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Spin-dependent photon echo for an ensemble of three-level systems

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Abstract. We consider the action of a polarized rectangular laser pulse on a three-level system as a model of the interaction of electromagnetic radiation with matter. We managed to get the analytical solution for non-resonant excitation by light of a system with degenerate excited energy states without taking relaxation into account. The found analytical expressions were applied then to model the signal of a two-pulse photon echo from an ensemble of three-level systems.

Keywords: coherent dynamics, computer modeling

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Спин-зависимое фотонное эхо для ансамбля трехуровневых систем

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Аннотация. В данной работе рассмотрена модель взаимодействия электромагнитного излучения с веществом на примере действия поляризованного прямоугольного лазерного импульса на трехуровневую систему. Получено решение для нерезонансного возбуждения светом системы с вырожденными возбужденными энергетическими состояниями без учета релаксации. Найденные аналитические выражения были применены при моделировании сигнала двухимпульсного фотонного эха от ансамбля трехуровневых систем.

Ключевые слова: когерентная динамика, компьютерное моделирование

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Introduction

The study of the interaction of electromagnetic radiation with matter is one of the fundamental aspects of physics. The investigations of processes occurring in a medium under the influence of radiation allows one to obtain unique information about the physical properties of the matter, its composition, and structure. This plays an important role in our understanding of the nature [1]. Currently, knowledge about the interaction of radiation with matter is actively used for the development of the technology. This is confirmed by advances in electronics, the creation of new ways of working with optical information, and the development of new types of lasers. Thus, a fundamentally important task is the creation of theoretical models. Despite the apparent complexity of a comprehensive description of the processes that occur in matter under the action of a radiation, there are models that explain important phenomena by simplifying them from a physical point of view. For example, the influence of the radiation on multielectron atoms in some cases can be reduced to the consideration of electron transitions between certain levels since the influence of other levels is much less significant or even negligible. Such models, in particular, make it possible to describe the effects of four-wave mixing and photon echo, which have promise as a physical implementation of quantum memory underlying quantum communication [2,3]. In recent studies of trions in charged InGaAs quantum dots it was shown that using control pulses it is possible to separate the photon echo signal into two circular components [4]. In the trion system transitions with different circular polarizations are not coupled and can be excited independently by control pulses because in the absence of a magnetic field the system represents in fact two two-level systems. For a three-level exciton system, transitions with different spins are connected through a common ground state. Therefore, it is interesting to compare the behavior of the excitonic system under the action of control pulses by analogy with what was obtained for trions. In our work a model of interaction between electromagnetic radiation and matter was studied. We examined the effect of a polarized rectangular laser pulse on a three-level system. For the case when a system with degenerate excited states is excited by non-resonant light analytical solutions were obtained. We analyzed the dependence of the probability of the system being in a certain state on the frequency detuning and the pulse area. Then we applied the resulting analytical expressions to model the two-pulse photon echo signal from an ensemble of three-level systems. We investigated the sequential action of two short polarized laser pulses on three-level systems and the free dynamics of the system. As a result, time profiles of the photon echo signal were obtained for various excitation protocols with pulses of different polarization and power.

Theoretical model

An important example of radiation that well describes the real effect on matter is a rectangular laser pulse. It is considered further and the expression for the pulse has the following form:

$$E_{\pm}(t) = E_0^{\pm}(t) \cdot e^{-i\omega t}, \tag{1}$$

here $E_0^{\pm}(t) = \begin{cases} E_0^{\pm}, 0 < t < t_p \\ 0, t > t_p \end{cases}$ and ω are the amplitude and the frequency of the electromagnetic wave, t_p is the pulse duration.

We consider a three-level system as a medium, i.e., a model of matter that has three energy states. The description of the behavior of such a medium is given by the Schrödinger equation. The use of perturbation theory allows us to take into account the influence of electromagnetic radiation on the system.

In this case, the Hamiltonian in the Schrödinger equation consists of the Hamiltonian H_0 of the unperturbed system and of the term which is responsible for the interaction with electromagnetic radiation V.

$$\widehat{H}_0 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & \hbar\omega_0 & 0 \\ 0 & 0 & \hbar\omega_0 \end{pmatrix}, \tag{2}$$

where ω_0 is the frequency of the transition between the ground and the excited states.

$$\hat{V} = \begin{pmatrix} 0 & (d_+E_+)^* & (d\_E\_)^* \\ d_+E_+ & 0 & 0 \\ d\_E\_ & 0 & 0 \end{pmatrix}, \quad (3)$$

here $d_+ = \langle 1|\hat{d}|0\rangle$, $d\_ = \langle 2|\hat{d}|0\rangle$ are matrix elements of the dipole moment operator. We can assume, for example, that transitions to different excited states can be allowed for different polarizations of an electromagnetic wave.

The Schrödinger equation can be written as follows:

$$i\hbar \begin{pmatrix} \dot{\alpha}_0 \\ \dot{\alpha}_1 \\ \dot{\alpha}_2 \end{pmatrix} = \begin{pmatrix} 0 & (d_+E_+)^* & (d\_E\_)^* \\ d_+E_+ & \hbar\omega_0 & 0 \\ d\_E\_ & 0 & \hbar\omega_0 \end{pmatrix} \begin{pmatrix} \alpha_0 \\ \alpha_1 \\ \alpha_2 \end{pmatrix}, \quad (4)$$

where $\psi = \begin{pmatrix} \alpha_0 \\ \alpha_1 \\ \alpha_2 \end{pmatrix}$ is the wave function expanded over the basis of eigenstates.

Thus, the Schrödinger equation for a three-level system interacting with a rectangular laser pulse is a system of the three ordinary differential equations. Taking into account the initial conditions (the value of the wave functions at the initial time), analytical solutions were obtained:

$$\alpha_0(t) = e^{-i\frac{\Delta}{2}t} \cdot \left[\alpha_0^0 \left(\cos\left(\frac{\Omega}{2}t\right) + i\frac{\Delta}{\Omega}\sin\left(\frac{\Omega}{2}t\right) \right) - 2i\frac{f_+^*\alpha_1^0 + f_-^*\alpha_2^0}{\Omega}\sin\left(\frac{\Omega}{2}t\right) \right], \quad (5)$$

here $\Delta = \omega_0 - \omega$ is the detuning from the laser carrier frequency, $\Omega = \sqrt{\Delta^2 + 4|f_+|^2 + 4|f_-|^2}$ is the generalized Rabi frequency, $f_\pm = \dfrac{d_\pm E_0^\pm}{\hbar}$ is the pulse area.

$$\alpha_1(t) = \left[\alpha_1^0 - \frac{M_+\alpha_1^0 + f_-^* f_+ \alpha_2^0}{M_+ + M_-} \right] \cdot e^{-i\omega_0 t} - 2i\frac{f_+\alpha_0^0}{\Omega}\sin\left(\frac{\Omega}{2}t\right) \cdot e^{i(\frac{\Delta}{2}-\omega_0)t} + \\ + \left[\frac{M_+\alpha_1^0 + f_-^* f_+ \alpha_2^0}{M_+ + M_-} \left(\cos\left(\frac{\Omega}{2}t\right) - i\frac{\Delta}{\Omega}\sin\left(\frac{\Omega}{2}t\right) \right) \right] \cdot e^{i(\frac{\Delta}{2}-\omega_0)t}, \quad (6)$$

$$\alpha_2(t) = \left[\alpha_2^0 - \frac{M_-\alpha_2^0 + f_+^* f_- \alpha_1^0}{M_+ + M_-} \right] \cdot e^{-i\omega_0 t} - 2i\frac{f_-\alpha_0^0}{\Omega}\sin(\tfrac{\Omega}{2}t) \cdot e^{i(\frac{\Delta}{2}-\omega_0)t} + \\ + \left[\frac{M_-\alpha_2^0 + f_+^* f_- \alpha_1^0}{M_+ + M_-} \left(\cos\left(\frac{\Omega}{2}t\right) - i\frac{\Delta}{\Omega}\sin\left(\frac{\Omega}{2}t\right) \right) \right] \cdot e^{i(\frac{\Delta}{2}-\omega_0)t}, \quad (7)$$

where $M_\pm = |f_\pm|^2$.

Fig. 1 shows the dependencies of the probabilities of the system being in the ground and excited states on the detuning and on the magnitude of the pulse area when interacting with light occurs for one excited state only ($f_- = 0$), that is, in fact, a two-level system. It is assumed that the system was initially in the ground state. As can be seen from the figure, the populations of the levels oscillate. The magnitude of the detuning has a great influence, so with the same pulse power but with different detuning the transitions are different. The analysis shows that if the initial condition is that the system is in an excited state, then the graphs, as expected, are inverted relative to each other. Fig. 2 shows the dependences of the probabilities of the system being in both excited states on the detuning and the magnitude of the pulse area ($f_+ = f_-$). It is assumed that initially the system was in one of the excited states. Calculations show that the probability of the system being in the ground state looks similar to the probability for a two-level system. Moreover, it is clear from Fig. 2 that the probabilities of being in excited states form a complex checkerboard structure, since now they are distributed. The oscillations in the probabilities of finding the system between these states can be observed.

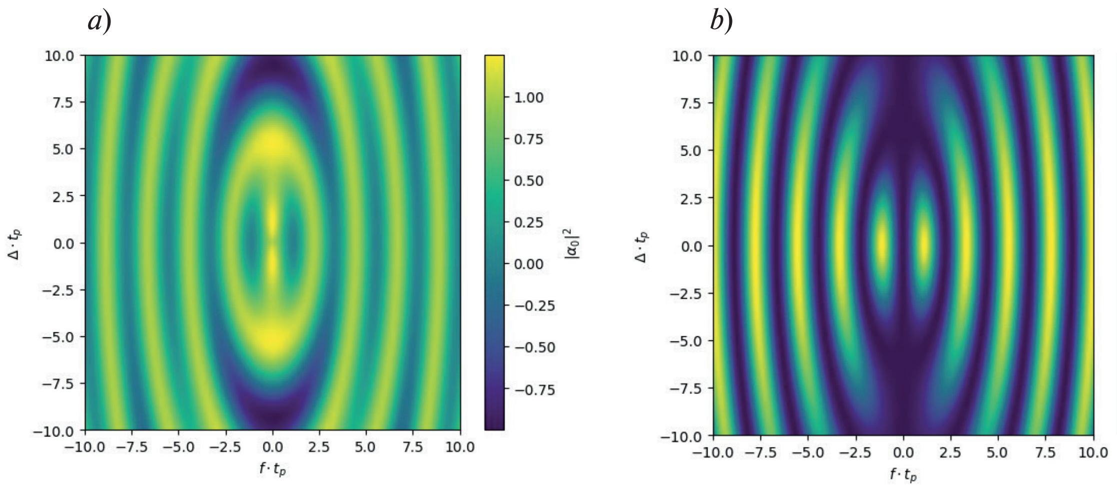

Fig.1. Dependences of the probabilities of the system being in the ground state (*a*) and in the excited state (*b*) on the pulse area and the detuning of the light frequency from the resonance of the system. Only one excited state interacts with light

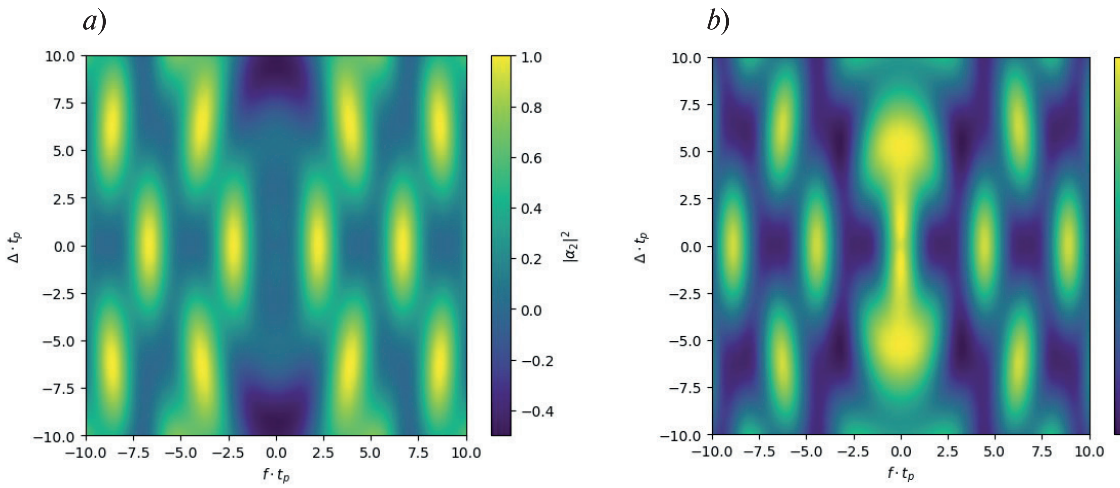

Fig. 2. Dependences of the probabilities of the system being in excited states 1 (*a*) and 2 (*b*) as a function of the pulse area and the detuning. Initially, the system was in an excited state 1

The density matrix formalism was used to model the photon echo signal. The evolution of the three-level system was determined by the Lindbland equation:

$$i\hbar \frac{\partial \rho}{\partial t} = \left[\widehat{H}, \rho\right] + i\hbar\Gamma, \tag{8}$$

here ρ is the density matrix, $\Gamma = \begin{pmatrix} (\rho_{11}+\rho_{22})/T_1 & -\rho_{01}/T_2 & -\rho_{02}/T_2 \\ -\rho_{10}/T_2 & -\rho_{11}/T_1 & -\rho_{12}/T_1 \\ -\rho_{20}/T_2 & -\rho_{21}/T_1 & -\rho_{22}/T_1 \end{pmatrix}.$

As in the previous part the Hamiltonian consisted of the Hamiltonian of the unperturbed system H_0 and the perturbation V for both impulses. The sequential action of two short laser pulses separated in time by a value of τ was considered taking into account the free precession between them. It was assumed that the time of the pulses action on the system is much shorter than the relaxation time which is in good agreement with experimental conditions in most cases. With such assumptions the problem can be divided into two parts: the consideration the interaction with light without taking into account relaxation during the pulses action; and the consideration the dynamics of the system. Using previously obtained expressions for the action of the pulse, we found the elements of the density matrix responsible for the contribution to the formation of the echo signal:

$$\tilde{\rho}_{01} \sim e^{-i\omega_0(t-2\tau)} e^{-t/T_2} \cdot \left|\alpha_0^0\right|^2 \cdot iL_1 \cdot L_2^2 \cdot M_1^* \left(f_{+1}^* f_{+2}^2 + f_{-1}^* f_{+2} f_{-2} \right), \tag{9}$$

$$\tilde{\rho}_{02} \sim e^{-i\omega_0(t-2\tau)} e^{-t/T_2} \cdot \left|\alpha_0^0\right|^2 \cdot iL_1 \cdot L_2^2 \cdot M_1^* \left(f_{-1}^* f_{-2}^2 + f_{+1}^* f_{+2} f_{-2} \right), \tag{10}$$

where $L_{1,2} = \dfrac{\sin(\Omega_{1,2} \cdot t_p)}{\Omega_{1,2} \cdot t_p}$, $\Omega_{1,2} = \sqrt{\nu_{1,2}^2 + \left|f_{-1,2}^2\right| + \left|f_{+1,2}^2\right|}$, $M_1 = \cos\left(\Omega_1 \cdot t_p\right) - i\dfrac{\nu}{\Omega_1}\sin\left(\Omega_1 \cdot t_p\right)$, $\nu = \dfrac{\Delta}{2}$.

The final expression for the polarization in the direction of four-wave mixing is:

$$P_\nu = \left\langle \hat{d} \right\rangle = Tr\left(\hat{d}\rho\right) \propto \mathrm{Re}\left(d_+ \tilde{\rho}_{01} + d_- \tilde{\rho}_{02}\right). \tag{11}$$

We consider the case when the spectral width of the pulse is comparable to the distribution width of the systems under study:

$$P = \int_{-\infty}^{\infty} P_\nu \cdot e^{-\frac{\nu^2}{2\sigma^2}} d\nu, \tag{12}$$

here σ is the variance of the normal distribution of three-level systems.

Numerical integration was used to obtain time profiles of the photon echo signal. The result is shown in Fig. 3.

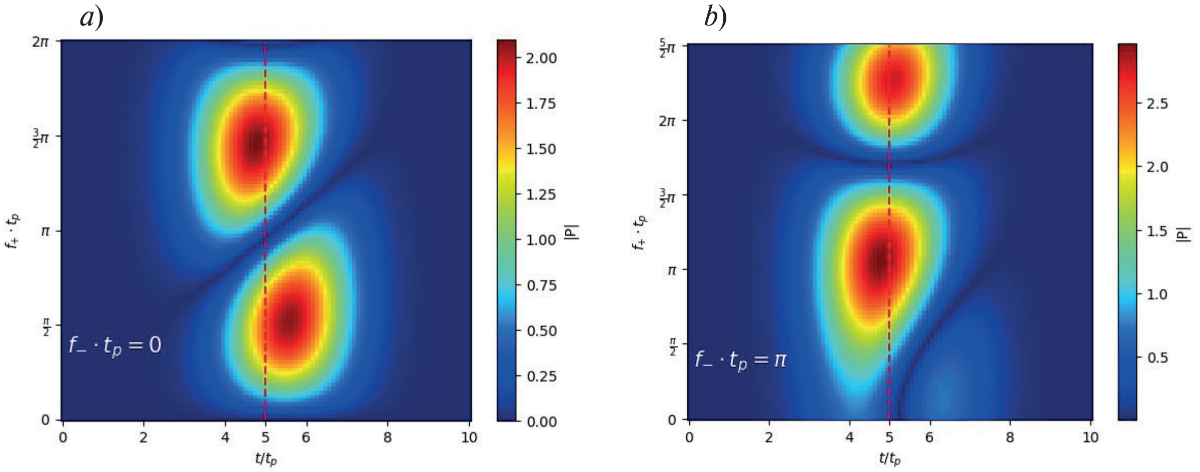

Fig. 3. Time profiles of the photon echo signal at $f_- = 0$ (*a*) and at $f_- = \pi$ (*b*)

It can be seen from the figure that the maximum of the photon echo signal is shifted in time relative to the time of occurrence of the echo signal at resonant excitation of the systems (marked by dashed line). The time of the echo signal occurrence can be controlled by changing the pulse areas. The analysis shows that, due to the common ground state, the excited states of the three-level exciton system is not separated by polarization in the photon echo protocol in contrast to the trion system, which in the absence of a magnetic field represents two two-level systems [4]. In the case when the transition to one of the excited states is prohibited, the results obtained coincide with the result for an ensemble of two-level systems [5].

Conclusion

In this work the action of a polarized rectangular laser pulse on a three-level system was studied. A solution was obtained for non-resonant excitation by light of a system with degenerate excited energy states without taking relaxation into account. The dependence of the probability of the system being in a certain state on the frequency detuning and on the pulse area was analyzed. The obtained analytical expressions were used to simulate the signal of a two-pulses photon echo from an ensemble of three-level systems. Time profiles of the echo signal were obtained for excitation protocols with pulses of different polarization and different powers. It turned out that if the detuning is taken into account the photon echo signal is shifted in time.

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