Raman scattering of an ultrashort resonant probe pulse in a monatomic gas

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We investigate the Stokes free induction and photon echo, and also the anti-Stokes free induction, which are produced in a gas after passage of an ultrashort probe pulse delayed in time relative to ultrashort biharmonic pumping and are at resonance with an allowed atomic transition adjacent to a Raman transition. The considered nonstationary four-photon process is given a physical interpretation, and the tensor of the cubic susceptibility is investigated. It is proved that the phenomena observed can be used in coherent spectroscopy in the case of homogeneously and inhomogeneously broadened transitions, for the study of the cubic-susceptibility tensor components, and for the identification of Raman transitions.

A process extensively investigated in picosecond coherent spectroscopy is nonstationary Raman scattering (NRS) of light under ultrashort biharmonic pumping, when the nonresonant probe pulse is long[1–4] or ultrashort but delayed in time.[3–5] This phenomenon was first used in Ref. 1 to study ultrafast processes and to observe the time dependence of the free decay of induced dielectric polarization in nondegenerate system. If the presence of nondegeneracy of the resonant levels, however, the NRS carries much more physical information if the numerous polarization properties of the scattered light are used together with the intensity-damping law.[6,7] In a gas at a pressure higher than one Torr, the atomic transitions are usually homogeneously broadened and the NRS, in addition to yielding other information, makes it possible to determine the damping of an individual multipole moment of a forbidden transition. Yet in a rarefied gas with pressure much lower than one Torr the atomic transitions are inhomogeneously broadened by the Doppler effect, so that the NRS previously obtained under biharmonic pumping[1–7] is suppressed by the Doppler dephasing of the active atoms and is not suitable for the investigation of irreversible relaxation.

We consider in the present paper new phenomena that appear in NRS only when an ultrashort resonant probe pulse is scattered; they are called Stokes free induction (SFI), Stokes photon echo (SPE), and anti-Stokes free induction (ASI). They appear after the action of the indicated probe pulse of frequency ω_3 close to the frequency of the atomic transition $E_b \to E_a$ ($E_c \to E_a$) that is adjacent to the Raman transition $E_b \to E_c$, while the frequencies ω_1 and ω_2 of the ultrashort biharmonic pump pulses satisfy the condition $\omega_1 - \omega_2 \approx (E_c - E_b)\hbar^{-1}$ of Raman resonance or $\omega_1 + \omega_2 \approx (E_c - E_b)\hbar^{-1}$ of two-photon resonance. If a probe pulse of frequency $\omega_3 \approx (E_a - E_b)\hbar^{-1}$ affects the lower level E_b of the Raman transition in a three-level system $E_b < E_c < E_a$, the anti-Stokes emission at each point of a gas is negligibly small compared with the Stokes emission during the passage of the probe pulse, and is absent after this passage, the source of the Stokes signal being the induced dielectric polarization in the allowed transition $E_c \to E_a$ (Fig. 1a). Passage of the probe pulse is followed by free decay of the induced Stokes dielectric polarization. This decay, regardless of the character of the atomic-transition broadening, is accompanied by spontaneous coherent emission in the form of SFI at the same frequency $\omega_s = \omega_3 - \omega_1 + \omega_2$ and with the same spatial synchronism condition for the wave vectors $\mathbf{k}_s = \mathbf{k}_3 - \mathbf{k}_1 + \mathbf{k}_2$ as the Stokes NRS produced during the passage of this probe pulse. In the case of homogeneously broadened transitions the SFI serves as the basis for the formation of the SPE. If an ultrashort probe pulse of frequency $\omega_3 \approx (E_a - E_c)\hbar^{-1}$ affects the upper level E_c of the Raman transition $E_b \to E_c$, the source of the NRS is the anti-Stokes dielectric polarization induced on the other allowed transition $E_b \to E_c$ (Fig. 1b). Its free decay after the passage of the probe pulse produces ASI at a frequency $\omega_a = \omega_3 + \omega_1 - \omega_2$ with a wave vector $\mathbf{k}_a = \mathbf{k}_3 + \mathbf{k}_1 - \mathbf{k}_2$. No anti-Stokes photon echo on homogeneously broadened transitions takes place, however. A similar picture is observed at a different arrangement of the resonance levels, $E_a < E_b < E_c$ (Figs. 2a, b). If the atomic transitions are uniformly broadened, the polarization properties of the SFI and ASI, and the laws governing their damping, make it possible to investigate ultrafast processes and determine the relaxation constant of an optically allowed as well as forbidden transition. In addition, the forbidden transitions are identified, and the parameters of the cubic-susceptibility tensor are determined. The same information for inhomogeneously broadened transition can be obtained with the aid of SPE. This expands appreciably the range of practical applications of the obtained new four-photon interaction of atoms with light waves.

1. BASIC EQUATIONS AND NOTATIONS

The propagation of a light pulse having a carrier frequency ω in a gas is described by the equations

$$\nabla\times\nabla\times\mathbf{E} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\hat{\varepsilon}\mathbf{E} = -\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}, \quad \operatorname{div}(\hat{\varepsilon}\mathbf{E}+4\pi\mathbf{P})=0, \tag{1}$$

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla - \Gamma\right)\rho = \frac{i}{\hbar}\left[\rho(H-\mathbf{E}\mathbf{d}) - (H-\mathbf{E}\mathbf{d})\rho\right] \tag{2}$$

where $\hat{\varepsilon}$ is a linear integral operator[8] acting on the electric field $\mathbf{E}$ of the light wave, $\varepsilon(\omega)$ is the dielectric constant which depends on the carrier frequency ω and takes into account the influence of the impurity gas and of the nonresonant levels of the active atoms, and $\Delta\omega$ is the width of the spectrum of the indicated light pulse. Next, $\mathbf{P}$ is the dielectric polarization vector of the gas medium and is due to the resonant levels of the active atoms, c is the speed of light in the vacuum, ρ is the density matrix, H is the Hamiltonian of the free active medium in its cms, $\mathbf{d}$ is the dipole-moment

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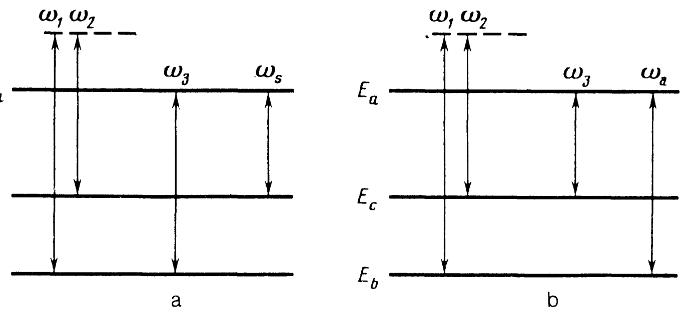

FIG. 1. Arrangement of levels E_a, E_b, and E_c for Stokes (a) and anti-Stokes (b) Raman scattering at frequencies $\omega_s = \omega_3 - \omega_1 + \omega_2$ and $\omega_a = \omega_3 + \omega_1 - \omega_2$.

operator, $\mathbf{v}$ is the thermal velocity of the active atom, and Γ_ρ is the collision integral. To write down the latter we recognize that the state of an atom with zero nuclear spin is characterized by the energy E, total angular momentum J, and its component M along the quantization axis. We consider below three resonant levels which are labeled, together with the other quantities of the complete set, by the subscripts a, b, and c.

The collision intergal Γ_ρ has a simple form if the density matrix ρ is expanded in terms of the irreducible tensor operators ($3J$-symbols[9]); if $\mathbf{E} = 0$ this transforms Eq. (2) into a system of independent equations

$$\left[\frac{\partial}{\partial t} + \mathbf{v}\nabla + i(\omega_{cb} + \Delta_{cb}^{(\varkappa)}) + \gamma_{cb}^{(\varkappa)}\right] \rho_q^{(\varkappa)}(J_cJ_b) = 0,$$

$$|J_c - J_b| \leqslant \varkappa \leqslant J_c + J_b, \tag{3}$$

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \gamma_b^{(\varkappa)}\right) \rho_q^{(\varkappa)}(J_bJ_b) = \gamma_b N_b f(v)\delta_{\varkappa 0}\delta_{q0}, \qquad 0 \leqslant \varkappa \leqslant 2J_b,$$

$$\gamma_{cb}^{(\varkappa)} = \gamma_{cb} + \Gamma_{cb}^{(\varkappa)}, \qquad \gamma_b^{(\varkappa)} = \gamma_b + \Gamma_b^{(\varkappa)}, \qquad \gamma_{cb} = (\gamma_c + \gamma_b)/2,$$

$$\omega_{cb} = (E_c - E_b)/\hbar, \qquad f(v) = (\pi^{1/2} u)^{-3} \exp(-v^2/u^2).$$

Here $\rho_q^{(\varkappa)}(J_cJ_b)$ is a multipole moment of rank $\varkappa$, which relates the state of the atom with the angular momenta J_c and J_b, and takes the form

$$\rho_q^{(\varkappa)}(J_cJ_b) = (2J_c+1)^{1/2} \sum_{M_bM_c} (-1)^{J_c - M_b} \begin{pmatrix} J_c & J_b & \varkappa \\ M_c & -M_b & q \end{pmatrix} \rho_{M_cM_b}, \tag{4}$$

where $\rho_{M_cM_b}$ is a component of the density matrix ρ. The multipole moment $\rho_q^{(\varkappa)}(J_bJ_b)$ describes the polarization state of an atom at the level E_b and is obtained from (4) by making the substitutions $J_c \to J_b$ and $M_c \to M'_b$. The quantities $\gamma_{cb}^{(\varkappa)}$ and $\gamma_b^{(\varkappa)}$ in the integral are the relaxation constants of these multipole moments, and $\Delta_{cb}^{(\varkappa)}$ describes the shifts of the levels E_b and E_c as a result of elastic collisions. The relaxation constants γ_{cb} and γ_b are due to spontaneous decay and to inelastic gaskinetic equations. Further, N_b is the stationary density of the active atoms on the level E_b without external fields, $f(v)$ is the Maxwell distribution, and u is the most probable velocity. The quantities $\Gamma_{cb}^{(\varkappa)}$, $\Gamma_b^{(\varkappa)}$ and $\Delta_{cb}^{(\varkappa)}$ governed by the elastic collisions were calculated theoretically for small angular momenta,[10] serve in the general case as certain relaxation characteristics that depend on the interatomic interaction potential of the resonant and impurity atoms, and must be determined by experiment. If the impurity-atom mass does not exceed that of the active atom, the quantities $\Gamma_{cb}^{(\varkappa)}$, $\Gamma_b^{(\varkappa)}$ and $\Delta_{cb}^{(\varkappa)}$ are smooth functions of the modulus v of the thermal velocity compared with $v^2 f(v)$ in the vicinity of the most probable velocity u, where they can be regarded as constants. Otherwise account must be taken of the dependence of these quantities on the velocities of the active atoms.

2. STOKES FREE INDUCTION AND PHOTON ECHO

Let the biharmonic pump consist of two ultrashort light pulses

$$\mathbf{E}_n = {}^1/{}_2\mathbf{l}_n a_n \exp[i(\mathbf{k}_n\mathbf{r} - \omega_n t - \varphi_n)] + \text{c.c.} \tag{5}$$

$$a_n = a_n(t'), \qquad t' = t - \mathbf{k}_n\mathbf{r}/\omega_n, \qquad 0 \leqslant t' \leqslant \tau_n, \qquad n = 1, 2,$$

where $\mathbf{l}_n$ is the vector of polarization of the electric field $\mathbf{E}_n$ of the light wave, $a_n(t')$ the slow amplitude, φ_n the constant phase shift, and let the pulse durations be equal, $\tau_1 = \tau_2$. The central frequency ω_n is connected with the wave vector $\mathbf{k}_n$ by the dispersion law $\varepsilon(\omega_n)\omega_n^2 = \mathbf{k}_n^2 c^2$. The frequency difference $\omega_1 - \omega_2$ is close to the frequency ω_{cb} of the optically forbidden transition $J_b \to J_c$, while ω_1 and ω_2 differ substantially from the frequencies of the atomic transitions. The leading front of the pulses (5) crosses the boundary point $\mathbf{r} = 0$ of the entry into the gas at $t = 0$.

The biharmonic pump (5) induces in the gas, on the forbidden atomic transition $J_b \to J_c$, an optical coherence described by a density matrix $\rho_{M_cM_b}$. The latter is determined

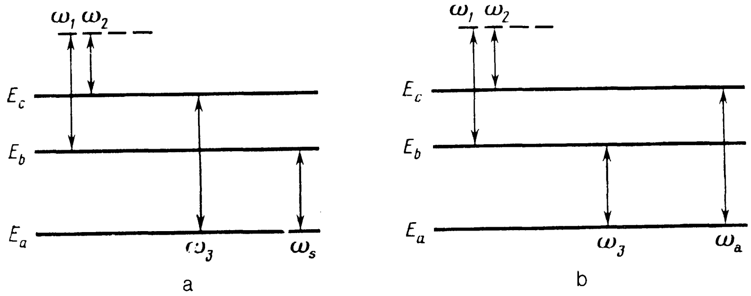

FIG. 2. Alternate level arrangement for Stokes (a) and anti-Stokes (b) Raman scattering.

by solving Eq. (2) during the time of the pumping (5) and by Eqs. (3) and (4) after the termination of the pump, when $\mathbf{E}_1 = \mathbf{E}_2 = 0$. To study the damping of the induced optical coherence after the action of the pump (5), one investigates the scattering of the ultrashort resonant probe pulse

$$\mathbf{E}_3={}^1/_2\mathbf{l}_3a_3\exp\left[i(\mathbf{k}_3\mathbf{r}-\omega_3t-\varphi_3)\right]+\text{c.c.},$$

$$a_3=a_3(t'),\quad t'=t-t_3-\mathbf{k}_3\mathbf{r}/\omega_3,\quad 0\leqslant t'\leqslant\tau_3,\quad \varepsilon(\omega_3)\omega_3^2=\mathbf{k}_3^2c^2, \tag{6}$$

where $t_3 = \tau + \tau_1$ is the instant of entry of this pulse into the gas, and the delay time τ is arbitrary. The carrier frequency ω_3 is chosen to accord with the location of the third resonant level E_a.

We assume first $E_b < E_c < E_a$ and stipulate $\omega_3 \approx \omega_{ab} = (E_a - E_b)/\hbar$ (Fig. 1a). An example is barium ^{138}Ba with atomic levels 6^1S_0, 5^3D_1 and $6^1P_1^0$, $E_c - E_b = 9034$ cm^{-1}, and wavelength corresponding to the $E_b \to E_a$ transition 5535.5 Å. In this case, owing to diffusion, the resonant probe pulse (6) induces at $\rho_{M_cM_b} \neq 0$ in the gas a Stokes dielectric polarization on the optically allowed transition $J_c \to J_a$. The latter is the source of Stokes NRS at a frequency $\omega_s = \omega_3 - \omega_1 + \omega_2 \approx \omega_{ac}$ in the time $t_3 \leqslant t \leqslant t_3 + \tau_3$ of action of the probe pulse (6). At $t_3 + \tau_3 \leqslant t$ and $\mathbf{E}_3 = 0$ the induced Stokes dielectric polarization undergoes a free decay accompanied by spontaneous coherent SFI. Physically, SFI constitutes Stokes NRS produced after the action of the resonant probe pulse (6) as a result of the frequency dispersion of the cubic-susceptibility tensor, at frequencies ω_s and $\omega_1 - \omega_2$ close to the frequencies ω_{ac} and ω_{cb} of the adjacent atomic transitions $E_c \to E_a$ and $E_b \to E_c$ (see the Appendix). This radiation can be easily distinguished from the preceding light pulses both by frequency and by polarization of the waves, and is therefore a convenient source of spectroscopic information. On the contrary, anti-Stokes NRS at the frequency $\omega_a = \omega_3 + \omega_1 - \omega_2$ is produced on account of gaseous-medium dielectric polarization that is proportional at each instant of time to the electric-field probe pulse at the same instant of time. In this case there is no frequency dispersion of the tensor of the cubic susceptibility at the frequency ω_a, so that the anti-Stokes radiation is produced only during the time $t_3 \leqslant t \leqslant t_3 + \tau_3$ of the probe-pulse action and vanishes at $t_3 + \tau_3 < t$. This radiation is of less interest and will not be discussed further in view of its negligible intensity.

The indicated NRS and SFI comprise the Stokes signal that is formed in the lowest order of perturbation theory as a result of four-phonon coherent interaction of light pulses with active atoms. We investigate it by assuming that the gas is an optically thin medium and that the light pulse durations are short:

$$\gamma_{ab}^{(1)}\tau_3\ll1,\quad \gamma_{ac}^{(1)}\tau_3\ll1,\quad \gamma_{cb}^{(\varkappa)}\tau_n\ll1,\quad n=1,2,3,$$

$$\varkappa=0,1,2.$$

Solving Eqs. (1)-(4) by perturbation theory, we obtain the electric field $\mathbf{E}_s$ of the indicated Stokes signal as it leaves the gas:

$$\mathbf{E}_s=\boldsymbol{\varepsilon}_s(t-\mathbf{k}_s\mathbf{r}_0/\omega_s)\exp\left[-i(\mathbf{k}_s\mathbf{r}_0-\omega_st-\varphi_s)\right]+\text{c.c.},$$

$$\omega_s=\omega_3-\omega_1+\omega_2,\qquad \varphi_s=\varphi_3-\varphi_1+\varphi_2, \tag{7}$$

where the subscript s of all the physical quantities corresponds to the Stokes signal. The point with the radius vector $\mathbf{r}_0$ is located on the gas boundary through which the pulse (7) emerges. The amplitude $\boldsymbol{\varepsilon}_s(t)$ for $t_3 \leqslant t$ is given by

$$\boldsymbol{\varepsilon}_s(t)=[\mathbf{M}-(\mathbf{M}\mathbf{k}_s)\mathbf{k}_s/k_s^2]G_sC_s(t)\exp[-(\gamma_{ac}^{(1)}-i\Delta_{ac}^{(1)})(t-\tau)].$$

$$\mathbf{M}={}^1/_3(M_0-M_2)(\mathbf{l}_1\mathbf{l}^{2*})\mathbf{l}_3^*+{}^1/_2(M_2+M_1)(\mathbf{l}_2^*\mathbf{l}_3^*)\mathbf{l}_1 \tag{8}$$

$$+{}^1/_2(M_2-M_1)(\mathbf{l}_3^*\mathbf{l}_1)\mathbf{l}_2^*, \tag{9}$$

$$M_\varkappa=\begin{Bmatrix}1&\varkappa&1\\J_c&J_a&J_b\end{Bmatrix}\Pi_\varkappa(\omega_1,\omega_2)\exp[-(\gamma_{cb}^{(\varkappa)}+i\Delta_{cb}^{(\varkappa)})\tau],$$

$$\varkappa=0,1,2. \tag{10}$$

$$\Pi_\varkappa(\omega_1,\omega_2)$$

$$=\frac{1}{\hbar}\sum_g d_{cg}d_{gb}\begin{Bmatrix}1&\varkappa&1\\J_c&J_g&J_b\end{Bmatrix}\left(\frac{1}{\omega_{gb}+\omega_2}+\frac{(-1)^\varkappa}{\omega_{gb}-\omega_1}\right).$$

$$G_s=\frac{2\sin(\boldsymbol{\Delta}_s\mathbf{L}_s/2)}{\boldsymbol{\Delta}_s\mathbf{L}_s}\exp(i\boldsymbol{\Delta}_s\mathbf{L}_s/2),\qquad \boldsymbol{\Delta}_s=\mathbf{k}_s-\mathbf{k}_3+\mathbf{k}_1-\mathbf{k}_2,$$

$$\mathbf{L}_s=\mathbf{k}_sL_s/k_s.$$

$$C_s(t)=\frac{-i\pi\omega_s^2d_{ca}^*d_{ab}^*L_sN_b}{4\hbar^2c^2k_s(2J_b+1)}\int_0^{\tau_1}d\xi\int_0^{t-t_3}d\eta\,a_1(\xi)a_2^*(\xi)a_3^*(\eta)$$

$$\times\exp\{i\Delta_{12}(t-\xi)-i\Delta_3(t-t_3-\eta)$$

$$-[\mathbf{k}_st+(\mathbf{k}_1-\mathbf{k}_2)\xi-\mathbf{k}_3(t_3+\eta)]^2u^2/4\},$$

$$\Delta_{12}=\omega_1-\omega_2-\omega_{cb},\quad \Delta_3=\omega_3-\omega_{ab},\quad \omega_{gb}=(E_g-E_b)/\hbar, \tag{11}$$

where L_s is the length of the Stokes-signal optical path in the active region of the gas, and the summation over g includes all the levels of the atom. The notation of Ref. 9 is used for the 6J-symbols and for the reduced dipole moments d_{ca} and d_{ab} of the atomic transitions $J_c \to J_a$ and $J_a \to J_b$. Equation (8) was calculated using the inequalities

$$\left|(\varepsilon(\omega_s))^{1/2}-(\varepsilon(\omega_n))^{1/2}\frac{\mathbf{k}_s\mathbf{k}_n}{k_sk_n}\right|\frac{L_s}{c}\ll\left(\tau_1,\frac{1}{|\Delta_{12}|},\frac{1}{|\mathbf{k}_1-\mathbf{k}_2|u}\right),$$

$$n=1,2.$$

$$\left|(\varepsilon(\omega_s))^{1/2}-(\varepsilon(\omega_3))^{1/2}\frac{\mathbf{k}_s\mathbf{k}_3}{k_sk_3}\right|\frac{L_s}{c}\ll\left(\tau_3,\frac{1}{|\Delta_3|},\frac{1}{k_3u}\right). \tag{12}$$

According to (11), the $\mathbf{k}_s$ direction is determined by the spatial synchronism condition

$$\mathbf{k}_s=\mathbf{k}_3-\mathbf{k}_1+\mathbf{k}_2,\qquad \varepsilon(\omega_s)\omega_s^2=\mathbf{k}_s^2c^2. \tag{13}$$

The electric field (7) with amplitude (8) describes Stokes NRS in the time interval $t_3 \leqslant t \leqslant t_3 + \tau_3$ and SFI damping in the interval $t_3 + \tau_3 \leqslant t$. If the two-photon resonance $\omega_1 + \omega_2 \approx \omega_{cb}$ obtains in place of the Raman resonance $\omega_1 - \omega_2 \approx \omega_{cb}$, the Stokes NRS and SFI are described by the same equations (8)-(13), but with the substitutions $\mathbf{k}_2 \to -\mathbf{k}_2$, $\varphi_2 \to -\varphi_2$ and $a_2 \to a_2^*$. For $\omega_3 < \omega_{cb}$ we have in this case $\omega_s = \omega_1 + \omega_2 - \omega_3$.

We assume next that the pulses (5) and (6) are linearly polarized, and the angles between the wave vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ are small and range from 10^{-3} to 10^{-2} rad, since this is frequently the case in experiments.[1] We direct the Cartesian axes X and Z along the vectors $\mathbf{l}_3$ and $\mathbf{k}_3$. In this case the terms $\mathbf{k}_s(\mathbf{M}\cdot\mathbf{k}_s)/k_s^2$ can be neglected and the amplitude

$\boldsymbol{\varepsilon}_s(t)$ is proportional to the vector (9), which takes the form

$$\mathbf{M}={}^1/_3\mathbf{l}_x[M_0\cos(\psi_1-\psi_2)+M_2(3\cos\psi_1\cos\psi_2-\cos(\psi_1-\psi_2))]$$
$$+{}^1/_2\mathbf{l}_y[M_1\sin(\psi_1-\psi_2)+M_2\sin(\psi_1+\psi_2)],\qquad(14)$$

where $\mathbf{l}_x$ and $\mathbf{l}_y$ are the unit vectors along X and Y, while the angles ψ_1 and ψ_2 between the polarizations of the probe pulse (6) and the pump pulses (5) are given, with the same accuracy, by the equations

$$\mathbf{l}_n=\mathbf{l}_x\cos\psi_n+\mathbf{l}_y\sin\psi_n,\quad \mathbf{l}_3=\mathbf{l}_x,\quad n=1,\,2.$$

The characteristic dependence of the amplitude (8) on the angles ψ_1 and ψ_2 makes it possible to select a Stokes signal with polarization properties that are independent of the elastic atomic collisions and of the relaxation processes, but are completely determined by the type of the forbidden atomic transition $J_b\to J_c$ and by the selection rules for the emission and absorption of a light photon in adjacent optically allowed transitions. These polarization properties of the Stokes signal are the basis for the method used to identify the forbidden transition $J_b\to J_c$, a method that coincides with that obtained in Ref. 6 for NRS with a long nonresonant probe pulse in the absence of elastic collisions.

If the atomic transitions are uniformly broadened,

$$(k_3-k_1+k_2)u<\gamma_{ac}^{(1)},\qquad(k_1-k_2)u<\gamma_{cb}^{(\varkappa)},\qquad(15)$$

the SFI intensity $I(t)=c|\boldsymbol{\varepsilon}_s(t)|^2/2\pi$ attenuates with the time t like $\exp(-2\gamma_{ac}^{(1)}t)$, and also decreases with increase of τ because of the presence of a sum of three exponentials (10) in the amplitude (8). However, the intensities $I_1(t)=c|\varepsilon_{sx}(t)|^2/2\pi$ and $I_2(t)=c|\varepsilon_{sy}(t)|^2/2\pi$ corresponding to the components of the electric field (7) along the X and Y axes are proportional, if the angles ψ_1 and ψ_2 are judiciously chosen, only to the square of one exponential from (10). For example, for $\psi_1=-\psi_2=\pi/4$ we have

$$I_1(t)=Q_1\exp[-2\gamma_{ac}^{(1)}(t-\tau)-2\gamma_{cb}^{(2)}\tau],$$
$$I_2(t)=Q_2\exp[-2\gamma_{ac}^{(1)}(t-\tau)-2\gamma_{cb}^{(1)}\tau],$$

whereas for $\psi_1=\psi_2=\arccos(3^{-1/2})$ we obtain

$$I_1(t)=Q_3\exp[-2\gamma_{ac}^{(1)}(t-\tau)-2\gamma_{cb}^{(0)}\tau],$$

where Q_1, Q_2, and Q_3 are independent of t and τ. The exponential damping of the SFI as a function of t as $t_3+\tau_3<t$ determines the relaxation constant $\gamma_{ac}^{(1)}$ of the allowed transition $J_c\to J_a$. At the same time, the indicated intensities measured at $t=t_3+\tau_3$ and regarded as functions of τ enable us to find the relaxation constants $\gamma_{cb}^{(\varkappa)}$ with $\varkappa=0$, 1, and 2 of the forbidden transition $J_b\to J_c$. If $\Delta_3\tau_3<1$, the instant of time $t=t_3+\tau_3$ corresponds to the maximum of the Stokes signal regardless of the form of the constant-sign amplitude $a_3(t)$ of the probe pulse (Fig. 3).

In the case of inhomogeneously broadened transitions

$$(k_3-k_1+k_2)u>\gamma_{ac}^{(1)},\qquad(k_1-k_2)u>\gamma_{cb}^{(\varkappa)},\qquad(16)$$

emission by the excited atoms is suppressed by Doppler diffraction, so that the Stokes NRS and the SFI are weaker. If the inequality (16) becomes quite strong, the Stokes NRS and the SFI are negligibly small. However, in view of the unique behavior of the phase of the Stokes dielectric polarization of each group of atoms having the same velocity component v_z, the SFI intensity increases with time and reaches at the instant

$$t_s=\tau\omega_3/(\omega_3-\omega_1+\omega_2)\qquad(17)$$

a maximum in the form of SPE if the inequality (16) is strong enough. The SPE is the result of the interference of the waves emitted by atoms with all possible velocities v_z. This phenomenon is related to the usual photon echo in an optically allowed transition (see the review[11]), although the mathematical description of the phase memory in (8) is much more complicated. We emphasize that SPE is produced after the passage of an ultrashort resonant probe pulse (6), in contrast to the Raman echo[12,13] and the Raman photon echo,[6,14] whose maxima are reached during the action of a prolonged resonant probe pulse.

According to (8) and (14), which hold for SPE at $t=t_s$, the intensities $I_1(t_s)=c|\varepsilon_{sx}(t_s)|^2/2\pi$ and $I_2(t_s)=c|\varepsilon_{sy}(t_s)|^2/2\pi$ corresponding to the components of the electric fields (7) along the axes X and Y take for the angles $\psi_1=-\psi_2=\pi/4$ the form

$$I_1(t_s)=P_1\exp\left[-2\left(\frac{(\omega_1-\omega_2)\gamma_{ac}^{(1)}}{\omega_3-\omega_1+\omega_2}+\gamma_{cb}^{(2)}\right)\tau\right],\qquad(18)$$

$$I_2(t_s)=P_2\exp\left[-2\left(\frac{(\omega_1-\omega_2)\gamma_{ac}^{(1)}}{\omega_3-\omega_1+\omega_2}+\gamma_{cb}^{(1)}\right)\tau\right],\qquad(19)$$

while for $\psi_1=\psi_2=\arccos(3^{-1/2})$ we obtain

$$I_1(t_s)=P_3\exp\left[-2\left(\frac{(\omega_1-\omega_2)\gamma_{ac}^{(1)}}{\omega_3-\omega_1+\omega_2}+\gamma_{cb}^{(0)}\right)\tau\right].\qquad(20)$$

where P_1, P_2, and P_3 are independent of τ. The relaxation

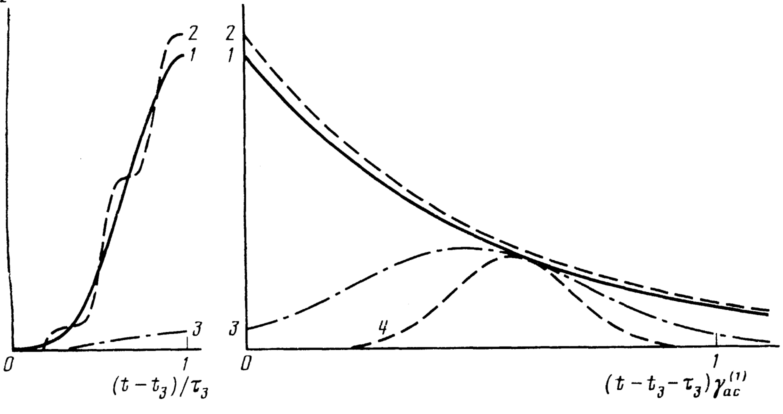

FIG. 3. Intensity $I_2(t)$ of Stokes signal in arbitrary units. It is assumed that $\Delta_3\tau_3=k_3u\tau_3=0.1$, $\psi_1=-\psi_2=\pi/4$, $\gamma_{cb}^{(1)}\tau=0.6$ and $(k_1-k_2)u/\gamma_{cb}^{(1)}=k_3u/\gamma_{ac}^{(1)}$. Curves 1 and 2 correspond to $k_3u/\gamma_{ac}^{(1)}=0.1$ and to probe-pulse profiles $a_3(t)$ equal to $a_0\{1-[2(t-t_3)/\tau_3-1]^2\}$ and $a_0\{1-[2(t-t_3)/\tau_3-1]^2\}\{1-\cos[6\pi(t-t_3)/\tau_3]\}$, respectively, where a_0 is a constant. They describe Stokes NRS in the time interval $0\leqslant t-t_3\leqslant\tau_3$ and SFI at $t_3+\tau_3\leqslant t$ on homogeneously broadened transitions (15). Curves 3 and 4 illustrate the appearance of SPE on inhomogeneously broadened transitions (16) for the parameter $k_3u/\gamma_c^{(1)}$ equal to 4 and 8, respectively.

constant $\gamma_{ac}^{(1)}$ of the allowed transition is determined with the aid of the usual photon echo.[11] The laws governing the damping of the intensities (18)–(20) as functions of τ make it next possible to find the relaxation constants $\gamma_{cb}^{(\varkappa)}$ ($\varkappa = 0$, 1, and 2) of an inhomogeneously broadened forbidden transition.

Let the spectrum of the probe pulse (6) be narrow enough, $k_3 u\tau_3 \gg 1$, and let the inequalities $(k_1 - k_2)u\tau_1 \ll k_3 u\tau_3$ and $\Delta_3 \ll k_3 u$ hold. The factor $C_s(t)$ in the amplitude (8) has for SPE the time dependence

$$a_3\left(\frac{\omega_s}{\omega_3}t - t_3\right)\exp\left[i\left(\frac{\omega_1-\omega_2}{\omega_3}\Delta_3 - \Delta_{12}\right)t\right].$$

Since the argument t of the function $a_3(t)$ is replaced by $t\omega_s/\omega_3 - t_3$, the SPE has the same profile as the probe pulse, but stretched in time on account of the factor $\omega_s/\omega_3 < 1$. The SPE central frequency differs then substantially from ω_3, so that the information entered into the modulation oscillations of the amplitude $a_3(t)$ of a probe pulse of frequency ω_3 is transferred to another frequency region ω_s.

Using the indicated rational choice of the angles ψ_1 and ψ_2, followed by investigation of the NRS emission intensities corresponding to the components of the electric field (7) along the X and Y axes in the cases (15) and (16), it is easy to determine experimentally the ratios

$$\left|\Pi_\varkappa(\omega_1,\omega_2)\begin{Bmatrix}1 & \varkappa & 1\\ J_c & J_a & J_b\end{Bmatrix}\Big/\Pi_0(\omega_1,\omega_2)\begin{Bmatrix}1 & 0 & 1\\ J_c & J_a & J_b\end{Bmatrix}\right|,$$

$$\varkappa = 1, 2. \tag{21}$$

On the other hand, using the method of ultrashort biharmonic pumping with a prolonged nonresonant probe pulse,[7] it is possible also to determine for a monatomic gas one more ratio

$$|\Pi_\varkappa^{(\pm)}\Pi_\varkappa(\omega_1,\omega_2)/\Pi_0^{(\pm)}\Pi_0(\omega_1,\omega_2)|, \qquad \varkappa = 1.2, \tag{22}$$

$$\Pi_\varkappa^{(\pm)} = \frac{1}{h}\sum_g d_{cg}{}^* d_{gb}{}^*\begin{Bmatrix}1 & \varkappa & 1\\ J_c & J_g & J_a\end{Bmatrix}\left(\frac{1}{\omega_{gb}\pm\omega_3} + \frac{(-1)^\varkappa}{\omega_{gc}\mp\omega_3}\right).$$

From the known experimental results (21) and (22) it is then easy to obtain the parameter ratios $|\Pi_\varkappa^{(\pm)}/\Pi_0^{(\pm)}|$ and $|\Pi_\varkappa(\omega_1,\omega_2)/\Pi_0(\omega_1,\omega_2)|$ with $\varkappa = 1$ and 2, which enter in the tensors of the cubic susceptibilities for the Stokes and anti-Stokes signals considered in Refs. 3, 4, and 15.

The characteristic angular dependence in (14) and (8) remains in force when the resonant probe pulse (6) is taken into account exactly (without perturbation theory), although in this case the coefficients $M_\varkappa$ with $\varkappa = 0$, 1, and 2 take on other values proportional to $\Pi_\varkappa(\omega_1,\omega_2)\exp(-\gamma_{cb}^{(\nu)}\tau)$. The described methods of determining the relaxation constants and the method of identifying the forbidden transitions for cases (15) and (16) remain valid here, as is also the method of determining the components of the cubic-susceptibility tensor.

The Stokes NRS, the SFI, and the SPE can also be observed if the level E_a lies lower than the two others $E_a < E_b < E_c$, and the frequency ω_3 of the probe pulse is close to the frequency $\omega_{ca} = (E_c - E_a)/\hbar$ of an adjacent transition (Fig. 2a). In this case, the preceding equations for the Stokes NRS, and SFI, and the SPE remain the same apart from the substitutions

$$M_\varkappa \to (-1)^{\varkappa+1}M_\varkappa, \qquad \Delta_3 \to \omega_3 - \omega_{ca}, \qquad \gamma_{ac}^{(1)} \to \gamma_{ba}^{(1)}, \qquad \Delta_{ac}^{(1)} \to \Delta_{ba}^{(1)}.$$

To determine the relaxation constant of $\gamma_{ba}^{(1)}$ an inhomogeneously broadened allowed transition we can use here either the usual photon echo[11] or the Raman photon echo.[6,14] Examples of transitions adjacent to $E_a \to E_c$ and $E_a \to E_b$ at $E_a < E_b < E_c$ are $4^1S_0 \to 4^1P_1^0$ and $4^1S_0 \to 4^3P_1^0$ in the calcium atoms ^{40}Ca–^{48}Ca with zero nuclear spin, corresponding to wavelengths 4226.7 and 6572.8 Å.

3. ANTI-STOKES FREE INDUCTION

Let the frequency ω_3 of the probe pulse (6) be close to the frequency ω_{ac} of a transition adjacent to the Raman transition $J_b \to J_c$, where $E_b < E_c < E_a$ (Fig. 1b). Anti-Stokes NRS is then produced during the time $t_3 \leqslant t \leqslant t_3 + \tau_3$ of action of this probe pulse, and ASI is produced after this pulse at $t_3 + \tau_3 \leqslant t$. The Stokes radiation is negligibly small here compared with the anti-Stokes one at $t_3 \leqslant t \leqslant t_3 + \tau_3$, and is nonexistent outside the indicated time interval. The electric field of the anti-Stokes signal at the exit from the gas is described at $t_3 \leqslant t$ by the equation

$$\mathbf{E}_a = \boldsymbol{\varepsilon}_a(t - \mathbf{k}_a\mathbf{r}/\omega_a)\exp[i(\mathbf{k}_a\mathbf{r} - \omega_a t - \varphi_a)] + \text{c.c.}, \tag{23}$$
$$\omega_a = \omega_3 + \omega_1 - \omega_2, \quad \varphi_a = \varphi_3 + \varphi_1 - \varphi_2,$$

where the amplitude $\boldsymbol{\varepsilon}_a(t)$ is of the form

$$\boldsymbol{\varepsilon}_a(t) = \mathbf{N}G_a C_a(t)\exp[-(\gamma_{ab}^{(1)} + i\Delta_{ab}^{(1)})(t-\tau)], \tag{24}$$

$$\mathbf{N} = {}^1/_2\mathbf{l}_x[N_0\cos(\psi_1-\psi_2) + N_2(3\cos\psi_1\cos\psi_2 - \cos(\psi_1-\psi_2))]$$
$$+ {}^1/_2\mathbf{l}_y[N_1\sin(\psi_1-\psi_2) + N_2\sin(\psi_1+\psi_2)],$$

$$N_\varkappa = (-1)^\varkappa\begin{Bmatrix}1 & \varkappa & 1\\ J_c & J_a & J_b\end{Bmatrix}\Pi_\varkappa(\omega_1,\omega_2)\exp[-(\gamma_{cb}^{(\varkappa)} + i\Delta_{cb}^{(\varkappa)})\tau],$$

$$\varkappa = 0, 1, 2,$$

$$C_a(t) = \frac{-i\pi\omega_a{}^2 d_{ca}{}^* d_{ab}{}^* L_a N_b}{4\hbar^2 c^2 k_a(2J_b+1)}\int_0^{\tau_1}d\xi\int_0^{t-t_3}d\eta\, a_1(\xi)a_2{}^*(\xi)a_3(\eta)$$

$$\times\exp\{i\Delta_3{}'(t-t_3-\eta) + i\Delta_{12}(t-\xi)$$

$$-[t-(\omega_1-\omega_2)\xi/\omega_a - \omega_3(t_3+\eta)/\omega_a]^2 k_a{}^2u^2/4\},$$

$$\varepsilon(\omega_a)\omega_a{}^2 = \mathbf{k}_a{}^2c^2, \qquad \Delta_3{}' = \omega_3 - \omega_{ac}.$$

The factor G_a is given here by Eq. (11) with the replacement $\boldsymbol{\Delta}_s \to -\boldsymbol{\Delta}_a$ under the conditions $\boldsymbol{\Delta}_a = \mathbf{k}_a - \mathbf{k}_3 - \mathbf{k}_1 + \mathbf{k}_2$ and $\mathbf{L}_a = \mathbf{k}_a L_a/k_a{}^*$, where L_a is the anti-Stokes optical path length in the active region of the gas.

In the case of the homogeneously broadened transitions $(k_3 + k_1 - k_2)u < \gamma_{ab}^{(1)}$ and $(k_1 - k_2)u < \gamma_{cb}^{(\varkappa)}$ the ASI can be used to determine the relaxation constant $\gamma_{ab}^{(1)}$ of the optically allowed transition $J_b \to J_a$, as well as of the values of $\gamma_{cb}^{(\varkappa)}$ with $\varkappa = 0, 1, 2$ pertaining to the optically forbidden transition $J_b \to J_c$, using the same methods as for SFI. The forbidden transitions are identified and the parameters of the cubic-susceptibility tensor are determined in similar fashion. If the atomic transitions are inhomogeneously broadened $(k_3 + k_1 - k_2)u > \gamma_{ab}$ and $(k_1 - k_2)u > \gamma_{cb}^{(\varkappa)}$ the maximum of the function $|C_a(t)|$ occurs at the instant $t_a = \omega_3\tau/(\omega_3 + \omega_1 - \omega_2)$. Relative to the instant t_3 of the entry of the probe pulse into the gas, the indicated maximum lies in the past $t_a < t_3$, so that the Doppler dephasing of the emitting atoms suppresses the anti-Stokes signal and no anti-Stokes photon echo is produced. When the two-photon resonance

$\omega_1+\omega_2\approx\omega_{cb}$ is produced, it is necessary to make the same change in Eqs. (23) and (24) as in the Stokes-signal case considered above.

If the level E_a lies lower then the two other $E_a<E_b<E_c$ and $\omega_3\approx\omega_{ba}$ (Fig. 2b), it is necessary to make in (23) the substitutions

$$N_\varkappa\to(-1)^{\varkappa+1}N_\varkappa,\quad \Delta_3'\to\omega_3-\omega_{ba},\quad \gamma_{ab}^{(1)'}\to\gamma_{ca}^{(1)'},\quad \Delta_{ab}^{(1)'}\to\Delta_{ca}^{(1)},$$

and the obtained ASI intensity makes it possible to obtain the values of $\gamma_{ca}^{(1)}$ and $\gamma_{cb}^{(\varkappa)}$ with $\varkappa=0$, 1, and 2.

4. CONCLUSION

The main advantage of the SPE over other NRS (Refs. 1-5) is that it can be used in practice to investigate inhomogeneously broadened atomic transitions, with an aim of studying ultrafast irreversible relaxation, and also to determine the parameters of the cubic-susceptibility tensor and to identify the type of the Raman transition, since the methods of Refs. 1-5 are inapplicable in this case. The use of SPE offers also advantages over the known three-level-echo method,[16,17] since the characteristic angular dependence (14) of the SPE amplitude is valid for arbitrary areas of the pulses (5) and (6), whereas to simplify the polarization properties of the three-level echo the first two exciting pulses must have small areas, and the area of the last (6) can be arbitrary. In addition, the SPE is easily distinguished from among the other light pulses by its different frequency, but separation of the three-level echo from the three-level echo is difficult because the central frequencies are equal. The range of validity of the SPE is larger, since it is produced not only in Raman resonance but also in two-photon resonance, as well as in excitation of four-level systems. Formation of a three-level echo calls for three resonant light pulses, whereas SPE can be produced by only one resonant probe pulse, and either of the two pump pulses $\omega_1-\omega_2\approx\omega_{cb}$ or $\omega_1+\omega_2\approx\omega_{cb}$ meets this condition.

We emphasize that for homogeneously and inhomogeneously transitions the NRS modification in the form SFI, SPE, and ASI make it possible to determine additionally the relaxation constants $\gamma_{ac}^{(1)}$ and $\gamma_{ab}^{(1)}$ of the allowed transitions, in addition with the three constants $\gamma_{cb}^{(\varkappa)}$ with $\varkappa=0$, 1, and 2 for a forbidden transition for a relatively great variety of arrangements of the levels E_a, E_b, and E_c (Figs. 1 and 2). Since the distance between the levels E_b and E_c is arbitrary, Raman transitions on formation of SFI, SPE, and ASI can be not only optical but also transitions between hfs components of the resonant level, or transitions between vibrational–rotational levels of a molecule.

APPENDIX

To be specific, we confine ourselves to a Stokes signal for which the components of the vector $\mathbf{P}(t,\mathbf{r})$ of the atomic dielectric polarization depend on the components of the electric field $\mathbf{E}=\mathbf{E}_1+\mathbf{E}_2+\mathbf{E}_3$ and of the momenta (5) and (6) as follows:

$$P_i(t,\mathbf{r})=\sum_{j,k,l=1}^{3}\int_{-\infty}^{t}\chi_{ijkl}(t-t',t-t'',t-t''')E_j(t',\mathbf{r})\times E_k(t'',\mathbf{r})E_l(t''',\mathbf{r})\,dt'\,dt''\,dt''',\qquad(25)$$

with expression (25) symmetric with respect to permutation of any symbol pair jt', kt'', and lt''', owing to the isotropy of the gaseous medium. Since the electric fields (5) and (6) are not monochromatic waves, we use the Fourier transforms for the amplitudes

$$a_n(\nu_n-\omega_n)=\int_{-\infty}^{+\infty}a_n(t)\exp[i(\nu_n-\omega_n)t]\,dt,\qquad n=1,2,3,$$

where ω_n is the central frequency, ν_n is the variable of the expansion into a Fourier integral, and the Fourier components $a_n(\nu_n-\omega_n)$ as functions of $\nu_n-\omega_n$ differ from zero in the narrow region $|\nu_n-\omega_n|\leqslant 1/\tau_n$ for all the amplitudes $a_n(t)$ with subscripts $n=1$, 2, and 3. Relation (25) makes is possible then to determine the frequency dispersion of the sought-for cubic-susceptibility tensor, using the equation

$$\begin{aligned}P_i(t,\mathbf{r})={}&\frac{3}{4(2\pi)^3}\sum_{j,k,l=1}^{3}\int_{-\infty}^{+\infty}\chi_{ijkl}(\nu_s,\nu_3,-\nu_1,\nu_2)\,l_{3j}l_{1k}^{*}l_{2l}a_3(\nu_3-\omega_3)\\&\times a_1^{*}(\nu_1-\omega_1)a_2(\nu_2-\omega_2)\delta(\nu_s-\nu_3+\nu_1-\nu_2)\exp[-i\nu_3(t-\mathbf{k}_3\mathbf{r}/\omega_3)\\&+i\nu(t-\mathbf{k}_1\mathbf{r}/\omega_1)-i\nu_2(t-\mathbf{k}_2\mathbf{r}/\omega_2)-i(\varphi_3-\varphi_1+\varphi_2)\\&+i(\nu_3-\omega_3)t_3]\,d\nu_1\,d\nu_2\,d\nu_3\,d\nu_s,\end{aligned}\qquad(26)$$

where the cubic-susceptibility tensor $\chi_{ijkl}(\nu_s,\nu_3,-\nu_1,\nu_2)$ for the Stokes signal is symmetric with respect to permutation of any pair of symbols $j\nu_3$, $k(-\nu_1)$ and $l\nu_2$. Here l_{1k}, l_{2l} and l_{3j} are the projections of the polarization unit vectors of the pump waves (5) and of the probe pulse (6).

According to the foregoing results, the atomic dielectric polarizability vector $\mathbf{P}(t,\mathbf{r})$ of a Stokes signal in a gaseous medium is connected with the electric fields of pulses (5) and (6) by the relation

$$\begin{aligned}\mathbf{P}(t,\mathbf{r})={}&\mathbf{M}\frac{N_b d_{ca}^{*}d_{ab}^{*}}{8\hbar^2(2J_b+1)}\int d\mathbf{v}\,f(v)\\&\times\int_0^t a_1\left(t'-\frac{\mathbf{k}_1\mathbf{r}}{\omega_1}\right)a_2^{*}\left(t'-\frac{\mathbf{k}_2\mathbf{r}}{\omega_2}\right)\\&\times\exp\{i[(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}-\Delta_{12}]t'\}\\&\times dt'\int_0^{t-t_3}a_3^{*}\left(t''-\frac{\mathbf{k}_3\mathbf{r}}{\omega_3}\right)\exp[i(\Delta_3-\mathbf{k}_3\mathbf{v})t'']\,dt''\\&\times\exp\{-i[(\mathbf{k}_3-\mathbf{k}_1+\mathbf{k}_2)(\mathbf{r}-\mathbf{v}t)-\omega_{ac}t-\varphi_s]\\&-(\gamma_{ac}^{(1)'}-i\Delta_{ac}^{(1)'})(t-\tau)\}+\text{c.c.}\end{aligned}\qquad(27)$$

We neglect for simplicity the elastic collisions and recast (27) in the form (26); this yields

$$\begin{aligned}&\chi_{ijkl}(\nu_s,\nu_3,-\nu_1,\nu_2)\\&=\frac{d_{ca}d_{ab}N_b}{6\hbar^2(2J_b+1)}A_{ijkl}\int f(v)\,[\nu_1-\nu_2-\omega_{cb}-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}\\&\quad-i\gamma_{cb}]^{-1}[(\nu_s-\omega_{ac}-\mathbf{k}_s\mathbf{v}+i\gamma_{ac})\\&\quad+(\nu_1-\nu_2-\omega_{cb}-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}-i\gamma_{cb})]^{-1}\,d\mathbf{v},\end{aligned}\qquad(28)$$

$$\begin{aligned}A_{xxyy}={}&\frac{1}{3}\left[\begin{Bmatrix}1&0&1\\J_c&J_a&J_b\end{Bmatrix}\Pi_0^{*}(\omega_1,\omega_2)\right.\\&\left.-\begin{Bmatrix}1&2&1\\J_c&J_a&J_b\end{Bmatrix}\Pi_2^{*}(\omega_1,\omega_2)\right],\end{aligned}$$

$$A_{xyxy}=\frac{1}{2}\left[\begin{Bmatrix}1 & 2 & 1\\ J_c & J_a & J_b\end{Bmatrix}\Pi_2^{*}(\omega_1,\omega_2)\right.$$
$$\left.+\begin{Bmatrix}1 & 1 & 1\\ J_c & J_a & J_b\end{Bmatrix}\Pi_1^{*}(\omega_1,\omega_2)\right],$$
$$A_{xyyx}=\frac{1}{2}\left[\begin{Bmatrix}1 & 2 & 1\\ J_c & J_a & J_b\end{Bmatrix}\Pi_2^{*}(\omega_1,\omega_2)\right.$$
$$\left.-\begin{Bmatrix}1 & 1 & 1\\ J_c & J_a & J_b\end{Bmatrix}\Pi_1^{*}(\omega_1,\omega_2)\right].$$

Of all the nonzero components of the tensor (28), the only independent ones are the three with the A_{ijkl} written out explicitly, while the other are obtained in accordance with the usual rules[3,4] with the aid of the symmetry properties of isotropic media. When ultrashort biharmonic pumping (5) and an ultrashort resonant probe pulse (6) are used, the dispersion of the cubic susceptibility tensor (28) at frequencies $\nu_1-\nu_2$ and ν_s close to the frequencies ω_{cb} and ω_{ac} of the atomic transitions (Fig. 1a) causes the electric field of the Stokes signal to be different from zero not only at the instants when the probe signal (6) acts, but also later, in SFI form. If the biharmonic pump and the resonant probe pulse are monochromatic waves, one must put in (26) $a_n(\nu_n-\omega_n)=2\pi a_n\delta(\nu_n-\omega_n)$ with constant values of the amplitudes a_n $(n=1,2,3)$. Equation (28) remains in force following the substitution $\nu_s\to\omega_s$ and the replacement of ν_n by the frequencies ω_n of the indicated monochromatic waves. The spatial dispersion in (28) is due to the Doppler effect.

We emphasize that for an ultrashort nonresonant trial pulse (6) and for an ultrashort pump (5) the cubic-susceptibility tensor for a Stokes signal in a monatomic gas can be obtained from the results of Ref. 7 in the form

$$\chi_{ijkl}(\nu_s,\nu_3,-\nu_1,\nu_2)$$
$$=\frac{N_bD_{ijkl}}{6\hbar(2J_b+1)}\int\frac{f(v)\,d\mathbf{v}}{\omega_{cb}-\nu_1+\nu_2+(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}+i\gamma_{cb}},\qquad(29)$$

$$D_{xxyy}={}^1/_3[\Pi_0^{(-)*}\,\Pi_0^{*}(\omega_1,\omega_2)-\Pi_2^{(-)*}\,\Pi_2^{*}(\omega_1,\omega_2)],$$

$$D_{xyxy}={}^1/_2[\Pi_2^{(-)*}\,\Pi_2^{*}(\omega_1,\omega_2)+\Pi_1^{(-)*}\,\Pi_1^{*}(\omega_1,\omega_2)],$$

$$D_{xyyx}={}^1/_2[\Pi_2^{(-)*}\,\Pi_2^{*}(\omega_1,\omega_2)-\Pi_1^{(-)*}\,\Pi_1^{*}(\omega_1,\omega_2)],$$

where the value of (29) must be used jointly with expression (26) in which the substitution $a_3(\nu_3-\omega_3)\to2\pi a_3\delta(\nu_3-\omega_3)$ is made. It follows from (26) and (29) that a Stokes signal is produced only in the time of action of the ultrashort nonresonant nonresonant pulse, and vanishes after the end of the pulse, so that there is no SFI or SPE here. In the case of monochromatic waves, the aforementioned substitutions $\nu_s\to\omega_s$ and $\nu_n\to\omega_n$ are made in (29), with $n=1,2$.

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