Difference in the behavior of the photon echo of excitons in InGaAs/GaAs quantum wells from the predictions of the model of two-level system ensemble

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Abstract. In this work, we studied the photon echo from heavy-hole excitons in a thin InGaAs/GaAs quantum well. To analyze the results, we used the model of an ensemble of two-level systems. The model allows us to describe the temporal profile and the moment of arrival of the echo signal, as well as the echo amplitude decay with increasing delay between pulses. In addition, excitation-induced dephasing effect was observed, that was beyond the limits of applicability of the model.

1. Introduction
Nonlinear optical experiments on the observation of the coherent dynamics of excitons and their complexes in semiconductor nanostructures are of special interest since they open new opportunities for creating devices working on optical effects. There are potential possibilities for creating optical memory based on the long-lived photon echo (PE) from charged exciton complexes [1].

Excitons in a thin InGaAs/GaAs quantum well (QW) could be used as a base for optical information processing due to the low inhomogeneous broadening of the exciton resonance in this system [2,3].

A simple description of the coherent excitons dynamics in QWs could be obtained in the model of an ensemble of two-level systems (TLS) [4]. This model allows one to describe temporal shifts in the PE signal from inhomogeneous ensembles [5] and Rabi-oscillations of the PE signal [5,6]. However, there are effects that cannot be described within the TLS ensemble model, such as excitation–induced dephasing (EID) [7], manifesting itself in the droop of the irreversible phase relaxation time with increasing intensity of light pulses.

In this paper, we study the photon echo from heavy-hole excitons in a thin InGaAs/GaAs QW and investigate the applicability limit of the TLS ensemble model.

2. TLS ensemble model
In this paper, the TLS ensemble model was used to theoretically describe the light–matter interaction. Each TLS has two states: ground (1) and excited (2), and can be described by a 2×2 Hermitian density matrix ρ. Its diagonal elements are states populations, and cross-diagonal ones are polarization of the system.
The interaction of light and a TLS and evolution in time $t$ could be described by the following Hamiltonian:

$$H = \left( \begin{array}{cc} 0 & \frac{dE_0 \exp(-i\omega_0 t)}{2} \\ \frac{dE_0 \exp(i\omega_0 t)}{2} & 0 \end{array} \right),$$

(1)

where $d$ is the transition dipole moment, $\omega_0$ is the transition frequency between states, $E_0$ is the electric field amplitude, $\omega$ is the excitation frequency.

The evolution of the density matrix can be described by the von Neumann equation:

$$i\dot{\rho} = [H, \rho].$$

(2)

It is more convenient to carry out further calculations in a rotating frame in the so-called Rotating Wave Approximation (RWA). To do this, we introduce the matrix $M$ and the rule connecting the density matrix in the rotating frame $\tilde{\rho}$ with $\rho$:

$$M = \left( \begin{array}{cc} 0 & \omega \\ 0 & \omega \end{array} \right), \quad \tilde{\rho} = e^{iMt} \rho e^{-iMt}.$$

(3)

With the matrix $M$, it becomes possible to introduce the time–independent Hamiltonian:

$$H_{eff} = e^{iMt}H e^{-iMt} - M = \left( \begin{array}{cc} 0 & \frac{dE_0}{\Delta} \\ \frac{dE_0}{\Delta} & 0 \end{array} \right),$$

(4)

where $\Delta = \omega - \omega_0$; we are not taking in account the field phase, considering that $E_0 = E_0^*$. In the RWA, the von Neumann equation could be rewritten in the following form:

$$i\dot{\tilde{\rho}} = [H_{eff}, \tilde{\rho}].$$

(5)

The solution of this equation with a known initial density matrix $\tilde{\rho}(t_0)$ allows one to obtain a density matrix at time $t$:

$$\tilde{\rho}(t) = e^{iH_{eff}(t-t_0)}\tilde{\rho}(t_0)e^{-iH_{eff}(t-t_0)}.$$  

(6)

2.1. Light action

Let us introduce the pulse area: $\Theta = dE_0 t_p$, where $t_p$ is the pulse duration. We consider an infinitely–short pulse ($t_p \to 0$) with a constant area ($\Theta = \text{const}$). In this case, we can neglect the precession, and introduce the dimensionless Hamiltonian of light action:

$$H_L = \left( \begin{array}{cc} 0 & \Theta/2 \\ \Theta/2 & 0 \end{array} \right).$$

(7)

According to the solution of the von Neumann equation (6), the action of the light pulse on the density matrix $\tilde{\rho}(t_0)$ can be written in the following form:

$$\tilde{\rho}(t_0 + 0) = \lim_{t_p \to 0} (\tilde{\rho}(t_0 + t_p)) = e^{iH_L} \tilde{\rho}(t_0)e^{-iH_L} =$$

$$= \begin{pmatrix} \tilde{\rho}_{11} \cos^2(\Theta/2) + \tilde{\rho}_{22} \sin^2(\Theta/2) + i \cos(\Theta/2) \sin(\Theta/2) (\tilde{\rho}_{21} - \tilde{\rho}_{12}) \\ \tilde{\rho}_{21} \cos^2(\Theta/2) + \tilde{\rho}_{12} \sin^2(\Theta/2) + i \cos(\Theta/2) \sin(\Theta/2) (\tilde{\rho}_{12} - \tilde{\rho}_{21}) \end{pmatrix},$$

(8)

where $\tilde{\rho}_{ij}$ are the elements of the initial density matrix $\tilde{\rho}(t_0)$. 


2.2. Temporal evolution

In the absence of light, the density matrix evolves with the Hamiltonian $H_T$:

$$H_T = \begin{pmatrix} 0 & 0 \\ 0 & \Delta \end{pmatrix}.$$  \hspace{1cm} (9)

The density matrix after time $t$ could be written in the following form:

$$\tilde{\rho}(t_0 + t) = \begin{pmatrix} \tilde{\rho}_{11} & \tilde{\rho}_{12}e^{-i\Delta t} \\ \tilde{\rho}_{21}e^{i\Delta t} & \tilde{\rho}_{22} \end{pmatrix}. \hspace{1cm} (10)$$

2.3. Photon echo experiment

Let us consider the primary (spontaneous) photon echo experiment. In this experiment, the TLS ensemble is excited by a sequence of two light pulses with a time delay $\tau_{12}$ between them. As a result, after a time delay $\tau_{12}$, a coherent response of the system is observed. This experiment can be described as a sequential action of the Hamiltonians $H_L$ and $H_T$ so it can be divided into the following steps:

1) Initially, a TLS with detuning $\Delta$ is in an unexcited state with a density matrix in the rotating frame $\tilde{\rho}(0)$:

$$\tilde{\rho}(0) = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}. \hspace{1cm} (11)$$

2) A light pulse with an are $\Theta_1$ acts on the system.

3) During the time $\tau_{12}$, the system evolves without external excitation.

4) A light pulse with an area $\Theta_2$ acts on the system.

5) The system evolves over time $\tau_{12}$.

The rules for transforming the density matrix $\tilde{\rho}(0)$ using the Hamiltonians $H_L$ and $H_T$ allow us to obtain $\tilde{\rho}(t)$ at any time $t$ after the arrival of the second pulse. The explicit form of the density matrix at each step with $\Theta_1 = \pi/2$ and $\Theta_2 = \pi$ is presented below:

$$\tilde{\rho}(0) \xrightarrow{H_L/\pi/2} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \xrightarrow{H_{\tau_{12}}/\pi} \begin{pmatrix} \frac{1}{2} e^{-i\Delta \tau_{12}} & \frac{i}{2} e^{i\Delta \tau_{12}} \\ -\frac{i}{2} e^{i\Delta \tau_{12}} & \frac{1}{2} \end{pmatrix} \xrightarrow{H_L/\pi} \tilde{\rho}(t).$$

$$\tilde{\rho}(0) \xrightarrow{H_L/\pi} \begin{pmatrix} \frac{1}{2} e^{i\Delta \tau_{12}} & \frac{i}{2} e^{-i\Delta \tau_{12}} \\ -\frac{i}{2} e^{-i\Delta \tau_{12}} & \frac{1}{2} \end{pmatrix} \xrightarrow{H_{\tau_{12}}/\pi} \begin{pmatrix} \frac{1}{2} e^{-i\Delta (t-\tau_{12})} & \frac{i}{2} e^{i\Delta (t-\tau_{12})} \\ -\frac{i}{2} e^{i\Delta (t-\tau_{12})} & \frac{1}{2} \end{pmatrix} \xrightarrow{H_L/\pi} \tilde{\rho}(t).$$

In the experiment, we can detect the polarization of the TLS, which is determined by the following expression:

$$P = \text{Tr}(\rho \hat{d}), \hspace{1cm} (13)$$

where $\hat{d}$ is the dipole moment matrix:

$$\hat{d} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \hspace{1cm} (14)$$

With the transition rule from RWA to the laboratory frame: $\rho = e^{-iMt} \tilde{\rho} e^{iMt}$, the final form of polarization can be found as:
The factor $e^{-t/T_2}$ is introduced phenomenologically, where $T_2$ is the irreversible phase relaxation time. Let us consider the Gaussian distribution of TLS frequencies in the ensemble:

$$S(\Delta) = \frac{T^*_2}{4\sqrt{\pi \ln 2}} \exp\left(-\frac{(\Delta - \omega')^2(T^*_2)^2}{16 \ln 2}\right),$$

(16)

where $T^*_2$ is the reversible phase relaxation time. This distribution is centered on the frequency $\omega'$. Let us now obtain the polarization of the entire ensemble for the case of coincidence of the frequencies of the exciting light and the central distribution frequency ($\omega = \omega'$) by integrating (15) with (16):

$$P_\Sigma(t) = \int_{-\infty}^{+\infty} P(\Delta)S(\Delta)d\Delta \sim \sin(\Theta_1)\sin^2\left(\frac{\Theta_2}{2}\right)e^{-t/T_2} e^{\frac{-4 \ln(2)(t - 2\tau_{12})^2}{(T^*_2)^2}}.$$  

(17)

First two terms in (17) describe the Rabi oscillations with the first and second pulse areas. The third term is responsible for the decay of the PE amplitude in time. The last term describes the Gaussian temporal profile of the echo signal centered at $2\tau_{12}$.

For small pulse areas, we can expand the first two factors of expression (17) in a Taylor series. Since the area of the pulse is proportional to the field amplitude $E_0$, this amplitude is proportional to the square root of intensity $E_0 \sim \sqrt{I}$. For the low pulse intensity case, we obtain the following form of polarization:

$$P_\Sigma(t) \sim \sqrt{I_1} I_2 e^{-t/T_2} e^{\frac{-4 \ln(2)(t - 2\tau_{12})^2}{(T^*_2)^2}}.$$  

(18)

3. Experiment

Figure 1: (a) PE profile with schematic excitation pulses (solid lines: experimental PE profile; dashed line: Gaussian fit); (b) PE temporal profiles for different delays between pulses $\tau_{12}$.

The sample P551 was grown on a GaAs:Si substrate with rotation and contains a 3 nm single In$_{0.03}$Ga$_{0.97}$As/GaAs quantum well. Optical excitation was performed by a Ti:Sapphire laser in
the picosecond regime. To reduce the thermal scattering of excitons, the sample was immersed in liquid helium in a closed–cycle cryostat at a temperature of 1.4 K. Two pulses were used for excitation and one for cross–correlation detection. More details on the experimental setup can be found in [8]. We have investigated the heavy-hole exciton resonance transition at 819 nm with co-linear excitation pulses and detection.

A typical PE profile with schematic first and second excitation pulses is shown in figure 1 (a). PE profiles at different delay times $\tau_{12}$ between excitation pulses are shown in figure 1 (b). Both pictures were obtained at relatively low pulse intensities $I_1 = 200 \, \mu W$ and $I_2 = 50 \, \mu W$.

According to figure 1 (a), the PE profile is close to the Gaussian, which is highlighted with the orange dashed line. In addition, the fact that the PE is centered exactly at $2\tau_{12}$ indicates the absence of a PE temporal shift [5] due to the low inhomogeneous broadening of the excitonic resonance [2]. The maximum PE amplitude decays exponentially (figure 1 (b)) which allowed us to extract the irreversible phase relaxation time $T_2 = 29$ ps. All of the above conclusions are consistent with the theoretically obtained equation (17).

![Figure 2](image1.png)

**Figure 2**: (a) PE decays normalized on $I_2$ for different second pulse intensities $I_2$; (b) the dependence of the irreversible phase relaxation time $T_2$ on intensities of the first and second pulses.

![Figure 3](image2.png)

**Figure 3**: (a) PE amplitude for different first pulse amplitudes at an intensity of the second pulse of $I_2 = 50 \, \mu W$; (b) PE amplitude for different second pulse amplitudes at an intensity of the first pulse of $I_1 = 50 \, \mu W$. 


The PE decays were measured at different intensities of the first ($I_1$) and the second ($I_2$) pulses. The data presented in figure 2 (a) were obtained at a constant $I_1 = 50 \mu W$ and normalized on $I_2$. All normalized PE decays overlap up to the second pulse intensities of around 200 $\mu W$ in accordance with equation (18). A further intensity increase leads to an acceleration of the PE decay. Extracted $T_2$ for different $I_2$ are depicted in figure 2 (b). It can be observed, that at high intensities, the irreversible phase relaxation time decreases by half. A similar trend is observed also for the case of constant $I_2$ and different $I_1$. Such behaviour is a manifestation of the EID effect, which cannot be described in the frame of an ensemble of non–interacting TLS.

Rabi–oscillations manifest themselves in the periodic behavior of the PE amplitude with a growing amplitude of excitation pulses, which is described by equation (17). However, the experimentally obtained dependences (figure 3 (a,b)) demonstrate the saturation of the PE signal with increasing intensity of the exciting pulses without a hint of oscillating behavior. Thus, the possibility of observing Rabi oscillations in this sample is limited by the EID effect.

4. Results and conclusion
In this work, we studied the PE from excitons in a thin InGaAs/GaAs quantum well. The TLS ensemble model was used for describing the PE and exploring the limits of its applicability for the system. It is found that the model could predict the PE arrival time, its temporal profile, and the exponential decay of the PE amplitude with $T_2$ time.

The irreversible relaxation time $T_2$ remains constant only in the low-power excitation regime. At higher excitation intensities, we have observed a decrease in $T_2$, known in literature as the EID effect [7]. This effect did not allow us to observe the Rabi-oscillations predicted in the TLS ensemble model.

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