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Coherence Restoring Effect in the Three-Level System by Spatially Inhomogeneous Electric Field

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Abstract: *Phase memory in a three-level system which is associated with isochromatic matching of inhomogeneously broadened lines excited by laser radiation at different frequency transitions having one common energy level at different time moments were examined. It is shown that the exposure on the system of the external spatially inhomogeneous electric field allows you to manage the system's coherence that can be used to significantly increase the intensity of the response stimulated photon echo in the three-level system.*

Keywords: *stimulated photon echo, the phase memory, frequency time correlation, the three-level system, inhomogeneously broadened line, spatially inhomogeneous electric field*

1. Introduction

The formation of transient optical processes in multilevel systems occurs when exposed to the medium of short laser pulses spaced in time and resonance to different transitions. Therefore, their interaction with each other becomes possible only through the medium and only in the case when the medium has a sufficiently long phase memory. In this situation, each laser pulse transmits information about its wave characteristics into the medium, and this information is saved here until the arrival of the next laser pulses. Therefore, when we speak of non-simultaneous interaction, it does not mean violation of causal relationships, despite the fact that the optical pump waves do not encounter each other; they cannot interfere directly. In this case, the recording medium is a kind of bridge connecting the time and space wave characteristics of laser fields.

In order to describe the degree of conservation of phase memory in the resonance system on different time intervals in [1-2] was first introduced by frequency-time correlation coefficient of inhomogeneous broadening. It has been shown that the formation of transient optical processes in multilevel systems in solids significantly depends on the degree of correlation of inhomogeneous broadening on different frequency transitions and different time intervals in connection with the possible destruction of the reversible phase memory of the system due to partial mutual fixation of the energies of transitions [3]. In [4] was an investigation of the correlation of inhomogeneous broadening on various frequency transitions in a three-level system depending on non-equidistant parameter of the system spectrum, which depends on the parameter of random interaction of an optical electron with a local field $m(\Gamma, x_1,..., x_n)$ where the parameters $x_1 ... x_n$ define the energy states $E_i(x_1,..., x_n)$, where Γ is the non-equidistant parameter of spectrum of the system. It has been shown that partial mutual fixation of the energies of transitions can lead to significant destruction of the phase memory of the resonant system and the inability observation of optical transient processes in multilevel systems.

The impact on the resonant medium of the external spatially inhomogeneous electric fields can lead to additional artificially created inhomogeneous broadening. In this case it is possible to control the phase memory of the system by changing parameters of the external spatially inhomogeneous electric fields. The most effective influence on the phase memory of the system is the case of laser excitation of narrow frequency regions of the inhomogeneously broadened line of the resonance transitions. This artificially created inhomogeneous broadening by external spatial inhomogeneous electric fields due to the stark effect can be comparable with the variation of frequency due to the partial mutual fixation of the energies of transitions that lead to a partial recovery of coherence of the multilevel system. This effect can lead to an increase in the intensity of the stimulated photon echo response (SPE) in a three-level system, that is the opposite of photon echo locking effect [5, 6].

In this paper we found optimal conditions to maximize SPE response intensity in a three-level system.

2. Optimal Conditions to Maximize SPE Response Intensity in a Three-Level System

The typical mechanisms of inhomogeneous broadening in solids are: the deformational broadening (dislocations and point defects in the crystal lattice); the broadening due to random electric fields and gradients of fields of charged defects; violation of order in the crystal lattice structure. This means that energies of states and frequencies of resonant transitions can be seen as functions of many parameters $f(x_1,...,x_n)$. The assumption that the number of parameters is more than one is essential. The range of variation of parameters is determined by the distribution function $g(\Delta(x_1,...,x_n))$ of the optical centers over frequencies, but even monochromatic excitation to an energy level $E(x_1,...,x_n)$ cannot lead to the separation of optical cents of the same type, since the fixation of a function value of many variables not lead to fixation of values of arguments (there are several local extrema) and only imposes the condition [7]

$$\Delta E_{ij}(x_1,...,x_n) = E_i(x_1,...,x_n) - E_j(x_1,...,x_n) = \hbar\omega_{ij}^{exitation} \tag{1}$$

When comparing different transitions non-fixed parameters on one transition are affected on energy of another transition.

The question therefore arises, under what conditions does the constraint (1) imposed for transition i-j fix the energy transition i-k or k-j. In turn, from the degree of such fixation of energy transition comes the dependence of preservation of coherence in the multilevel system under the excitation of various transitions.

On the formation of optical transients processes affect in particular the relaxation processes. Lowering the temperature of the sample allows to increase the relaxation time and is used in the experiments for the observation of photon echo. Uncorrelated inhomogeneous broadening in the different resonance transitions does not depend on temperature and affect the intensity of the SPE response at low temperatures [3, 8].

The situation can be changed by affecting, on the three-level medium, by spatially inhomogeneous electric fields.

In describing the interaction of a quantum system with laser radiation limit ourselves only the case of short laser pulses duration Δt with much less time of irreversible relaxation. During the action of the η-th of the exciting laser pulse, the equation for the single-particle density matrix in the rotating coordinate system can be written in the form

$$\frac{\partial\tilde{\rho}}{\partial t} = -\frac{i}{\hbar}\left[B_\eta, \tilde{\rho}\right], \tag{2}$$

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 - the transition matrix in a rotating coordinate system. In the case of a three-level system $A = P_{22}\omega_{12} + P_{33}\omega_{13}$.

The Hamiltonian of the three-level optical center in the crystal matrix in the presence of external spatial inhomogeneous electric field with the gradient $\vec{\nabla}E$ we write in the form

$$\tilde{\mathrm{H}}_0 = \hbar\left(\Delta + C_s\vec{\nabla}E\vec{r}\right)\mathrm{P}_{22} + \mathrm{P}_{33}\hbar\left(\left(\Delta + C_s'\vec{\nabla}E\vec{r}\right)\Gamma + \Delta'\Gamma m\right), \tag{3}$$

where C_s, C_s' are the stark constants for the transitions 1-2 and 1-3 respectively, $\vec{r}$ is the radius – vector of location of the optical center, P_{ij} – projective matrix (element ij is equal to one and the other equal to zero), $\Gamma = \frac{\Omega_{13}}{\Omega_{12}}$ - a non-equidistant parameter of system's spectrum, $m(\Gamma, x_1 ... x_n)$ - defines the different interaction of the optical electron with the local crystal field in different states ($\lim_{\Gamma \to 1} m(\Gamma, x_1 ... x_n) = 0$), $\Delta = \hbar^{-1} E_{12}(x_1 ... x_n) - \Omega_{12}$ - initial frequency shift of an individual isochromatic grouping, Ω_{12} is the central frequency of the in homogeneously broadened line on the transition 1-2, Ω_{13} is the central frequency of the in homogeneously broadened line on the transition 1-3, Δ' - additional frequency shift due to the partial fixation of the energy of the 1-2 transition relative to the energy of the transition 1-3.

The solution of equation (2) we write in the form

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

Where t_η - time of exposure to the η-th pulse. The bordering exponents in (4) can be calculated by the methods of functions of matrices [9].

The density matrix after effect of η-th laser pulse we find from (4) and the matrix of interaction operator with the η-th laser pulse we write in the form

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

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

Where $\vec{r}$ - the radius vector of the location of the optical center, d_{ij} is the dipole moment of the transition $i - j$, $\varepsilon_{ij}^{(\eta)}$ the electric field Fourier-component of the η- th laser pulse, $\vec{k}_\eta$ - the wave-vector.

Consider the formation of the stimulated photon echo: when the first and second excitatory laser pulses act on the transition 1-2 and the third pulse on the transition 1-3. We assume that the spectral region of excitation of the inhomogeneously broadened line $\varepsilon_{12}^{(1)}(\Delta)$ is determined by the first laser pulse and spectra of the second and third pulses overlap the entire in homogeneously broadened line. The electric field intensity in this case we will receive as:

$$E(t) \sim \left[\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_V \varepsilon_{12}^{(1)*}(\Delta) \Phi(t, \Delta, \Delta', \vec{r}) g_1(\Delta) g_2(\Delta') d\Delta d\Delta' d\vec{r} \right], \tag{5}$$

Where V is the volume of excited part of the sample,

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

τ_{12} - the time interval between the first and second exciting pulses τ_{23} is the time interval between the second and third pulses. The distribution of optical centers over frequencies $g_1(\Delta)$ and $g_2(\Delta')$ we assume Gaussian with dispersions σ^2 and σ'^2 respectively.

In this case, to describe the phase memory of the resonance system we will introduce the coefficient of time-frequency correlation of inhomogeneous broadening on different frequency transitions and on different time intervals:

$$R_{12,13}(\tau_\eta, \tau'_{\eta'}) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \int_V \varepsilon_{12}^{(1)}(\Delta) \frac{(f_{12} - \bar{z}_{12})(f_{13} - \bar{z}_{13})}{\sigma_{12}^2 \sigma_{13}^2} g_1(\Delta) g_2(\Delta') d\Delta d\Delta' d\vec{r}, \tag{6}$$

where

$$f_{12} = \Delta + C_s \vec{\nabla} E(\tau_\eta) \vec{r},$$

$$f_{13} = \Gamma\left(\left(\Delta + C_s' \vec{\nabla} E(\tau_{\eta'}') \vec{r}\right) + \Delta' m(\Gamma, x_1 ... x_n)\right),$$

$$\bar{z}_{12} = \int d\vec{r} \iint \varepsilon_{12}^{(1)}(\Delta) f_{12} g_1(\Delta) g_2(\Delta') d\Delta d\Delta',$$

$$\bar{z}_{13} = \int d\vec{r} \iint f_{13} g_1(\Delta) g_2(\Delta') d\Delta d\Delta',$$

$$\sigma_{12}^2 = \int d\vec{r} \iint \varepsilon_{12}^{(1)} (f_{12} - \bar{z}_{13})^2 g_1(\Delta) g_2(\Delta') d\Delta d\Delta',$$

$$\sigma_{13}^2 = \int d\vec{r} \iint (f_{13} - \bar{z}_{13})^2 g_1(\Delta) g_2(\Delta') d\Delta d\Delta',$$

$\tau_\eta, \tau_{\eta'}'$ - time intervals in which there are the influence of external spatial inhomogeneous electric fields.

Thus, the coefficient of the time-frequency correlation of inhomogeneous broadening on different frequency transitions depends on the ratio of the magnitudes and directions of gradients of an external spatially inhomogeneous electric fields at different time moments allowing one to manage its value.

As resonant transitions we can take a three-level system of optical centers with nonequidistant levels |1›, |2›, |3› which correspond to energy levels of Pr^{3+} ions in the LaF_3 matrix in a $LAF_3{:}Pr^{3+}$ doped crystal. Transition |1›-|2› corresponds to a ${}^3H_4 \to {}^1D_2$ with a wavelength of 5925 Å, and the transition |1›-|3› corresponds to ${}^3H_4 \to {}^3P_0$ with the wavelength 4777 Å. On these transitions the stimulated photon echo was observed in [10, 11]. Further, we assume that the region of excitation of the inhomogeneously broadened line by the first laser pulse is equal to $k\sigma$, where $k < 1$.

Fig.1 presents the results of a numerical calculation of the relative intensity $I = |E(t)|^2 / |E_{\max}(t)|^2$ of the SPE response in the presence of spatially inhomogeneous electric field acting on the resonance medium after the third exciting laser pulse. From the analysis of fig.1 it is revealed that in the ranges of values of the gradient of a spatially inhomogeneous electric field ($200 V/cm^2 < |\vec{\nabla} E_2| < 400 V/cm^2$) there is an increase in the intensity of the SPE response is more than 10 times as compared to the case $|\vec{\nabla} E_2| = 0$. At the same time, the value of the coefficient time-frequency correlation of inhomogeneous broadening on different frequency transitions fig.2 do not experience a sharp change in this area. The slow change of the correlation coefficient leads to the photon echo response locking effect.

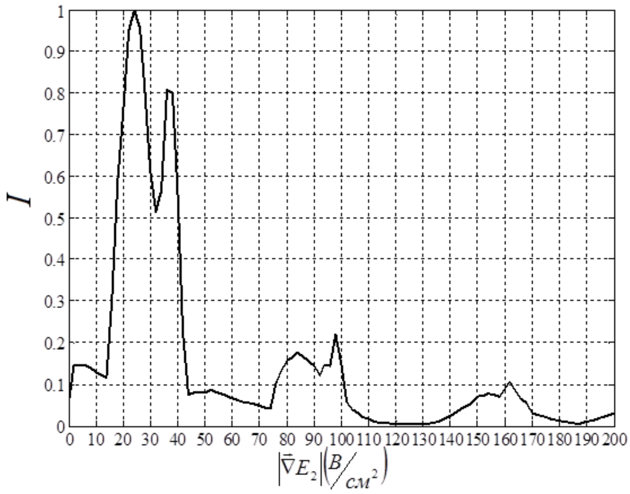

Figure1. *The SPE response intensity dependence formed in a three-level system of the value of the electric field gradient imposed on the sample in the time interval between third excitation pulse and the anticipated time of occurrence of SPE response. m=1, σ' =0.1 ns^{-1}, Γ=1.26, k=0.1, the sample size L=0.1m, σ=5 ns^{-1},*

C_s =100kHz/(V/cm), C_s' =100kHz/(V/cm)), $|\vec{\nabla} E_1| = 0 V/cm^2$, $\tau_{12} = \tau_{23} = 10\,ns$

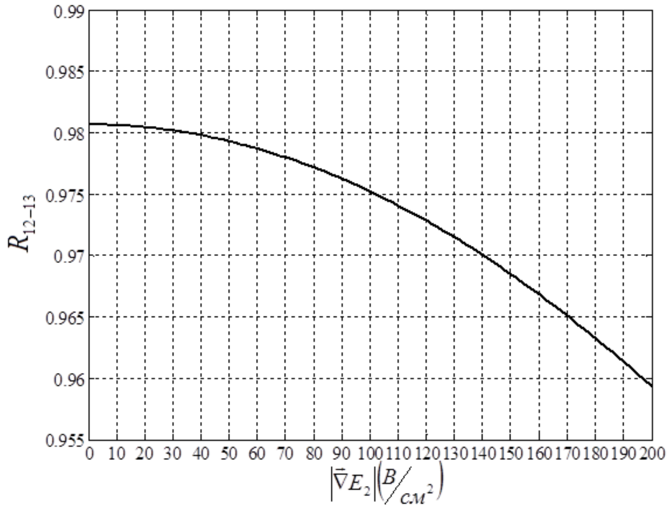

Figure2. *The dependence of the correlation coefficient of inhomogeneous broadening of different energy transitions in the three-level system of the value of the electric field gradient imposed on the sample in the time interval between third excitation pulse and the anticipated time of occurrence of SPE response. m=1,* σ' *=0.1 ns*$^{-1}$*, Γ=1.26, k=0.1, the sample size L=0.1m, σ=5 ns*$^{-1}$*,* C_S *=100kHz/(V/cm),* C'_S *=100kHz/(V/cm)),*

$$\left|\vec{\nabla}E_1\right| = 0V/cm^2,\ \tau_{12} = \tau_{23} = 10\,ns$$

Thus, for a small value of the gradient of a spatially inhomogeneous electric field there is the effect of partial recovery of coherence in a three-level system, which was destroyed due to the partial mutual fixation of the energies of the transitions, which is manifested in the increase in the intensity of the SPE response.

3. Conclusions

Under the influence of the spatially non-uniform electric field at the resonant three-level medium one can observed the photon echo response locking effect and effect of coherence recovery in a three-level system.

The optimal conditions of increase of the SPE response intensity in a three-level system is the influence after the third excitation laser pulse of spatially inhomogeneous electric field with a insufficient magnitude gradient for the manifestation of the effect of locking.

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