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Correlation effect of Rabi oscillations of excitons in quantum dots

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Abstract

We performed a transient four-wave mixing experiment on a strain-compensated InAs quantum dot (QD) ensemble over a wide range of excitation intensities. Under the resonant excitation of an exciton ground state, an extremely long dephasing time of 1 ns was found. By increasing the areas of the excitation pulses, Rabi oscillations of excitonic polarizations were clearly observed. The corresponding Rabi frequency is three orders of magnitude higher than the measured dephasing rate. For larger pulse areas, we found that the deviation of experimental data from two-level predictions became significant. The deviations cannot be explained by taking into account, as has been suggested in other research, excitation density-dependent dephasing or Hartree–Fock-type Coulomb interactions between excitons.

Keywords: Quantum dots; Rabi oscillation; Four-wave mixing spectroscopy

1. Introduction

Optical Rabi oscillations (ROs) play a crucial role in processing and storing quantum information in two-level systems [1]. Semiconductor quantum dots (QDs) are promising solid-state candidates for use in quantum information devices due to their atomic-like density of states of excitons. Moreover, QD excitons provide an opportunity to investigate a wide variety of ROs, since QD excitons often cannot be treated as an ideal two-level system.

Recently, excitonic ROs have been demonstrated experimentally in QDs using interferometric photoluminescence [1–3], transient pump–probe [4,5] and four-wave mixing (FWM) [6,7] spectroscopies. The most previous experiments were performed on single QDs [1–4,6] to demonstrate a coherent manipulation of a single exciton state. On the other hand, there is an interest in investigating ROs in QD ensembles because the phenomenon has important applications in quantum devices, such as scalable quantum computers, self-induced transparency solitons, and photon echo optical memories. However, coherent interactions between a QD exciton and light become difficult to observe due to a large inhomogeneous broadening of excitonic transition energies in QD ensembles [5,7]. Therefore, there have been only a few experimental demonstrations of excitonic ROs in a QD ensemble [5,7].

We have investigated excitonic ROs in QD ensembles using a transient FWM technique to simultaneously excite a large number of QDs. The sample contained multilayer-stacked InAs QDs fabricated using a strain-compensation technique [8]. They showed an extremely long exciton dephasing time, as reported in our papers [9–11]. We observed clear ROs as pulse area-dependent FWM intensities and a significant deviation of the ROs' characteristics from the predictions for an ideal two-level system.

2. Experiment

A sample was composed of a stack of 150 layers of InAs self-assembled QDs embedded in 60-nm-thick $In_{0.52}Ga_{0.1}Al_{0.38}As$ spacers grown on an InP(3 1 1)B substrate. The sample was fabricated to compensate for a QD strain by tailoring the composition of the spacer layer as well as using an InP(3 1 1)B substrate instead of a GaAs substrate [8]. The strain compensation enabled us to stack over a

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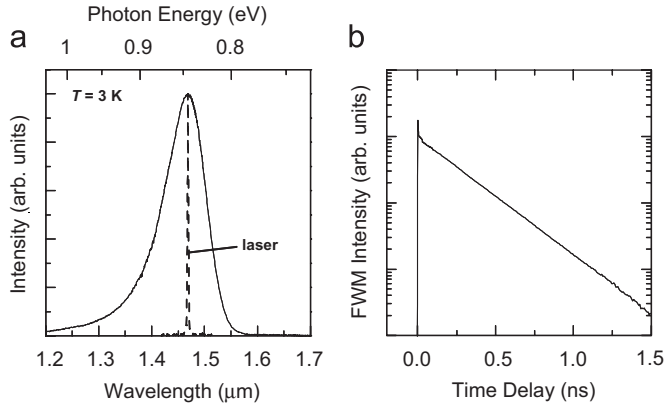

Fig. 1. (a) Solid line: Photoluminescence spectrum at 3 K under nonresonant laser excitation. Dashed line: Spectrum of excitation pulses used in FWM experiment. (b) FWM intensity measured as a function of time delay τ between two excitation pulses. Pulse areas are $\Theta_1 = \pi/2$ and $\Theta_2 = \pi$, respectively.

hundred QD layers, thereby to enhance FWM signal intensity [9]. The area density of QDs per single layer was $9 \times 10^{10}\,\mathrm{cm}^2$/layer.

Fig. 1 shows a photoluminescence spectrum at 3 K under nonresonant excitation. The photoluminescence attributed to an exciton ground-state transition is observed at 1.468 μm with an inhomogeneous broadening of 44 meV.

A two-pulse FWM experiment in transmission geometry was performed using 1.1 ps optical pulses at a 76-MHz-repetition rate emitted by a Ti:S-pumped optical parametric oscillator. The FWM intensity I_{FWM} in the $2\boldsymbol{k}_2$–$\boldsymbol{k}_1$ direction was measured as a function of time delay τ or pulse areas Θ_i ($i = 1, 2$). The pulse areas were estimated by measuring excitation intensity and using the transition dipole moment (58 debye) calculated from the radiative lifetime (1.01 ns) [11]. The radius of both pulses was approximately 86 μm. In the FWM experiment, an exciton ground state was resonantly excited by a narrowband pulse with a central wavelength of 1.468 μm (see the dashed line in Fig. 1). Due to the narrower pulse bandwidth, a maximum of 2% of the QDs in the irradiated area (corresponding to 10^7 QDs) was resonantly excited. The polarization directions of excitation pulses were aligned in the $[\bar{2}\,3\,3]$ direction to selectively excite either of the two nondegenerated exciton ground states [12]. All measurements were performed at 3 K.

3. Results and discussion

Fig. 1(b) shows FWM intensity, I_{FWM}, measured as a function of τ at $\Theta_1 = \pi/2$ and $\Theta_2 = \pi$. The excitation density corresponding to a π pulse was $0.6\,\mu\mathrm{J/cm}^2$/pulse. The observed signal decayed with a long time constant, which indicates an extremely long dephasing time of QD excitons [9,13]. The corresponding dephasing time is approximately 1 ns. This is three orders of magnitude longer than the pulse width that meets the requirement to observe an RO. The spike-like signal observed for $\tau < 20$ ps was attributed to the two-photon coherence between the crystal ground state and the biexciton state. This signal rapidly decayed due to the biexcitons' relatively fast dephasing time and the dispersion of their binding energies. The dephasing of the biexcitons did not affect the ROs of excitonic polarization that we investigated.

In FWM, an RO of excitonic polarization can be observed as a sinusoidal oscillation of I_{FWM} with a pulse area that follows the relationship $I_{\mathrm{FWM}} \propto \sin^2(\Theta_1)\sin^4(\Theta_2/2)$, assuming that QD excitons can be treated as an ideal two-level system [6]. Fig. 2 shows the measured I_{FWM} as a function of the pulse area Θ_1, when $\Theta_2 = \pi$ and $\tau = 20$ ps. The solid line in Fig. 2 represents the wave generated by $\sin^2(\Theta_1)$, whose amplitude is adjusted to reproduce the first maximum I_{FWM} at $\Theta_1 = \pi/2$. For $\Theta_1 < 0.7\pi$, the measured I_{FWM} is in an excellent agreement with the two-level prediction in spite of the fact that the fitting parameter in the calculation is only an amplitude. Therefore, the observed oscillation of I_{FWM} is due to an RO but not caused by saturation. To our knowledge, this is the first observation of excitonic ROs in the optical communication wavelength region. The corresponding Rabi frequency (~THz) is three orders of magnitude higher than the measured dephasing rate (refer to Fig. 2). For $\Theta_1 > 0.7\pi$, the deviation of amplitude from predictions becomes significant while the oscillation period remains in agreement with the predictions. Such deviation has been previously observed in pump–probe experiments and was interpreted as an increased dephasing rate induced by an optically excited carrier density [4]. However, in our experiment, the measured dephasing time does not change with increasing Θ_1, as shown by the filled triangles in

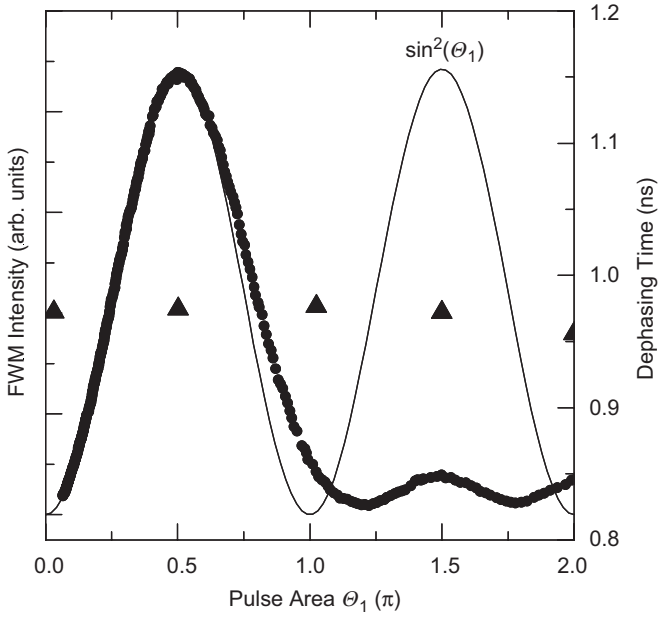

Fig. 2. Filled circles: FWM intensity at $\tau = 20$ ps measured as a function of pulse area Θ_1. Θ_2 is fixed to π. Solid line: Expected two-level result $\propto \sin^2(\Theta_1)$. Filled triangles: Dephasing time of exciton state estimated from decay time constant of FWM intensity.

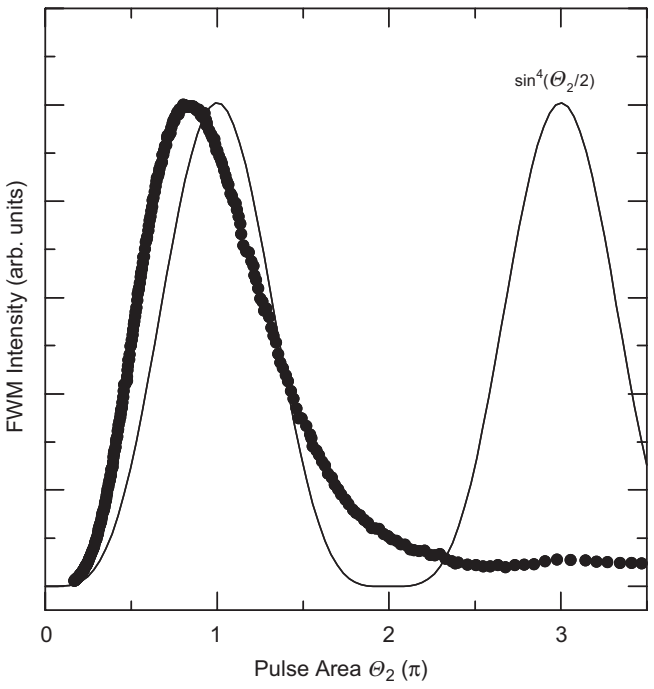

Fig. 3. Filled circle: FWM intensity at $\tau = 20$ ps measured as a function of pulse area Θ_2 where $\Theta_1 = \pi/2$. Solid line: Expected two-level result $\propto \sin^4(\Theta_2/2)$.

Fig. 2. This means that another reason, such as correlation effects of excitons, should be considered as a cause of the deviation.

Next, we measured I_{FWM} at various Θ_2's when Θ_1 and τ were fixed at $\pi/2$ and 20 ps, respectively. Fig. 3 clearly indicates that the relationship $\mathrm{RO} \propto \sin^4(\Theta_2/2)$ exists and that there are deviations from the predictions. The deviations are more significant than those illustrated in Fig. 2. This implies that the effective Θ_2 is influenced by excitons or carriers photogenerated by an initial incident pulse. Previous studies of excitonic ROs in quantum wells showed that Hartree–Fock-type Coulomb interactions between excitons result in density-dependent Rabi frequencies [14]. Therefore, in our experiments, the Rabi frequency was expected to increase with increasing excitation density based on these interpretations; however, the contrary occurred. At present, the mechanism causing the deviation from the two-level prediction for our QD remains unresolved. The identification of the underlying mechanism will play a crucial role in the future development of quantum information technology in semiconductors.

4. Conclusions

We observed clear excitonic ROs in strain-compensated InAs QDs using FWM techniques under resonant excitation of an exciton ground state. The observed ROs showed deviations from predictions for an ideal two-level system. The deviations became more significant as the pulse areas increased.

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