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CONDENSED-MATTER SPECTROSCOPY

The Effect of Uncorrelatedness of Inhomogeneous Broadening on the Formation of Transient Optical Processes in Multilevel Systems

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Abstract—The correlation of inhomogeneous broadening at various frequency transitions in three-level systems is investigated, as is its effect on the intensity of a stimulated photon echo. It is shown that the correlation coefficient of inhomogeneous broadening at two different transitions and the response intensity of the stimulated photon echo depend on the parameter of the random interaction of an optical electron with a local field, as well as on the distribution width of additional frequency shifts owing to a partial fixation of transition energies. In this case, an insignificant variation of the correlation coefficient results in a considerable decrease in the response intensity.

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INTRODUCTION

The resonance interaction of laser radiation with matter is one of the fundamental problems of contemporary physics. Coherent fields can induce interference of two or more quantum states. This interference can manifest itself both in the coherent response of a system of particles and at the level of a single particle, which is a quantum object. The study of these processes is of interest not only for basic science; it also has relevance for application. For example, coherent processes can be used for storage and processing of information [1–4]. Investigation of the interaction of several resonance fields with multilevel quantum systems (atoms, molecules, impurity ions, etc.) is of interest. This interest is determined by possible applications of different effects that are observed as a result of multifrequency excitation of quantum objects. Among them, one can note color echo holography [5], information compression in three-level media [6], quantum information copying [4], and multilevel quantum gates performing logical operations. Recording and reconstruction of echo holograms in multilevel systems result in the possibility of performing, along with logical operations, variation of real time scale and sequence of events, information from which was included in the spatiotemporal structure of an object pulse [7, 8]. In the listed processes, information from the object pulse is transformed into structural (potential) information, of which transient dynamic lattices of populations and polarizations of the resonant medium are the carrier, i.e., the spatial frequency distribution of q-bits in the range of inhomogeneously broadened lines of resonance transitions. Therefore, the formation of optical transient processes in multilevel systems depends significantly on the degree of correlation of the inhomogeneous broadening at different frequency transitions in connection with possible destruction of the reversible phase memory of the system.

Spectral inhomogeneity is more or less inherent in all real media. In the gas, it is caused by the Doppler effect; in this case, inhomogeneous broadening at different resonance transitions is completely intercorrelated. Deformation broadening (dislocations and point defects in the lattice), broadening caused by random electric fields and gradients of fields of charged defects, and irregularities of the lattice structure are typical mechanisms of inhomogeneous broadening in solids. Thus, both energies of states and energies (frequencies) of resonant transitions can be considered as functions of many parameters x_i, the number of which is more than one. Changes in these parameters result in a distribution of optical centers over frequencies, which is described by a multiparametric function $g(\Delta(x_1...x_n))$. In this case, even a monochromatic excitation of the system to a level with the energy $E_i(x_1...x_n)$ may not result in separation of optical centers of the same type, since the fixation of the value of a multivariable function does not fix the values of its arguments (there can be several local extrema). Therefore, only the following condition can be imposed [9]:

$$\Delta E_{ij}(x_1...x_n) = E_i(x_1...x_n) - E_j(x_1...x_n) = \hbar\omega_{ij},$$

from which it follows that, upon comparison of two different resonant transitions having one common level, parameters that remained unfixed at one transition affect the energy (frequency) of the other transition. The degree of preserving the coherence in a multilevel system upon its excitation at different resonant transitions depends on the degree of this fixation.

In this paper, we study the formation of stimulated photon echo in a three-level system in relation to the correlation of inhomogeneous broadening at different resonant transitions.

BASIC EQUATIONS

An equation for single-particle density matrix in a particle-fixed coordinate system has the form

$$i\hbar \frac{\partial \rho}{\partial t} = [H, \rho], \tag{1}$$

where $H = H_0 + H_C + U + V$, H_0 and H_C are the Hamiltonians of the quantum system and the environment, U is the operator of their interaction, and V is the operator of interaction of the quantum system with radiation. Considering the relaxation processes to be Markovian, we can find from Eq. (1) [10]

$$\frac{\partial \rho_{ij}}{\partial t} = -\frac{i}{\hbar}[V, \rho]_{ij} - \sum_{kl} K_{ijkl}\rho_{kl}, \tag{2}$$

where K_{ijkl} are the relaxation coefficients.

Upon description of the interaction of the quantum system with the radiation, we restrict our consideration to the case of short laser pulses the duration Δt of which is much shorter than the time of irreversible relaxations. Thus, during the action of the ηth pulse, the equation for the density matrix in the rotating coordinate system can be written in the form

$$\frac{\partial \tilde{\rho}}{\partial t} = -\frac{i}{\hbar}[B_\eta, \tilde{\rho}], \tag{3}$$

where

$$B_\eta = \tilde{H}_0 + \tilde{V}_\eta - \hbar A,$$

$$\tilde{H}_0 = e^{iAt} H_0 e^{-iAt},$$

$$\tilde{V}_\eta = e^{iAt} V_\eta e^{-iAt},$$

A is the transfer matrix to the rotating coordinate system. The solution of Eq. (3) is written as

$$\begin{aligned} &\tilde{\rho}(t - t_\eta) \\ &= \exp\{-i\hbar^{-1} B_\eta (t - t_\eta)\}\tilde{\rho}(t_\eta)\exp\{i\hbar^{-1} B_\eta (t - t_\eta)\}, \end{aligned} \tag{4}$$

where t_η is the time moment of the beginning of the action of the ηth pulse. Enveloping exponentials in Eq. (4) can be derived by the methods of matrix functions. As an example, we find the matrix function $f(\alpha_\eta B_\eta)$, where B_η is the matrix of the mth order, $\alpha_\eta = i\hbar^{-1}\Delta t_\eta$. The roots of the matrix $\alpha_\eta B_\eta$ are found from the secular equation

$$|\lambda I - \alpha_\eta B_\eta| = 0,$$

where I is the unit matrix of the mth order. Assuming that the roots are $\lambda_{s\eta}^{m_s}$, where m_s is the multiplicity of the root $\lambda_{s\eta}$, $\sum_s m_s = m$, then

$$\begin{aligned} f(\alpha_\eta B_\eta) = \sum_{k=1}^{s} \Big[& f(\lambda_{k\eta}) Z_{k1}^{\eta} \\ & + f'(\lambda_{k\eta}) Z_{k2}^{\eta} + \ldots. + f^{(m_k - 1)}(\lambda_{k\eta}) Z_{km_k}^{\eta} \Big], \end{aligned} \tag{5}$$

where Z_k^η are the components of the matrix $f(\alpha_\eta B_\eta)$, which are found from the system of equations

$$g_i(\alpha_\eta B_\eta) = \sum_{k=1}^{s} \Big[g_i(\lambda_{k\eta}) Z_{k1}^{\eta} + q_i'(\lambda_{k\eta}) Z_{k2}^{\eta} + \ldots \Big], \tag{6}$$

where

$$g_1(\lambda) = \lambda^0 = 1, \quad g_2(\lambda) = \lambda, \quad \ldots, \quad g_m(\lambda) = \lambda^{m-1},$$

$$g_1(\alpha_\eta B\eta) = I, \quad g_2(\alpha_\eta B_\eta) = \alpha_\eta B_\eta, \quad \ldots,$$

$$g_m(\alpha_\eta B_\eta) = \alpha_\eta^{m-1} B_\eta^{m-1}.$$

It is convenient to represent the density matrix in Eq. (4) in the form

$$\tilde{\rho}(t) = \sum_{\alpha\beta} \tilde{\rho}_{\alpha\beta}(t) P_{\alpha\beta}, \tag{7}$$

where $P_{\alpha\beta}$ are projective matrices. Substituting Eq. (7) into Eq. (4), we obtain

$$\begin{aligned} \tilde{\rho}(t_\eta + \Delta t_\eta) &= \sum \tilde{\rho}_{\alpha\beta}(t_\eta) e^{-\alpha_\eta \beta_\eta} P_{\alpha\beta} e^{\alpha_\eta \beta_\eta} \\ &= \sum_{\alpha\beta\gamma\delta} \tilde{\rho}(t_\eta) p_{\gamma\delta\alpha\beta}^{\eta} P_{\gamma\delta}. \end{aligned} \tag{8}$$

Here,

$$\sum_{\gamma\delta} p_{\gamma\delta\alpha\beta}^{\eta} P_{\gamma\delta} = e^{-\alpha_\eta B_\eta} P_{\alpha\beta} e^{\alpha_\eta B_\eta}. \tag{9}$$

Thus, it is necessary to calculate the coefficients $p_{\gamma\delta\alpha\beta}^{\eta}$ in order to find the density matrix after the action of the ηth pulse.

Within time intervals between exciting pulses, the evolution of the density matrix in the rotating coordinate system is described by the kinetic equations of the form

$$\frac{d\tilde{\rho}_{nn}}{dt} = \sum (K_{n'n}\tilde{\rho}_{n'n'} - K_{nn'}\tilde{\rho}_{nn}), \tag{10}$$

$$\frac{d\tilde{\rho}_{nn'}}{dt} = \left(-i\Delta_{nn'} - \frac{1}{\xi_{nn'}}\right), \tag{11}$$

where $K_{nn'}$ are the relaxation coefficients of the diagonal part of the density matrix and $\xi_{nn'}$ are the coefficients of the off-diagonal part. The solution of Eq. (10) has the form

$$\rho = e^{Qt}\rho_0, \tag{12}$$

where ρ_0 is the initial value of the density matrix. In the case of a three-level system,

$$Q = -K_1P_{11} + K_{32}P_{12} - K_2P_{22},$$

where $K_1 = K_{21}$ and $K_2 = K_{32} + K_{31}$. The calculation of $F = \exp(Qt)$ matrix by the methods of matrix functions yields

$$F_{11} = e^{-K_1t}, \quad F_{12} = -\frac{K_{32}}{\gamma}K_2K_1\left(e^{-K_1t} - e^{-K_2t}\right),$$

$$F_{21} = F_{31} = F_{32} = 0, \quad F_{22} = e^{-K_2t}, \quad F_{33} = 1,$$

$$\gamma = -K_1K_2^2 + K_2K_1^2.$$

Correspondingly, for the off-diagonal part of the density matrix, we have

$$\rho_{nn'} = \exp\left(-i\Delta_{nn'} - \frac{1}{\xi_{nn'}}\right)\rho^0_{nn'}, \tag{13}$$

where

$$\Delta_{nn'} = \hbar^{-1}\Delta E_{nn'}(x_1 \ldots x_n) - \omega_{nn'}.$$

Therefore, the destruction of the coherence in the resonance system can occur not only due to irreversible relaxations but also as a result of a partial fixation of the energy of one resonant transition relative to the energy of the other transition.

EFFECT OF UNCORRELATEDNESS OF INHOMOGENEOUS BROADENING ON THE FORMATION OF A STIMULATED PHOTON ECHO IN A THREE-LEVEL SYSTEM

The Hamiltonian of a three-level optical center in a crystal matrix in the rotating coordinate system is written in the form

$$\tilde{H}_0 = \hbar\Delta P_{22} + P_{33}\hbar\Gamma\left(\Delta + \Delta' m(\Gamma, x_1 \ldots x_n)\right), \tag{14}$$

where $\Delta = \hbar^{-1}E_{12}(x_1 \ldots x_n) - \omega$, P_{ij} are projective matrices (the ij element of which is unity, while the remaining elements are zero), Γ is the parameter of nonequidistance of the system, $m(\Gamma, x_1 \ldots x_n)$ is the parameter that determines the inequality of interaction of an optical electron with a local crystal field in different states $\left(\lim\limits_{\Gamma\to 1} m(\Gamma, x_1 \ldots x_n) = 0\right)$ and Δ' is an additional frequency shift related to the partial fixation of the energy of the transition 1–3 relative to the energy of the transition 1–2.

The correlation coefficient of inhomogeneous broadening at different frequencies is introduced similarly to [11],

$$R_{ik,i'k'} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{(f_{ik} - z_{ik})(f_{i'k'} - z_{i'k'})}{\sigma_{ik}\sigma_{i'k'}} \times g(\Delta)g_1(\Delta')d\Delta d\Delta',$$

$$f_{12} = \Delta, \quad f_{13} = \Gamma\left(\Delta + \Delta' m(\Gamma, x_1 \ldots x_n)\right), \quad f_{23} = f_{13} - \Delta, \tag{15}$$

$$z_{ik} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} f_{ik}g(\Delta)g_1(\Delta')d\Delta d\Delta',$$

$$\sigma^2_{ik} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}(f_{ik} - z_{ik})^2 g(\Delta)g_1(\Delta')d\Delta d\Delta',$$

and the distributions of optical centers over the frequencies $g(\Delta)$ and $g_1(\Delta')$ are considered to be Gaussian with the dispersions σ^2 and σ'^2, respectively. Thus, for each isochromate of the inhomogeneously broadened line at one transition, there is a set of isochromates of the inhomogeneously broadened line at the other transition, which can lead to loss of the phase memory of the system under consideration. In its turn, the phase memory loss can affect significantly the formation of optical transient processes in multilevel systems.

Figure 1 presents the results of numerical calculation of the correlation coefficient of inhomogeneous broadening at different frequency transitions (1–2 and 1–3).

It follows from Fig. 1 that the R_{12-13} correlation coefficient of inhomogeneous broadening at different frequency transitions decreases with an increase in the $m(\Gamma, x_1 \ldots x_n)$ and σ' parameters, which results in the loss of the phase memory of the system.

We will consider the formation of a stimulated photon echo (SPE) in a system of three-level optical centers with nonequidistant levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ with energies $\hbar\Omega_0, \hbar\Omega_1, \hbar\Omega_2$, respectively (Fig. 2).

The durations of exciting laser pulses Δt_η are considered to be sufficiently small for their frequency spectrum to completely overlap the inhomogeneously broadened lines of the resonant medium.

After the action of the ηth laser pulse, the density matrix can be found from Eq. (3), where the transition matrix into the rotating coordinate system will have the form of $A = P_{22}\omega_{12} + P_{33}\omega_{13}$; the matrix of the

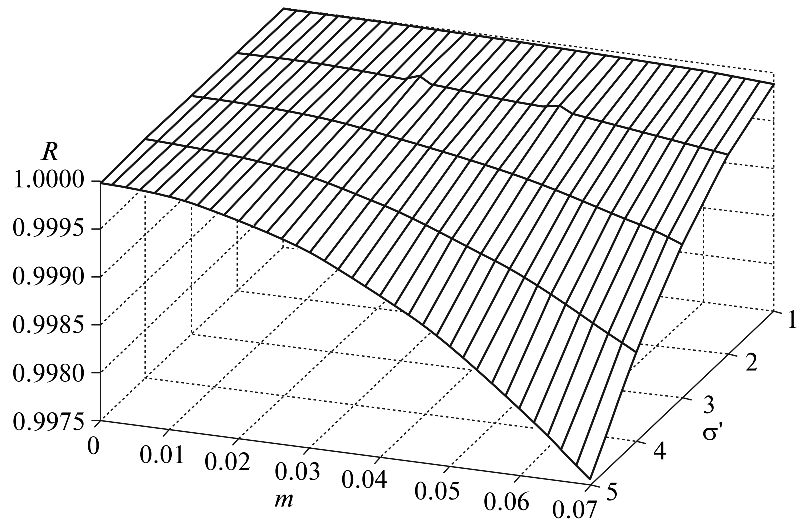

Fig. 1. Dependence of the correlation coefficient R_{12-13} of inhomogeneous broadening at two different transitions on the parameter $m(\Gamma, x_1...x_n)$ of random interaction of an optical electron with a local field and on the width σ of the distribution $g(\Delta)$ of additional frequency shifts Δ' owing to the partial fixation of the energy of the transition 1—3 relative to the energy of the transition 1—2.

interaction operator with the ηth laser pulse is written in the form

$$\tilde{V}^{(\eta)} = P_{12}V_{12}^{(\eta)}e^{-i\omega_{12}t} + P_{21}V_{21}^{(\eta)}e^{i\omega_{12}t} + P_{13}V_{13}^{(\eta)}e^{-i\omega_{13}t} + P_{31}V_{31}^{(\eta)}e^{i\omega_{13}t},$$

$$V_{ik}^{(\eta)} = -\frac{1}{2}d_{ik}\varepsilon_{ik}^{(\eta)}e^{i\omega_{ij}t - i\mathbf{k}_{(\eta)}\mathbf{r}}.$$

Here, $\mathbf{r}$ is the radius-vector of the position of the optical center, d_{ik} is the dipole moment of the ik transition, and $\varepsilon_{ij}^{(\eta)}$ is the electric field strength of the Fourier-component of the ηth laser pulse.

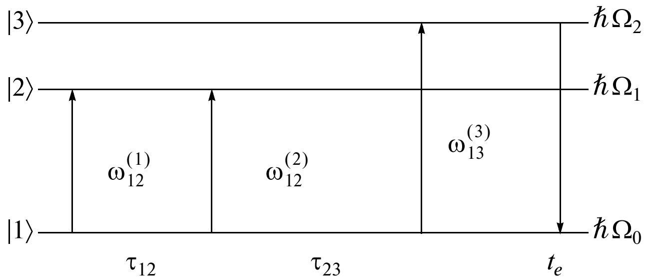

Fig. 2. Excitation spectrum of a SPE in a three-level system. $\omega_{ij}^{(\eta)}$ is the carrier frequency of the η laser pulse at the transition ij, t_e is the time of the appearance of the SPE response.

The electric field strength of the response in the wave zone at the observation point with radius-vector $\mathbf{R}$ is written in the form

$$\mathbf{E} = \sum_j \frac{1}{c^2|\mathbf{R} - \mathbf{r}_j|}\left[\langle \ddot{\mathbf{d}}_j(t') \rangle \times \mathbf{n}\right] \times \mathbf{n}, \quad (17)$$

where $\mathbf{r}_j$ is the radius-vector of the position of the jth optical center,

$$\mathbf{n} = \frac{\mathbf{R}}{|\mathbf{R}|}, \quad t' = t - \frac{|\mathbf{R} - \mathbf{r}_j|}{c}, \quad \langle \mathbf{d}_j(t') \rangle = \mathrm{Sp}(\mathbf{d}_j\rho(t)),$$

and the solution for the density matrix $\rho(t)$ is derived from Eqs. (4), (7), (9), (12), and (13). In the case of $K_1 \ll \tau_2$ and $K_2 \ll \tau_2$, the effect of the longitudinal relaxation on the intensity of the stimulated photon echo response can be neglected and the dependence of the electric field strength of the response on the uncorrelatedness of inhomogeneous broadening at different frequency transitions and longitudinal irreversible relaxation follows from Eq. (17),

$$E(t) \sim \left[\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \Phi(t, \Delta, \Delta')g(\Delta)g_1(\Delta')d\Delta d\Delta'\right] \times \exp\left\{-\frac{\tau_{12}}{\xi_{12}} - \frac{t - \tau_{12} - \tau_{23}}{\xi_{13}}\right\}, \quad (18)$$

where

$$\Phi(t, \Delta, \Delta') = \exp\left\{i\Delta\Gamma\left[(t - \tau_{12} - \tau_{23})\left(1 - \frac{\Delta'}{\Delta}m(\Gamma, x_1...x_n) - \frac{\tau_{12}}{\Gamma}\right)\right]\right\}.$$

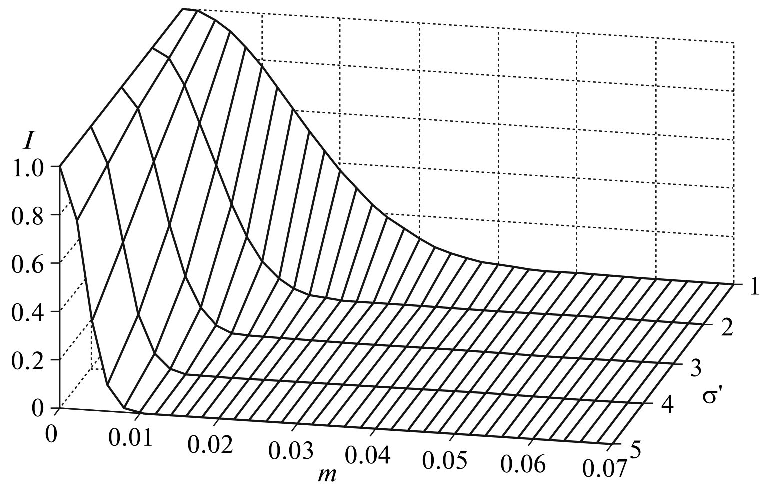

Fig. 3. Dependence of the intensity of the response of a stimulated photon echo on the parameter $m(\Gamma, x_1 \ldots x_n)$ of the random interaction of an optical electron with a local field and on width σ of the distribution $g_1(\Delta')$ of additional frequency shifts Δ' owing to the partial fixation of the energy of the transition 1–3 relative to the energy of the transition 1–2.

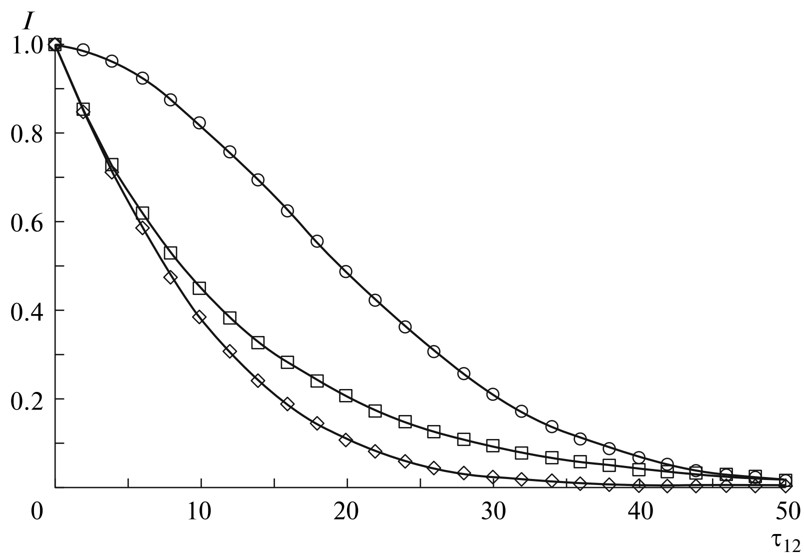

Fig. 4. Dependence of the intensity of the response of a stimulated photon echo on the time between the first and second laser pulses τ_{12} at different times of the longitudinal relaxation and different parameters of random interaction of an optical electron with a local field m. Open circles represent the dependence of the intensity of the SPE response on τ_{12} at $\xi_{ij} \gg \tau_{12}$, $m = 0.04$; squares correspond to the dependence of the intensity of the SPE response on τ_{12} at $\xi_{ij} \sim \tau_{12}$, $m = 0$; and diamonds show the dependence of the intensity of the SPE response on τ_{12} at $\xi_{ij} \sim \tau_{12}$, $m = 0.04$.

Figure 3 presents the results of numerical calculations of the relative intensity of the SPE response $I_{\rm rel} = |E(t)|^2 / |E_{\max}(t)|^2$.

Comparison of Figs. 3 and 1 shows that small variations of the R_{12-13} correlation coefficient of inhomogeneous broadening at different frequency transitions result in a significant decrease in the SPE intensity with an increase in the parameters $m(\Gamma, x_1 \ldots x_n)$ and σ', which is due to the loss of the phase memory of the system.

Variation of the time interval τ_{12} results in variation of the intensity of the SPE response, which was the result of two factors: first, by the relaxation decay (ξ_{12}, ξ_{13}) and, second, by the decay at the expense of the uncorrelatedness of inhomogeneous broadening (Fig. 4).

CONCLUSIONS

It has been shown that, in the considered excitation scheme, the correlation coefficient of inhomogeneous broadening at two different transitions R_{12-13} and the intensity of the stimulated photon echo response depend on the parameter of random interaction of an optical electron with local field m and on frequency shifts σ'. Furthermore, small variations of the correlation coefficient result in a significant decrease in the response intensity. A change in time τ_{12} between laser pulses gives rise to an additional decay, which is similar to the relaxation at the expense of the uncorrelatedness effect.

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