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Multiple-frequency quantum beats of quantum confined exciton states

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Multiple frequency quantum beats of a system of the coherently excited quantum confined exciton states in a high-quality semiconductor structure containing a wide InGaAs/GaAs quantum well are experimentally detected by the spectrally resolved pump-probe method. The beat signal is observed both at positive and at negative delays between the pump and probe pulses. Several quantum beat (QB) frequencies are observed in the experiments, which coincide with the interlevel spacings in the exciton system. A theoretical model is developed, which allows one to attribute the QBs at negative delay to the four-wave mixing (FWM) signal detected at the nonstandard direction. The beat signal is strongly enhanced by the interference of the FWM signal with the secondary emission induced by the probe pulse. At positive delays, the QBs are due to the interference of the quantum confined exciton states. The decay time for QBs is of the order of several picoseconds both at positive and negative delays. This is close to the relaxation time of the exciton population that allows one to consider the exciton depopulation as the main mechanism of the coherence relaxation in the system under study.

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Optical quantum beats (QBs) are oscillations in the optical response of a quantum system that appear if two or several excited states of the system are mutually coherent [1–3]. QBs can be detected as oscillations of intensity of the emitted or absorbed light induced by a short pulse of light that coherently excites the system in a superposition of quantum states.

In semiconductors, QBs have been observed for discrete energy states of excitons interacting with light [4]. The majority of experiments have been devoted to QBs of heavy- and light-hole excitons [5–9] or QBs between spin states of excitons or free carriers [10–17]. Semiconductor quantum wells (QWs) represent a convenient model system for studies of QBs between confined exciton states. Transitions between excited electronic and excitonic states in QWs are exploited for terahertz lasing [18–20].

From the fundamental point of view, QWs offer an opportunity of band engineering and allow for studies of the multilevel coherences. QBs in multilevel systems have been theoretically described in Refs. [21,22]. A multilevel system of Landau level magnetoexcitons has been experimentally studied in Ref. [23] while only QBs between two lowest energy levels have been observed. No experimental evidence of multifrequency QBs in QWs has been reported so far, to the best of our knowledge.

In this Rapid Communication, we report on the pump-probe optical study of time evolution of a coherently excited system of several quantum confined exciton states in a high-quality InGaAs/GaAs QW. We have found that the excitation of several exciton states by a short laser pulse gives rise to multifrequency oscillations of the time-resolved reflection intensity both for positive and negative delays between pump and probe pulses. The density matrix model allows for attributing of these oscillations to QBs of several coherently coupled exciton states.

The sample studied represents a single 95-nm $In_{0.02}Ga_{0.98}As$/GaAs QW grown by molecular beam epitaxy. The photoluminescence spectrum of this structure reveals a number of resonances related to the quantum confined exciton states in the QW (left inset in Fig. 1). The detailed optical characterization of the structure can be found in Ref. [24].

The kinetics of the secondary emission of this QW has 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. The 1.7-ps pump pulses were spectrally selected from 100-fs laser pulses, so that they have additional spectral maxima. The spectral shape of the pump pulses can be well described by function of type $(\sin x/x)^2$ (see Ref. [24] for details). The main peak of the pump intensity has been tuned to the third and fourth exciton energy levels in the QW. The spectrum of femtosecond probe pulses was much broader than the pump ones and covered all the studied exciton transitions with almost equal amplitude. The modulated reflectivity of the probe pulse has been spectrally resolved in a 0.5-m spectrometer and detected by a photodiode connected with a lock-in amplifier and computer.

The exciton secondary emission dynamics for the spectral range of four lowest energy exciton transitions is shown in Fig. 1. The maximum of the emitted intensity corresponds to the fourth exciton energy level, which is most efficiently excited by the pump beam. Figure 1 shows the nontrivial dependence of the amplitudes of exciton spectral resonances on the pump-probe delay. The peak intensities exhibit rapidly decaying oscillations superimposed with the slowly varying background signal. The slow component of the signal is due to the long-lived reservoir of dark excitons (see Ref. [24] for details). Here we will discuss the oscillating component that is characteristic of exciton QBs. It is important to note that the oscillations are observed both at positive and negative pump-probe delays, which is unusual.

Figure 2 shows the reflectance kinetics for several exciton transitions as well as the corresponding Fourier spectra. The

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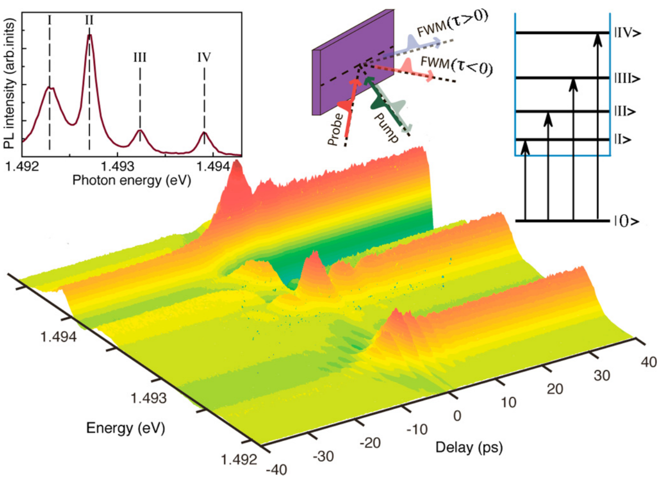

FIG. 1. (Color online) Spectrally resolved kinetics of pump-probe signal under coherent excitation of four exciton states. Left inset: photoluminescence spectrum of the heterostructure with four exciton resonances (I–IV). Middle inset: geometry of experiment. Right inset: a simplified scheme of the exciton transitions.

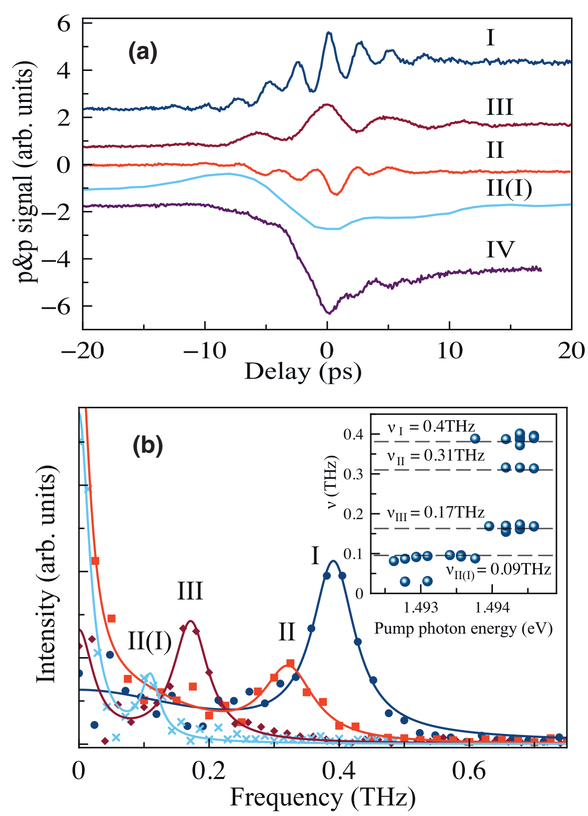

FIG. 2. (Color online) (a) Examples of reflectance kinetics detected at different spectrally selected exciton transitions noted near each curve. Curves I, II, III, and IV are detected under predominant excitation of state IV. Curve II(I) is detected when only I and II states are excited. (b) Fourier analysis of the kinetics. Solid lines are the fits by Lorentzians. Inset shows the frequencies of QBs obtained from the experiment (solid balls) and expected beat frequencies obtained from the energy spacing between the exciton states (dashed lines).

detected frequencies of QBs measured at the spectral maxima correspond to the energy spacing between respective exciton levels and level IV [see inset in Fig. 2(b)]. The oscillating component in the signal detected at the optical frequency corresponding to level IV is relatively weak. However, its Fourier analysis allows one to reliably extract the QB frequency and to attribute it to the beats of the I and IV exciton states. In the case of a coherent excitation of only I and II exciton states by the pump beam, QBs with lowest frequency, $\nu_{12} = 0.094$ THz, have been observed.

A general theoretical analysis [4] shows that the oscillations in a transient response of a multilevel quantum system may be caused by one of two competing processes. The first one is related to the optical interference of polarizations created by the coherent excitation of the *independent* quantum systems. The oscillations are observed while the *polarization coherence* at optical frequencies is saved. The second process is the QBs of coherently excited states of the *single* quantum system. In this case, the polarization coherence is not required and the QBs are observed while the *mutual* coherence of excited states persists.

Discrimination of these two processes is a challenging problem [25,26]. Pump-probe experiments, however, allow one to identify the processes because the interference of polarizations created by the pump beam in independent quantum systems is not detected in these experiments. On the contrary, the QBs of states in the single quantum system can be detected because, once the mutual coherence of excited states is created, the probability of optical transition to the ground state of the quantum system is an oscillating function of time [27–29]. These oscillations of the probability give rise to the beating signal observed in kinetics of photoluminescence [1,2] and of modulated reflectance [9]. For this reason, the oscillations observed in our experiments can be definitely treated as QBs of quantum confined exciton states in a*single* quantum system.

A theoretical analysis shows that, once the coherent superposition of several exciton states is prepared, the beating signal detected in the reflected probe beam direction consists of several components for each particular exciton transition j:

$$I_{pp}(\omega_j) \sim \sum_{k\neq j} |d_k|^2 \cos(\omega_{jk}\tau) e^{-\tau/T_{jk}} \tag{1}$$

[see Eq. (12) in the Supplemental Material [30]], where d_k is the dipole moment of exciton transition $|0\rangle \rightarrow |k\rangle$ and T_{jk} is the decay time of mutual coherence of the j and k exciton states. Each component k contains an oscillating function of time delay, τ, between the pump and probe pulses. The oscillation frequency, $\omega_{jk} = (\varepsilon_j - \varepsilon_k)/\hbar$, is governed by the energy spacing between the states j and k, which may be detected in the reflectivity or transmission experiment.

The detailed theoretical model describing the observed QBs is presented in the Supplemental Material. According to the supplementary Eq. (12), the contribution of each component depends on the energy spectrum of exciting pulses. Indeed, when the IVth quantum confined exciton state has been predominantly excited in the experiment (see Fig. 2), the QB frequencies obtained from the pump-probe signal at the frequencies of excitonic transitions I–III were governed by the energy difference between the corresponding exciton state and state IV. At the same time, the QBs obtained at the IVth exciton transition are mainly affected by the interference of states IV and I, because the optical transition from state I is characterized by the largest oscillator strength [24].

The above discussion has been focused on QBs observed at the positive delays, i.e., if the probe pulse comes to the sample after the pump pulse. However, as one can see in Fig. 2(a), the QBs are clearly present also at the negative delays, i.e., when the probe pulse precedes the pump one. This phenomenon is unusual and needs to be carefully studied. One of possible mechanisms of QBs at the negative delay has been proposed in Ref. [31]. It assumes that the probe pulse creates an oscillating polarization due to coherent excitation of several exciton states. The strong enough pump pulse coming with some delay may destroy the coherence because it generates new excitons and free carriers. This coherence breaking gives rise to the steplike decrease of the secondary emission intensity in the direction of the reflected probe beam, which results in the signal oscillating as a function of the pump-probe time delay [28]. However, as our analysis shows, oscillations in the detected signal should be weak in this case.

We propose a different formation mechanism for the oscillating signal at the negative delays. We argue that it appears due to the diffraction of the secondary emission induced by the pump pulse on the population grating created by the joint action of the probe and pump pulses. This represents a four-wave mixing (FWM) signal that can be detected in the direction of the reflected probe beam. We should mention that the standard direction for the FWM detection is determined by $2k_1 - k_2$, where k_1 and k_2 are the projections of wave vectors of the pump and probe beams on the QW plane, respectively [4] (see middle inset in Fig. 1). We have used the nonstandard direction (determined by k_2) for the FWM detection, which brings an important advantage over the standard direction of the FWM observation, as we discuss below.

To verify the assumptions formulated above, we have studied the dependence of the detected signal on the pump and probe intensities. The FWM signal detected in the standard direction is expected to depend linearly on the intensity of the pulse coming first and to depend quadratically on the intensity of the delayed pulse [4]. At first glance, one would expect the similar behavior of the FWM signal detected also in the nonstandard direction. However, the experiment demonstrates a linear dependence of the FWM signal on both the pump and probe powers. This tendency has been confirmed by varying the pump and probe by one order of magnitude (Fig. 3). The further increase of pump and probe intensities gives rise to a rapid decay of QBs, most probably due to the accumulation of nonradiative excitons [24], so that the beat amplitude cannot be reliably determined.

To understand the obtained seeming contradiction, we should take into account the supplementary contribution to the signal detected in the nonstandard direction that comes from the secondary emission of excitons generated by the probe beam. It is a relatively intense signal, which interferes with the weaker FWM signal so that the total detected intensity $I_{pp} \propto |E_{pu}|^2 |E_{pr}|^2$ (see Supplemental Material), where E_{pu} and E_{pr} are the pump and probe amplitudes, respectively. The interference of the FWM signal with the reflected probe explains the linear dependencies of the detected signal on the pump and probe intensities.

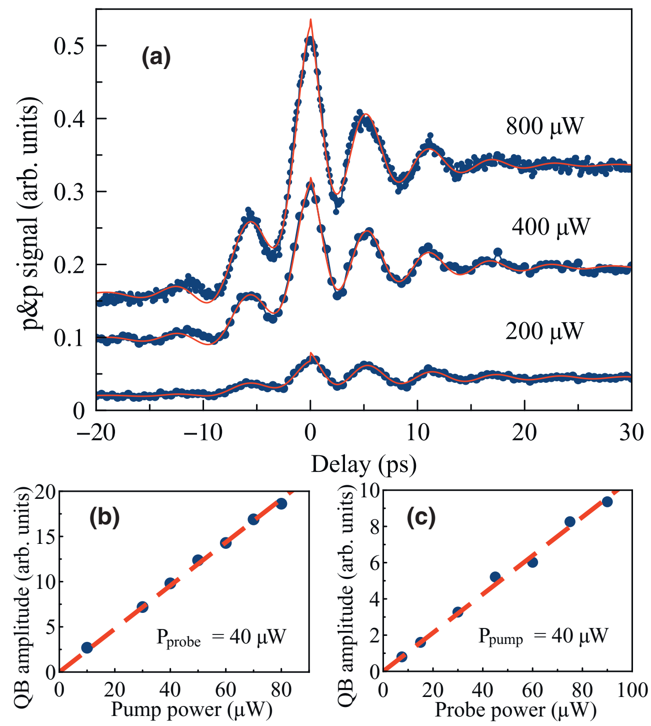

FIG. 3. (Color online) (a) Pump-probe signals measured at the IIIrd exciton transition at different pump powers given near each curve. $P_{probe} = 40\ \mu$W. The observed QBs are due to the interference of the IIIrd and IVth exciton states. Dashed lines are the fits by function 2 superimposed on a smooth function describing the background signal. (b) and (c) show the dependencies of beat amplitude, Q_0, on the pump and probe powers, respectively.

The experimentally detected time evolution of the QBs allows us to directly estimate the decay time of the mutual coherence of quantum confined exciton states. In order to obtain it, we fit the oscillating component of the signal by a function

$$Q(\tau) = Q_0 \cos(\omega\tau) \exp(-|\tau|/T_{jk}). \quad (2)$$

The decay times obtained in this way are $T_{\mathrm{I-IV}} = 3.5 \pm 1$ ps for the Ist exciton state and $T_{n-\mathrm{IV}} = 7 \pm 2$ ps for all other states. These values appear to be in good agreement with those obtained from the analysis of the line broadening studied in Ref. [24]. The decay constant for each exciton transition is found to be the same for positive and negative delays within the experimental accuracy.

The latter result is nontrivial because, as discussed above, the signals observed at positive and negative delays are formed by different processes. The QB decay in the pump-probe signal (at positive delay) is controlled by the relaxation of the *mutual* coherence of excitonic states, whereas the decay in the FWM signal (negative delay) is due to the relaxation of *polarization* coherence at the optical frequency. Typically, the optical frequency dephasing is the fastest process in a quantum system controlled by interaction of light with different quasiparticles (nonradiative excitons, carriers, phonons) as well as by the inhomogeneous broadening of the quantum ensemble [4].

At the same time, the mutual coherence of exciton states is expected to survive much longer and eventually decay with the population relaxation time.

Almost total coincidence of the decay times for positive and negative delays points out that, in the structure under study, the decays of mutual (quantum) coherence and of the coherence of exciton transitions are dominated by the same processes. Since the QB decay times are close to the exciton depopulation time, $\tau \approx 6.5$ ps, reported in Ref. [24] for this QW structure, we may conclude that the decoherence of exciton states is governed by the exciton population decay. The rapid depopulation of exciton states is mainly due to the high rate of exciton radiative recombination.

In conclusion, the experimental study of exciton dynamics in a high-quality heterostructure with a wide InGaAs/GaAs QW allowed us to detect a new type of QBs, which are due to the quantum interference of several quantum confined exciton states in the QW. The beat signal is detected both at positive pump-probe delays and at negative ones. A model describing the QBs of coherently excited multiple quantum states is developed. The theoretical analysis shows that the QBs at negative delays are observed due to the FWM effect detected in the nonstandard direction of the reflected probe beam. The amplitude of the secondary emission is strongly enhanced by the interference of the FWM signal with the reflected probe pulse. Surprisingly, the decay time of QBs detected at the positive and negative delay is the same within our experimental accuracy although the relaxation mechanisms seem to be different in these two regimes. At positive delays, the decay time is governed by the relaxation of the mutual coherence of exciton states, while at negative delays the decay is caused by the dephasing of optical waves. The origin of this unexpected coincidence is in the high quality of the QW structure under study. In our structure, the depopulation of exciton states due to radiative recombination and scattering constitutes the main mechanism of the coherence relaxation.

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