Long dephasing time in self-assembled InAs quantum dots at over 1.3 μm wavelength

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Excitonic dephasing is investigated in InAs self-assembled quantum dots fabricated by the strain-compensation technique. The exciton ground-state emission is centered at the wavelength of 1420 nm at 5 K. Transient four-wave mixing measurements under resonant excitation clearly demonstrate a long dephasing time of 1.09 ns at 5 K, corresponding to the homogeneous broadening of 1.2 μeV. The extrapolated zero-temperature homogeneous broadening is limited only by the population lifetime of the exciton ground state. At slightly increased temperatures, the acoustic-phonon broadening becomes dominant on dephasing. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2217156]

Self-assembled quantum dots (SAQDs) emitting over 1.3 μm light are of great interest, because they have enabled development of devices for optical[1,2] and quantum[3,4] telecommunications. Fabrication techniques such as a strain compensation[5,6] and a double-cap method[7,8] have been developed to redshift the emission wavelength of SAQDs in the last few years. Subsequently, single photon emission,[6,9] light slowing based on electromagnetically induced transparency,[2] and QD lasing[10,11] have been proposed and demonstrated at longer wavelengths.

To improve the performance of these applications, long coherence time (characterized by homogeneous dephasing time T_2) of an optically generated exciton is indispensable. In addition, long T_2 is essential for a larger number of quantum operations utilizing QD excitons. This dephasing should be suppressed drastically in QDs by quantizing the energy levels. Although a subnanosecond[12,13] or more[14] T_2 and an extraordinarily narrow linewidth[15,16] of the exciton ground-state (GS) transition have been observed in SAQDs, there has thus far been no report on excitonic dephasing in SAQDs at over 1.3 μm. This is because previous experiments have been done mainly on In(Ga)As SAQDs embedded in a GaAs matrix where, because of the large strain, the emission wavelength is limited to 1.3 μm or shorter. Moreover, though transient four-wave mixing (FWM) experiments are a powerful tool to measure T_2 in the nanosecond range,[17] they are difficult to carry out on QDs when the FWM signal is weak due to low QD density.

In this letter, we investigate excitonic dephasing and its mechanism using the FWM technique at an excitation wavelength of over 1.3 μm. Our strain compensation technique in the fabrication process enables us to control the emission wavelength. Simultaneously, strain compensation enables us to stack over a hundred QD layers, thereby enhancing FWM signal intensity.

The investigated sample contains 150 layers of InAs SAQDs embedded in 20 nm thick $In_{0.46}Ga_{0.11}Al_{0.43}As$ spacers [see Fig. 1(a)]. The sample is fabricated using molecular beam epitaxy as detailed elsewhere.[5] To compensate for QD strain, the composition of spacers is precisely tailored, and an InP(311)B substrate is used instead of a GaAs one. The

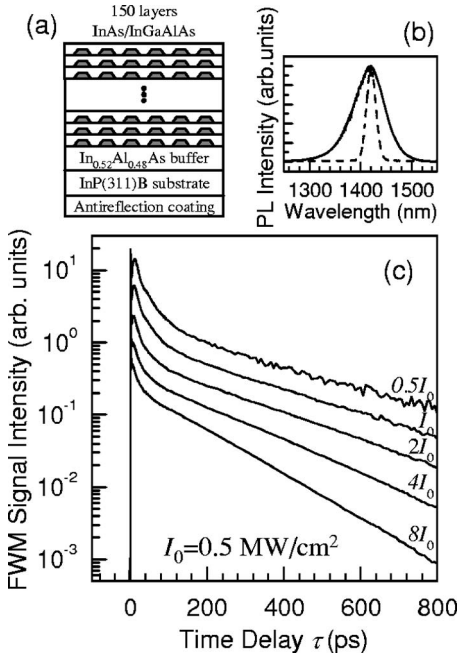

FIG. 1. (a) Schematic of our sample structure. (b) Photoluminescence spectrum at 5 K for nonresonant laser excitation (solid line) and spectrum of excitation pulse (dashed line). (c) Normalized time-integrated FWM signals vs τ at 5 K for various excitation densities of incident pulses I. I_0 = 0.5 MW/cm^2 corresponds to incident photon density of 1×10^8 photons per pulse.

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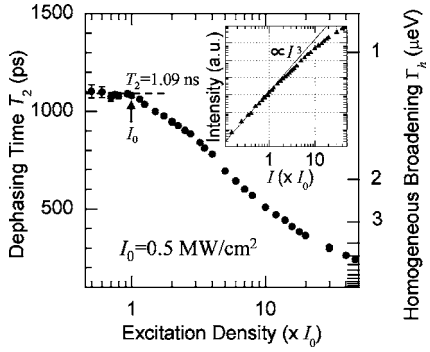

FIG. 2. Estimated dephasing time T_2 and signal intensity (inset) for various excitation densities. T_2 at low excitation density is 1.09 ns corresponding to Γ_h=1.2 μeV.

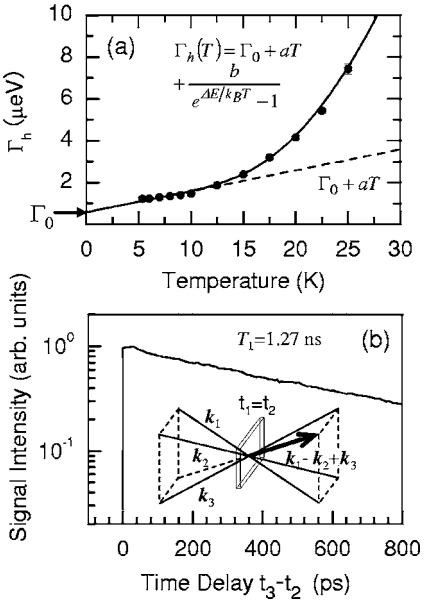

FIG. 3. (a) Temperature dependence of Γ_h with theoretical curve (solid line). (b) Transient-grating signal vs time delay t_3-t_2 when $t_1=t_2$. The inset illustrates the experimental setup of three-pulse transient grating.

backside of our sample is antireflection-coated to prevent multiple reflections. The photoluminescence spectrum at 5 K shown in Fig. 1(b) indicates that the exciton GS emission peaks at 1420 nm. This wavelength is much longer than that in conventionally self-assembled In(Ga)As/GaAs SAQDs. In addition, the strain compensation enables us to stack up to 150 QD layers,[18] corresponding to the effective QD density of $\sim 6\times 10^{12}$ dots/cm^2.

The T_2 is measured using a two-pulse self-diffraction FWM technique in the transmission geometry. A time-integrated FWM signal in the direction of $2\mathbf{k}_1-\mathbf{k}_2$ is measured as a function of time delay τ between two excitation pulses with wave vectors $\mathbf{k}_{1,2}$. A FWM experiment is done using 170 fs pulses from an optical parametric oscillator pumped by a mode-locked Ti:sapphire laser. The central wavelength of the excitation pulses is set at 1420 nm [refer to the dashed line in Fig. 1(b)]. In all the measurements, the excitation intensities of $\mathbf{k}_1$ and $\mathbf{k}_2$ pulses are identical, and both pulses are linearly polarized along the $[\bar{2}33]$ direction.

Figure 1(c) shows the normalized FWM signals versus τ at 5 K for various excitation densities of the incident pulses I. At I_0=0.5 MW/cm^2, it is estimated that 1×10^8 photons per pulse are incident on our sample. We confirmed that the InP substrate and wetting layers do not significantly contribute to FWM signals. The observed FWM signals decay nonexponentially and very fast for a few tens of picoseconds. Then, they decay exponentially with a much longer time constant (τ_{slow}) of several hundred picoseconds. This behavior is in good agreement with previous FWM experiments.[12,14] Therefore, it is likely that the fast decay originates from the interaction between excitons and acoustic phonons with relatively higher energies. This interaction was previously observed as sidebands around a narrow zero-phonon line in single-QD photoluminescence spectroscopy.[16] The slow decay reflects the broadening of the zero-phonon line with a Lorentzian profile.[14,16] The corresponding T_2 is given by $T_2=4\times\tau_{\mathrm{slow}}$ since the FWM signals are emitted as photon echoes.[12,17]

Figure 2 shows T_2 measured at various excitation densities I. T_2 is almost independent of the excitation density for $I<I_0$ (=0.5 MW/cm^2), while it drastically decreases with increasing excitation density for $I>I_0$. We can estimate from the absorbance that the exciton density corresponding to I_0 is no more than one exciton per QD. This implies that the dephasing in the weak excitation regime for $I<I_0$ is caused by two mechanisms: population decay and pure dephasing due to exciton-phonon interactions. In the higher excitation regime for $I>I_0$, exciton-exciton interactions become prominent, accelerating the dephasing. The signal intensity of the slow decay component increases in a perfectly proportional way to I^3 below I_0, as shown in the inset of Fig. 2. It starts to deviate from I^3 and eventually saturates for $I>I_0$. This suggests that exciton-exciton interactions prevent coherent FWM processes in the higher excitation regime.

The T_2 unaffected by exciton-exciton interactions can be estimated from the results in the weak excitation regime. The average of T_2 for $I<I_0$ reaches $T_2=1.09\pm0.02$ ns, which is much longer than typical values of T_2 in SAQDs, i.e., from several tens to several hundreds of picoseconds.[4,12,13,15,16] The corresponding homogeneous broadening, Γ_h ($\Gamma_h=2\hbar/T_2$), is calculated to be only 1.2 μeV, which is clear evidence of the extremely sharp zero-phonon line in our sample.

To investigate the dephasing processes in the weak excitation regime more closely, we measured the temperature dependence of Γ_h at the threshold excitation density I_0. The results are summarized in Fig. 3(a). With increasing temperature, Γ_h increases slowly below 10 K and shows nonlinear growth at 10–25 K. This result is well reproduced by the standard thermoactivated broadening,[14,15] $\Gamma_h(T)=\Gamma_0+aT+b/(e^{\Delta E/k_BT}-1)$. The second linear term in the equation represents acoustic-phonon broadening, while the last term represents thermal activation from GS to a higher energy state by a phonon absorption process. The best fit is obtained for $\Gamma_0=0.6\pm0.2$ μeV, $a=0.10\pm0.02$ μeV/K, $b=210\pm70$ μeV, and $\Delta E=8.3\pm0.9$ meV. Because the contribution of the last term is still insignificant at temperatures below 10 K, the dephasing at 5 K is determined by zero-temperature dephasing Γ_0 and the acoustic-phonon broadening characterized by a.

We independently measured the population decay using a three-pulse transient-grating technique[19] at I_0. Figure 3(b) shows the signal trace measured at 5 K, and the inset illustrates the experimental setup. The population lifetime T_1 is

estimated to be 1.27 ns, which is almost constant up to at least 25 K. Theoretically, the population decay's contribution to dephasing is given by $\hbar/T_1$, i.e., 0.51 μeV. This value is almost identical to the zero-temperature broadening Γ_0. Therefore, Γ_h is limited only by the population decay at zero temperature. However, the dominance of pure dephasing related to aT is already established at a temperature of 5 K. Nevertheless, the value of a for our sample is much smaller than that in most of SAQDs.[12–16] This is why our SAQDs show such a long T_2 (over a nanosecond). Further investigations are needed to clarify why our strain-compensated SAQDs have a small a.

Our results clearly demonstrate that acoustic-phonon broadening plays a crucial role in excitonic dephasing in SAQDs at low temperature, as extensively discussed in previous papers.[12–16] Thus, control of acoustic-phonon broadening is important in obtaining a longer T_2. More systematic study on dephasing in SAQDs will be needed to control exciton-acoustic phonon interactions by changing the QD structure. In addition, actively controlling dephasing by irradiating an optical pulse sequence provides an effective method of elongating the dephasing time,[20] but its experimental demonstration in QDs remains a challenge.

We have demonstrated a dephasing time T_2 of over a nanosecond at an excitation wavelength of 1420 nm in strain-compensated InAs SAQDs. This extremely long coherence is due to the suppression of acoustic-phonon broadening.

The emission wavelength of our sample can be controlled by slightly changing the composition and the thickness of the spacer layers. Thus, our SAQDs show great promise for operation at telecommunication wavelengths.

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