Transient nonlinear optical spectroscopy studies involving biexciton coherence in single quantum dots

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Transient differential transmission measurements were performed to study coherent dynamics involving the biexciton resonance confined in a single GaAs quantum dot. When laser pulses with sufficient bandwidth were tuned to cover both the exciton and biexciton transitions, an oscillatory behavior in the probe transmission was observed. These oscillations persist only as long as the pump and probe pulses overlap in time, in contrast to the usual quantum beat phenomena.

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Biexcitons in semiconductor quantum dots (QD's) have been proposed as the physical realization of two-bit logic gates.[1–3] In a single QD, quantum confinement greatly enhances the higher-order Coulomb interaction, leading to the formation of a bound state of two orthogonally polarized excitons. The excitation of one exciton leads to a different excitation energy of the other exciton, which corresponds to the characteristic conditional dynamics needed for building a controlled-NOT (CNOT) gate. In fact, such a two-bit quantum gate has been demonstrated recently.[4] Further studies of coherence between states in the computational basis would provide a more comprehensive understanding of the quantum dynamics governing the performance of these elementary quantum devices.

The focus of the current study is the biexciton dipole coherence and the nonradiative coherence between the biexciton and the crystal ground state. Such quantum coherence induced by optical excitation can be quickly lost due to scattering and coupling to other modes in the semiconductor environment. The loss of probability amplitude as well as phase coherence between states without population loss (pure dephasing) can lead to decoherence in the system, thereby leading to errors in quantum information processing.

Biexciton transitions are not observable in single-photon processes. However, it is well known that biexcitons dramatically modify the nonlinear optical properties of semiconductor heterostructures.[5–9] The biexcitons represent easily identified genuine many-body correlation beyond mean-field theory.[10] Therefore, understanding dephasing dynamics related to biexcitons in single quantum dots is important for developing a more comprehensive microscopic theory for semiconductor quantum-confined structures.

Oscillatory signals observed in both differential transmission (DT) and four-wave-mixing experiments performed on GaAs quantum wells were attributed to biexciton transitions.[11–13] These oscillations only appear when the pump and probe beams overlap in time, contrary to the well-known quantum beat phenomenon.[14,15] In the usual quantum beat experiments, two electronic states sharing a common ground state are close in energy and are coherently driven by the same laser field. This is clearly not true for the case of the biexciton, whose energy is approximately twice that of the exciton. The oscillations involving the biexciton transition are related to the interference of two laser pulses, mediated by the oscillating nonlinear susceptibility $\chi^{(3)}$.[16]

In this Brief Report, we performed transient DT measurements to study biexcitons confined in single GaAs interface fluctuation quantum dots. We obtained information on both the biexciton dipole coherence and the nonradiative coherence between the biexciton and ground state in the strong-field regime. We were able to extract such information by using a broadband femtosecond laser combined with simple grating-based pulse shapers, which function as tunable spectral filters. When shaped laser pulses with just enough bandwidth were tuned to cover both the exciton and biexciton transitions, oscillatory behavior was observed. The oscillations persist only as long as the pump and probe pulses overlap in time, similar to experiments performed on GaAs quantum wells.[11,13] The temporal characteristics of the pulses are convolved in the nonlinear response, making it difficult to extract the nonradiative coherence time between the biexciton and ground state. Nevertheless, the observation of these distinct spectroscopy features provides valuable information about the relevant quantum dynamics.

In interface fluctuation QD's, the exciton states are excited with linearly polarized light reflecting the reduced symmetry of the QD potential. The biexciton is then excited by collinearly polarized light.[17,18,20] If the polarizations of the excitation fields are chosen to be collinear, the problem is then reduced to a three-level ladder system as shown in Fig. 1, in which the biexciton state can also decay to the exciton state with orthogonal polarization.

The proper formalism to describe the quantum dynamics of the ladder system is based on the master equation for the density matrix operators[21]

$$i\hbar \frac{d\rho}{dt} = [H,\rho] + i\hbar \left.\frac{d\rho}{dt}\right|_{relaxation}, \qquad (1)$$

where $H=H_0+V=H_0-\hat{\boldsymbol{\mu}}\cdot\mathbf{E}$, the first term corresponds to the diagonalized Hamiltonian for the eigenstates of the excitation level diagram, and the second term describes the coherent coupling of the laser field. The nonzero ele-

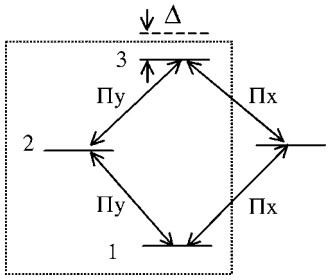

FIG. 1. Excitation energy level diagram of a biexciton. Δ is the binding energy of the biexciton, and Π_x and Π_y indicate orthogonally linearly polarized light coupled to the dipole-allowed transitions. If collinearly polarized excitation fields are used, the relevant states are reduced to a three-level ladder system enclosed in the dotted square.

ments of the dipole moment operator correspond to the transitions labeled in Fig. 1. The last term in Eq. (1) is a generalized decay operator that accounts for the decay of the population of different states (the diagonal density matrix terms) and the dephasing of the optically induced coherence (the off-diagonal density matrix terms).

There are two different types of terms contributing to the third-order nonlinear signal of a ladder system. One type results from population saturation effects, which goes through the following perturbations:

$$\rho_{11}^{(0)} \rightarrow \rho_{21}^{(1)} \rightarrow \begin{Bmatrix} \rho_{11}^{(2)} \\ \rho_{22}^{(2)} \end{Bmatrix} \rightarrow \begin{Bmatrix} \rho_{21}^{(3)} \\ \rho_{32}^{(3)} \end{Bmatrix}. \tag{2}$$

These terms are derived from the diagonal terms in the second order, often referred to as "stepwise" (SW) terms. Depending on the time ordering of the excitation fields, the SW terms may or may not give rise to nonlinear signal at large delay positions where the pump and probe beams no longer overlap temporally. Another type of possible term is the coherent term that goes through the perturbation path

$$\rho_{11}^{(0)} \rightarrow \rho_{21}^{(1)} \rightarrow \rho_{31}^{(2)} \rightarrow \begin{Bmatrix} \rho_{21}^{(3)} \\ \rho_{32}^{(3)} \end{Bmatrix}. \tag{3}$$

There are only off-diagonal density matrix terms in these perturbation paths, often referred to as "two-photon coherence" (TPC) terms. The TPC terms only contribute to the nonlinear signal near zero delay in the case of a ladder system.

The laser system used in this experiment is a 76 MHz mode-locked Ti:sapphire laser producing nearly transform-limited pulses of ~200 fs. Two passive pulse shapers in the folded geometry were set up outside the laser cavity with a mechanical slit at their Fourier planes. By simply adjusting the width of the slit, pulses with the proper bandwidth are obtained. The wavelength of the laser can be scanned within the original laser bandwidth [7.2 meV full width at half maximum (FWHM)] by moving the mechanical slit on a translation stage.

In DT measurements, two laser pulses, the pump and probe derived either from the same pulse shaper or two different pulse shapers, were focused onto the same submicrometer apertures on the sample held at 7 K in a liquid helium flow cryostat. The apertures were created using electron beams on an aluminum mask deposited on top of the sample. The excitation area is then limited by the size of the apertures, achieving the high spatial resolution needed to resolve exciton and biexciton resonances from single QD's. Two traveling-wave acousto-optic modulators (AOM's) were used to modulate the pump and probe beams at 1 and 1.05 MHz, respectively. The nonlinear signal was homodyne detected at the difference frequency using a lock-in amplifier.

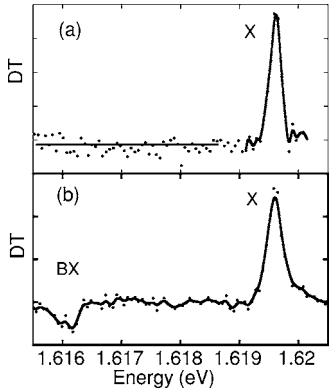

FIG. 2. Single-exciton and biexciton spectra taken from a submicrometer aperture using narrowband excitation pulses. (a) A single-exciton resonance was mapped out in a degenerate DT measurement. There are other exciton resonances at higher energy (data not shown). (b) A single exciton and the biexciton related to it were identified in a nondegenerate DT measurement.

A simple way often used to understand the DT measurement is the following: the pump pulse $[\mathbf{E}_1(t), \omega_1]$ promotes the electronic population from a lower level $\rho_{gg}(t)$ to a higher level $\rho_{ee}(t)$ of a dipole-allowed transition. The probe pulse $[\mathbf{E}_2(t), \omega_2$, delayed from the pump by $\tau]$ measures the changes induced by the pump pulse by creating a nonlinear optical polarization field. However, this explanation only accounts for the SW terms. The TPC terms resulting from quantum interference cannot be explained in such a simple picture.

Biexciton resonances in quantum dots can be identified by a variety of spectroscopy features such as the superlinear power dependence of photoluminescence, two-photon absorption, and induced absorption.[19] We follow previously demonstrated nonlinear optical spectroscopy methods in natural quantum dots to identify biexciton and the relevant exciton transitions.[20,22] First, the pulse shapers were set to produce pulses with a FWHM bandwidth of ~0.35 meV. A degenerate DT ($\omega_1 = \omega_2$) measurement was performed with the pump and probe beams derived from the same pulse shaper. The wavelength of the pulses was scanned with the delay (τ) between the two beams fixed around zero. A single-exciton resonance under the submicrometer aperture was mapped out as shown in Fig. 2(a). The typical value of the fractional change of the probe transmission induced by the pump is $\sim 10^{-4}$, determined mainly by the oscillator strength, dephasing dynamics, aperture size, excitation level, and the ratio between the absorption cross section of the dot and the aperture size. Next, in a nondegenerate DT ($\omega_1 \neq \omega_2$) configuration, the wavelength of the pump pulse derived from

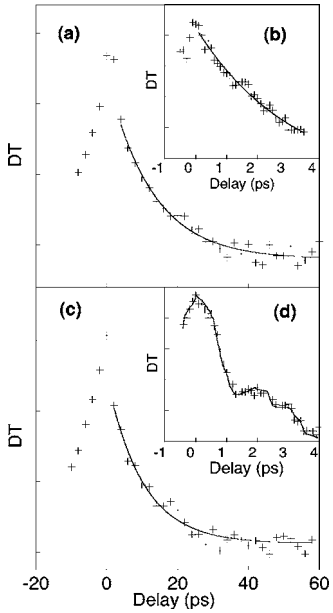

FIG. 3. DT measurements with broadband pulses tuned to different wavelengths. (a), (b) The wavelength was tuned to cover the exciton only. (c), (d) The wavelength was tuned to cover both the exciton and biexciton transitions. (b) and (d) are fine scans over the first few picoseconds. Solids lines in (a)–(c) are exponential functions fitted to the data, and the solid line in (d) is a guide to the eye.

one pulse shaper was tuned to the exciton, and the wavelength of the probe pulse from the second pulse shaper was scanned with delay τ fixed at $\sim$5 ps. The data show a strong positive peak at the exciton transition as a result of population saturation. An induced absorption dip appeared at the lower-energy end in Fig. 2(b) due to the excitation of the biexciton transition arising from both the SW and TPC terms discussed earlier. The probe tuned to the biexciton transition can only be absorbed following the excitation of the exciton by the pump. The energy splitting between the exciton and biexciton transitions, $\sim$3.5 meV, is the biexciton binding energy.

In order to study quantum dynamics involving the biexciton transition, degenerate DT measurements were performed with the slit in the pulse shaper opened to produce pulses with a bandwidth broad enough ($\sim$3.7 meV) to cover both the exciton and biexciton transitions. First, the center wavelength of the shaped pulse was tuned so that only the exciton resonance was excited. As the delay was varied, a smooth exponential curve with a decay constant of $\sim$13 ps was observed as shown in Fig. 3(a). This decay constant measures the exciton population relaxation time Γ_{12} under the current excitation condition. However, this relaxation time is shorter compared to that measured with the narrowband pulses.[23] The shorter decay time could result from an increased scattering rate between the resonantly excited exciton and the nearly degenerate delocalized states as discussed in a previous study.[24]

The wavelength of the broadband pulse was then tuned to cover both the exciton and biexciton transitions. The DT signal as a function of delay seemed to remain as a smooth decay with a decay constant of $\sim$10 ps over the range of tens of picoseconds as shown in Fig. 3(c). As discussed previously, only SW terms contribute to the DT signal at delay times greater than the pulse duration. Close examination of the master equation solutions for a ladder system indicates that the decay constants in both Figs. 3(a) and 3(c) are determined by the exciton population relaxation time Γ_{12}.[23]

When a fine scan was taken over the first few picoseconds with the laser bandwidth covering both the exciton and biexciton transitions, an oscillatory signal was observed as shown in Fig. 3(d). In contrast, there were no beats seen in Fig. 3(b) if the wavelength was tuned to cover the exciton transition only. We note that the observed oscillatory behavior is not simply due to the temporal sidebands of the shaped pulse since it depends sensitively to tuning relative to the biexciton resonance as seen in Figs. 3(b) and 3(d).

The period of the beats should depend not only on the binding energy of the biexciton (corresponding to a beat period of order of 1 ps) but also on the laser frequency detuning in the case of a homogeneously broadened system. If an ensemble of quantum dots with inhomogeneous broadening were excited, the period of the beats would be equal to the biexciton binding energy under certain approximations.[16] The oscillatory behavior observed in Fig. 3(d) is not a pure sinusoid, which may reflect the possibility that the system is more complex than the simple energy level structure assumed in the theory based on Fig. 1. While it would be useful in terms of analysis if more oscillation periods were observable, the oscillations only exist when the pump and probe pulses temporally overlap. Due to the broad spectral width required to cover both the exciton and the biexciton resonances, the temporal duration of the pulse is necessarily short.

We also note that the qualitative features of the temporal oscillations reported here do not depend on the pump intensity in the power regime used in these measurements (where the Rabi frequency is much less than the dipole coherence rate), and therefore, they do not arise from dynamic Stark effects.[25] In addition, early experiments have shown that many-body effects such as screening as seen in higher-dimensional structures have no detectable contribution to the nonlinear optical response in quantum dot structures under excitation conditions that do not lead to coupling to continuum states or states in nearby dots.[20,26]

The amplitude decay of the beats is related to the biexciton dipole coherence γ_{32} as well as the nonradiative coherence γ_{31}. It is difficult to extract any qualitative information about the decay parameters due to the fact that multiple quantum mechanical pathways contribute and the temporal pulse shape is convolved with the nonlinear signal. We have performed numerical calculations similar to those in Ref. 16 and determined a lower bound for the biexciton dephasing times of 1 ps. If biexciton dephasing times lower than 1 ps were used in the calculation, the oscillatory signal would disappear. Such dephasing parameters are comparable to the population relaxation times of the dipole-allowed transitions, around 10 ps.

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