Excitonic and biexcitonic decoherence in self-assembled GaAs quantum dots as observed by phase-locked interferography

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We observe single-photon interferograms for emission of a single self-assembled GaAs quantum dot. A phase stabilizer is applied to the interferometer, enabling us to trace autocorrelations of weak emissions with a photon-counting level. At low excitation, where the average number of excitons in a dot is less than one, interferography reveals a single exponential decay, reflecting excitonic decoherence. At moderately high excitation, polarization interference between the exciton-biexciton transitions is found to appear on the interferogram. The decoherence time and the binding energy of biexcitons are determined simultaneously. © 2006 American Institute of Physics.

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Optical transitions in semiconductor quantum dots (QDs) are known to exhibit ultranarrow resonance lines that are similar to those of isolated atoms.1 The sharp resonance, corresponding to the long decoherence time, suggests that excitons in QDs are good candidates for elemental devices in quantum information processing. So far, one-qubit2–5 and two-qubit6 operations have been demonstrated, in which the quantum information processing. So far, one-qubit2–5 and two-qubit6 operations have been demonstrated, in which the quantum state of excitons and biexcitons inside a single QD (SQD) is manipulated using short laser pulses. For integration of multiple quantum gates in this manner, a system of QDs that holds robust excitonic transitions should be designed. Basic research is, therefore, needed to characterize the decoherence mechanism of excitons and biexcitons.

A straightforward approach to evaluate the decoherence time is the measurement of photoluminescence (PL) linewidth. Making use of near-field optics7 or a micro-objective setup,8–11 emissions from SQD can be captured. The decoherence time is, thus, estimated as the inverse of the PL linewidth. Typically, however, such a procedure is not followed because of its linewidth being smaller than the instrumental limit. A large-scale monochromator equipped with multiple gratings—engendering low signal throughputs and long integration times—is essentially required for determination of the PL line shape. Another approach to resolve the narrow PL linewidth is interferometric spectroscopy, whereby the autocorrelation of SQD emissions can be observed. The line shape is presented by the Fourier transform of the correlation function. In this case, spectral resolution is limited by the optical path difference inside the interferometer, being as low as the sub-μeV level. The versatility of this technique was first demonstrated by Kammerer et al.12 studying InAs QDs;12 later, it was applied by Zwiller et al.13 to InP QDs and their temperature dependence.

In this letter, we report decoherence features of excitons and biexcitons in self-assembled GaAs QDs. Their homogeneous linewidths are determined precisely by phase-locked interferometry. We adopt a phase stabilizer into the interferometer, allowing long-time accumulation of weak emitted signals obtained at low pumping condition. Consequently, we can confirm the negligible contribution of multiple carrier effects, which are sometimes difficult to eliminate in conventional SQD experiments.

Experiments were performed with self-assembled GaAs QDs embedded in Al0.3Ga0.7As grown by modified droplet epitaxy.14,15 Atomic-force microscopy and high-resolution scanning electron microscopy demonstrated the lens-shaped formation of QDs with a 16-nm height and 20-nm base; their surface density was 6 × 108 cm−2. Figure 1(a) shows a schematic of the experimental setup. The sample was excited using a continuous-wave He–Ne laser emitting at 543 nm. Emissions were collected using an objective lens and were passed through an aperture that was designed for 1.0-μm detection spot. Then, the emission beam was guided to a Michelson-type interferometer. The optical path length was manipulated using short laser pulses. For integration of multiple gratings—engendering low signal throughputs and long integration times—is essentially required for determination of the PL line shape. Another approach to resolve the narrow PL linewidth is interferometric spectroscopy, whereby the autocorrelation of SQD emissions can be observed. The line shape is presented by the Fourier transform of the correlation function. In this case, spectral resolution is limited by the optical path difference inside the interferometer, being as low as the sub-μeV level. The versatility of this technique was first demonstrated by Kammerer et al.12 studying InAs QDs;12 later, it was applied by Zwiller et al.13 to InP QDs and their temperature dependence.

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varied using a mechanical stage and a piezotransducer (PZT). During signal integration, a phase-locking loop (PLL) was applied to retain the optical path difference. For this purpose, the optical path length was modulated by λ/100; harmonic components of laser reflections were measured using a lock-in amplifier. By changing the offset voltage of PZT and maintaining the lock-in signal unchanged, we stabilized the optical path difference to an accuracy of ±10 nm over 10 min. Interferometer outputs were carefully collimated and introduced into a low-dispersion spectrometer that was equipped with a charge-coupled device camera. All measurements were performed at 4 K.

Figure 1(b) shows two examples of micro PL spectra of our sample. Emission bands at 1.96 eV and 1.94 eV were assigned by donor-acceptor recombination at the barrier layer. Several sharp lines were detected in the range from 1.65 to 1.85 eV, which are attributed to excitonic recombination of QDs. The number of emission lines (~5) agrees with the expected number of QDs inside the detection spot. The spectral width of each QD emission was determined by the resolution of our spectrometer (~1 meV).

To evaluate the linewidth of an SQD emission, we selected one PL line and observed its dependence on the optical path difference. For the interferometric measurement, we adopted a low dispersive spectrometer, and spectrally integrated the signal over 5 meV, so as to remove the influence of a finite spectral window on the interferogram. Outputs of the interferometer are shown in Fig. 1(c), where the SQD emission at 1.78 eV is portrayed. The excitation density is limited to 3.2 W/cm². In this case, we can estimate the mean number of excitons inside a QD as 0.09, according to the carrier capturing cross section determined for the present sample. In the top panel, the optical path difference is varied around 0 mm in 20 steps, each of 60 nm. It shows a continuous change in intensity between constructive and destructive phase relations. Visibility of the modulation is estimated as >0.97. With increasing optical path difference, we find a pronounced decrease in visibility (0.66 at 4.8 mm, and 0.34 at 11.6 mm), while sinusoidal modulation with the same interval is still present, as shown in the middle and bottom panels of Fig. 1(c). These results suggest the finite coherent length of the electric field emitted by the SQD, reflecting the phase disturbance of excitonic dipoles.

We can produce the autocorrelation function by plotting the visibility as a function of the optical path length. The diamonds in Fig. 2 represent the correlation function for the SQD emission at 3.2 W/cm². The data well fit to a single exponential decay (a solid line), implying a relevant PL spectrum consisting of a single Lorentzian curve. We estimate the exciton decoherence time as 35 (±3) ps, corresponding to the homogeneous width of 38 (±3) µeV, full width at half maximum.

When the excitation density increases to 32 W/cm², i.e., 0.9 excitons being in a dot on average, the correlation function changes to an oscillatory shape, as shown by circles in Fig. 2. A solid line shows a fit with a model consisting of two decaying dipoles with different frequencies. Through the fit, we deduce the oscillation period as 4.2 ps; the respective decay times for the slow and fast components are 32 (±5) ps and 5.4 (±1) ps. Note that the decay time of the slow component agrees well with that at low excitation. For that reason, the slow component comes from excitons, and the fast component is related to biexcitons, i.e., the electron-hole recombination of the two-exciton state leaving an exciton. A small change in the exciton decoherence time suggests that the presence of multiple carriers does not influence the coherence of excitons, but it produces a well-defined biexciton state. The oscillatory profile shown in the interferogram reflects an energy split between the excitonic and biexcitonic transitions. The inset depicts a Fourier transform of the autocorrelation function—a reconstructed spectrum of the SQD emission. It shows a biexcitonic satellite appearing at 0.95 meV from the exciton line, which determines the biexciton binding energy.

We emphasize that the biexcitonic emission is present even when the number of excitons is less than one. According to Poisson statistics, the probability to find N excitons in a QD is represented as p(N)=e−n0N/N!, where n0 is the mean number of excitons. Therefore, the intensity ratio of the biexciton line to the exciton one is expected to be p(2)/p(1)=n0/2×0.45 for the present condition of n0=0.9. The experimental ratio is found to be 0.33, showing reasonable agreement with this argument. It further suggests that the condition to find just one exciton, free from multiple carrier effects, is only possible in conditions of extremely low pumping.

The biexciton binding energy is also confirmed by timeresolved observation of SQD emissions, as shown in Fig. 3. It is excited by fs pulses of 47 nJ/cm², corresponding to 1.8 excitons injected in a dot. Figure 3(a) shows that the SQD emission starts at 0 ps and decays with time. Temporal changes in the emission spectrum are summarized in Fig. 3(b). The transient spectrum reflects the population of excitons depending on time. A broadband at 1825 meV in the center energy–low energy side of a bright line is assigned by transitions from a three-exciton complex. We should remark that the bright line at 1828 meV shifts to high energy with time. At the relevant excitation, it is the most probable to produce two excitons in a QD. Thus, the biexcitonic transition appears in the initial stage of emission; it is later replaced by the single-exciton transition. Because of a low instrumental resolution (0.8 meV), the two components of
excitons and biexcitons are not resolved in spectral domain, but they result in the blueshift in the peak energy with time. The energy shift, i.e., the biexciton binding energy, is evaluated as 0.9 meV, which agrees well with the interferometric result. Note that the transient spectral shift comes from the cascade-type relaxation from biexcitons, and is not interpreted in terms of a trion dynamics. Moreover, the same biexcitonic transition was also observed in high-resolution PL spectra.16,17

It is noted that the observed biexciton binding energy (0.95 meV) is relatively smaller than reported values (a few milli-electron-volts). This is due to the high aspect ratio of our QD shape, which is characteristic of the QDs grown by droplet epitaxy. Numerous studies on GaAs quantum wells suggest that the biexciton binding enhances from the bulk value (0.45 meV) for well widths of less than 20 nm.7,18 Such conditions are fulfilled for QDs grown by the Stranski-Krastanow (SK) method, where their height is a few nanometers. In contrast, the height of our QDs is \(~16\) nm, which should reduce quantum confinement along the vertical direction, resulting in weak biexciton binding. Characteristics of our droplet epitaxial QDs reflect in the PL linewidth which is much larger than that of SK-grown QDs (a few micro-electron-volts).19,20: Our sample is expected to contain relatively high density of imperfections and/or external carriers. These result in low-frequency fluctuations in the local field surrounding QDs, and efficient broadening in the SQD spectra.

In summary, we reported high-resolution measurements of exciton decoherence in single GaAs QDs. We obtained single exponential decay signals, whose homogeneous linewidth is 37 \(\mu\)eV. The exciton-biexciton beat is found to appear at high excitation intensity. Through fitting, we estimated the biexciton decoherence time and its binding energy. The technique presented in this report is useful for evaluation of long decoherence times, i.e., narrow linewidths of emission, which is expected to contribute to development of novel SQD devices.

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