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Radiatively limited dephasing of quantum dot excitons in the telecommunications wavelength range

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The extremely long dephasing time of excitons in strain-compensated quantum dots at telecommunications wavelengths was measured using a polarization-dependent four-wave mixing technique. The use of a 150-layer-stacked structure enabled them to measure a four-wave mixing signal with a high signal-to-noise ratio, in spite of the fact that a high-sensitive heterodyne detection was not used. The large anisotropy of the dephasing time indicates the dominance of the radiative recombination process on dephasing. By simultaneously measuring the radiative lifetime using a pump-probe technique, they could directly estimate pure dephasing with an accuracy of better than 0.1 μeV. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2780120]

Semiconductor quantum dots (QDs) are promising solid-state candidates for quantum logic gates[1] and single-photon or entangled-photon sources.[2] A critical issue for their implementation is how to suppress nonradiative dephasing of optically allowed excitonic transitions in QDs. In QDs, solid-state environments strongly interact with excitons, which causes significant nonradiative dephasing. Even for the lowest investigated temperature and excitation density, nonradiative dephasing is larger than dephasing via radiative recombination processes in most QDs.[3–6] The underlying mechanism of nonradiative dephasing is still under debate;[7] therefore, the control of nonradiative dephasing remains a challenging problem.

It has recently been found that nonradiative dephasing is significantly suppressed in a few self-assembled In(Ga)As QDs,[7–9] which are attractive for quantum communication. In these QDs, the dephasing time T_2 approaches the upper limit determined by the radiative lifetime, i.e., $2T_r$. The authors in Ref. 8 indirectly confirmed that T_2 was dominated by T_r, by analyzing the anisotropy of T_2, which reflected the anisotropic transition dipole moment of asymmetric QDs. The anisotropic nature of T_2 was measured by a polarization-dependent heterodyne four-wave mixing (FWM) technique, which is the most powerful tool to measure T_2 in the nanosecond range.[3,4] Up to now, Ref. 8 was the only paper to report values for radiatively limited T_2 taking into account its anisotropic property. Since their experiment used In(Ga)As/GaAs QDs grown on a GaAs(100) substrate, the radiatively limited T_2 was observed only in the short wavelength range (<1.3 μm). Moreover, they did not perform a direct comparison between T_2 and T_r, which allows a precise measurement of nonradiative dephasing.

In this letter, we report on an anisotropic T_2 very close to the radiative limit at 3 K for *strain-compensated* InAs QDs in the telecommunications wavelength range. The values of T_2 and T_r were independently obtained from polarization-dependent FWM and pump-probe (PP) measurements with a high degree of accuracy, which was significantly improved using the 150-layer-stacked structure. Consequently, the pure dephasing was directly estimated with an accuracy of better than 0.1 μeV.

The sample consisted of 150 layers of InAs self-assembled QDs embedded in 60 nm thick $In_{0.52}Ga_{0.1}Al_{0.38}As$ strain-compensation spacers grown on an InP (311)*B* substrate.[10] The sample structure was similar to that used in our previous experiment, but the thickness and the composition of the spacers were different.[11] The average-lateral size of the QDs was estimated to be 39 nm in the $[01\bar{1}]$ direction and 51 nm in the $[\bar{2}33]$ direction. The exciton ground-state emission reached maximum intensity at 1.468 μm at 3 K, as shown in Fig. 1(a). The ground and first-excited states of the exciton were separated by 53 meV.[12] The energy separation was sufficiently larger than the inhomogeneous broadening of the transition energies (half-width at half maximum ~22 meV).

We used a two-pulse FWM scheme in a transmission geometry to measure T_2 directly in an inhomogeneously broadened QD ensemble [see Fig. 1(b)]. The polarizations of the excitation pulses were set in either the $\boldsymbol{x}\|[01\bar{1}]$ or $\boldsymbol{y}\|[\bar{2}33]$ directions to estimate an individual T_2 for the orthogonally polarized ground-state doublets[8,13] [refer to Fig. 1(c)]. The splitting energy of the doublets was approximately 180 μeV. The FWM experiments were performed using 1.1 ps optical pulses at a 76 MHz repetition rate in resonance with the center of the inhomogeneously broadened ground-state transition, as shown in Fig. 1(a). The intensities of the excitation pulses were adjusted to 16 kW/cm^2, where the

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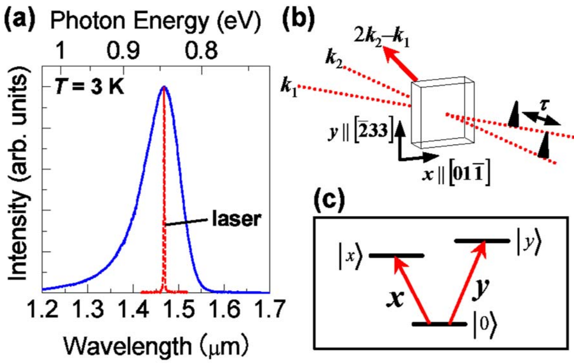

FIG. 1. (Color online) (a) Solid line: photoluminescence spectrum at 3 K under nonresonant laser excitation. Dashed line: spectrum of excitation pulses used in the FWM and pump-probe experiments. (b) Experimental setup of polarization-dependent FWM. (c) Energy-level diagram and optically allowed transitions related to an exciton ground-state doublet in an elongated QD. $\boldsymbol{x}$ and $\boldsymbol{y}$ correspond to the $[01\bar{1}]$ and $[\bar{2}33]$ directions.

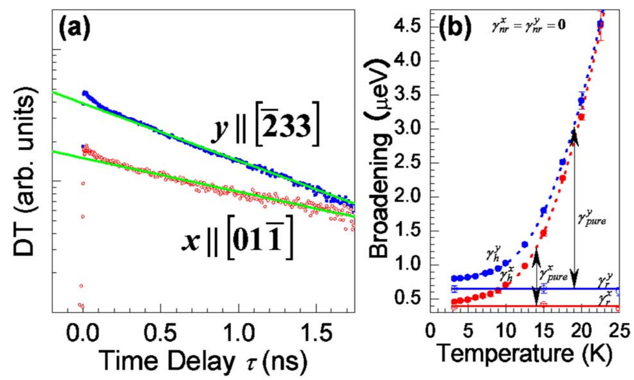

FIG. 3. (Color online) (a) Transient differential transmission of the probe pulse at 3 K for $\boldsymbol{x}$ (open circles) and $\boldsymbol{y}$ (filled circles) polarizations. Solid lines represent the single exponential curves. (b) Polarized γ_h (filled circles) and γ_r (open circles) measured at various temperatures.

FWM signal intensity is proportional to the cube of the excitation intensity (the $\chi^{(3)}$ region).

Figure 2 shows time-integrated intensities of FWM signals measured at various time delays (τ) between the two excitation pulses at 3 K. This study focuses on the exponentially decaying slow component corresponding to the broadening of the zero-phonon line.[14] The decay time constant of the slow component significantly depends on the polarizations of the excitation pulses. This result indicates that the T_2 for the $\boldsymbol{x}$- and $\boldsymbol{y}$-polarized transitions are different from each other. We estimate them to be T_2^x=2.86 ns and T_2^y=1.64 ns, respectively. The T_2^x value is longer than any other T_2 reported for QD excitons so far.[3–9] The homogeneous broadenings γ_h $(=2\hbar/T_2)$ were calculated to be only $\gamma_h^x = 0.46 \pm 0.01$ μeV and $\gamma_h^y = 0.80 \pm 0.01$ μeV.

The error of the γ_h estimation was as small as 0.01 μeV, as described above, in spite of the fact that the excitation intensity was weak and that a high-sensitive heterodyne detection was not used. This is at least one order of magnitude smaller than the errors obtained by interferometric correlation photoluminescence spectroscopy on a single QD (Ref. 6) and by spectral hole burning spectroscopy on a QD ensemble.[9] The 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 150-layer-stacking QDs,[11] plays an important role in improving the accuracy. High accuracy is crucial for precise measurement of γ_h values less than 1 μeV.

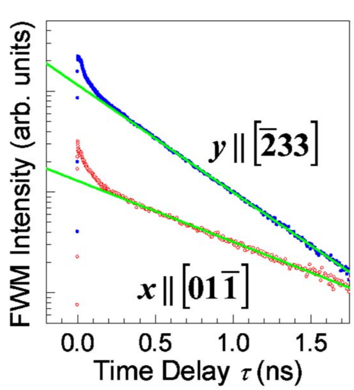

FIG. 2. (Color online) Time-integrated FWM signals at various time delays τ at 3 K for $\boldsymbol{x}$ (open circles) and $\boldsymbol{y}$ (filled circles) polarizations. Solid lines represent the single exponential curves.

To roughly evaluate the radiative contribution to γ_h, we note the polarization dependence of the FWM signal intensity in Fig. 2. In the $\chi^{(3)}$ excitation regime, the FWM signal intensity is proportional to the eighth power of the transition dipole moment $|\mu|^8$ at zero delay.[8] Therefore, the ratio of $|\mu|$ can be determined to be $|\mu_y|/|\mu_x| = 1.32 \pm 0.02$ from the extrapolated zero-delay FWM intensities. The ratio of the radiative dephasing γ_r is thus $\gamma_r^y/\gamma_r^x = 1.75 \pm 0.04$, because $\gamma_r \propto T_r^{-1} \propto |\mu|^2$. This ratio coincides with $\gamma_h^y/\gamma_h^x = 1.75 \pm 0.07$. This agreement implies that γ_h is dominated by γ_r and that the contribution of nonradiative dephasing is quite small.

The analysis mentioned above is similar to that in Ref. 8 for InAs QDs grown on a GaAs(100) substrate. The anisotropy of $|\mu|^2$ of our QDs is larger than that of the QDs in Ref. 8 (i.e., 1.32 ± 0.015). This leads to a larger anisotropy for the radiatively limited T_2. One of the possible origins of the large anisotropy of our QDs is the lower symmetry of the high-index substrate. Sanguinetti *et al.* reported that QDs on a (311)*B* substrate show a larger anisotropy in the photoluminescence spectrum compared with that for QDs on a (100) substrate.[15] However, the optical anisotropy of QDs depends not only on the substrate orientation but also on the other fabrication conditions. Therefore, further systematic study will be required to clarify the origin of the large anisotropy.

To estimate the radiative contribution more precisely, we measured the absolute value of T_r by using a polarization-dependent PP technique. The experiment was performed using optical pulses with the same wavelength, pulse width, and pump intensity as those in the FWM experiment. The effect of biexciton formation can be ignored due to the narrow band and weakness of the pump pulse. The probe intensity was set to 0.5% of the pump intensity.

Figure 3(a) shows the transient differential transmission (DT) at 3 K for $\boldsymbol{x}$ and $\boldsymbol{y}$ polarizations. From the exponential fitting of the DT, the DT decay times τ_{DT} are determined as τ_{DT}^x=1.7 ns and τ_{DT}^y=1.0 ns. The ratio $\tau_{DT}^x/\tau_{DT}^y = 1.7 \pm 0.2$ is in quantitative agreement with the ratio $|\mu_y|^2/|\mu_x|^2$ deduced from the FWM experiment. Therefore, we expected τ_{DT} to correspond directly to T_r for our QDs. This was confirmed by the fact that the values of τ_{DT} exactly coincided with the values of T_r calculated using the absolute values of $|\mu|$ obtained from the measurements of the Rabi oscillations. As a result, $\gamma_r(=\hbar/T_r)$ becomes $\gamma_r^x=0.38\pm0.01$ μeV and $\gamma_r^y=0.65\pm0.06$ μeV. The results of the PP experiment demonstrates that the nonradiative population decay is negligible compared with radiative population decay for our QDs. Therefore, dephasing caused by nonradiative population decay, γ_{nr} $(=\hbar/T_{nr})$, can be set to be zero.

The relationship between γ_h, γ_r, and γ_{nr} is expressed by the equation, $\gamma_h=\gamma_r+\gamma_{nr}+\gamma_{pure}$, where γ_{pure} represents pure dephasing. The total nonradiative dephasing is given by $\gamma_{nr}+\gamma_{pure}$. From the results of the FWM and PP experiments, the values of γ_{pure} are estimated to be $\gamma_{pure}^x=0.08\pm0.02$ μeV and $\gamma_{pure}^y=0.15\pm0.07$ μeV, respectively. Thus, the obtained γ_{pure} is much smaller than γ_r. Nevertheless, small but non-negligible values of γ_{pure} still exist for our QDs, though they are much smaller than those for other In(Ga)As QDs. As shown in Fig. 3(b), the value of γ_{pure} significantly increases with increasing temperature.[16] This means that the main cause of pure dephasing is exciton-phonon interactions.[3–5,7] Only at temperatures lower than 10 K, γ_{pure} is smaller than γ_r while γ_{nr} remains at zero at higher temperatures. At $T>20$ K, γ_h is dominated by γ_{pure}, which results in an isotropic γ_h. This demonstrates that the contribution of exciton-phonon interactions to γ_h is isotropic.

Since the minimal γ_{pure} is smaller than 0.1 μeV, it is difficult to estimate from the polarization-dependent FWM signals alone because of the measurement errors. Moreover, the polarization dependence of the FWM signals is not sensitive to γ_{pure}, which shows an anisotropy similar to that of T_r^{-1}. On the other hand, the combination of FWM and PP measurements as demonstrated enables us to estimate a value of γ_{pure} with an accuracy of 0.01 μeV, better than that from a FWM measurement alone. We believe that high accuracy measurement of γ_{pure} will have great advantages for investigating open questions regarding pure dephasing mechanisms, such as exciton-phonon interactions.

In conclusion, we have discovered extremely long dephasing times of excitons, which are very close to the radiative limit at low temperatures, in strain-compensated InAs QDs on an InP $(311)B$ substrate at telecommunications excitation wavelengths. The measured dephasing time T_2 shows a large anisotropy, reflecting the anisotropic radiative lifetime, which seems to be influenced by the substrate orientation. Consequently, the value of T_2 approaches 3 ns at 3 K for the $[01\bar{1}]$ polarization, which is the longest ever reported for QDs. Our simultaneous measurements of FWM and PP signals with a high signal-to-noise ratio enabled us to directly estimate the pure dephasing values with an accuracy of better than 0.1 μeV. A long T_2 is expected to be obtained over a wide range of excitation wavelengths from 1.4 to 1.55 μm because of the large inhomogeneous broadening of the transition wavelengths of QD excitons. Therefore, strain-compensated QDs are excellent candidates for quantum information devices operating at telecommunications wavelengths.

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