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On the possibility of measuring the population, orientation, and alignment relaxation times by the photon echo method

I. V. Evseev, V. M. Ermachenko, and V. A. Reshetov

Moscow Engineering-Physics Institute

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Stimulated photon echo in a resonant gaseous medium is investigated theoretically. It is found that the echo intensity and the polarization depend markedly on the relaxation characteristics responsible for the relaxation of the population, magneto-dipole (orientation relaxation) moment, and quadrupole (alignment relaxation) moment of the resonance levels. These dependences make it possible to carry out experimental measurements of these relaxation times by the photon echo method. The observed effect should stimulate the setting up of new experiments on the photon echo in gases with the aim of measurement of the above-mentioned characteristics of the resonance levels.

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A photon echo is formed in a medium after passage of two exciting light pulses separated by a time interval τ_s , and represents spontaneous coherent radiation from a superradiant state created by the first exciting pulse.¹ By increasing τ_s and observing experimentally the attenuation of the intensity of the photon echo, we can effectively measure the relaxation characteristics of the resonance transition by the photon echo method. Thus, in experiments on the photon echo in gases (see, for example, Refs. 2-5), this method was used successfully to find relaxation characteristic $\gamma^{(1)}$ responsible for the damping of the component of the optical coherence matrix, which is proportional to the polarization of the medium. There are also theoretical works,^{6,7} showing the possibility of measurement by the photon echo method of other relaxational characteristics $\gamma^{(n)}$ ($n \neq 1$) of the optical coherence matrix.

In the present work, it is shown that we can also measure the relaxation characteristics of the resonance levels themselves by the photon echo method: the relaxation of the population, orientation, and alignment. For this purpose, it is proposed to observe not the ordinary but the so-called stimulated photon echo.

The stimulated echo was predicted and observed by Hahn⁸ in the radiofrequency range. This echo is cre-

ated at an instant of time approximately equal to $2\tau_1 + \tau_2$ upon successive passage through the medium of three exciting pulses, separated by the respective time intervals τ_1 and τ_2 . We note that the three-pulse method of excitation of resonant media has already progressed at the present time from the radiofrequency to the optical range (see, for example, Trgd. 9-11). In particular, stimulated photon echo in ruby has been observed in the work of Samatsev *et al.*¹¹ Thus, the fact of the possibility of observation of a stimulated echo in the optical range raises no doubts whatever.

In the present work, we have carried out a calculation of the intensity and polarization of the stimulated photon echo produced in a gaseous medium. In this case we have taken into account the degeneracy of the resonance levels of the considered transition and the effect of elastic depolarizing atomic (molecular) collisions on the interaction of the atoms (molecules) of the gas with the resonance electromagnetic field. The calculations that have been carried out show that it is possible to select such experimental conditions under which the damping of the components of the maximum of intensity of the stimulated photon echo in the direction of polarization of the third exciting pulse and in the perpendicular direction will be determined either only by the relaxation times of the population of the resonan-

ce levels, or only by the relaxation times of the orientation or only by the alignment relaxation times. Thus, the photon echo method can take its place alongside the classical methods of the spectroscopy of resonance levels, such as the Hanle effect, the effect of level crossings, and so on.¹²

1. SMALL AREAS OF EXCITING PULSES

We consider the formation of a stimulated photon echo in a gas by three linearly polarized exciting-light pulses of duration T_1 , T_2 and T_3 , respectively, propagating along the Y axis with carrier frequency ω and resonance frequency ω_0 of the atomic (molecular) transition with change in total angular momentum $j_a \rightarrow j_b$. Let the polarization vectors of the first and third exciting pulses form an angle ψ , and those of the second and third, an angle ψ_2 .

As the basic equations, we write down the d'Alembert equation and the quantum mechanical equation for the components of the density matrix, taking into account the interaction of the atoms (molecules) of the gas with the resonance electromagnetic field and the irreversible relaxation, due to the radiation decay and to the inelastic gaskinetic and elastic depolarizing collisions.

The method of finding the intensity of the electromagnetic field of the stimulated photon echo is similar to that used earlier⁷ for finding the intensity of the electric field of an ordinary photon echo. In this case, an expansion is carried out of the components of the density matrix in irreducible tensor operators and a solution is obtained of the set of equations from Ref. 7 for the slow functions in the given field approximation. Moreover, it is assumed that the durations T_i ($i = 1, 2, 3$) of the exciting light pulses are small in comparison with the intervals of time τ_1 and τ_2 between them, and also in comparison with the times of irreversible relaxation. The intensity of the external magnetic field has been assumed to be zero.

In the present section, we consider the formation of the stimulated photon echo in the optically allowed transition with arbitrary angular momenta j_a and j_b of the upper a and lower b resonance levels. It is obvious that without simplifying assumptions the solution of the similar problem can apparently not be obtained analytically. Therefore, as in Ref. 13, the approximation of small areas of the exciting pulses is used in this section.

The action on the resonant medium of the first two light pulses leads to the result that up to the instant of incidence of the third pulse, the medium preserves a phase memory of the first two exciting pulses. For the stimulated photon echo in the absence of the ordinary one, the coherence created by these pulses in the density-matrix components $\rho_{mm'}^{(a\phi)}$ and $\rho_{\mu\mu'}^{(b\phi)}$ of the resonance levels themselves. Here m and μ are respectively the projections of the total angular momentum of the upper and lower resonance levels.

At the instant

$$t = \tau_1 + \tau_2 + T_1 + T_2 + y/c,$$

when the third exciting pulse reaches the point y of the gaseous medium, the parts of the amplitudes $f_q^{(x)}$ and $q_q^{(x)}$ of the expansion of the components $\rho_{mm'}^{(a\phi)}$ and $\rho_{\mu\mu'}^{(b\phi)}$ in irreducible tensor operators, which make a contribution to the stimulated photon echo, have the form

$$f_q^{(x)}(\tau_2) = B \sum_{q', q_1} C_{qq'q_1}^{x1} C_{q'}(\psi_1) C_{q_1}(\psi_2) \exp\{-i(kv\tau_1 - \Phi_2 + \Phi_1) - \gamma^{(1)}\tau_1 - \gamma_a^{(x)}\tau_2\}, \quad (1)$$

$$\begin{aligned} \varphi_q^{(x)}(\tau_2) = & B \sum_{q', q_1} (-1)^{1+n} B_{qq'q_1}^{x1} C_{q'}(\psi_1) C_{q_1}(\psi_2) \\ & \times \exp\{-i(kv\tau_1 - \Phi_2 + \Phi_1) - \gamma^{(1)}\tau_1 - \gamma_b^{(x)}\tau_2\}, \end{aligned} \quad (2)$$

where

$$B = (-1)^{j_b - j_a + x + 1} \frac{|d|^2}{3\hbar^2} e^{(1)} T_1 e^{(2)} T_2 I_1 I_2 N_0 f(v), \quad (3)$$

$$I_i = [\sin kvT_i - i(1 - \cos kvT_i)] / kvT_i, \quad i=1, 2,$$

$$C_q(\psi) = [\cos \psi + 2^{-1/2} \sin \psi (\delta_{q,1} - \delta_{q,-1})].$$

Here d is the reduced matrix element of the dipole operator of the angular momentum transition $j_a \rightarrow j_b$, N_0 is the density of overpopulation of the Zeeman sublevels up to the incidence on the medium of the first exciting pulse, $f(v)$ is the Maxwellian distribution function in the projections v of the velocity of the atoms on the Y axis; $e^{(i)}$ is the constant amplitude of the intensity of the electric field and Φ_i is the constant phase shift of the i th exciting pulse. The quantities $C_{qq'q_1}^i$ and $B_{qq'q_1}^i$, which depend on the angular momenta of the considered energy levels and which enter into (1) and (2), are given in Ref. 7. Further, the relaxational characteristic

$$\gamma^{(1)} = (\gamma_a^{(0)} + \gamma_b^{(0)}) / 2 + \Gamma^{(1)},$$

which describes the damping of the component of the optical coherence matrix, and which is proportional to the polarization of the medium, is expressed in terms of the relaxation times $1/\gamma_a^{(0)}$ and $1/\gamma_b^{(0)}$ of the populations of the levels a and b , due to gaskinetic inelastic collisions and radiation decay, and also in terms of the quantity $\Gamma^{(1)}$, which describes the broadening of the spectral radiation line under the action of elastic depolarizing collisions. Finally,

$$\gamma_{a,b} = \gamma_{a,b}^{(0)} + \Gamma_{a,b}^{(x)},$$

where $\Gamma_a^{(x)}$ and $\Gamma_b^{(x)}$ describe the collisional relaxation of the upper and lower levels, due to elastic depolarizing collisions. We emphasize that since we are dealing with elastic collisions that do not change the total population of the level, it follows that $\Gamma_a^{(0)} = \Gamma_b^{(0)} = 0$. In the writing of (1) and (2), it was assumed that the polarization of the first exciting pulse makes an angle ψ , with the Z axis and the second the angle ψ_2 .

As follows from (1) and (2), the amplitudes $f_q^{(x)}$ and $\varphi_q^{(x)}$, through the factor

$$\exp\{-i(kv\tau_1 - \Phi_2 + \Phi_1)\} \quad (4)$$

preserve the phase memory of the first two exciting pulses.

The expressions (1) and (2) play the role of initial conditions in consideration of action on the medium of the third exciting pulse. This action leads to the result that the term containing the phase factor (4) appears also in the polarization vector of the medium P_ν , which

relates to the group of atoms having a projection of velocity on the Y axis equal to v . Therefore, after passage of the third exciting pulse by the point y of the gaseous medium, the part of P_y responsible for the formation of the stimulated photon echo oscillates as a function of v according to the law

$$P_y \propto f(v) F(v) \exp\{ikv(t' - \tau_1)\}, \quad (5)$$

where

$$t' = t - \tau_1 - \tau_2 - T_1 - T_2 - T_3 - y/c, \quad F(v) = I_1 I_2 I_3^*.$$

Here the quantity I_3 is obtained from (3) at $i = 3$.

Thus, after passage of the third exciting pulse through the medium, the vectors P_y created by the different groups of atoms with different projections v of the velocity on the Y axis have different phases; therefore, the radiated or electromagnetic waves are noncoherent. However, at the instant of time approximately equal to

$$t = 2\tau_1 + \tau_2 + T_1 + T_2 + T_3 + y/c, \quad (6)$$

as follows from (5), synchronization of the different P_y takes place, and the system of excited atoms in each point y undergoes a transition into the superradiant state. As a result, spontaneous coherent radiation takes place—a stimulated photon echo, the independent pulses of which from the different portions of the medium are combined, with account of the delay, to form a common electromagnetic pulse.

Omitting the intermediate calculations, we write down the final expression for the intensity of the electric field of the stimulated photon echo

$$E_e = -\frac{\pi}{\hbar^2} \omega \frac{L}{c} |d|^2 e^{(1)} T_1 e^{(2)} T_2 e^{(3)} T_3 N_e e^{\theta}$$

$$\times \exp\{i(\omega t - ky + \Phi_3 + \Phi_2 - \Phi_1) - \gamma^{(1)}(t' + \tau_1)\} + c.c., \quad (7)$$

where

$$I = \int dv F(v) f(v) \exp\{ikv(t' - \tau_1)\}. \quad (8)$$

Here L is the extent of the gaseous medium, and the vector e^{θ} , which characterizes the polarization properties of the stimulated photon echo, has the form

$$e_x^{\theta} = \frac{2}{3} [U \cos(\psi_1 - \psi_2) + W (2 \cos \psi_1 \cos \psi_2 - \sin \psi_1 \sin \psi_2)], \quad (9a)$$

$$e_z^{\theta} = -V \sin(\psi_1 - \psi_2) + W \sin(\psi_1 + \psi_2), \quad (9b)$$

$$e_y^{\theta} = 0, \quad (9c)$$

$$U = M(j_a, j_b) \exp(-\gamma_a^{(1)} \tau_2) + M(j_b, j_a) \exp(-\gamma_b^{(1)} \tau_2), \quad (10)$$

$$V = N(j_a, j_b) \exp(-\gamma_a^{(1)} \tau_2) + N(j_b, j_a) \exp(-\gamma_b^{(1)} \tau_2),$$

$$W = L(j_a, j_b) \exp(-\gamma_a^{(2)} \tau_2) + L(j_b, j_a) \exp(-\gamma_b^{(2)} \tau_2).$$

In writing down (9a) and (9b), it was assumed that the polarization vector of the third exciting pulse is directed along the Z axis. The quantities $M(j_a, j_b)$, $N(j_a, j_b)$ and $L(j_a, j_b)$ for the different types of transitions entering into (9a) and (b) have the form

$$M(j, j) = M(j, j+1) = \frac{1}{3(2j+1)}, \quad N(j, j) = \frac{1}{6j(j+1)(2j+1)},$$

$$L(j, j) = \frac{(2j-1)(2j+3)}{30j(j+1)(2j+1)}, \quad M(j+1, j) = \frac{1}{3(2j+3)},$$

$$L(j, j+1) = \frac{j(2j-1)}{30(j+1)(2j+1)(2j+3)}, \quad L(j+1, j) = \frac{(j+2)(2j+5)}{30(j+1)(2j+1)(2j+3)},$$

$$N(j, j+1) = \frac{j}{6(j+1)(2j+1)}, \quad N(j+1, j) = \frac{j+2}{6(j+1)(2j+3)}.$$

The echo (7)–(10) propagates the carrier frequency ω and is linearly proportional to it. The shape of the pulse of the stimulated photon echo is determined by the expression (8). In the limiting case of a narrow spectral line ($1/T_0 \ll 1/T_i$) we have from (8)

$$I = \exp\{-(t' - \tau_1)^2 / 4T_0^2\}. \quad (11)$$

Here $T_0 = 1/ku$ is the time of the reversible Doppler relaxation and u is the mean thermal velocity of the atoms of the gas. Thus, the maximum of the intensity of the stimulated photon echo, formed on the narrow spectral line, takes place at the moment of time (6), and the width of the echo pulse is of the order of T_0 .

In the limiting case of a wide spectral line ($1/T_0 \gg 1/T_i$) we get from (8) at $T_1 = T_2 = T_3 = T$

$$I = \sqrt{\pi} \frac{T_0}{T} \{ (a+1)^2 [\theta(a+1) - \theta(a)] - (2a^2 - 2a - 1) [\theta(a) - \theta(a-1)] + (a-2)^2 [\theta(a-1) - \theta(a-2)] \}, \quad (12)$$

where

$$a = \frac{(t' - \tau_1)}{T}, \quad \theta(x) = \begin{cases} 1, & x \geq 0 \\ 0, & x < 0 \end{cases}.$$

It is seen from Eq. (12) that the maximum intensity of the stimulated photon echo on the wide spectral line is shifted relative to the instant of time $t = 2\tau_1 + \tau_2 + 3T + y/c$ by $0.5T$, while the width of the pulse echos is of the order of T .

We emphasize that the stimulated photon echo, both on a wide and on a narrow spectral line, is formed at an instant of time which is considerably different from the time of formation of the ordinary photon echo, and also of other possible responses of a resonant medium excited by three light pulses.

Formulas (7)–(10) allow us to find the intensity and polarization of the stimulated photon echo on an optically allowed transition with arbitrary conditions of the angular momenta of the levels in the approximation of small areas of exciting pulses. We note that the intensity of the stimulated echo is proportional to the product of the amplitudes of the intensities of all the exciting light pulses.

As follows from (7)–(12), the entire dependence of the maximum intensity of the stimulated photon echo on the time interval τ_2 is determined by the dependence of only the components of the vector e^{θ} on τ_2 . Therefore, the component of the maximum intensity of this photon echo in the direction of the polarization vector of the third exciting pulse depends on τ_2 through $(e_x^{\theta})^2$, and in the perpendicular direction, through $(e_z^{\theta})^2$. Using the observed property of the intensity maximum, we shall show in what fashion one can obtain information of the times determining the relaxation of the population, orientation, and alignment of the resonance levels.

As follows from (9) and (10), at $\psi_2 = \psi_1$, the investigation of the damping of the X component of the intensity maximum of the stimulated photon echo as a function of τ_2 allows us to carry out the direct measurement of the time $1/\gamma_{a,b}^{(2)}$ of the alignment relaxation. At $\psi_2 = -\psi_1$, the investigation of the damping of the maximum of this component allows us to perform the measurement of the times $1/\gamma_{a,b}^{(1)}$ of the relaxation of the orientation. Final-

ly, if ψ_1 and ψ_2 are such that $\tan\psi_1 \tan\psi_2 = 2$, then the study of the damping of the Z component of the maximum intensity of the stimulated photon echo makes it possible to carry out the direct measurement of the times $1/\gamma_{a,b}^{(0)}$ of the population relaxation. We note that inasmuch as $\gamma_{a,b}^{(0)}$ enter into the relaxation characteristic $\gamma^{(1)}$, which is well measured by the usual photon echo method, the possibility develops of the direct extraction, from the experiments on the photon echo, of the quantity $\Gamma^{(1)}$, which is due only to the elastic depolarizing collisions.

As follows from (9a) and (9b), there are other relations between ψ_1 and ψ_2 which allows us to separate in (9a) or (9b) damping with only any one of the relaxation times (population, orientation or alignment).

We turn to the experimentally interesting case of large angular momenta of the resonance levels, which is realized in experiments on the photon echo in molecular gases (see, for example, Refs. 2-5). In this case, for the transitions $j \rightarrow j$ ($j \gg 1$) we get from (9a) and (9b)

$$e_z^e = \frac{1}{9j} \left[\cos(\psi_1 - \psi_2) A_0(\tau_2) + \frac{2}{5} (2 \cos \psi_1 \cos \psi_2 - \sin \psi_1 \sin \psi_2) A_2(\tau_2) \right], \quad (13)$$

$$e_x^e = \frac{1}{15j} \sin(\psi_1 + \psi_2) A_2(\tau_2), \quad (14)$$

where

$$A_\kappa(\tau_2) = \exp(-\gamma_a^{(\kappa)} \tau_2) + \exp(-\gamma_b^{(\kappa)} \tau_2), \quad \kappa=0, 1, 2. \quad (15)$$

Consequently, in the transitions $j \rightarrow j$ at $j \gg 1$, the relaxation times of the population and alignment of the resonance levels can be measured by the stimulated photon echo method.

For the transitions $j \rightarrow j+1$ at $j \gg 1$, we have from (9a) and (9b)

$$e_z^e = \frac{1}{9j} \left[\cos(\psi_1 - \psi_2) A_0(\tau_2) + \frac{1}{10} (2 \cos \psi_1 \cos \psi_2 - \sin \psi_1 \sin \psi_2) A_2(\tau_2) \right], \quad (16)$$

$$e_x^e = \frac{1}{60j} [\sin(\psi_1 + \psi_2) A_2(\tau_2) - 5 \sin(\psi_1 - \psi_2) A_1(\tau_2)]. \quad (17)$$

Here $A_\kappa(\tau_2)$ are given by the formulas (15). Thus, in the transitions $j \rightarrow j+1$ at $j \gg 1$, we can measure by the stimulated photon echo method all three relaxation times of the resonance levels: population, orientation, and alignment.

We note that the formulas (13), (14), (16) and (17) can also be used for identification of the type of resonance transition ($j \rightarrow j$ or $j \rightarrow j+1$). Such an identification will be especially effective at $\gamma_a^{(\kappa)} \tau_2 \ll 1$ and $\gamma_b^{(\kappa)} \tau_2 \ll 1$ since these formulas are greatly simplified in the case of such parameters.

For clarification of the way in which the increase in the areas of the exciting pulses affect the considered effects, we have considered transitions in the next section with small levels moments, which can serve to find the intensities of the electric field of the stimulated photon echo in the case of arbitrary areas of the exciting pulses.

2. TRANSITIONS WITH SMALL LEVEL ANGULAR MOMENTA

First we consider a transition with the level angular momenta $j_a = \frac{1}{2}$ and $j_b = \frac{1}{2}$. This transition is realized in experiments on the photon echo in alkali-metal vapors. Inasmuch as $0 \leq \kappa \leq 2j_{a,b}$, $\gamma_{a,b}^{(0)}$ and $\gamma_{a,b}^{(1)}$ are possible in the transition $\frac{1}{2} \rightarrow \frac{1}{2}$. However, because of the proportionality of the S matrices of the elastic scattering of each of the levels to the Kronecker symbol, we have in this transition $\gamma_{a,b}^{(1)} = \gamma_{a,b}^{(0)}$.

The method of finding the intensity of the electric field of the stimulated photon echo in the transition $\frac{1}{2} \rightarrow \frac{1}{2}$ is analogous to that used in the previous section. As a result, we obtain that the intensity of the electric field of the stimulated echo in this transition is given by the formulas (7) and (8), where the quantity I_i is obtained not from formula (3), but from the expression

$$I_i = \frac{1}{\Omega_i T_i} \left[\sin \Omega_i T_i - i \frac{kv}{\Omega_i} (1 - \cos \Omega_i T_i) \right]. \quad (18)$$

Here the frequency Ω_i is connected with the area

$$\theta_i = \sqrt{2} |d| e^{(i)} T_i / \sqrt{3} \hbar$$

of the i -th exciting light pulse in the following way:

$$\Omega_i^2 T_i^2 = \theta_i^2 + (kv T_i)^2.$$

As follows from (8) and (18), in the transition $\frac{1}{2} \rightarrow \frac{1}{2}$ the shape of the pulse of the stimulated photon echo in the narrow spectral line at $\theta_i \gg T_i/T_0$ is given by equation (11) and has the same form as in the limiting case of small areas of the exciting pulses. However, on the broad spectral line, the shape of the pulse of the stimulated echo is made complicated in comparison with (12) in the case of an increase in the areas θ_i .

In the transition $\frac{1}{2} \rightarrow \frac{1}{2}$ the nonzero components of the vector e^e that enter in (7) and characterize the polarization properties of the echo have the form

$$e_z^e = \cos(\psi_1 - \psi_2) A_0(\tau_2), \quad (19)$$

$$e_x^e = -\sin(\psi_1 - \psi_2) A_0(\tau_2), \quad (20)$$

where $A_0(\tau_2)$ is determined by the formula (15). Thus, the angle which the polarization vector of the stimulated echo makes with the polarization echo of the third exciting pulse is found in this transition from the equation

$$\operatorname{tg} \varphi_{se} = -\operatorname{tg}(\psi_1 - \psi_2).$$

Consequently, in the transition $\frac{1}{2} \rightarrow \frac{1}{2}$ the angle φ_{se} does not depend on the relaxation characteristics of the resonance levels. Such a behavior of the angle φ_{se} is a specific property only of this transition.

By investigating the decay of the maximum intensity of the stimulated photon echo with growth of τ_2 in the transition $\frac{1}{2} \rightarrow \frac{1}{2}$, we can carry out the experimental measurement of the relaxation times of the level populations. If the characteristics that govern the radiation decay of the levels are known, we can measure the contribution of the inelastic collisions to the relaxation times of the level populations by this method. We note that attention was first called to this circumstance in Ref. 14.

We emphasize that the formulas (19) and (20) are ob-

tained from the general formulas (9) and (10) at $j = \frac{1}{2}$. This indicates that the polarization properties of the stimulated photon echo in the transition $\frac{1}{2} \rightarrow \frac{1}{2}$ do not depend on the value of the areas of the exciting light pulses. The noted property of the polarization of the stimulated echo occurs only in the transition $\frac{1}{2} \rightarrow \frac{1}{2}$.

We now consider the transition with level angular momenta $j_a = 0$ and $j_b = 1$. Here the level a is characterized by a single relaxation time: $1/\gamma_a^{(0)}$, while the level b is characterized by three: $1/\gamma_b^{(0)}$, $1/\gamma_b^{(1)}$ and $1/\gamma_b^{(2)}$. We note that, as is obtained as a result of calculations in Ref. 15, the quantities $\Gamma_b^{(1)}$ and $\Gamma_b^{(2)}$, due to inelastic depolarizing collisions and entering into the relaxation characteristics $\gamma_b^{(1)}$ and $\gamma_b^{(2)}$ of the level with moment $j = 1$, are connected, for an interaction of the van der Waals type, by the relation $\Gamma_b^{(1)}/\Gamma_b^{(2)} = 1.13$.

In the case of the transition $0 \rightarrow 1$, we consider the formation of the stimulated echo on the narrow spectral line in the case $\theta_i \gg T_i/T_0$. Omitting the intermediate calculations, we obtain the result that the intensity of the electric stimulated echo in this transition is given by the expression (7) with the quantity I determined by formula (11), and by the vector e^e , which has the following components different from zero:

$$e_z^e = \frac{2 \sin \theta_1 \sin \theta_2}{9\theta_1\theta_2\theta_3} \left[\cos(\psi_1 - \psi_2) \sin \theta_2 \left(\exp\{-\gamma_a^{(0)} \tau_2\} + \frac{1}{3} \exp\{-\gamma_b^{(0)} \tau_2\} \right) + \frac{1}{2} \exp\{-\gamma_b^{(2)} \tau_2\} \left[\cos(\psi_1 - \psi_2) \left(\frac{1}{3} + \cos 2\psi_2 \right) \sin \theta_2 - 2 \sin(\psi_1 - \psi_2) \sin 2\psi_2 \sin \frac{\theta_2}{2} \right] \right], \quad (21)$$

$$e_x^e = \frac{2 \sin \theta_1 \sin(\theta_2/2)}{9\theta_1\theta_2\theta_3} \left\{ -2 \sin(\psi_1 - \psi_2) \sin \frac{\theta_2}{2} \exp\{-\gamma_b^{(1)} \tau_2\} + \exp\{-\gamma_b^{(2)} \tau_2\} \left[\cos(\psi_1 - \psi_2) \sin \theta_2 \sin 2\psi_2 + 2 \sin(\psi_1 - \psi_2) \sin \frac{\theta_2}{2} \cos 2\psi_2 \right] \right\}, \quad (22)$$

where

$$\theta_i = 2|d|e^{(i)}T_i/\sqrt{3}\hbar$$

are the areas of the exciting pulses.

It follows from a comparison of formulas (21) and (22) with formulas (9a) and (9b), taken at $j_a = 0$ and $j_b = 1$, that the polarization properties of the stimulated photon echo in the transitions $0 \neq 1$ depend on the areas θ_2 and θ_3 of the second and third exciting pulses.

As follows from (21) and (22), observation of the damping of the stimulated photon echo with increase in τ_2 of the X component of the maximum intensity in the case $\psi_2 = \psi_1$ makes it possible to measure the time $1/\gamma_b^{(2)}$ of alignment relaxation. In the case of ψ_1 and ψ_2 such that

$$2 \sin(\theta_2/2) \operatorname{tg}(\psi_1 - \psi_2) + \sin \theta_2 \operatorname{tg} 2\psi_2 = 0,$$

observation of the damping of the maximum of this same term makes it possible to observe the time $1/\gamma_b^{(1)}$ of the orientation relaxation. Finally, for ψ_1 and ψ_2 such that

$$2 \sin(\theta_2/2) \sin 2\psi_2 \operatorname{tg}(\psi_1 - \psi_2) = \sin \theta_2 (\frac{1}{3} + \cos 2\psi_2),$$

the times $1/\gamma_b^{(0)}$ of the population relaxation can be observed from the damping of the Z component of the maximum intensity of the stimulated echo.

In conclusion, we note that, as considerations given in the present section have shown, there is no sense in greatly increasing the areas of the exciting light pulses for spectroscopic purposes, since the small gain obtained in such a case in the intensity of the echo leads to a significant complication of the formulas and makes more difficult the extraction of experimental information from them.

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