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# Quantum beats of light-hole and heavy-hole excitons in reflection spectra of GaAs/AlGaAs quantum well

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In this work we experimentally study dynamics of the heavy-hole and light-hole exciton states in a quantum well under coherent excitation of both excitonic resonances using co- and cross-linear and circular polarizations of pump and probe beams. Oscillations of basic parameters of excitonic resonances are observed. Their dependence on the polarization of the pump and probe beams is used to determine the structure of states of the heavy-hole and light-hole excitons by means of theoretical modelling of experimental data.

**Keywords:** quantum well, excitons, quantum beats, light-hole exciton, heavy-hole exciton.

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## 1. Introduction

When a multilevel quantum system is excited into a superposition state, quantum beats can be observed. This requires the existence of a common ground state. If the heavy-hole and light-hole excitons in a quantum well form the system with a common state, their quantum beats can be observed in oscillations of the dipole moment of optical transitions. Such oscillations were found experimentally, for example, in signals of four-wave mixing [1,2] and differential transmission [3,4]. However, the exact structure of the states of a system consisting of the heavy-hole and light-hole excitons with different spins has not been elucidated. This leaves open the possibility of alternative explanation of the oscillations by the interference of polarizations, which is observed in the secondary radiation of two independent two-level systems during their coherent excitation. Therefore, some authors explained the oscillations observed in the experiment by interference of polarizations [1], while others did it by quantum beats [2]. Another factor is the interaction of excitons with each other. It is pointed out in the works [5–7] that, in the presence of exciton-exciton interaction, the effect of quantum beats and the effect of polarization interference in four-wave mixing are indistinguishable. An illustrative experiment can be the resonant excitation of one of the exciton states. In the work [8], when studying the differential absorption spectrum, it was shown that, when only a heavy-hole exciton is pumped, the level of a light-hole exciton also shifts. The authors interpret this effect in terms of intersubband coherence in the valence band in quantum well. Thus, questions concerning the nature of the observed oscillations of the signal intensity, i.e., quantum beats or interference of polarizations, as well as about the influence of the exciton-exciton interaction on the observed effect, have not yet been fully resolved.

In this work the experimental pump-probe method with spectral resolution [9,10] and polarization selection is used. This method makes it possible to distinguish the observed effects by analyzing the shape of exciton resonances within the framework of the nonlocal dielectric response model [11]. Theoretical simulation of possible configurations of exciton states makes it possible to determine the real energy scheme of exciton levels from experimental data.

## 2. Experimental results

High-quality heterostructure with a GaAs/AlGaAs quantum well 14 nm wide with small aluminum concentration (3%) in the barrier layers was studied. The structure was grown by molecular beam epitaxy on the GaAs substrate. The sample was placed in a closed-cycle helium cryostat and studied at temperature of 4 K.

Specific feature of the pump-probe method used in this work is a measurement of total reflection spectra, rather than differential reflectance as in earlier studies. Such a method makes it possible to obtain quantitative data on the change in the exciton-light interaction in absolute units. In addition, the measurements were carried out in different polarizations with respect to the polarization of the pump beam, which made it possible to determine the system of levels of the heavy-hole and light-hole excitons.

Fig. 1 shows the block diagram of the experimental setup. For the formation of pump and probe beams, the radiation of a femtosecond titanium-sapphire laser with wide spectrum (the half-width at half-maximum $\sim 10$ nm) covering the resonances of the heavy-hole and light-hole excitons was used. The polarization of the pump beam

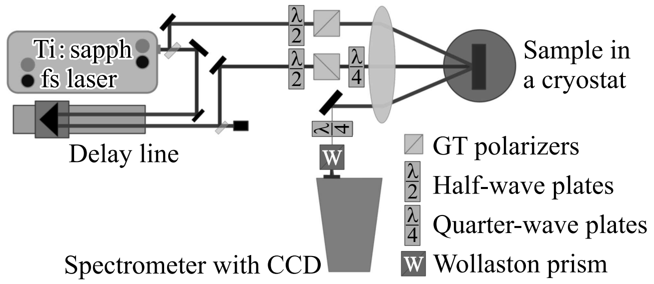


**Figure 1.** Scheme of the experimental pump-probe setup with spectral resolution and polarization selection.

was set to be linear or circular. The polarizer axes in the probe beam were set at 45° relative to the polarizer axes in the pump beam. Thus, the linear polarization of the probe beam could be represented either as the sum of co- and cross-linear polarizations for the linear pump polarization, or co- and cross-circular polarizations for the circularly polarized pumping. Reflection spectra were detected in various polarizations using the quarter-wave plate and the Wollaston prism. During the measurements in circular polarizations, this system divided the reflected beam into two beams with orthogonal circular polarizations, and during the measurements in linear polarizations, two orthogonal linear polarizations. Both beams were focused on the monochromator slit and, after decomposition into a spectrum, were simultaneously recorded by different parts of the CCD-matrix.

The measured spectra were analyzed within the framework of the theory of nonlocal dielectric response, and such characteristics of the heavy-hole and light-hole exciton states as the resonance energy, the radiative and nonradiative broadening were extracted from the spectra. The basic formulas used for fitting of the spectra have the form [11]:

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

$$R(\omega) = \left| \frac{r_s + r_{QW}(\omega)e^{i2\phi}}{1 + r_s r_{QW}(\omega)e^{i2\phi}} \right|. \quad (2)$$

Here $r_{QW}(\omega)$ describes the amplitude reflection from the exciton resonance, $\Gamma_R$ and $\Gamma_{NR}$ are the rates of the radiative and non-radiative decay of the exciton polarization, $\omega_x$ is the frequency of the exciton transition, $\phi$ is the phase acquired by the light wave on the path 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 reflection from the sample, taking into account the resonant exciton reflection.

The analysis of reflection spectra by fitting exciton resonances by formulas (1) and (2) makes it possible to determine all 4 exciton resonance parameters, $\Gamma_R$, $\Gamma_{NR}$, $\omega_x$ and $\phi$ [9,10,12–15]. In this work, we investigate the rate of radiative decay $\Gamma_R$ of the heavy-hole and light-hole excitons at each delay step between the pump and probe pulses. The dependence of $\Gamma_R$ on the delay makes it possible to observe its dynamics.

Fig. 2 shows the characteristic reflection spectrum of the sample in the absence of pumping. One can see exciton resonances with light and heavy holes in the form of reflection peaks. This form of resonances is the result of special design of the sample structure. The figure also shows the curve fitting the resonances, which demonstrates very good description of the experimental spectra by formulas (1), (2). In addition, the figure shows the spectrum of the laser pulses. Its center is shifted to the region of lower energies in order to reduce the probability of excitation of transitions between free electrons and holes, as well as the probability of creation of excitons in barriers by the high-frequency edge of the laser spectrum. Such transitions lead to filling of the reservoir of non-radiating excitons and free carriers [15], which reduces the amplitude of quantum beats.

Figure 3 shows the dynamics of radiative broadenings $\hbar\Gamma_R$ extracted from the reflection spectra of excitons with

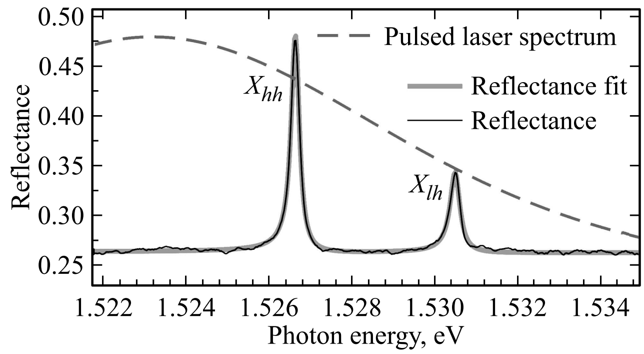


**Figure 2.** Reflection spectrum of the studied sample in the region of resonances of the heavy-hole ($X_{hh}$) and light-hole ($X_{lh}$) exciton and fitting of the spectrum by formulas (1) and (2). The dashed line shows the emission spectrum of the laser.

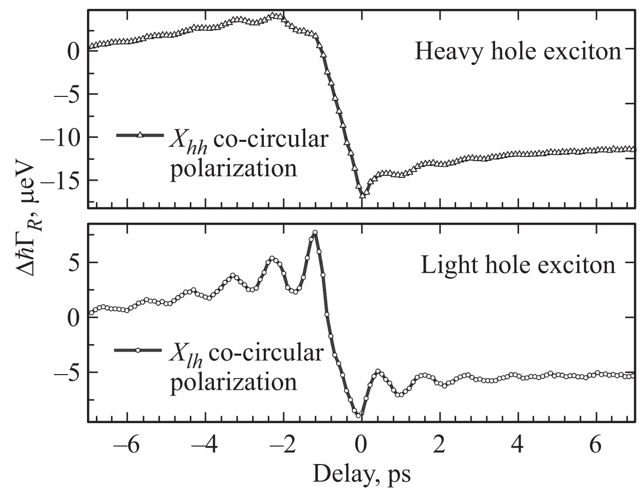


**Figure 3.** Dynamics of the photoinduced change in the radiative broadening of excitons with heavy holes (upper panel) and light holes (lower panel) in co-circular polarizations. Sample temperature $T = 4\,\text{K}$. Pumping power density $P = 30\,\text{W/cm}^2$.

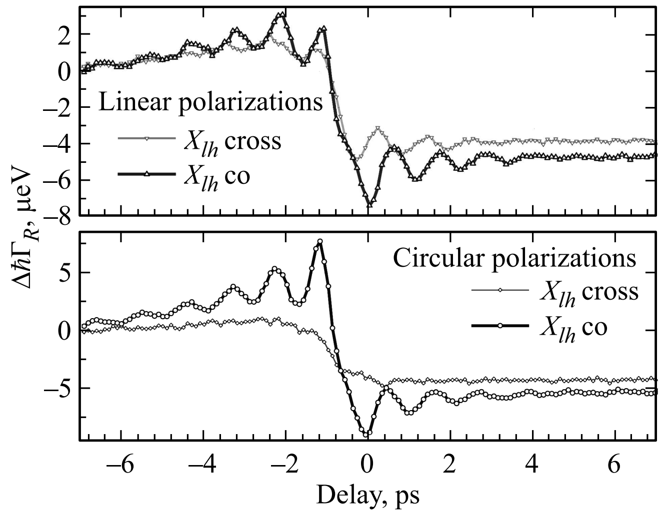


**Figure 4.** Dependence of the photoinduced change in the radiative broadening of the light-hole exciton on the time delay for linear (upper panel) and circular polarizations (lower panel). Curves for co- and cross-polarization of detection are shown. Sample temperature $T = 4$ K. Pumping power density $P = 30$ W/cm$^2$.

heavy and light holes in co-circular polarization. Only the part of the radiative broadening induced by the optical pumping is shown. In the absence of pumping, the radiative broadenings are equal to $\hbar\Gamma_{hh} = 30$ mkeV for the exciton with heavy holes and $\hbar\Gamma_{lh} = 15$ mkeV for the exciton with light holes. As can be seen from the figure, the broadening dynamics has a rather complex form. At small negative delays, the slight increase in the broadening is observed, then the broadening sharply decreases in the region of near-zero delays. The experiment shows that the decrease in the radiative broadening, which means the bleaching of the optical transition, persists for a long time (nanoseconds). This effect requires a separate study. Against the background of the indicated broadening behavior, oscillations of $\hbar\Gamma_R$ are clearly observed. We can interpret these oscillations as a quantum beats signal. Their frequency corresponds to the energy splitting between the states of excitons with light and heavy holes.

Fig. 4 shows the result of processing the individual reflection spectra as a function of the time delay for the radiative broadening of the light-hole exciton in various polarizations. It is important to note that the quantum beats are observed in all combinations of pumping and probing polarizations, except for the cross-circular polarization. Such a set of the experimental data, together with the corresponding data for the heavy-hole exciton, makes it possible to separate the effect of quantum beats from the effects of exciton-exciton interaction, the overlap of which was a problem for previous studies on this topic.

## 3. Simulating

We perform a theoretical study of the dynamics of coherent excitation of exciton states with the light and heavy holes. The purpose of the simulation is to explain the polarization properties of the observed oscillations of the radiative broadening of exciton resonances. The simulation is carried out in a semiclassical model, when the electromagnetic wave is described by the Maxwell's equations, and the excited exciton system is described by a quantum-mechanical way.

The action of two laser pulses of low intensity and short duration on an exciton system is considered. The low pulse intensity means a weak perturbation of the system, which can be described in terms of successive approximations according to perturbation theory. The short duration of the pulses allows us to not consider the relaxation of the system during the action of the pulse, which corresponds to the experimental conditions.

The action of a light pulse on the system is described by the optical Bloch equations. Several model configurations of levels of the exciton system with different initial states are considered. It is assumed in the calculation that the excited states are always four exciton levels with heavy and light holes, $|\text{exc}, \pm 3/2\rangle$, $|\text{exc}, \pm 1/2\rangle$. For the ground states, the following options are considered (see the level diagrams in Fig. 5):

– Diagram ($a$), the following basic states are pairwise mixed: $|+3/2\rangle$ and $|-3/2\rangle$; $|+1/2\rangle$ and $|-1/2\rangle$.

– Diagram ($b$), the following basic states are pairwise mixed: $|+3/2\rangle$ and $|+1/2\rangle$; $|-3/2\rangle$ and $|-1/2\rangle$.

– Diagram ($c$), there is one basic state (no excitons).

– Diagram ($d$), there are four independent ground states for an electron in the valence band.

For each of the diagrams, the von Neumann equation is solved for the density matrix $\rho$ in the interaction representation:

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}[V, \rho]. \quad (3)$$

The matrix of interaction with light $V$ consists of off-diagonal elements corresponding to allowed transitions in the system under study. If the transition $j-k$ is allowed, the corresponding perturbation matrix element has the form

$$V_{jk} = -d_{jk}E_{jk}(t)e^{-i(\omega-\omega_{jk})t}, \quad (4)$$

where $d_{jk}$ is the dipole moment, $E_{jk}(t)$ is the electromagnetic wave envelope function, $\omega_{jk}$ is the transition frequency, and $\omega$ is the carrier frequency of the electromagnetic wave.

The optical polarization of an exciton system is the sum of the off-diagonal elements:

$$P = Tr(d\rho) = \sum_{j,k} d_{jk}\rho_{jk}. \quad (5)$$

The simulation shows that the ($a$) and ($d$) diagrams in Fig. 5 do not give quantum beats between the states of

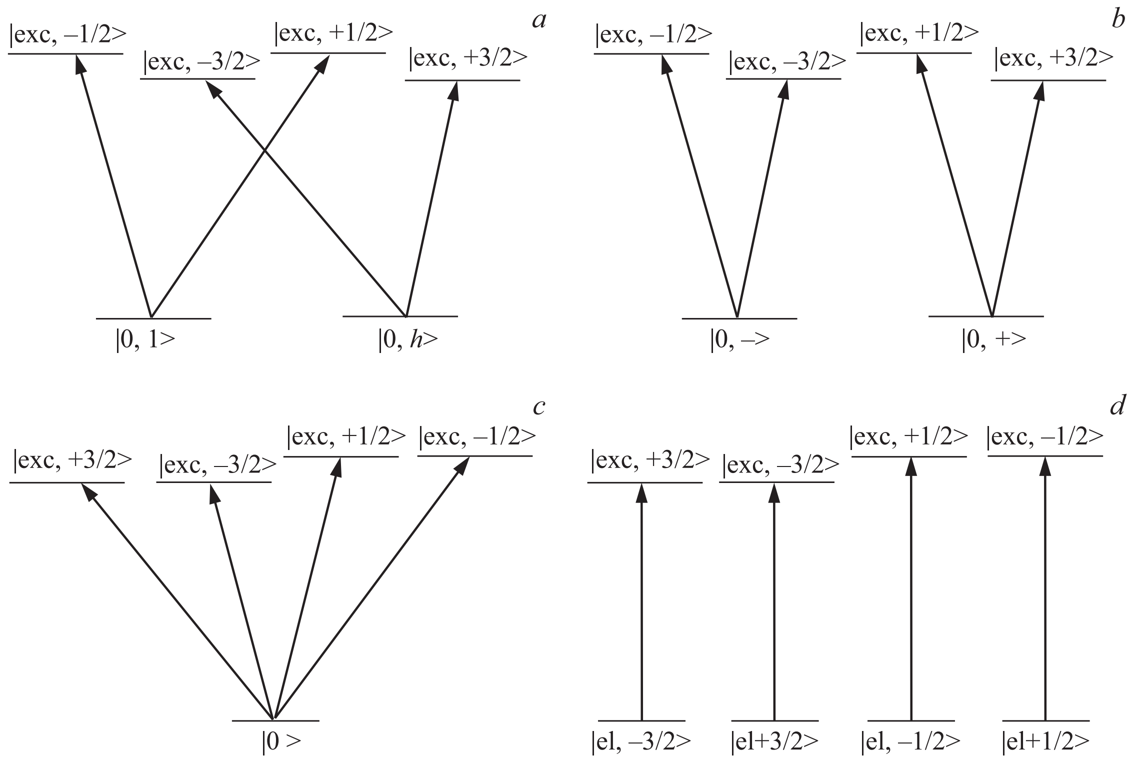


**Figure 5.** Level diagrams for theoretical simulating (see explanations in the text).

the heavy-hole and light-hole excitons (there is no common ground state). For the $(b)$ diagram, the circular components of the optical polarization at one of the exciton transitions have the form:

$$P_+ \sim |E^+_{pu}|^2 E^+_{pr} e^{-i\Delta_{hl}t} + c.c., \tag{6a}$$

$$P_- \sim |E^-_{pu}|^2 E^-_{pr} e^{-i\Delta_{hl}t} + c.c. \tag{6b}$$

Here $E^{\pm}_{pu}$ and $E^{\pm}_{pr}$ are the circular components of the pump and probe light pulses, $\Delta_{hl}$ is the splitting between the states of excitons with heavy and light holes. Quantum beats for the $(b)$ scheme are possible in all combinations of pump and probe polarizations, except for cross-circular polarization.

For the $(c)$ diagram, the polarization components have the form:

$$P_+ \sim |E^+_{pu}|^2 E^+_{pr} e^{-i\Delta_{hl}t} + E^+_{pu}(E^-_{pu})^* E^-_{pr} e^{-i\Delta_{hl}t} + c.c., \tag{7a}$$

$$P_- \sim |E^-_{pu}|^2 E^-_{pr} e^{-i\Delta_{hl}t} + E^-_{pu}(E^+_{pu})^* E^+_{pr} e^{-i\Delta_{hl}t} + c.c. \tag{7b}$$

The $(c)$ scheme gives rise to the beats for the circular and linear co–polarized pumping and probing, and no beats for any cross-polarization.

Thus, agreement with the experiment turned out to be possible only for one of the considered diagrams — are diagram $(b)$, where the ground states are pairwise mixed states: $|+3/2\rangle$ with $|+1/2\rangle$, $|-3/2\rangle$ with $|-1/2\rangle$.

## 4. Conclusion

The dynamics of the radiative broadening of excitons with the light and heavy holes in the 14-nm GaAs/AlGaAs quantum well under their coherent excitation has been studied by the „pump–probe“ method with spectral resolution. Oscillating component of the radiation broadening of both excitons is found in different polarization configurations of the experiment. The oscillation frequency corresponds to the splitting value of the exciton states. The observed effect is interpreted as a manifestation of quantum beats of the states of excitons with light and heavy holes. The experiment shows that the quantum beats appear in co-circular and co- and cross-linear polarizations, but they are not observed in cross-circular polarization.

Comparing data of the pump–probe experiments with the simulation results for various schemes of levels of the heavy-hole and light-hole excitons, we have found that the calculation for one of the frequently used configurations with common initial state (scheme $(c)$ in Fig. 5) does not explain the experiment. Agreement with experiment turned out to be possible only with the initial pairwise mixed states of light and heavy holes ($|+3/2\rangle$ with $|+1/2\rangle$ and $|-3/2\rangle$ with $|-1/2\rangle$).


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### Conflict of interest

The authors declare that they have no conflict of interest.

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