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Modeling of coherent dynamics of excitons in a GaAs quantum well in the pump-probe experiment

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Abstract. We investigated polarization-dependent quantum beats of excitons with light and heavy holes observed in the pump-probe experiment. Oscillations in the energy of exciton resonances have been experimentally observed upon simultaneous excitation of exciton levels. To explain this effect, a theoretical model was formulated based on the consideration of a five-level scheme with the inclusion of nonlinearity in the form of an exchange exciton-exciton interaction. It has been found that the shift of energy resonances can be described only if the exciton-exciton exchange interaction is taken into account. The results of theoretical calculations of the coherent dynamics of excitons in this model are in agreement with the experimental data.

Keywords: quantum well, excitons, coherent dynamics, computer modeling

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Материалы конференции
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Моделирование когерентной динамики экситонов в квантовой яме GaAs, наблюдаемой в эксперименте «накачка-зондирование»

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Аннотация. В данной работе рассмотрены поляризационно-зависимые квантовые биения экситонов с лёгкой и тяжёлой дыркой, наблюдаемые в эксперименте «накачка-зондирование». Экспериментально обнаружены осцилляции энергии экситонных резонансов при одновременном возбуждении экситонных уровней. Для объяснения этого эффекта сформулирована теоретическая модель, основанная на рассмотрении пятиуровневой схемы с введённой нелинейностью в виде обменного экситон-экситонного взаимодействия. Обнаружено, что сдвиг энергетических линий удается описать только при учете обменного экситон-экситонного взаимодействия. Результат теоретических расчетов когерентной динамики экситонов в данной модели находятся в согласии с экспериментальными данными.

Ключевые слова: квантовая яма, экситоны, когерентная динамика, компьютерное моделирование

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Introduction

Studies of low-dimensional systems are promising areas of semiconductor physics. In particular, in GaAs/AlGaAs quantum wells, there are a number of effects that are not fully understood and are of considerable interest due to the possibility of using them for the purposes of optoelectronics. Investigation of exciton resonances in such systems can show such effects as polarization-sensitive dynamic shifts of the transition energy and quantum beats [1].

In this work we use an experimental pump-probe technique with spectral resolution and polarization selection to study coherent nonlinear dynamics of heavy and light hole excitons in GaAs/AlGaAs quantum well. This technique makes it possible to distinguish the observed effects by analyzing the shape of exciton resonances with high precision. We developed a theoretical model that allowed us to describe the observed coherent phenomena and obtained that the energy oscillations appear due to the exchange interaction of excitons in quantum well created in a superposition state.

Experimental Details

The experimental sample used in this work is a high-quality GaAs/AlGaAs 14-nm quantum well grown by molecular beam epitaxy. The sample was cooled in a closed-cycle helium cryostate to the temperature of 4 K. The kinetics of the secondary emission of this quantum well have been studied by the pump-probe method where the time-integrated reflectivity is studied as a function of the time delay between the pump and probe pulses [1]. In the experiment both heavy-hole and light-hole exciton resonances are excited with a spectrally wide pulses generated by a fs Ti:Sapphire laser. The pump beam has either linear or circular polarization. The probe beam has linear polarization which is split into either co- and cross-linear components or co- and cross-circular components with respect to the pump beam by a quarter-wave plate and a Wollaston prism. Both probe beam components are then detected at the same time by a spectrometer with a CCD camera. The probe beam spectra are detected at small time delays between pump and probe pulses with a sub-picosecond step.

Fig. 1, *a* shows the typical reflection spectrum of the sample in the absence of a pump pulse. Resonances of light and heavy hole excitons are seen in the form of reflection peaks and marked by Xhh and Xlh labels. Due to the small width and small depth of the well, it was possible to achieve a situation where the well contains only one exciton level, which is split into a levels with a heavy hole angular momentum 3/2 and with a light hole angular momentum 1/2. This splitting is clearly seen in the spectrum and is equal to 3.6 meV. The shape of the resonances is the result of a special design of the sample structure. The figure 1 also shows the spectrum fitting. All measured spectra were analyzed within the framework of the theory of non-local dielectric response. This allows to extract such characteristics of the states of heavy and light excitons as the resonance energy, radiative and non-radiative broadening [2–6]. The basic formulas used to fit the spectra are [7]:

$$r_{QW}(\omega) = \frac{i\Gamma_R}{\omega_x - \omega - i(\Gamma_R + \Gamma_{NR})}, \tag{1}$$

$$R(\omega) = \left| \frac{r_s + r_{QW}(\omega)e^{iz\phi}}{1 + r_s r_{QW}(\omega)e^{iz\phi}} \right|. \tag{2}$$

Here, rQW(ω) describes the amplitude reflection from the exciton resonance, Γ_R and Γ_{NR} are the rates of radiative and nonradiative decay of the exciton polarization, ω_x is the frequency of the exciton transition, ϕ is the phase shift of the light from the sample surface to the center of the quantum well, r_s is the amplitude reflection from the sample surface, R is the intensity of the reflection from the sample.

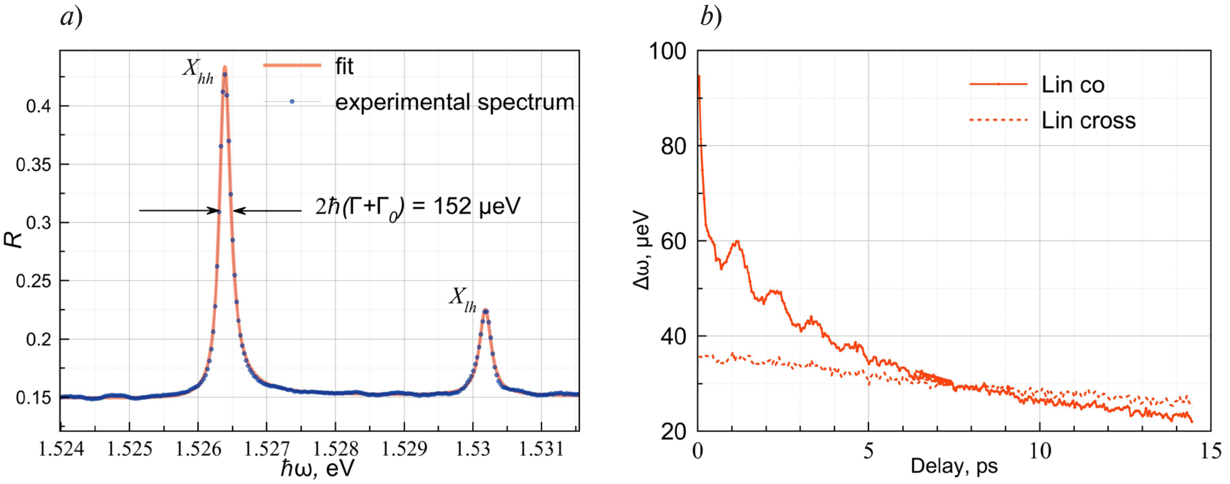

Fig. 1. Reflection spectrum of the GaAs/AlGaAs quantum well studied in the experiment (*a*). X_{hh} and X_{lh} correspond to optical transitions for heavy-hole and light-hole excitons respectively. Energy shift oscillations for the heavy-hole exciton in the linear co and cross configurations (*b*)

The dynamics of light-hole and heavy-hole exciton resonances have been studied in co- and cross- circular and linear polarizations of the pump pulse and the probe pulse. The results indicate the presence of an oscillating energy shift of the exciton lines in the reflection spectrum (see Fig. 1, *b*), as well as amplitude beats depending on the delay. Observation of these effects is associated with significant difficulties due to the need for a very high spectral resolution (of the order of 1 μeV). However, by using the high quality sample (ideal surface geometry, absence of defects) and modern technology such resolution can be achieved. The frequency of the energy shift oscillations is equal to the energy splitting of the exciton states with heavy and light holes. However, the observed beats are not present in all configurations, but only in those where the polarizations of the pump pulse and the probe pulse are coincided. As well, a very small energy shift has been observed in the cross-linear-polarizations configuration.

Theoretical model

For a theoretical description of the observed effect, a five-level exciton model is proposed, in which there are one ground state and four states of excitons with heavy and light holes, separated by spin. The action of light pulses on the system was described by the optical Bloch equations. It turned out that the detected oscillating component of the radiative broadening of exciton resonances in various polarization configurations can be interpreted as a manifestation of quantum beats of the states of excitons with light and heavy holes [8]. However, the energy shift can only be described if the exchange exciton-exciton interaction is taken into account, which has not been considered before. In our model the nonlinearity in the form of the exchange interaction between pump-produced excitons and probe beam excitons is considered in accordance with Ref. [1], and generalized to a five-level energy scheme. Such consideration required a transition to the second quantization formalism for excitons. During the simulation, the dynamics of excitons created by light pulses was considered sequentially: 1) A pump pulse creates a coherent superposition of exciton states in dependence on the polarization of light. 2) The probe pulse also produces an exciton in the superposition state and its further dynamics is determined by the exciton-exciton interaction with excitons produced by pump. It should be emphasized that the effect of energy shift oscillations is due to the exciton-exciton exchange interaction and arises only if the excitons are in the superposition quantum state. 3) Next, the intensity of reflection of the probe beam from the sample is calculated taking into account the resonant exciton reflection. Such consideration leads to an oscillating shift of exciton resonances for the same linear or circular polarizations of light beams, and to a small non-oscillating shift in crossed polarizations.

Let us consider in more detail the theoretical description of exciton coherent nonlinear dynamics.

The Hamiltonian of the system (without the interaction with light) is

$$H_{tot} = \hbar \sum_i \omega_i c_i^\dagger c_i + H_{int}, \tag{3}$$

where H_{int} is the exciton-exciton interaction Hamiltonian.

The interaction with light is described in the dipole approximation. The wavefunction of the exciton created by the pump pulse is

$$| \psi_{pump} >= \sum_i a_i c_i^\dagger \, | 0 >, a_i = \alpha_i e^{-i\omega_{0i} t}, \alpha_i \sim \frac{id_i}{\hbar} \int_{-\infty}^{t} E_{pump}(t') e^{-i\omega_{0i}(t-t')} dt', \tag{4}$$

where E_{pump} is the strength of the electric field of the pump pulse, d_i and ω_{0i} are the dipole moments and frequencies of exciton optical transitions, c_i^+ are creation operators. Likewise, the probe exciton wavefunction is

$$| \psi_{pr} >= \left(1 + \sum_i a_i' c_i'^\dagger \right) | 0 > . \tag{5}$$

Assuming that only identical excitons interact, the interaction Hamiltonian takes the form of the exchange interaction

$$H_{int} = \hbar \omega_{ex} \sum_i c_i^\dagger c_i c_i'^\dagger c_i'. \tag{6}$$

Here c_i and $c_i^{'}$ (c_i^+ and $c_i^{'+}$) are the annihilation (creation) operators for the pump and probe exciton, respectively. Since the pump and probe pulses are spectrally wide, superposition states of the heavy-hole and the light-hole state are excited. This corresponds to a linear transform of the complex amplitudes and the field operators

$$\gamma_j = \sum_k m_{jk} \alpha_k, c_j' = \sum_k m_{jk} c_k, \tag{7}$$

where m_{jk} is the transform matrix.

Applying the field operators to the pump and probe wavefunction, we can write down the Schrödinger equations for the complex amplitudes

$$\dot{\gamma}_j = -i \left(\left(\omega_0 + \omega_{ex,j} \right) \gamma_j + \sum_{k \neq j} \gamma_k \sum_l \omega_l m_{lj} m_{jk} \right), \tag{8}$$

where

$$\begin{cases} \omega_{ex,1} = \omega_{ex0} \left(\tilde{N}_0 + \frac{1}{2} Re[\left(\alpha_1^* + \alpha_3^* \right) \left(\alpha_2 + \alpha_4 \right) e^{i(\omega_h - \omega_l)(t+\tau)} + \alpha_1^* \alpha_3 + \alpha_2^* \alpha_4] e^{-\Gamma_c (t+\tau)} \right), \\ \omega_{ex,2} = \omega_{ex0} \left(\tilde{N}_0 + \frac{1}{2} Re[-\left(\alpha_1^* + \alpha_3^* \right) \left(\alpha_2 + \alpha_4 \right) e^{i(\omega_h - \omega_l)(t+\tau)} + \alpha_1^* \alpha_3 + \alpha_2^* \alpha_4] e^{-\Gamma_c (t+\tau)} \right), \\ \omega_{ex,3} = \omega_{ex0} \left(\tilde{N}_0 + \frac{1}{2} Re[\left(\alpha_1^* + \alpha_3^* \right) \left(\alpha_2 + \alpha_4 \right) e^{i(\omega_h - \omega_l)(t+\tau)} - \alpha_1^* \alpha_3 - \alpha_2^* \alpha_4] e^{-\Gamma_c (t+\tau)} \right), \\ \omega_{ex,4} = \omega_{ex0} \left(\tilde{N}_0 + \frac{1}{2} Re[-\left(\alpha_1^* + \alpha_3^* \right) \left(\alpha_2 + \alpha_4 \right) e^{i(\omega_h - \omega_l)(t+\tau)} - \alpha_1^* \alpha_3 - \alpha_2^* \alpha_4] e^{-\Gamma_c (t+\tau)} \right), \end{cases} \tag{9}$$

$\Gamma_0 = 2\Gamma_R$, $\Gamma_c = 2(\Gamma_R + \Gamma_{NR})$, τ is the delay time between pump and probe pulses,

$$\tilde{N}_0 = \frac{1}{4}\sum_i \left|\alpha_i\right|^2 e^{-\Gamma_0(t+\tau)}. \tag{10}$$

Integrating (8), we can obtain the reflectance of the quantum well

$$r_{QW}(\omega) = \int_0^\infty \sum_j d_j \gamma_j(t) e^{i\omega t} dt. \tag{11}$$

The exciton reflectance can then be obtained using (2).

Results and Discussion

Fig. 2 shows the results of theoretical calculation using the Runge-Kutta method for linearly polarized beams (similar results are also obtained for circular polarization). The reflection spectra of heavy hole exciton are shown in Fig. 2, *a* as the color map. These spectra calculated by using of the Eq. (2) with taking into account the obtained r_{QW} (Eq. (11)) in dependence on the delay between the pump and probe pulses. The reflection amplitude is shown in color. Zero energy shift corresponds to the position of the exciton line without pumping.

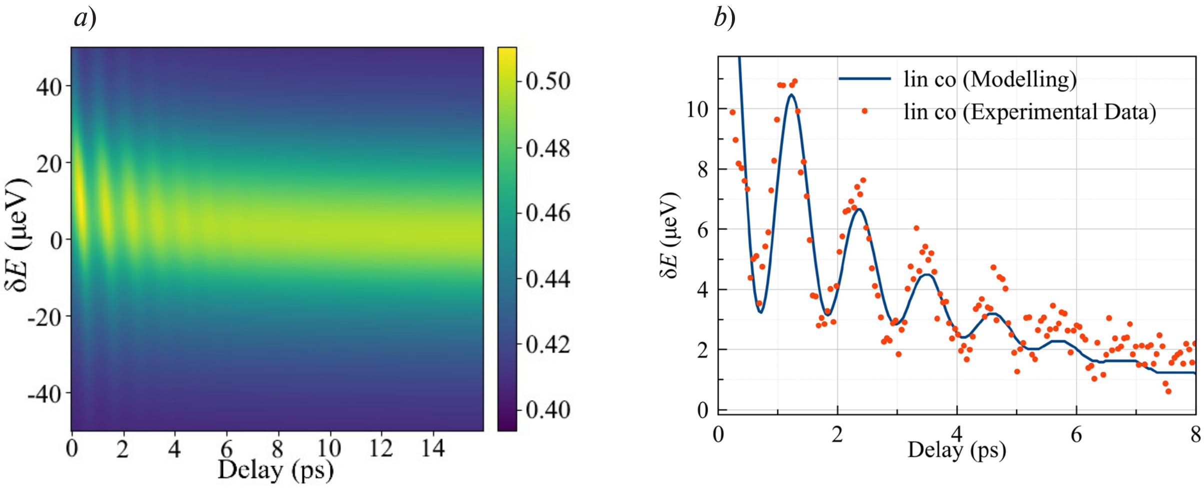

Fig. 2. Modelled reflectance spectrum of heavy hole exciton in dependence on delay between the pump and the probe pulses (*a*). Comparison of the energy shift in the experiment and the theoretical model (*b*)

The energy position of the exciton resonance as a function of the delay is shown in Fig. 2, *b*. The red dots show the experimental data, the blue curve is the simulation result. Calculation parameters such as the splitting between exciton states (it corresponds to $\Omega = 5.47$ ps^{-1}) and the values of radiative and nonradiative broadenings $\Gamma_R = 0.02\Omega$ and $\Gamma_{NR} = 0.04\Omega$ are obtained from the direct fitting of experimental spectra within the framework of the theory of nonlocal dielectric response as described above. The exchange interaction energy was an adjustable parameter and is equal to $\omega_{ex0} = 0.0175\Omega$. There is still a long-lived energy shift in the dynamics of the experimental spectra due to the incoherent interaction with the reservoir of non-radiating excitons. To compare with the experimental dependence, we added this long-lived bias to the calculated curve in Fig. 2, *b*. One can see that the results of theoretical calculations of the coherent exciton dynamics in the frame of our model are in agreement with the experimental data.

Conclusion

The coherent nonlinear dynamics of heavy and light hole excitons in GaAs/AlGaAs quantum well upon coherent excitation of exciton resonances by short laser pulses is investigated experimentally and theoretically. We found delay-dependent oscillations of the exciton resonance energies. Theoretical model for the interaction of excitons with light in a GaAs/AlGaAs quantum well in the pump-probe experiment has been proposed. It was found that the energy shift can only be described if the nonlinearity in the form of the exchange exciton-exciton interaction is taken into account, which has not been considered before. The results of the modeling using the Runge–Kutta method agree with the experimental data. Further study may include the overview of negative delays, where four-wave mixing effects are prevalent, as well as taking "dark" exciton states into account, which are less likely to interact with the pulse and have much lower decay rates.

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