Polarization features of coherent transition phenomena in two-photon resonance

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A mathematical apparatus is formulated for the study of polarization properties of a large class of two-photon coherent transition phenomena at any degeneracy multiplicity of the atomic or molecular levels. Polarization features are investigated of two-photon optical nutation, of third-harmonic generation in a previously excited medium, and of two-photon echo. The possibility is demonstrated of determining the parameters of the two-photon interaction operator, of identifying optically forbidden transitions, and of investigating atomic relaxation by starting from the polarization properties of the considered coherent transition phenomena.

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Nonlinear spectroscopy is a new trend in the physics of interaction of coherent radiation with matter under resonance conditions.[1] Methods of nonstationary coherent spectroscopy are being intensively developed in recent years for the investigation of atoms, molecules, and condensed media.[2,3] The most widely used is the method of Doppler-free two-photon spectroscopy.[4]

Two alternate techniques have by now been proposed and are under development for spectroscopic research into two-photon (optically forbidden) atomic and molecular transitions. One is based on trilevel echo[5] and two-photon coherent transition phenomena (such as optical nutation[6,7] and two-photon echo[8]). Up to now only trilevel echo has been exhaustively studied theoretically, since its analysis is in essence based on the physical premises and analytic results of the theory of one-photon resonance.[11] The reported theoretical investigations of coherent transition phenomena in two-photon resonance usually disregard the resonant-level degeneracy that always takes place in experiments with all media, including condensed ones. A multiphoton vector model was used in Ref. 19 to consider optical nutation on vibrational-rotational transitions in a diatomic molecule, neglecting thermal motion of the molecules. The vector model, however, does not reflect the polarization properties of the coherent transition phenomena. In addition, the calculation method in Ref. 19 is restricted to two molecules.

We construct in this paper a theory that covers a large class of two-photon coherent phenomena from a unified viewpoint. We consider an ensemble of atoms or molecules that move in the field of linearly or circularly polarized light waves under conditions of two-photon resonance with degenerate levels. The degeneracy is due only to the arbitrary orientation of the total angular momentum. The behavior of the resonant system is described by an equation for the density matrix, whose solution is obtained in the approximation of a given field of arbitrary intensity. The expressions obtained make it possible to investigate various two-photon coherent transition phenomena with allowance for thermal motion of the atoms or molecules, for Stark shifts of resonant levels, for level degeneracy of arbitrary multiplicity, and for the polarization state of the exciting light pulses. On the basis of this approach we analyze in this article, for the first time ever, the polarization properties of two-photon optical nutation and consider the polarization features of third-harmonic generation in a pre-excited medium and of two-photon echo. As a result, new features of these transition phenomena are observed and extend substantially the possibility of using these effects for the investigation of two-photon transitions; they yeild more spectroscopic information than trilevel echo.[5,9,10]

It is established that by comparing linearly polarized two-photon nutation with circular-polarization optical nutation it is possible to distinguish between the Q, P (or R), and O (or S) branches of vibrational-rotational transitions. Simple expressions are obtained that make it possible to determine from the period of the optical nutation the parameters of the operator of two-photon interaction at any multiplicity of the level degeneracy. It is shown that polarization investigations of third-harmonic generation and of two-photon echo can yield extensive information on the relaxation characteristics of two-photon transitions and energy levels whose number increases appreciably if elastic depolarizing atomic collisions are significant besides the radiative decay.

1. CALCULATION METHOD AND BASIC RELATIONS

Assume that the nth ($n = 1,2,...$) exciting light pulse propagates through a medium in the Y-axis direction. The pulse electric-field intensity is of the form

$$\mathbf{E}_n = \mathbf{l}^{(p)} a_n e^{-i\Phi_n} + \text{c.c.}, \quad \Phi_n = \omega t - ky + \varphi_n. \tag{1}$$

Here a_n and φ_n are respectively the amplitude and phase, while the polarization unit vector $\mathbf{l}(p)$ is equal to $\mathbf{l}^{(0)} = \mathbf{l}_z$, $\mathbf{l}^{(-1)} = 2^{-1/2}(i\mathbf{l}_x - \mathbf{l}_z)$, and $\mathbf{l}^{(1)} = 2^{-1/2}(i\mathbf{l}_x + \mathbf{l}_z)$ for linear ($p = 0$), left-circular ($p = -1$), and right-circular polarization ($p = 1$) respectively, with $\mathbf{l}_x$ and $\mathbf{l}_z$ the unit vectors of the Cartesian axes. The carrier frequency $\omega = kc$ of the wave (1) is at two-photon resonance $|2\omega - \omega_{ca}| \ll \omega$ with the frequency $\omega_{ca} = (E_c - E_a)/\hbar$ of some two-photon (optically forbidden) atomic or molecular transition between levels with energies E_c and E_a, which are also characterized by total angular momenta j_c and j_a and by their projections ν and m on the quantization axis.

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The interaction of the light wave (1) with the resonant atoms is considered on the basis of the quantum-mechanical equation for the density matrix ρ:

$$i\hbar\left(\frac{\partial}{\partial t}+v_y\frac{\partial}{\partial y}-\hat{\Gamma}\right)\rho=(H-\mathbf{E}_n\mathbf{d})\rho-\rho(H-\mathbf{E}_n\mathbf{d}), \qquad (2)$$

where H is the Hamiltonian of the free atom in its c.m.s., $\mathbf{d}$ is the dipole-moment operator, and v_y is the projection of the atom velocity $\mathbf{v}$ on the Y axis. We assume that the reaction of the atoms to the light wave (1) is small [the case (13) of an optically thin medium] and therefore Eq. (2) contains the given external field $\mathbf{E}_n$. The relaxation operator takes into account the radiative decay, the elastic and inelastic atomic collisions, and the pumping[20]

$$(\hat{\Gamma}\rho)_{vm}=-(\gamma_c+\gamma_a)\rho_{vm}/2$$

$$-\sum_{\varkappa q\nu'm'}(-1)^{2j_c+\nu+\nu'}(2\varkappa+1)\mathcal{T}_{ca}^{(\varkappa)}\times\begin{pmatrix}j_c & j_a & \varkappa\\ \nu & -m & q\end{pmatrix}\begin{pmatrix}j_c & j_a & \varkappa\\ \nu' & -m' & q\end{pmatrix}\rho_{\nu'm'},$$

$$(\hat{\Gamma}\rho)_{\nu\nu'}=\frac{\gamma_c N_c}{2j_c+1}f(v)\delta_{\nu\nu'}-\gamma_c\rho_{\nu\nu'}$$

$$-\sum_{\varkappa q\nu_1\nu_1'}(-1)^{2j_c+\nu+\nu_1}(2\varkappa+1)\Gamma_c^{(\varkappa)}\times\begin{pmatrix}j_c & j_c & \varkappa\\ \nu & -\nu' & q\end{pmatrix}\begin{pmatrix}j_c & j_c & \varkappa\\ \nu_1 & -\nu_1' & q\end{pmatrix}\rho_{\nu_1\nu_1'},$$

while the quantity $(\hat{\Gamma}\rho)_{mm'}$ is obtained from the expression for $(\hat{\Gamma}\rho)_{\nu\nu'}$ by the interchanges $c\to a$ and $\nu\to m$ of the indices. Here $\rho_{\nu\nu'}$ and $\rho_{mm'}$ are the density matrices of the atoms in states with energies E_c and E_a, respectively, N_c and N_a are the stationary densities of the atoms on these energy levels, $f(v)=(\pi u^2)^{-3/2}\exp(-v^2/u^2)$ is the Maxwell distribution, and u is the most probable velocity of the atom. The constants $\hbar\gamma_c$ and $\hbar\gamma_a$ are the partial widths of the levels E_c and E_a, and are governed by the radiative decay and inelastic gaskinetic collisions. The real quantities $\Gamma_c^{(\varkappa)}$ $(0\leqslant\varkappa\leqslant 2j_c)$ and $\Gamma_a^{(\varkappa)}$ $(0\leqslant\varkappa\leqslant 2j_a)$ represent the contributions of the elastic depolarizing collisions and the level broadening. The complex quantities

$$\mathcal{T}_{ca}^{(\varkappa)}=\Gamma_{ca}^{(\varkappa)}+i\Delta_{ca}^{(\varkappa)}, \quad |j_c-j_a|\leqslant\varkappa\leqslant j_c+j_a$$

describe the relaxation of the optical-coherence matrix ρ_{vm} of the atomic transition $j_c\to j_a$.

Following Ref. 12, we introduce a new matrix R connected with the density matrix ρ by the unitary transformation

$$R=e^{-iQ}\rho e^{iQ}=\rho-i(Q\rho-\rho Q)-\ldots,$$

where we have in the approximation linear in the field $\mathbf{E}_n$

$$Q_{v\mu}=-i\frac{\mathbf{l}^{(p)}\mathbf{d}_{v\mu}a_n}{\hbar(\omega_{cb}-\omega)}e^{-i\Phi_n}-i\frac{\mathbf{l}^{(p)*}\mathbf{d}_{v\mu}a_n}{\hbar(\omega_{cb}+\omega)}e^{i\Phi_n},$$

$$Q_{\nu\nu'}=Q_{mm'}=Q_{vm}=0.$$

Here μ is the projection, on the quantization axis, of the angular momentum j_b of the nonresonant level with energy E_b,

$$\omega_{bc}=-\omega_{cb}=(E_b-E_c)/\hbar, \quad \omega_{ab}=-\omega_{ba}=(E_a-E_b)/\hbar.$$

The matrix element $Q_{m\mu}$ is obtained from $Q_{v\mu}$ by interchanging the indices, $c\to a$ and $\nu\to m$.

In the resonance approximation, the slowly varying functions

$$r_{vm}=R_{vm}\exp(2i\Phi_n), \quad r_{\nu\nu'}=R_{\nu\nu'}, \quad r_{mm'}=R_{mm'}$$

satisfy the system of equations

$$i\hbar\left[\frac{\partial}{\partial t}+v_y\frac{\partial}{\partial y}+i(\eta-\Delta)\right]r_{vm}=i\hbar(\hat{\Gamma}r)_{vm}+V_{vm'}r_{m'm}-r_{\nu\nu'}V_{\nu'm}+V_{\nu\nu'}r_{\nu'm}-r_{\nu m'}V_{m'm}, \qquad (3)$$

$$i\hbar\left(\frac{\partial}{\partial t}+v_y\frac{\partial}{\partial y}\right)r_{\nu\nu'}=i\hbar(\hat{\Gamma}r)_{\nu\nu'}+V_{vm}r_{\nu'm}^{*}-r_{vm}V_{\nu'm}^{*}+V_{\nu\nu''}r_{\nu''\nu'}-r_{\nu\nu''}V_{\nu''\nu'}, \qquad (4)$$

$$i\hbar\left(\frac{\partial}{\partial t}+v_y\frac{\partial}{\partial y}\right)r_{mm'}=i\hbar(\hat{\Gamma}r)_{mm'}+V_{vm}^{*}r_{vm'}-r_{vm}^{*}V_{vm'}+V_{mm''}r_{m''m'}-r_{mm''}V_{m''m'}, \qquad (5)$$

$$\eta=2kv_y, \quad \Delta=2\omega-\omega_{ca}.$$

We sum here over the repeated indices.

It is convenient to choose the quantization axis along the polarization vector (the Z axis) in the case of linear polarization and along the Y axis in the case of circular polarization. The interaction operator takes then the simplest form

$$V_{vm}=(-1)^{j_c-\nu+1}\frac{\hbar U_{n\nu}}{2}e^{-2i\Phi_n}\delta_{\nu,m+2p},$$

$$V_{mm'}=U_{nm}^{(a)}\delta_{mm'}, \qquad V_{\nu\nu'}=U_{n\nu}^{(c)}\delta_{\nu\nu'},$$

where

$$U_{n\nu}=\frac{a_n^2}{\hbar^2}(2j_a+1)^{1/2}\sum_{\varkappa}(2\varkappa+1)\times\begin{pmatrix}j_c & j_a & \varkappa\\ \nu & 2p-\nu & -2p\end{pmatrix}\begin{pmatrix}1 & 1 & \varkappa\\ p & p & -2p\end{pmatrix}\Pi_{\omega}^{(\varkappa)},$$

$$U_{n\nu}^{(c)}=(-1)^{j_c-\nu+p}\frac{a_n^2}{\hbar}(2j_c+1)^{1/2}\sum_{\varkappa}(2\varkappa+1)\times\begin{pmatrix}j_c & j_c & \varkappa\\ \nu & -\nu & 0\end{pmatrix}\begin{pmatrix}1 & 1 & \varkappa\\ -p & p & 0\end{pmatrix}\Pi_c^{(\varkappa)},$$

$$\Pi_{\omega}^{(\varkappa)}=\sum_b\frac{d_{cb}d_{ba}}{(2j_a+1)^{1/2}}\left[\frac{1}{\omega_{ba}-\omega}+\frac{(-1)^{\varkappa}}{\omega_{bc}+\omega}\right]\begin{Bmatrix}1 & \varkappa & 1\\ j_c & j_b & j_a\end{Bmatrix}(-1)^{j_c+j_a},$$

$$\Pi_c^{(\varkappa)}=\sum_b\frac{|d_{cb}|^2}{(2j_c+1)^{1/2}}\left[\frac{1}{\omega_{cb}-\omega}+\frac{(-1)^{\varkappa}}{\omega_{cb}+\omega}\right]\begin{Bmatrix}1 & \varkappa & 1\\ j_c & j_b & j_c\end{Bmatrix}(-1)^{j_c+j_b},$$

and the quantities $U_{nm}^{(a)}$ and $\Pi_a^{(\varkappa)}$ are obtained from $U_{n\nu}^{(c)}$ and $\Pi_c^{(\varkappa)}$ by interchanging the indices, $c\to a$ and $\nu\to m$. The quantity $U_{n\nu}$ characterizes the frequency of the two-photon transitions induced by the field (1) between the levels $E_a^{\cdot}$ and

E_c, while $U_{nm}^{(a)}$ and $U_v^{(c)}$ are the Stark shifts of the Zeeman sublevels of these levels. By d_{cb} and d_{ba} we denote the reduced dipole moments of the optically allowed transitions $j_c \to j_b$ and $j_b \to j_a$ (Ref. 21). The subscript b in the expressions for $\Pi_\omega^{(\varkappa)}$, $\Pi_c^{(\varkappa)}$, and $\Pi_a^{(\varkappa)}$ numbers all the nonresonant levels connected with the levels E_a and E_c by optically allowed transitions.

Equations (3)–(5) describe two-photon resonance of the light wave (1) with degenerate atomic or molecular energy levels whose degeneracy is due only to the difference in the orientation of the total angular momentum of the atom or of the molecule relative to the fixed quantization axis. In this case the quantities $\Pi_\omega^{(\varkappa)}$, $\Pi_a^{(\varkappa)}$, and $\Pi_c^{(\varkappa)}$ are parameters of the two-photon interaction, are difficult to calculate, and must be determined from experiment. In a number of cases, however, the parameters $\Pi_\omega^{(\varkappa)}$ for diatomic molecules can be expressed in terms of the probabilities of two-photon vibrational-rotational transitions, calculated in Ref. 19. For the vibrational-rotational transitions in a diatomic molecule, without change of the total angular momentum, the results of Ref. 19 leads to the relation $\Pi_\omega^{(0)} = \sqrt{10}\,\Pi_\omega^{(2)}$.

It can be seen from (3)–(5) that the level degeneracy complicates appreciably the initial equations and the subsequent solution compared with the nondegenerate case.[12,18] Nonetheless, allowance for the level degeneracy is essential in principle for the description of the polarization effects. It must be emphasized that Eqs. (3)–(5) do not reduce to the equations obtained in Ref. 19 for the multiphoton vector model, even in the simplest case of strict resonance $\Delta = 0$, in the absence of thermal motion of the molecules ($\eta = 0$), and neglecting the irreversible relaxation ($\hat{\Gamma} = 0$) and the linear polarization ($p = 0$) of the light wave (1). They can be reduced if it assumed in addition that the matrices r_{vm}, $r_{vv'}$, and $r_{mm'}$ are diagonal:

$$r_{vm} \sim \delta_{vm}, \quad r_{vv'} \sim \delta_{vv'}, \quad r_{mm'} \sim \delta_{mm'}.$$

This assumption is patently not satisfied in experiments in which the exciting light pulses have different polarizations, or in which additional static fields, say a magnetic one, are present.

In investigations of coherent transition phenomena the durations of the exciting pulses (1) are usually shorter (much shorter) than the irreversible-relaxation time, and the time intervals between pulses are comparable with the irreversible-relaxation time. Under these conditions we need a solution of Eqs. (3)–(5) at $\hat{\Gamma} = 0$; we write this solution in the form

$$r(t) = S(t, t_0) r(t_0) S^+(t, t_0), \tag{6}$$

where t_0 is a certain initial instant of time. The matrix elements of the evolution operator $S(t,t_0)$ are obtained by solving the corresponding differential equations that follow from (3)–(5). It is easy to find that

$$S_{vv'}(t, t_0) = \delta_{vv'} A_{nv}^*(t-t_0) \exp[-iO_{nv}(t-t_0)],$$
$$S_{mm'}(t, t_0) = \delta_{mm'} A_{n,m+2p}(t-t_0) \exp[-iQ_{n,m+2p}(t-t_0)],$$
$$S_{mv}(t, t_0) = i\delta_{v,m+2p}(-1)^{j_c-v} B_{nv}^*(t-t_0) \exp[-iO_{nv}(t-t_0)],$$
$$S_{vm}(t, t_0) = i\delta_{v,m+2p}(-1)^{j_c-v} B_{nv}(t-t_0) \exp[-iO_{nv}(t-t_0)],$$

$$A_{nv}(t-t_0) = \cos\frac{\Omega_{nv}(t-t_0)}{2} + i\frac{\eta-\Delta_{nv}}{\Omega_{nv}}\sin\frac{\Omega_{nv}(t-t_0)}{2},$$

$$B_{nv}(t-t_0) = \frac{U_{nv}}{\Omega_{nv}}\sin\frac{\Omega_{nv}(t-t_0)}{2}, \quad O_{nv} = \frac{(U_{nv}^{(c)} + U_{n,v-2p}^{(a)})}{2\hbar},$$

$$\Omega_{nv} = [(\eta-\Delta_{nv})^2 + |U_{nv}|^2]^{1/2}, \quad \Delta_{nv} = \Delta - (U_{nv}^{(c)} - U_{n,v-2p}^{(a)})/\hbar.$$

The solution of Eqs. (3)–(5) at $\hat{\Gamma} \neq 0$, in the time intervals between the pulses (1), can be easily found by expanding in periodic tensor operators.[20,21]

Resonant interaction with the atoms outside the light wave (1) induces in the medium an electric field **E** that can be obtained by starting from the d'Alambert equation

$$\left(\frac{\partial^2}{\partial y^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{E} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}. \tag{7}$$

The polarization **P** of the medium can be represented in the form

$$\mathbf{P} = \int \mathrm{Sp}(\rho \mathbf{d})\, d\mathbf{v} = \int \mathrm{Sp}(R\mathbf{D})\, d\mathbf{v}, \tag{8}$$

in which

$$\mathbf{D} = e^{-iQ}\mathbf{d}e^{iQ} \approx \mathbf{d} - i(Q\mathbf{d} - \mathbf{d}Q). \tag{9}$$

The matrix elements of the spherical component D_s of the vector (9) are given by the expressions

$$(D_s)_{mv} = \frac{a_n}{\hbar}(-1)^{j_c-v+p}(2j_a+1)^{1/2}\sum_{\varkappa q}(2\varkappa+1)\begin{pmatrix} j_c & j_a & \varkappa \\ v & -m & q \end{pmatrix}$$
$$\times\begin{pmatrix} 1 & 1 & \varkappa \\ s & p & -q \end{pmatrix}\Pi_{-\omega}^{(\varkappa)*} e^{-i\Phi_n} + \frac{a_n}{\hbar}(-1)^{j_c-v}(2j_a+1)^{1/2}\sum_{\varkappa q}(2\varkappa+1)$$
$$\times\begin{pmatrix} j_c & j_a & \varkappa \\ v & -m & q \end{pmatrix}\begin{pmatrix} 1 & 1 & \varkappa \\ s & -p & -q \end{pmatrix}\Pi_\omega^{(\varkappa)*} e^{i\Phi_n}, \tag{10}$$

$$(D_s)_{vv'} = (\bar{D}_s)_{vv'} + (-1)^s(\bar{D}_{-s})_{v'v}, \tag{11}$$

$$(\bar{D}_s)_{vv'} = -\frac{a_n}{\hbar}(-1)^{j_c-v+s}(2j_c+1)^{1/2}\sum_{\varkappa q}(2\varkappa+1)\begin{pmatrix} j_c & j_c & \varkappa \\ v & -v' & q \end{pmatrix}$$
$$\times\begin{pmatrix} 1 & 1 & \varkappa \\ s & p & q \end{pmatrix}\Pi_c^{(\varkappa)} e^{-i\Phi_n},$$

and $(D_s)_{mm'}$ is obtained from $(D_s)_{vv'}$ by letting $c \to a$ and $v \to m$.

Expressions (7), (8), and (10) can be used also to determine the Raman scattering of a test wave

$$\mathbf{E}_0 = \mathbf{l}^{(p)} a_0(t-y/c) \exp(-i\Phi_0) + \text{c.c.}, \quad \Phi_0 = \omega_0 t - k_0 y + \varphi_0$$

in a pre-excited medium. In this case it is necessary to replace a_n, Φ_n, and $\Pi_{\pm\omega}^{(\varkappa)}$ in (10) by $a_0(t-y/c)$, Φ_0, and $\Pi_{\pm\omega_0}^{(\varkappa)}$, respectively.

Expressions (6), (8), (10), and (11) are necessary for the investigation of arbitrary transition phenomena in two-photon resonance with transitions between levels that are degenerate in the projections of the total angular momenta. They make it possible to analyze the polarization properties of these phenomena as functions of the degeneracy multiplicity of the resonant levels, of the type of transition, and of the polarization state of the exciting light waves.

2. OPTICAL NUTATION

If a powerful light pulse in the form of a step

$$\mathbf{E}_1=\mathbf{l}^{(p)}a_1\exp\left[i(ky-\omega t-\varphi_1)\right]+\text{c.c.},\quad 0\leqslant t-y/c,$$
$$\mathbf{E}_1=0,\quad t-y/c<0 \tag{12}$$

is incident on the entrance ($y=0$) into a two-photon exciting medium, a phenomenon known as optical nutation takes place, wherein the alternating processes of absorption and induced emission cause the amplitude of the pulse to oscillate near its leading front. For a distinct observation of the effect it is necessary that the leading-front growth time be short compared with the period of the optical nutation. In addition, to prevent violation of the condition that the interaction with the medium be coherent, the irreversible-relaxation time must be long compared with the nutation period. We therefore assume below that there is no irreversible relaxation, $\hat{\Gamma}=0$.

Let the length L of the resonant medium be such that

$$\frac{a_1\omega|\Pi_\omega^{(\varkappa)}|}{\hbar c}\left|\frac{N_a}{2j_a+1}-\frac{N_c}{2j_c+1}\right|L\ll 1,\quad \varkappa=0.2. \tag{13}$$

In this case the intensity of the electric field due to the reemission of photons by the resonant atoms is small compared with (12) and is obtained with the aid of Eqs. (6) and (7). Ultimately, after the pulse (12) leaves the medium, its intensity time-averaged over the period $2\pi/\omega$ of the fast oscillation is of the form

$$I\left(t-\frac{y}{c}\right)=\frac{ca_1^2}{2\pi}\left[1+2\varepsilon'\left(t-\frac{y}{c}\right)a_1^{-1}\right],\quad 0\leqslant t-\frac{y}{c},\ L\leqslant y, \tag{14}$$

where the amplitude $\varepsilon'(t)$ of the nutation oscillations (14) is given for linear ($p=0$), left-circular ($p=-1$), and right-circular ($p=1$) polarization of the pulse (12) by the expression

$$\varepsilon'(t)=-\varepsilon_0\sum_{\nu=-j_c}^{j_c}\tilde{\Pi}_\nu|U_{1\nu}|\int dv f(v)\frac{\sin\Omega_{1\nu}t}{\Omega_{1\nu}}, \tag{15}$$

$$\varepsilon_0=\pi L\left(\frac{N_a}{2j_a+1}-\frac{N_c}{2j_c+1}\right)\frac{\omega}{c}\left(\frac{a_1\Pi_\omega}{\hbar}\right),$$

$$\tilde{\Pi}_\nu=\left|\sum_{\varkappa}(2\varkappa+1)(2j_a+1)^{1/2}\begin{pmatrix} j_c & j_a & \varkappa \\ \nu & 2p-\nu & -2p\end{pmatrix}\times\begin{pmatrix}1 & 1 & \varkappa\\ -p & -p & 2p\end{pmatrix}\frac{\Pi_\omega^{(\varkappa)}}{\Pi_\omega}\right|,$$

$$\Pi_\omega=(|\Pi_\omega^{(0)}|^2+|\Pi_\omega^{(2)}|^2)^{1/2}.$$

In the absence of thermal motion of the atoms, when

$$\frac{a_1^2\Pi_\omega}{ku\hbar^2}\gg 1,\quad \frac{a_1^2\Pi_a^{(\varkappa)}}{ku\hbar^2}\gg 1,\quad \frac{a_1^2\Pi_c^{(\varkappa)}}{ku\hbar^2}\gg 1, \tag{16}$$

at strict resonance $\Delta=0$ with the vibrational-rotational transition in a diatomic molecule ($\Pi_\omega^{(0)}=\sqrt{10}\Pi_\omega^{(2)}$) and for linear polarization ($p=0$) of the optical nutation, Eq. (15) coincides with the result of Ref. 19. In the indicated region (16) the character of the optical nutation differs substantially from optical nutation on single-photon transitions.[22,23] In particular, under certain conditions[19] it becomes possible to observe undamped nutations, and in some cases the response of the system is reminiscent of random noise.[19]

We consider now nutation in a region where the thermal motion of the atoms cannot be neglected. This is precisely the situation at optical and infrared frequencies:

$$\frac{a_1^2\Pi_\omega}{ku\hbar^2}\ll 1,\quad \frac{a_1^2\Pi_a^{(\varkappa)}}{ku\hbar^2}\ll 1,\quad \frac{a_1^2\Pi_c^{(\varkappa)}}{ku\hbar^2}\ll 1. \tag{17}$$

In the region (17), the period of the nutation oscillations (unlike in Ref. 19) is independent of the Stark shifts of the levels $\Pi_a^{(\varkappa)}$ and $\Pi_c^{(\varkappa)}$, but is determined entirely by the field broadening $a_1^2\Pi_\omega^{(\varkappa)}/\hbar^2$. This circumstance can serve as a basis for an experimental determination of the indicated quantity.

Computer calculations[1)] show that in the case of the optically forbidden transitions $j_c\to j_a$, which are characterized by large numerical values of the angular momenta, $j_c\gg 1$ and $j_a\gg 1$, the period of the optical nutation is well described by the quantity $2\pi/\Omega_{\text{nut}}$, where

$$\Omega_{\text{nut}}=\max|U_{1\nu}|.$$

For each of the transitions $j\leftrightarrow j+2$, $j\leftrightarrow j+1$, and $j\to j$, the quantity indicated is different, depending on the polarization p of the light wave:

a) for the transitions $j\leftrightarrow j+2$ ($j\gg 1$)

$$\Omega_{\text{nut}}=\frac{\sqrt{5}}{2}\frac{a_1^2|\Pi_\omega^{(2)}|}{\hbar^2},\quad p=0,$$

$$\Omega_{\text{nut}}=\frac{\sqrt{5}a_1^2|\Pi_\omega^{(2)}|}{\hbar^2},\quad p=\pm 1;$$

b) for the transitions $j\leftrightarrow j+1$ ($j\gg 1$)

$$\Omega_{\text{nut}}=\frac{\sqrt{5}}{2}\frac{a_1^2|\Pi_\omega^{(2)}|}{\hbar^2},\quad p=0,$$

$$\Omega_{\text{nut}}=\frac{3\sqrt{15}}{8}\frac{a_1^2|\Pi_\omega^{(2)}|}{\hbar^2},\quad p=\pm 1;$$

c) for the transition $j\to j$ ($j\gg 1$)

$$\Omega_{\text{nut}}=\frac{a_1^2}{2\sqrt{3}\hbar^2}\max_\nu|2\Pi_\omega^{(0)}+\sqrt{10}\Pi_\omega^{(2)}(1-3\nu^2/j^2)|,\quad p=0,$$

$$\Omega_{\text{nut}}=\sqrt{\frac{15}{8}}\frac{a_1^2|\Pi_\omega^{(2)}|}{\hbar^2},\quad p=\pm 1.$$

The circumstance noted should make it possible in practice to determine the parameters $\Pi_\omega^{(0)}$ and $\Pi_\omega^{(2)}$ of the two-photon interaction operator by starting from the experimentally obtained value of the period of the optical nutation. In addition, by investigating experimentally the change of the nutational oscillations when the linear polarization of the pulse (12) is replaced by circular, we can distinguish between the optically forbidden transitions $j\leftrightarrow j+2$, $j\to j+1$, and $j\to j$. Thus, in the case of $j\leftrightarrow j+2$ transitions with $j\gg 1$ the period of the linearly polarized nutation is double the period of the nutational oscillations with circular polarization and furthermore the latter attenuate more rapidly (Fig. 1). Opti-

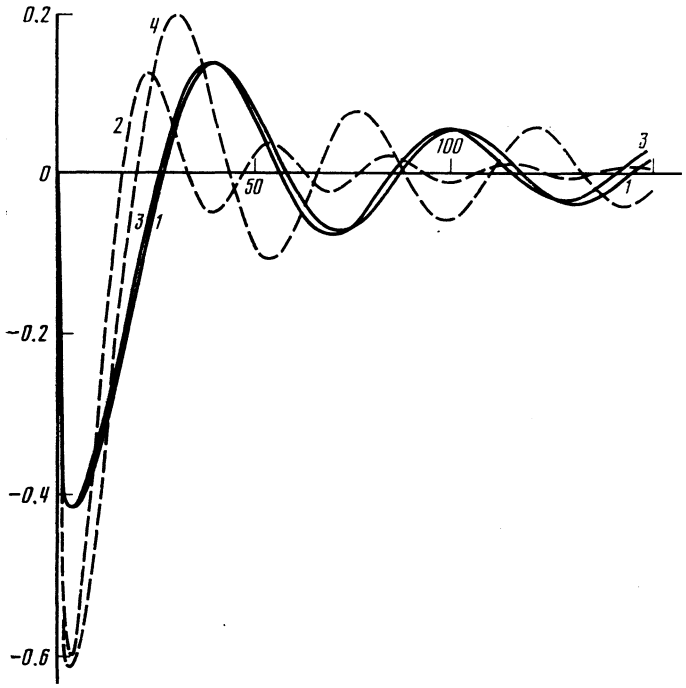

FIG. 1. Two-photon optical nutation near the leading edge of a rectangular exciting pulse in the region (17) for large values of the angular momenta of the resonant levels. Cases of the quantum transitions $j \leftrightarrow j+2$ (curves 1 and 2) and $j \leftrightarrow j+1$ (curves 3 and 4). Curves 1 and 3 describe nutation with linear polarization, and curves 2 and 4 with circular polarizations. The abscissas and the ordinates are respectively $2kut$ and $(\pi^{1/2}a_1^2\Pi_\omega/2ku\hbar^2)(\varepsilon'(t)/\varepsilon_0)$. It is assumed that $a_1^2\Pi_\omega^{(2)}/2ku\hbar^2 = 0.1$, $\Delta = 0$.

cal nutation on the transitions $j \leftrightarrow j+1$ ($j \gg 1$) with linear and circular polarizations attenuates approximately at the same rate, whereas the period of the nutation with linear polarization exceeds that with circular polarization by 1.3 times. The laws of optical nutation on the transition $j \to j$ are less universal, since they are determined also by the relation between the quantities $\Pi_\omega^{(0)}$ and $\Pi_\omega^{(2)}$. For example, for vibrational-rotational transitions in a diatomic molecule, when $\Pi_\omega^{(0)} = \sqrt{10}\Pi_\omega^{(2)}$, the period of the linearly polarized nutation is half the period of the nutation with circular polarization. If $\Pi_\omega^{(0)} = \Pi_\omega^{(2)}$, the periods of nutation with circular and linear polarizations are approximately equal (Fig. 2). The regularities indicated can serve as a basis for a method of identifying two-photon (optically forbidden) atomic and molecular

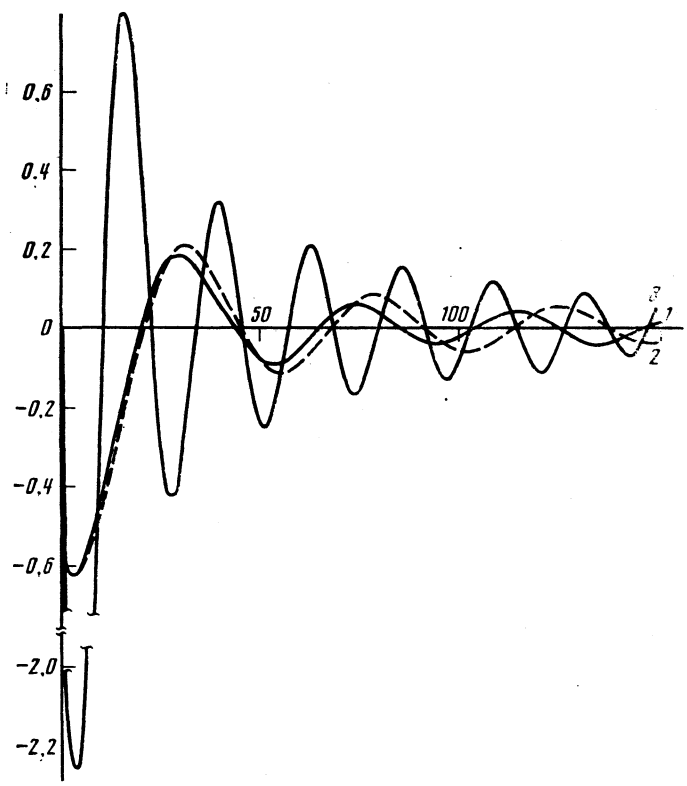

FIG. 2. Transition $j \to j$. Curves 1 and 2 correspond to the case $\Pi_\omega^{(0)} = \Pi_\omega^{(2)}$. Curves 2 and 3 describe optical nutation on the vibrational-rotational transition in a diatomic molecule (when $\Pi_\omega^{(0)} = \sqrt{10}\Pi_\omega^{(2)}$). All the value of the parameters are the same as in Fig. 1.

transitions.

3. THIRD-HARMONIC GENERATION

When the high-power linearly polarized pulse (12) propagates through a two-photon absorbing medium, optical nutation is accompanied also by generation of electromagnetic radiation at the frequency[18,24-27] 3ω (third-harmonic generation). This radiation is linearly polarized in the same plane as the exciting pulse. We shall consider third-harmonic generation in a medium taken beforehand out of equilibrium by a two-photon interaction with a perturbing pulse

$$\mathbf{E}_1=\mathbf{l}_1 a_1 \exp\left[i(ky-\omega t-\varphi_1)\right]+\text{c.c.}, \quad 0\leqslant t-y/c\leqslant\tau_1. \tag{18}$$

Then the pulse

$$\mathbf{E}_2=\mathbf{l}_2 a_2 \exp\left[i(ky-\omega t-\varphi_2)\right]+\text{c.c.}, \quad \tau_1+\tau\leqslant t-y/c\leqslant\tau_1+\tau_2+\tau, \tag{19}$$

which propagates in the medium at a time τ after the passage of the perturbing pulse (18), generates radiation whose electric field intensity is

$$\mathbf{E}_3=\boldsymbol{\varepsilon}_3(t-y/c)\exp\left[i(3ky-3\omega t-3\varphi_2)\right]+\text{c.c.}, \quad \tau_1+\tau\leqslant t-y/c\leqslant\tau_1+\tau_2+\tau, \tag{20}$$

where

$$\varepsilon_3(t)=-\varepsilon_{03}\int dv f(v)\,(\mathbf{l}_z V_0+2^{1/2}\mathbf{l}_x V_{-1}), \tag{21}$$

$$\varepsilon_{03}=2\pi L\frac{N_a}{2j_a+1}\frac{3\omega}{c}\left(\frac{a_2\Pi_{-\omega}}{\hbar}\right),$$

$$V_q=\sum_{\varkappa\varkappa'}\{[F^{(a)}_{\varkappa\varkappa' q}A^{(a)}_{\varkappa'}\exp(-\gamma_a^{(\varkappa')}\tau)-F^{(c)}_{\varkappa\varkappa' q}A^{(c)}_{\varkappa'}\times\exp(-\gamma_c^{(\varkappa')}\tau)]d^{\varkappa'}_{q0}(\psi)$$

$$+\delta_{q0}F_\varkappa[1-\exp(-\gamma_a^{(0)}\tau)]\}\begin{pmatrix}1&1&\varkappa\\ q&0&-q\end{pmatrix}\frac{\Pi^{(\varkappa)*}_{-\omega}}{\Pi_{-\omega}},$$

$$A^{(a)}_\varkappa=\sum_m(-1)^{j_a-m}\begin{pmatrix}j_a&j_a&\varkappa\\ m&-m&0\end{pmatrix}|A_{1m}(\tau_1)|^2,$$

$$A^{(c)}_\varkappa=\sum_\nu(-1)^{j_c-\nu}\begin{pmatrix}j_c&j_c&\varkappa\\ \nu&-\nu&0\end{pmatrix}|B_{1\nu}(\tau_1)|^2,$$

$$F^{(a)}_{\varkappa\varkappa' q}=(2\varkappa+1)(2\varkappa'+1)\times(2j_a+1)^{1/2}\sum_{\nu m}\begin{pmatrix}j_c&j_a&\varkappa\\ \nu&-m&q\end{pmatrix}\begin{pmatrix}j_a&j_a&\varkappa'\\ \nu&-m&q\end{pmatrix}\times(-1)^{j_a-m}A_{2m}{}^*(t')B_{2\nu}(t')\exp[i(O_{2m}-O_{2\nu})t'],$$

$$F^{(c)}_{\varkappa\varkappa' q}=(2\varkappa+1)(2\varkappa'+1)(2j_a+1)^{1/2}\sum_{\nu m}\begin{pmatrix}j_c&j_a&\varkappa\\ \nu&-m&q\end{pmatrix}\begin{pmatrix}j_c&j_c&\varkappa'\\ \nu&-m&q\end{pmatrix}\times(-1)^{j_c-\nu}A_{2\nu}{}^*(t')B_{2m}(t')\exp[i(O_{2m}-O_{2\nu})t'],$$

$$F_\varkappa=(2\varkappa+1)(2j_a+1)^{1/2}\sum_\nu\begin{pmatrix}j_c&j_a&\varkappa\\ \nu&-\nu&0\end{pmatrix}A_{2\nu}{}^*(t')B_{2\nu}(t'),$$

$$t'=t-\tau-\tau_1, \quad \gamma_a^{(\varkappa)}=\gamma_a+\Gamma_a^{(\varkappa)}, \quad \gamma_c^{(\varkappa)}=\gamma_c+\Gamma_c^{(\varkappa)}.$$

Here τ_1 and τ_2 are the durations of the pulses (18) and (19), ψ is an arbitrary angle between the polarization vectors $\mathbf{l}_1=\mathbf{l}_z\cos\psi+\mathbf{l}_x\sin\psi$ and $\mathbf{l}_2=\mathbf{l}_z$ of these pulses, $d^{\varkappa}_{qq'}(\beta)$ is the Wigner D function $D^{\varkappa}_{qq'}(\alpha,\beta,\gamma)$ at $\alpha=\gamma=0$ (Ref. 21), and the remaining quantities are defined in Sec. 1.

Expressions (20) and (21) were obtained under the assumption that the durations of the pulses (18) and (19) are small compared with the irreversible-relaxation time, and the time interval τ between the pulses is long compared with τ_1 and τ_2 as well as with the time $T_0=1/2ku$ of the Doppler dephasing of the emitters. In addition, it is assumed that the level E_c is not populated: $N_c=0$.

The third-harmonic amplitude (21) contains the relaxation parameters $\gamma_a^{(\varkappa)}$ and $\gamma_c^{(\varkappa)}$ with even $\varkappa$ and with $\varkappa=0$. These parameters describe the relaxations of the population, of the alignment, and of the other multipole moments of the resonant levels.[20] If the time interval τ is longer than the irreversible-relaxation time, the third-harmonic polarization plane coincides with the polarization plane of the exciting pulse (19). If τ does not exceed the irreversible-relaxation time, the parametric frequency transformation takes place with strongly inhomogeneous population of the Zeeman sublevels of both the upper and lower levels. Under these conditions the linearly polarized wave (19), which is an assembly of equal numbers of left- and right-polarized photons, generates unequal numbers of left- and right-polarized photons of frequency 3ω. As a result, at $\psi\neq 0$ the polarization of the third harmonic becomes elliptic: a component (20) appears in a direction (X axis) perpendicular to the plane of polarization of the exciting pulse (19). This makes it also possible, by measuring the damping (with changing τ) of the third-harmonic intensity along the x projection of the electric field intensity (20), to investigate the relaxation of the alignment and of the other multipole moments of the levels E_a and E_c.

We shall illustrate the foregoing using as an example an atomic transition with change of total angular momentum $j_c=5/2\rightarrow j_a=1/2$. If the angle ψ between the planes of polarization of the momenta (18) and (29) are chosen to satisfy the condition $\cos\psi=(3/7)^{1/2}$, i.e., $d^4_{-10}(\psi)=0$, the projection of the amplitude (21) on the X axis attenuates with increasing τ like

$$\exp(-\gamma_c^{(2)}\tau),$$

so that it is possible to determine the rate $\gamma_c^{(2)}$ of the relaxation of the alignment of the level E_c. The same quantity, but at $\cos\psi\neq(3/7)^{1/2}$, $\psi\neq 0$, and $\psi\neq\pi/2$, varies like

$$F^{(c)}_{22-1}A^{(c)}_2 d^2_{-10}(\psi)\exp(-\gamma_c^{(2)}\tau)+F^{(c)}_{24-1}A^{(c)}_4 d^4_{-10}(\psi)\exp(-\gamma_c^{(4)}\tau),$$

where $\gamma_c^{(2)}$ is known. This law enables us to determine $\gamma_c^{(4)}$ in experiment.

We note that an investigation of the damping of the third-harmonic intensity on the Z axis makes it possible to determine from (21) the relaxation rates $\gamma_a^{(0)}$ and $\gamma_c^{(0)}$ of the populations of the levels E_a and E_c. In this case, however, the reduction of the experimental results becomes somewhat more complicated. It is important to emphasize that in the general case of arbitrary angular momenta of the levels, Eq. (21) makes it possible, in principle, to determine the constants $\gamma_a^{(\varkappa)}$ and $\gamma_c^{(\varkappa)}$ with even $\varkappa$ and $\varkappa=0$ from experimental

investigations of the damping of the intensity of the third harmonic (20) as a function of τ for different values of the angle ψ.

4. PHOTON ECHO

In contrast to optical echo at one-photon resonance (see the reviews[18,28]), the propagation of two ultrashort resonant pulses (18) and (19) in a two-photon absorbing medium does not lead under ordinary conditions to the onset of an echo phenomenon, inasmuch as in this case the single-photon transition is forbidden in the dipole approximation. To observe a photon-echo signal it is necessary to illuminate the excited medium by nonresonant radiation

$$\mathbf{E}_0=\mathbf{l}_z a_0 \exp\left[i(k_0 y-\omega_0 t-\varphi_0)\right]+\text{c.c.}, \tag{22}$$

or apply external static fields. In this case a photon echo is produced at the instant of time $t=2\tau$ both at the Stokes frequency $\omega_s=\omega_0-2\omega$ and at the anti-Stokes frequency $\omega_{an}=\omega_0+2\omega$ (we assume for the sake of argument that $\omega_0>2\omega$). This process is usually considered within the framework of the two-photon vector model.[12,13,19] This model, however, cannot describe the photon echo if the exciting pulses and the nonresonant extraneous illumination are differently polarized. The solution of the problem is based here on Eqs. (6), (8), and (10) above. By using them it is easy to obtain the intensities $\mathbf{E}_s$ and $\mathbf{E}_{an}$ of the photon-echo electric field at both the Stokes and the anti-Stokes frequencies when the sounding wave (22) is scattered in a medium excited by pulses (18) and (19) with polarization vectors

$$\mathbf{l}_1=\mathbf{l}_z\cos\psi_1+\mathbf{l}_x\sin\psi_1 \text{ and } \mathbf{l}_2=\mathbf{l}_z\cos\psi_2+\mathbf{l}_x\sin\psi_2.$$

Leaving out the intermediate calculations, we write down the photon-echo electric-field intensity

$$\mathbf{E}_{s,an}=\boldsymbol{\varepsilon}_{s,an}(t-y/c)\exp\left[\mp i(k_{s,an}y-\omega_{s,an}t-\varphi_{s,an})\right]+\text{c.c.}, \tag{23}$$

where the upper and lower signs correspond to the Stokes and anti-Stokes frequencies, $\omega_{s,an}=ck_{s,an}$, $\varphi_{s,an}=\varphi_0 \pm 2(\varphi_1-2\varphi_2)$, and the photon-echo amplitudes are given by the expressions

$$\boldsymbol{\varepsilon}_{s,an}(t)=\mp\varepsilon_0^{s,an}\int dv f(v)\,(\mathbf{l}_z W_0^{s,an}+2^{1/2}\mathbf{l}_x W_{-1}^{s,an}) \times \exp\left[-i(\eta-\Delta)(t-t_e)\right], \tag{24}$$

$$\varepsilon_0^{s,an}=2\pi L\left(\frac{N_a}{2j_a+1}-\frac{N_c}{2j_c+1}\right)\frac{\omega_{s,an}}{c}\frac{a_0\Pi_{\pm\omega_0}}{\hbar},$$

$$W_q^{s,an}=\sum_{\varkappa q'}\begin{pmatrix}1 & 1 & \varkappa\\ q & 0 & -q\end{pmatrix}\frac{\Pi_{\pm\omega_0}^{(\varkappa)*}}{\Pi_{\pm\omega_0}}d_{-qq'}^{\varkappa}(\psi_2)F_{\varkappa q'},$$

$$F_{\varkappa q'}=\sum_{\varkappa'}H_{\varkappa\varkappa' q'}G_{\varkappa'}{}^{*}d_{q'0}^{\varkappa'}(\psi_2-\psi_1) \times\exp\{-[\gamma_{ca}^{(\varkappa)}+\gamma_{ca}^{(\varkappa')}+i(\Delta_{ca}^{(\varkappa)}-\Delta_{ca}^{(\varkappa')})]\tau\},$$

$$H_{\varkappa\varkappa' q'}=(2\varkappa+1)\times(2\varkappa'+1)(2j_a+1)^{1/2}\sum_{m\nu}\begin{pmatrix}j_a & j_c & \varkappa\\ m & -\nu & q'\end{pmatrix}\begin{pmatrix}j_c & j_a & \varkappa'\\ m & -\nu & q'\end{pmatrix} \times B_{2m}(\tau_2)B_{2\nu}(\tau_2)\exp[i(O_{2\nu}-O_{2m})\tau_2],$$

$$G_{\varkappa'}=\sum_m\begin{pmatrix}j_a & j_c & \varkappa'\\ m & -m & 0\end{pmatrix}A_{1m}{}^{*}(\tau_1)B_{1m}(\tau_1),$$

$$\gamma_{ca}^{(\varkappa)}=(\gamma_a+\gamma_c)/2+\Gamma_{ca}^{(\varkappa)}, \qquad t_e=2\tau+\tau_1+\tau_2.$$

The photon-echo signals (23) occur near an instant of time $t=t_e$. With increasing τ, they attenuate as a result of irreversible relaxation. In contrast to the results of the preceding section, this damping is due only to the relaxation constants $\gamma_{ca}^{(\varkappa)}$ and $\Delta_{ca}^{(\varkappa)}$ with even $\varkappa$ and $\varkappa=0$ and 1; these constants describe the relaxation of the optical-coherence matrix of the optically forbidden transition $j_c\to j_a$ as a result of radiative decay and of the elastic depolarizing atomic collisions. The photo-echo damping law (23) varies with the values of ψ_1 and ψ_2. It makes it possible to estimate the relaxation constants $\gamma_{ca}^{(\varkappa)}$ and $\Delta_{ca}^{(\varkappa)}$ with even $\varkappa$ and $\varkappa=0$ from the measured damping of the photon echo as a function of τ at the different values of the angles ψ_1 and ψ_2. The possibility of using the general formula (24) for the investigation of the relaxation can be illustrated with the simple example of the atomic transition $j_c=3\to j_a=1$.

If we put $\psi_2=0$ and $\cos\psi_1=(3/7)^{1/2}$, the projection of the amplitude (24) on the X axis in the case $j_a=1$ and $j_c=3$ depends on τ in the following manner:

$$\exp(-2\gamma_{ca}^{(2)}\tau).$$

A comparison of this dependence with the experimental one makes it possible to determine $\gamma_{ca}^{(2)}$. On the other hand, at $\psi_2=0$ and $\cos\psi_1=1/\sqrt{3}$ the projection of the amplitude (24) on the Z axis attenuates with changing τ like

$$\exp[-(\gamma_{ca}^{(2)}+\gamma_{ca}^{(4)})\tau-i(\Delta_{ca}^{(2)}-\Delta_{ca}^{(4)})\tau].$$

This damping law makes it possible to determine $\gamma_{ca}^{(4)}$. The quantity $\Delta_{ca}^{(2)}-\Delta_{ca}^{(4)}$ can be measured by investigating the damping of the echo intensity as a function of τ at $\psi_1=\psi_2=0$; this damping is determined by the factor

$$\exp(-4\gamma_{ca}^{(2)}\tau)\{1+\alpha_1\cos[(\Delta_{ca}^{(2)}-\Delta_{ca}^{(4)})\tau+\alpha_2] \times\exp(\gamma_{24}\tau)+\alpha_3\exp(2\gamma_{24}\tau)\},$$

where $\gamma_{ca}^{(2)}$ and $\gamma_{24}=\gamma_{ca}^{(2)}-\gamma_{ca}^{(4)}$ are known in accordance with the foregoing, while the quantities α_1, α_2, and α_3 do not depend on τ and follow from the general formula (24).

5. DISCUSSION OF RESULTS

The main difference between the present paper and the known researches is that account is taken of the degeneracy of the resonance levels and of the polarization state of the exciting pulses. In the case of optical nutation with linear and circular polarizations, we were able to establish a definite dependence of the period of the nutation oscillations on the parameters of the two-photon-operator interaction and on the type of the optically forbidden transition. Undoubted interest attaches to the use of the observed regularities in practice, with an aim at investigating optically forbidden transitions and determining the parameters of the two-photon-interaction operator. Notice must be taken, however, of

the following. It is not desirable to investigate optical nutation with an aim of obtaining spectroscopic information by the method of Stark pulses, for in this case there occurs a complicated transition process which constitutes in the best case a superposition of nutation oscillations and a decay of free induction, in analogy with the phenomena occurring in single-photon resonance.[28] It seems advisable to set up experiments on two-photon nutations using the recently proposed[29] method, already experimentally realized for one-photon resonance,[30] of producing optical nutation by shifting the phase of the light wave. In this case, as shown by calculations, all the regularities of optical nutations formulated in the present article remain in force near the leading edge of a rectangular exciting pulse. Optical nutation was excited in an experiment[7] by using a pulse of a standing wave in whose field the thermal motion of the atoms is insignificant, so that the regularities obtained do not hold.

Attention must be called to the promise offered by the use of third-harmonic generation in spectroscopy. Usually generation of harmonics is regarded as a means of obtaining intense coherent radiation in the short-wave region of the spectrum.[18,24-27] Yet in the study of ordinary echo effects, where the resonant medium is excited by two and more ultrashort pulses, definite interest attaches to the investigation of parametric conversion of the frequency of the second exciting pulse, since it makes it possible to determine the quantum-state relaxation times which are determined both by radiative decay and by elastic depolarizing atomic collisions. The polarization features of third-harmonic generation by the second pulse can then be used to separate the third-harmonic signal from the exciting pulses. To this end we need counterpropagation of the first pulse

$$\mathbf{E}_1=(\mathbf{l}_z\cos\psi+\mathbf{l}_x\sin\psi)\,a_1\exp[-i(ky+\omega t+\varphi_1)]+\text{c.c.},$$

$$0\leqslant t+y/c\leqslant\tau_1 \qquad (25)$$

and of the second pulse (19). The third-harmonic electric-field intensity is described in this case as before by expressions (20) and (21), from which it can be seen that at $\psi\neq 0$ the projection of the third harmonic on the X axis is completely separated from the exciting pulses (25) and (19).

We emphasize in conclusion that the mathematical formalism developed in the article [Eqs. (6), (8), (10), and (11)] can be used also to study the polarization peculiarities of coherent transition phenomena in two-photon resonance of two light waves traveling in the same direction and having frequencies ω_1 and ω_2 satisfying the condition $\omega_1+\omega_2\approx\omega_{ca}$. The equations obtained can cover also the case of nonstationary SRS, when $\omega_1-\omega_2\approx\omega_{ca}$. In the case of two-photon resonance $\omega_1+\omega_2\approx\omega_{ca}$ the components of the waves making up the two-frequency exciting wave pulse must be of like polarization, while in Raman resonance $\omega_1-\omega_2\approx\omega_{ca}$ these waves should have either like linear or unlike circular polarizations. All that changes in all the indicated cases are then the expressions for $\Pi_\omega^{(\varkappa)}$, $\Pi_a^{(\varkappa)}$, and $\Pi_c^{(\varkappa)}$.

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