^{m}\rho_{mm}{}^{0}-\tilde{u}_{ml}{}^{l}\rho_{ll}{}^{0}],\\(\Gamma_{nl}-i\varepsilon')r_{nl}&=A_{nl}r_{nl}-i\bar{U}_{nm}{}^{l}r_{ml}+iU_{ml}{}^{n}R_{nm},\\\rho_{ml}&=r_{ml}e^{-i(\omega_\mu t-\mathbf{k}_\mu \mathbf{r})}, \quad \rho_{nl}=r_{nl}e^{-i(\omega t-\mathbf{pr})}.\end{aligned} \tag{4.2}$$

The induced polarization at the frequency of the trial field is determined by the element r_{ml}. For this element, using Green's functions in analogy with (3.5), we obtain (cf. (3.6)):

$$\begin{aligned}r_{ml}=if_{ml}\{&(u_{ml}{}^{m}\rho_{mm}{}^{0}-\tilde{u}_{ml}{}^{l}\rho_{ll}{}^{0})\\-\bar{U}_{mn}{}^{l}f_{nl}[&u_{ml}{}^{n}F_{nm}(\bar{U}_{nm}{}^{m}\rho_{mm}{}^{0}-U_{nm}{}^{n}\rho_{nn}{}^{0})\\+\bar{U}_{nm}{}^{l}f_{ml}&(u_{ml}{}^{m}\rho_{mm}{}^{0}-\tilde{u}_{ml}{}^{l}\rho_{ll}{}^{0})]\\-u_{ml}{}^{m}F_{mm}[&U_{nm}{}^{m}F_{mn}(U_{mn}{}^{m}\rho_{mm}{}^{0}-U_{mn}{}^{n}\rho_{nn}{}^{0})\\+(\bar{U}_{mn}{}^{m}&F_{nm}(\bar{U}_{nm}{}^{m}\rho_{mm}{}^{0}-U_{nm}{}^{n}\rho_{nn}{}^{0})]\}.\end{aligned} \tag{4.3}$$

Assuming that the Green's function has the structure (3.7) and that the density matrix not perturbed by the field takes the form (3.8), we arrive at the following analog of formula (3.9):

$$r_{ml}(1\alpha)\equiv\int r_{ml}(1\alpha v)\,dv=iN\Big\{G_\alpha^\mu a_{ml}-9\sum_{\varkappa q\alpha_1}(-1)^{1-\alpha_1}c(11\varkappa|\alpha-\alpha_1 q)$$
$$\times[G_{\alpha_1}J(\varkappa q)\beta_{ml}(\varkappa)+G_{\alpha_1}^\mu I(\varkappa q)B_{ml}(\varkappa)]\Big\}, \tag{4.4}$$
$$a_{ml}=\langle f_{ml}(\mathbf{v}|\mathbf{v}_1)W(\mathbf{v}_1)\rangle(N_m-N_l)/N,$$

$$\beta_{ml}(\varkappa)=\langle f_{ml}(\mathbf{v}|\mathbf{v}_1)f_{nl}^{(\varkappa)}(\mathbf{v}_1|\mathbf{v}_2)[f_{ml}(\mathbf{v}_2|\mathbf{v}_3)+F_{mn}{}^{*}(\mathbf{v}_2|\mathbf{v}_3)]W(\mathbf{v}_3)\rangle c_\beta^{(\varkappa)},$$
$$B_{ml}(\varkappa)=\langle f_{ml}(\mathbf{v}|\mathbf{v}_1)F_{mm}^{(\varkappa)}(\mathbf{v}_1|\mathbf{v}_2)[F_{mn}(\mathbf{v}_2|\mathbf{v}_3)+F_{mn}{}^{*}(\mathbf{v}_2|\mathbf{v}_3)]W(\mathbf{v}_3)\rangle c_B^{(\varkappa)},$$
$$c_\beta^{(\varkappa)}=\begin{Bmatrix}1&\varkappa&1\\J_l&J_m&J_n\end{Bmatrix}^2, \quad c_B^{(\varkappa)}=\begin{Bmatrix}1&\varkappa&1\\J_m&J_l&J_m\end{Bmatrix}\begin{Bmatrix}1&\varkappa&1\\J_m&J_n&J_m\end{Bmatrix}(-1)^{J_n-J_l}. \tag{4.5}$$

We proceed to the analysis of formulas (3.9) and (4.4).

5. DISCUSSION. POLARIZATION METHODS

The choice of the model (3.7) for the Green's functions has resulted in separation, in explicit form, of the polarization structure in (3.9) and (4.4), i.e., of the dependence on the states of the polarization of the trial and strong fields, and the structure turned out to be the same for r_{mn} and r_{ml}. All the subsequent deductions concerning the polarization properties will consequently pertain equally well to two-level and three-level systems. We note that in the derivation of formula (4.4) for the three-level system we did not use specific features of the system, such as Raman scattering (Fig. 1b). The results are therefore equally applicable to schemes of the two-quantum absorption (luminescence) type.

The terms proportional to $\beta(\varkappa)$ in (3.9) and (4.4) describe the so-called nonlinear interference effects[11] (interference terms). The terms proportional to $B(\varkappa)$ are connected with the changes of the distributions in the velocities and in the magnetic sublevels on the combining levels m and n under the influence of the strong field. We shall call them "population" terms. We note that the "population" and interference terms differ substantially in their polarization structure (this will become particularly clear later on).[3)]

Terms having different values of $\varkappa$ in the nonlinear increment to formula (3.9) are due to the relaxation of the polarization moments of the states m and n; this relaxation is described by the Green's functions $f_{jj}^{(\varkappa)}$ and $F_{jj}^{(\varkappa)}$. The terms contributing to the sum are those with $\varkappa = 0, 1, 2$, a fact dictated by the properties of the Clebsch-Gordan coefficients. At $\varkappa = 0$ the Green's functions describe the relaxation of the total population, at $\varkappa = 1$ and at $\varkappa = 2$ they describe the relaxations of the orientation and of the alignment, respectively.[6] In the case of a three-level system (formula (4.4)) the interference term is connected with the relaxation of the "polarization moments" of the forbidden transition n-l (the Green's function $f_{nl}^{(\varkappa)}$), which are constructed in analogy with the polarization moments of the levels (see (2.3)).

In the general case, the expressions for r_{mn} and r_{ml} contain all the terms with $\varkappa = 0, 1, 2$, i.e., a rather large number of relaxation characteristics. We shall show how to separate with the aid of the polarization method the terms with separate values of $\varkappa$, and will indicate the optimal way of successively investigating the relaxation characteristics.

Consider the following experimental setup: the strong field is circularly polarized and weak one linearly, while the wave vectors $\mathbf{k}$ and $\mathbf{k}_\mu$ are collinear. Figures 2a illustrate the transition scheme (for the sake of simplicity we show the transition $J_m = 1 \to J_n = 0$) in a coordinate system haveing a quantization axis along the wave vectors. In accordance with (3.9) and (4.4) we have

$$r(1\alpha)=iNG_\alpha^\mu\{a-3|G|^2[B(0)+{}^3/_2\alpha B(1)+{}^1/_2B(2)$$
$$+\beta(0)\delta_{1\alpha}+{}^3/_2\beta(1)\delta_{1\alpha}+{}^1/_2\beta(2)(\delta_{1\alpha}+6\delta_{-1\alpha})]\}, \quad \alpha=\pm1. \tag{5.1}$$

The indices numbering the energy states have been left out here, since this structure is common to the two-level and three-level systems; $\delta_{k\alpha}$ is the Kronecker symbol. Since $r(1\alpha)\propto G_\alpha^\mu$, the cyclic components of the trial field are propagated independently of one another, i.e., are natural waves. The propagation of each of them can consequently be described with the aid of a complex vector

$$E_\alpha^\mu(t, x)=E_{\alpha 0}^\mu \exp[-i(\omega_\mu t-k_\mu x)-i\Delta k_\alpha^\mu x],\ r(1\alpha)\propto G_\alpha^\mu \Delta k_\alpha^\mu. \qquad (5.2)$$

In the course of the propagation, the amplitudes and phases of the different natural waves vary essentially in different fashions (dichroism and birefringence). In the general case this difference is determined both by "population" and the interference parts of r. In the "population" part of the terms with $\varkappa=0$ and $\varkappa=2$ influence the propagation of the left and right "circles" ($\alpha=\pm1$) in like manner. The difference is determined by the term with $\varkappa=1$. In the interference part, the right "circle" ($\alpha+1$) is influenced by terms with both values of $\varkappa$, while the left circle only with $\varkappa=2$. Let us explain the reason for these singularities. We start with the "population" terms. The strong field, characterized by a polarization tensor $I(\varkappa q)$, induces in the states m and n the elements $\rho_{jj}(\varkappa q\nu)\propto I(\varkappa q)$[10] ($j=m, n$). In our case we have

$$I(\varkappa q)=I(\varkappa 0)\delta_{q0}.\quad I(00)=2^{1/2}I(20)=\left(\frac{2}{3}\right)^{1/2}I(10)=3^{-1/2}|G|^2, \qquad (5.3)$$

i.e., the elements $\rho_{jj}(00\nu)$ are induced. Taking into account the connection between $\rho_{jj}(\varkappa q\nu)$ and $\rho_{J_jM;J_jM'}$ (formula (2.3)), we obtain

$$\rho_{J_jM;J_jM'}(\nu)=\delta_{MM'}\{\rho_{jj}(00\nu)+3^{1/2}[J_j(J_j+1)]^{-1/2}M\rho_{jj}(10\nu)$$
$$+5^{1/2}[J_j(J_j+1)(2J_j-1)(2J_j+3)]^{-1/2}[3M^2-J_j(J_j+1)]\rho_{jj}(20\nu)\}(2J_j+1)^{-1/2}. \qquad (5.4)$$

Thus, $\rho_{jj}(00\nu)$ represents uniform distributions of the populations over the sublevels, while $\rho_{jj}(10\nu)$ and $\rho_{jj}(20\nu)$ represent linear and quadratic distributions, respectively. The Green's functions with $\varkappa=0, 1$, and 2 describe independent relaxation of the corresponding distributions. The relaxation of the symmetrical distributions ($\varkappa=0, 2$), naturally, does not offer any advantages in interactions with any particular "circle" of the trial field. The difference lies only in the asymmetrical distribution, i.e., at $\varkappa=1$ as is reflected in (5.1).

The understanding of the polarization structure of the interference part is facilitated by Fig. 2a. For the right-hand "circle" of the trial wave ($\alpha=1$) an important role is played by the relaxation of the sublevel populations (more accurately, the beats of the populations). This is precisely why at $\alpha=1$ the quantities $\beta(\varkappa)$ in (5.1) are represented in the same proportion as $B(\varkappa)$. For the left-hand "circle," only the relaxation of the coherence between the magnetic sublevels with $\Delta M=2$ is significant, and this relaxation is connected with the quantity $\varkappa=2$. This fact also agrees with the structure of (5.1).

Let us examine another experimental setup (Fig. 2b): the strong field is linearly polarized and the trial field is either circularly or also linearly polarized, but the plane of polarization is rotated (say through 45°). The wave vectors are collinear as before, and the quantization axis is parallel to the electric field of the strong field. In this case we obtain

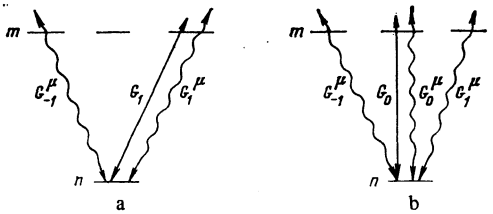
$$r(1\alpha)=iNG_\alpha^\mu\{a-3|G|^2[B(0)+B(2)(1+\delta_{0\alpha})$$
$$+\beta(0)\delta_{0\alpha}+{}^3/_2\alpha^2\beta(1)+{}^1/_2\beta(2)(\delta_{0\alpha}+3)]\},\quad \alpha=0, \pm1. \qquad (5.5)$$

In this choice of the coordinate system, the circular components ($\alpha=0, \pm1$) are also natural waves whose propagation is described by formula (5.2). The conditions for the propagation of the waves with $\alpha=\pm1$ are the same, so that in actual fact the natural waves are waves with orthogonal linear polarizations, one of which is polarized in the same manner as the strong field.

In this case, the polarization tensor of the strong field $I(\varkappa q)$ also changes the structure of $I(\varkappa 0)\delta_{q0}$ and, in addition, $I(10)=0$. Consequently, the elements $\rho_{jj}(10\nu)$ are not induced in the system, and this has led to the vanishing of the term with $\varkappa=1$ in the "population" part. The fact that the terms proportional to $B(2)$ are not the same at $\alpha=0$ and $\alpha=\pm1$ is caused by the non-uniform populations of the sublevels on account of the element $\rho_{jj}(20\nu)$ (see (5.4)) which leads, as is illustrated in Fig. 2b, to different positions of the propagation for the waves with $\alpha=0$ and $\alpha=\pm1$. The term proportional to $B(0)$ is the same for all the waves for the same reasons as in the preceding scheme. The interference part contains a term with $\varkappa=1$. It is connected with the coherence between the magnetic sublevels, a coherence which is substantial for the waves with $\alpha=\pm1$. For the waves with $\alpha=0$, the interference terms $\beta(\varkappa)$ are present in the same proportion as the "population" terms, and in particular, there is no term with $\varkappa=1$. For waves with $\alpha=\pm1$ (orthogonal polarization), there is no term with $\varkappa=0$, inasmuch as here, in accord with Fig. 2b, only the coherence between the sublevels is of importance.

We proceed now to the question of separating the individual relaxation characteristics. We confine ourselves here to systems with large Doppler broadening ($k\bar{v}\gg\Gamma$, where Γ is the characteristic homogeneous width). The organization of the experiment is assumed to be the following.[1,2] After passing through the medium, the trial field is made to pass through a polaroid crossed with the polarization of the field parallel to its interaction with the medium. The signal passing through the polaroid is then registered. It is known[11] that when waves propagate in opposite directions in systems with large Doppler broadening, there are no interference terms in the nonlinear susceptibility, i.e., there are no $\beta(\varkappa)$ terms in formulas (5.1) and (5.5).[4)] As the field polarization state characterized by Fig. 2a, the signal past the crossed polaroid is proportional, in accordance with (5.1) and the propagation law (5.2), to the quantity

$$|\Delta k_1^\mu-\Delta k_{-1}^\mu|^2\propto N^2|G|^2|9B(1)|^2. \qquad (5.6)$$



a b

FIG. 2. Transitions between sublevels of a two-level system ($J_m=1$, $J_n=0$). The strong field is circularly (a) or linearly (b) polarized.

For the case corresponding to Fig. 2b and formula (5.5), the signal is proportional to

$$|\Delta k_0^{\mu}-\Delta k_{\pm 1}^{\mu}|^2 \propto N^2|G|^2|3B(2)|^2. \tag{5.7}$$

We can consequently separate only one term from each of the sets of terms $B(\varkappa)$ and $\beta(\varkappa)$ in (5.1) and (5.5), which differ in their relaxation characteristics. Thus, it is possible to create experimental conditions under which the terms $B(\varkappa)$ with $\varkappa=1$ and $\varkappa=2$ can be separately investigated.

Unfortunately, the remaining terms $B(0)$ and $\beta(\varkappa)$ cannot be separated experimentally, and only a program whereby they are turned on in succession can be realized. To this end, after separately studying $B(1)$ and $B(2)$ by the described method, it is advantageous to use the ordinary methods of nonlinear spectroscopy—investigation of nonlinear absorption (amplification). In an experiment with opposing waves there will likewise be registered only the "population" part of the nonlinear increment. For the case described by formula (5.5) this means that in the registered signal there will be present only terms $B(0)$ and $B(2)$. It is next possible to include the interference terms $\beta(\varkappa)$. For waves with orthogonal circular polarizations and propagating in the same direction (formula (5.1)) the only intereference term present is $\beta(2)$, while the entire set of the "population" terms is present. Next, the term $\beta(1)$ is added to the term $\beta(2)$ in the case of waves with orthogonal linear polarizations and propagating in the same direction (formula (5.5)), and a combination of $\beta(0)$ and $\beta(1)$ enters in the case of identical linear polarizations. We note that in a two-level system $\beta(\varkappa)$ and $B(\varkappa)$ are not completely independent, inasmuch as in the equations for the Green's functions f_{jk} and F_{jk} the collision operator A_{jk} is one and the same (see (3.3))and (3.5)). This circumstance facilitates the investigation of $\beta(\varkappa)$ after $B(\varkappa)$.

So far we did not touch upon the question of the explicit forms of the quantities $\beta(\varkappa)$ and $B(\varkappa)$. The structure of these quantities, i.e., the dependence of the frequency detunings and the change of this dependence with changing pressure, is closely connected with the relaxation model, in other words, it is determined by the concrete experimental object and by the experimental conditions. In the investigation of $\beta(\varkappa)$ and $B(\varkappa)$ as functions of the frequency detunings and of the pressure yields information on the parameters of the model (relaxation characteristics). By way of example, we shall discuss here two relaxation models.

In the first of them, which is the simplest and most widely used, the change of the velocity is described by the model of strong collisions and is accompanied by a uniform mixing of the sublevels.[12] In addition, it is assumed that the change of the projection of the angular momentum occurs in the collisions without change of velocity.[5-7] This model describes well atomic systems, which are characterized by short lifetimes of the states and also molecular systems at moderate pressures.[9] The Green's functions in this case are

$$F_{jj}^{(\varkappa)}(\mathbf{v}|\mathbf{v}_1)=\frac{1}{\Gamma_{jj}^{(\varkappa)}}[\delta(\mathbf{v}-\mathbf{v}_1)+n_j\delta_{\varkappa 0}W(\mathbf{v})], \quad (j=m,n),$$

$$F_{mn}(\mathbf{v}|\mathbf{v}_1)=\delta(\mathbf{v}-\mathbf{v}_1)[\Gamma_{mn}^{(1)}-i(\Omega-\mathbf{kv})]^{-1}. \tag{5.8}$$

Here $\Gamma_{jk}^{(\varkappa)}$ are relaxation constants of the polarization moment,[6] n_j is the effective number of collisions during the lifetime on the level j. The Green's functions (5.8) lead to the following expressions for the "population" terms:

$$B_{mn}(\varkappa)=\frac{\pi^{1/2}}{k\bar{v}}e^{-(\Omega_\mu/k\bar{v})^2}\Big[\frac{1}{2\Gamma_{mn}^{(1)}-i(\Omega_\mu+\Omega)}(c_m^{(\varkappa)}/\Gamma_{mm}^{(\varkappa)}+c_n^{(\varkappa)}/\Gamma_{nn}^{(\varkappa)})$$

$$+\frac{\pi^{1/2}}{k\bar{v}}e^{-(\Omega/k\bar{v})^2}(c_m^{(\varkappa)}n_m+c_n^{(\varkappa)}n_n)\delta_{\varkappa 0}\Big], \quad \Omega_\mu=\omega_\mu-\omega_{mn}. \tag{5.9}$$

$$B_{ml}(\varkappa)=\frac{\pi^{1/2}}{k\bar{v}}e^{-(\Omega_\mu/k\bar{v})^2}\Big[\frac{1}{\Gamma_{mm}^{(\varkappa)}}\frac{1}{\Gamma_{mn}^{(1)}+\Gamma_{ml}^{(1)}-i(\Omega_\mu+\Omega)}+\frac{\pi^{1/2}}{k\bar{v}}e^{-(\Omega/k\bar{v})^2}n_m\delta_{\varkappa 0}\Big]c_B^{(\varkappa)},$$

$$\Omega_\mu=\omega_\mu-\omega_{ml}, \quad |\mathbf{k}-\mathbf{k}_\mu|\ll k. \tag{5.10}$$

Each quantity $B(\varkappa)$ is a superposition of a narrow dispersion term of homogeneous width and a broad homogeneous-saturation "pad." We call attention to the fact that the "pad" contains the Kronecker symbol $\delta\varkappa_0$ as a factor. Consequently, it will not be registered in the experiment with the crossed polaroid. The registered signal takes the following form (for example, for the case (5.10)):

$$|B_{ml}(\varkappa)|^2=e^{-2(\Omega_\mu/k\bar{v})^2}\frac{1/[\Gamma_{mm}^{(\varkappa)}]^2}{(\Gamma_{mn}^{(1)}+\Gamma_{ml}^{(1)})^2+(\Omega_\mu+\Omega)^2}\Big[\frac{\pi^{1/2}}{k\bar{v}}c_B^{(\varkappa)}\Big]^2, \quad \varkappa=1,2. \tag{5.11}$$

The value $\varkappa=1$ corresponds to circular polarization of the strong field, and $\varkappa=2$ to linear polarization. The registered signal has a Lorentz profile with a half-width $\Gamma_{mn}^{(1)}+\Gamma_{ml}^{(1)}$ and with an amplitude proportional to $1/[\Gamma_{mm}^{(\varkappa)}]^2$. In this case one can determine experimentally both $\Gamma_{mn}^{(1)}+\Gamma_{ml}^{(1)}$ (by measuring the width of the line profile) and $\Gamma_{mm}^{(\varkappa)}$ ($\varkappa=1, 2$) by investigating the pressure dependence of the amplitude of the profile (or the area).

The reason for the absence of a "pad" in the signal past the crossed polaroid is connected with the assumed strong mixing of the sublevels in collisions, and is quite clear in light of the arguments given above. On the other hand, the appearance of a "pad" would be evidence of incomplete sublevel mixing, which would yield additional information on the processes of disorientation and would lead to a refinement of the relaxation model.

For molecular systems and atomic metastable systems at low pressures, the analyzed relaxation model becomes no longer valid, since in this case elastic scattering through small angles plays an important role.[9] The influence of collisions on the nonlinear-absorption spectrum in systems with degenerate levels was analyzed in[9]. The basis used was the isotropic–perturbation model, and the change of velocity was described with the aid of collision-integral kernels that depend on the difference between the velocities before and after the collisions. It follows from the result of[9] that $B(\varkappa)$ contains in place of the dispersion terms (as in (5.9) and (5.10)) structures that are spectrally more complicated and whose parameters are connected not only with the relaxation constants but also with the characteristics of the differential cross section of the scattering. In addi-

tion, the terms $B(\varkappa)$ have different spectral structures at different values of $\varkappa$, so that in this case the most pressing problem is the experimental separation and investigation of each of the terms $B(\varkappa)$. Thus, under the experimental conditions that lead to formulas (5.6) and (5.7), the quantities $|B(1)|^2$ and $|B(2)|^2$ are individually separated. Investigating the dependence of the signal on the frequency of the trial field, we can obtain on the basis of the results of[9] information on the parameters of the model that describe small-angle elastic scattering.

In conclusion, let us dwell on one important practical application of the results. We have in mind high-accuracy stabilization of laser frequencies. In our opinion, the polarization method using a crossed polaroid is in this respect highly promising, and not merely because of its sensitivity. Namely, in the development of lasers with high frequency stability interest attaches only to long-lived systems (molecules and atomic metastable systems), and in these systems the homogeneous saturation due to the translational relaxation is exceptionally large. The important role in molecules is played also by rotational relaxation, which also leads to homogeneous saturation. It is natural to expect translational and rotational relaxation to be accompanied also by a strong disorientation (mixing over the magnetic sublevels). Consequently, just as in (5.8), the Green's functions $F_{jk}^{(\varkappa)}$ describing the relaxation in these systems will contain, with a large weight, a term proportional to $\delta_{\varkappa 0}W(\mathbf{v})$ and responsible for the homogeneous saturation and containing the Kronecker symbol $\delta_{\varkappa 0}$. The remaining part of the Green's function, which gives the inhomogeneous saturation, can either correspond to the model (5.8), or, at low pressure, to a more complicated model.[9] Thus, if we take a trial wave having the same frequency (or the same source) as the strong wave, then in the experiment with a crossed polaroid, corresponding to Fig. 2a or 2b, the signal (for example for the model (5.8)) will be proportional to the quantity

$$e^{-2(\Omega/k\bar{v})^2}\frac{1}{[\Gamma_{mn}^{(1)}]^2+\Omega^2}[c_m^{\varkappa}/\Gamma_{mm}^{(\varkappa)}+c_n^{(\varkappa)}/\Gamma_{nn}^{(\varkappa)}]. \quad \varkappa=1,2. \qquad (5.12)$$

As a function of the frequency of the field, the signal is a contour with homogeneous half-width $\Gamma_{mn}^{(1)}$. In the model of[9], Eq. (5.12) is replaced by a more complicated spectral structure, but an important fact is that in either case the "pad" of the homogeneous saturation is completely missing.

Formula (5.12) and its equivalent in the model of[9] pertain, strictly speaking, to systems in which each state can be described by one rotational quantum number—the total angular momentum J. It is obvious, however, that the conclusion that the homogeneous saturation is substantially "suppressed" is valid also for complicated systems (molecules of low symmetry): the homogeneous saturation is accompanied in either case by strong disorientation of the particles, at which the anisotropy of the medium vanishes.

I am deeply grateful to S. G. Rautian for a useful discussion of the result.

[1)]The velocity $\mathbf{v}$ enters in the matrices and the vectors on the par with indices, i.e., integration with respect to the velocity is implied in the corresponding cases.

[2)]We note that this model incorporates all the presently known relaxation models for degenerate states.

[3)]We note in this connection that the distinction between the interference and population terms, which is made in[11] on the basis of the frequency-correlation properties of the nonlinear susceptibility, turns out to be natural also from the point of view of the polarization structure.

[4)]In (luminescence) schemes, to the contrary, the interference terms drop out if the waves propagate in the same direction.[11]

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