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Negligible Pure Dephasing in InAs Self-Assembled Quantum Dots

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We measured the dephasing time and radiative lifetime of excitons in InAs quantum dots fabricated using the strain compensation technique. The dephasing time at 3 K was as long as 2.86 ns using transient four-wave mixing measurements at an excitation wavelength of 1.468 μm. This ultralong dephasing time was due to the significant suppression of pure dephasing. [DOI: 10.1143/JJAP.46.6352]

KEYWORDS: dephasing, quantum dot, coherent nonlinear spectroscopy, exciton–phonon interaction

1. Introduction

Self-assembled quantum dots (SAQDs) show great potential for use as essential building blocks in solid-state-based quantum logic devices.[1)] The long dephasing time (T_2) of excitons is of crucial importance with respect to their implementation since T_2 limits the number of possible quantum operations. In most SAQDs, however, the measured T_2 is shorter than the upper dephasing limit determined by the exciton population lifetime (T_1).[2–5)] The most frequent cause of additional dephasing is non-negligible interactions between excitons and phonons, which reduce phase coherence among excitons without affecting the exciton population. Therefore, it is necessary to control exciton–phonon interactions to obtain a long T_2.[6)]

Exciton–phonon interactions can be partly suppressed by decreasing the temperature. However, previous research has shown these interactions are still predominant over population decay even at low temperatures (<10 K).[2,3)] Two groups have recently succeeded in obtaining a T_2 limited only by the exciton radiative lifetime of In(Ga)As SAQDs, consequently, a T_2 as long as 2 ns was obtained.[7–9)]

In this study, we demonstrated a long T_2, approaching 3 ns at 3 K, in InAs SAQDs fabricated using the strain compensation technique.[10,11)] The emission wavelength of the exciton ground states measured in our QDs was 1.468 μm, which is much longer than the emission wavelengths for QDs fabricated using conventional self-assembly. Transient four-wave mixing (FWM) and pump–probe (PP) measurements showed that the T_2 is very close to the upper dephasing limit determined by the $2T_1$. The result of temperature-dependent measurement suggests that the remaining pure dephasing was caused by exciton–acoustic phonon interactions.

2. Experimental Procedure

2.1 Sample preparation

The sample used consisted of 150 layers of InAs SAQDs embedded in 60-nm-thick InGaAlAs spacers grown on an InP(311)B substrate.[10,11)] A schematic of the sample structure is shown in Fig. 1. To reduce QD strain, the composition of the spacers was precisely tailored, and an InP(311)B substrate was used. The strain compensation technique enables us to stack up to 150 QD layers, thereby significantly improving the signal-to-noise ratios in the FWM and the PP measurements.[12)] Both sides of our sample contained an antireflection coating to prevent multiple reflections. Figure 2 shows the photoluminescence spectrum of our sample at 3 K under nonresonant excitation. The exciton ground-state emission peaked at 1.468 μm, which is much longer than that of conventional In(Ga)As/GaAs SAQDs. The inhomogeneous broadening of the emission energy was 44 meV.

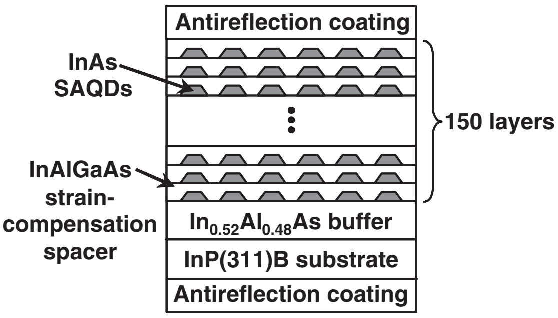

Fig. 1. Schematic of sample structure. To compensate QD strain, the composition of the spacers was precisely tailored and an InP(311)B substrate was used.

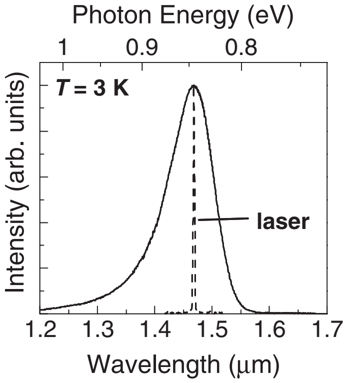

Fig. 2. Photoluminescence spectrum at 3 K under nonresonant laser excitation (solid line). Exciton ground state emission peaks at wavelength of 1.468 μm. The dashed line represents spectrum of the excitation pulses used in the FWM and PP measurements.

2.2 Transient FWM and PP measurements

T_2 was measured using a two-pulse self-diffraction FWM technique in the transmission geometry. The experimental setup is illustrated in the inset of Fig. 3. The time-integrated

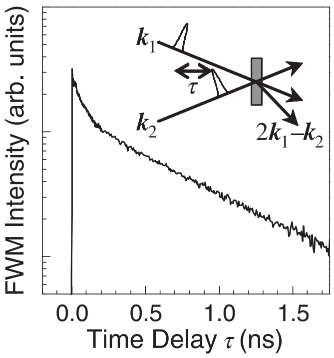

Fig. 3. Time-integrated FWM signals versus τ at 3 K. The inset illustrates the experimental setup used for the FWM measurements.

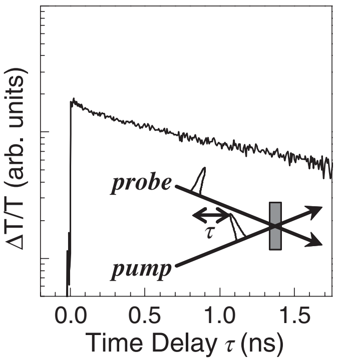

Fig. 4. Differential transmission of probe pulse measured at various τ values. The PP signal decays mainly owing to the radiative recombination of excitons. The inset illustrates the experimental setup used for the PP measurements.

FWM signal intensity in the direction of $2\boldsymbol{k}_1 - \boldsymbol{k}_2$ was measured as a function of the time delay τ between two excitation pulses. The intensities of the excitation pulses were adjusted to 16 kW/cm^2, for which the excitation-dependence of T_2 was not significant.

T_1 was measured using a PP technique in the transmission geometry (see the inset of Fig. 4). The differential transmission of the probe pulse was detected at various τ values between the pump and probe pulses. The intensity of the pump pulse was fixed at 16 kW/cm^2, and the intensity of the probe pulse was 0.5% of that of the pump pulse.

Both FWM and PP measurements were performed using 1.1 ps optical pulses at a repetition rate of 76 MHz produced from an optical parametric oscillator pumped by a mode-locked Ti:sapphire laser. The central wavelength of the excitation pulses was tuned to 1.468 μm. The spectrum of excitation pulses is shown in Fig. 2. The polarization of the excitation pulses was in the $[01\bar{1}]$ direction so that one of the nondegenerate exciton ground states was selectively excited.[13)]

3. Results and Discussion

Figure 3 shows a typical FWM signal at 3 K. For long time delays, the observed FWM signal decays exponentially with a long time constant. The T_2 is 4-fold longer than the decay time constant since the FWM signals of an inhomogeneously broadened QD ensemble act as photon echos in real time.[2)]

The estimated T_2 was 2.86 ± 0.07 ns, which is the longest value ever reported for SAQDs. Such a long-lived coherence reflects the high quality of our QDs. The T_2 can be converted into the homogeneous broadening $\Gamma_\mathrm{h} = 2\hbar/T_2$. The Γ_h was calculated to be only 0.46 ± 0.01 μeV. The obtained Γ_h is much smaller than typical Γ_h values in In(Ga)As SAQDs, i.e., from 2 to 100 μeV.[2–4,14)]

Now, we will comment on the accuracy of determination of Γ_h since it is crucial for the quantitative investigation of Γ_h values less than 1 μeV. In our FWM measurements, Γ_h was determined with an accuracy of 0.01 μeV even for weak excitation intensities. This is at least one order of magnitude higher than those obtained by interferometric correlation photoluminescence spectroscopy on a single QD[14)] and spectral hole burning spectroscopy on a QD ensemble.[9)] This difference comes from the physical and mechanical differences between FWM and the other techniques. In addition, the high signal-to-noise ratio in the present FWM experiment, which was achieved with a stack of 150 QD layers,[12)] plays an important role in improving the accuracy. To obtain an accuracy of 0.01 μeV using the same QDs, it would be necessary to stack more than 90 QD layers assuming that the FWM signal intensity is proportional to the square of the number of QDs.

To estimate the contribution of pure dephasing on Γ_h, we measured the T_1 using a PP technique. Figure 4 shows the differential transmission of a probe pulse measured at 3 K. The many-particle effect can be neglected at the pump intensity since the exciton density generated by the pump pulse is no more than one exciton per QD. The decay time constant was estimated to be 1.7 ns from the fitting using a single exponential function. We also measured the differential transmission for orthogonal polarization in the $[01\bar{1}]$ direction in which the other exciton state was excited (data not shown here). The decay time constant in this case was 1.0 ns, which is shorter than that for parallel polarization in the $[01\bar{1}]$ direction. The ratio of the decay time constant is in good agreement with the ratio of the square of the transition dipole moments obtained using polarization-dependent FWM measurements.[15)] This comparison demonstrates that the differential transmission decays mainly owing to a radiative recombination process of excitons and that other population relaxation processes are not significant in our QDs. Therefore, the decay time constant of the differential transmission coincides with the radiatively-limited T_1. Consequently, the T_1 for the $[01\bar{1}]$ polarized exciton state was estimated to be 1.7 ± 0.2 ns. The lifetime broadening Γ_0 given by the T_1 (i.e., $\hbar/T_1$) was 0.38 ± 0.06 μeV, which shows good agreement with previously reported values.[16)] The relationship between Γ_h and Γ_0 is expressed by the equation $\Gamma_\mathrm{h} = \Gamma_0 + \Gamma_\mathrm{pure}$, where Γ_pure is the pure dephasing rate. The FWM and PP measurements clearly demonstrated that the Γ_pure was much smaller than Γ_0 at 3 K in our QDs. The Γ_pure was approximately 0.08 μeV, which is much smaller than that of QD excitons previously reported.[3)] Owing to the small Γ_pure, the T_2 is just below the upper limit determined by $2T_1$, which results in the ultralong T_2 for our QDs.

To investigate the pure dephasing process in detail, we measured the temperature dependence of Γ_pure under the same excitation conditions. Figure 5 shows the Γ_pure (solid

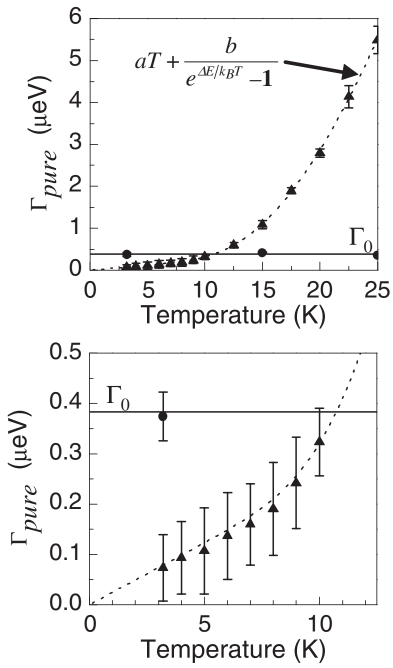

Fig. 5. Temperature dependence of pure dephasing rate Γ_{pure} (closed triangle) with theoretical curves (dashed lines) calculated using the equation shown in upper graph. The solid circles represent the lifetime broadening Γ_0 and the solid line represents an average of Γ_0 in the range of 3–25 K. The lower graph shows a magnified view for temperatures lower than 12.5 K.

triangles) and Γ_0 (solid circles) measured at various temperatures (T). At $T < 8$ K, Γ_{pure} is smaller than Γ_0, even though the Γ_{pure} increases linearly with increasing temperature. The Γ_{pure} increases significantly at temperatures exceeding 8 K and predominates over the Γ_{h} at higher temperatures. This behavior is very different from that of Γ_0, which is almost constant up to 25 K.

The measured Γ_{pure} is well reproduced by the equation[17] $\Gamma_{\mathrm{pure}}(T) = aT + b/(e^{\Delta E/k_{\mathrm{B}}T} - 1)$, as shown by the dashed lines in Fig. 5. The extrapolated zero-temperature Γ_{pure} is reduced to almost zero. A line of best fit was obtained for $a = 0.025 \pm 0.005\,\mu\mathrm{eV/K}$, $b = 72 \pm 5\,\mu\mathrm{eV}$, and $\Delta E = 5.9 \pm 0.2\,\mathrm{meV}$. The a obtained in this experiment is one order of magnitude smaller than the smallest a ever reported in QDs.[3] This is why the Γ_{pure} is very small even at 3 K and why the long T_2 is observed in our QDs. The most probable origin of Γ_{pure} is an exciton–acoustic phonon interaction, as theoretically and experimentally investigated in the literature.[17] For a complete understanding of the underlying mechanism in our QDs, more strict analysis on exciton–phonon interactions in QDs is needed.

4. Conclusions

We measured the dephasing time, T_2, and the radiative lifetime, T_1, of excitons in InAs SAQDs fabricated using the strain compensation technique. T_2 and T_1 were evaluated using four-wave mixing and pump–probe techniques, respectively. Even at 3 K, the homogeneous broadening, Γ_{h}, was predominated by the lifetime broadening, Γ_0, since pure dephasing is considerably suppressed in our QDs. This small pure dephasing resulted in an ultralong T_2, approaching 3 ns, which is the longest value ever reported.

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