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Spectral broadening and population relaxation in a GaAs interfacial quantum dot ensemble and quantum well nanostructure

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Optical two-dimensional Fourier transform spectroscopy (2DFTS) is used to measure thermal broadening of the ground state exciton homogeneous linewidth and energy relaxation in a single layer of interfacial GaAs quantum dots (QDs). We observe a nonlinear increase in the homogeneous linewidth with temperature from 6 to 50 K and find that a phonon activation term + offset fit the data well. The absence of an activation peak in the 2D spectra indicates that elastic exciton–phonon scattering via virtual transitions between the ground and excited states significantly contributes to the thermal broadening. Measurements of the linewidth across the inhomogeneous distribution show that excitons localized in smaller QDs experience stronger thermal broadening. We record 2D spectra for a population time up to 500 ps and temperature up to 50 K to reveal excitonic relaxation from quantum well to QD states. As the temperature increases from 6 to 50 K, the relaxation time decreases from 180 to 30 ps, respectively. This behavior is consistent with a phonon-mediated scattering process.

1 Introduction The dephasing and coupling mechanisms governing the coherent optical response of semiconductor quantum dots (QDs) provide insight into fundamental processes that limit the lifetime of the excitonic polarization. The dephasing time (inversely proportional to the homogeneous linewidth) is affected by the QD surroundings, such as lattice temperature and exciton population density. In QDs, exciton–phonon interactions were thought to be suppressed because of the discrete density of states [1], yet a strong thermal component of the homogeneous linewidth is observed. Thermal broadening was initially attributed to phonon activation of the exciton population to higher-lying states [2]; however more recent results indicate that population decay contributes weakly to the thermal broadening and that the dominant broadening mechanism is *elastic* exciton–phonon coupling [3, 4]. The majority of studies have focused on measuring the temperature dependence of the lineshape or dephasing rate to deduce the coupling mechanisms, but have not provided direct evidence of the nature of the exciton–phonon coupling. In addition to thermal broadening, relaxation of delocalized quantum well (QW) excitons into QD states can strongly affect the coherent response.

In order to elucidate the dominant thermal broadening and coupling mechanisms in a QD ensemble, we use optical two-dimensional Fourier transform spectroscopy (2DFTS) to unfold three-pulse transient four-wave mixing (TFWM) signals onto two spectral dimensions. By correlating the phase of the signal with the phases of the excitation pulses, we are able to separate the homogeneous and inhomogeneous linewidths and we can observe incoherent relaxation processes unambiguously. We present measurements of the homogeneous linewidth and show that it increases nonlinearly with temperature from 6 to 50 K. Our experiment records the dynamics of the entire QD ensemble, allowing us to determine the homogeneous linewidth as a function of QD size, and we show that excitons localized in smaller QDs couple more strongly to acoustic phonons. We record 2D spectra at various population times and observe relaxation of excitons from QW to QD states. An increase in the relaxation rate with temperature suggests that the process is mediated by exciton–phonon scattering of the QW excitons.

2 Sample and experimental technique The epitaxially grown sample consists of a single GaAs layer 4.2 nm thick (15 monolayers), confined between two 35 nm thick $Al_{0.3}Ga_{0.7}As$ barriers, shown as a schematic in Fig. 1a. Growth interruption wait times on the order of tens of seconds result in monolayer width fluctuations in the QW thickness, and these fluctuations form island-like features that are the interfacial QDs. Upon optical excitation, excitons are created in the QD ensemble and are weakly confined in three-dimensions, while excitons created in the QW are delocalized, only confined in one-dimension. The QW spectral features are inhomogeneously broadened due to high-frequency width fluctuations at the bottom $Al_{0.3}Ga_{0.7}As$/GaAs interface, inherent from the growth process. Inhomogeneity of the QD ensemble is due to the sample having a distribution of QD sizes.

The excitonic wavefunction in a QD is confined in the growth direction by the GaAs/$Al_{0.3}Ga_{0.7}As$ band mismatch and in the lateral directions by the monolayer width difference in the QW thickness. Lateral localization further confines the exciton relative to the QW by ≈10 meV based on the QD/QW peak energy separation. Comparing energy eigenvalues obtained from a solution to Schrödinger's equation with observed photoluminescence excitation (PLE) spectra indicates that the QDs have a lateral size in the range of 25–50 nm.

Multiple techniques have been used to measure the homogeneous linewidth of the ground state exciton, but they are limited to studying systems with zero or initially known inhomogeneity. Spectral broadening of excitons in a single QD has been studied using PLE, but these experiments necessarily neglect interactions between QDs. Spectrally resolved or time-integrated TFWM can measure the homogeneous linewidth in an inhomogeneously broadened ensemble as long as knowledge of the nature of the broadening is known [5]; however, distinguishing between the numerous spectral contributions to the signal is difficult.

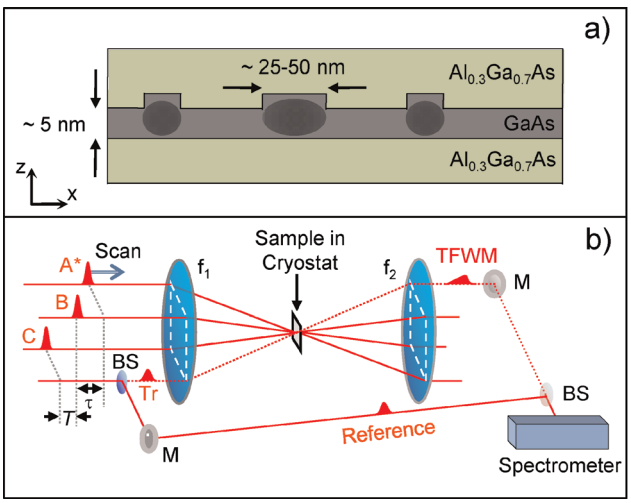

Figure 1 (online color at: www.pss-b.com) Schematic diagram of (a) the sample structure and (b) the 2DFTS experimental setup.

2DFTS has the advantage of being able to unfold complicated TFWM signals onto 2D, allowing for the measurement of the homogeneous and inhomogeneous linewidths simultaneously and for the separation of the contributing mechanisms to the signal.

In 2DFTS, the phase of the signal is correlated with the phases of the excitation pulses, which permits a Fourier transform of the time delays to be taken. The phase-stabilized pulses generate a nonlinear multi-dimensional time-domain signal in the phase-matched direction $\boldsymbol{k}_s = -\boldsymbol{k}_a + \boldsymbol{k}_b + \boldsymbol{k}_c$. In the experiments reported in this paper, the signal is recorded as the mixing time τ (time between the first two pulses A^* and B) is stepped with interferometric precision. The population time T (time between pulses B and C) is held constant for a given spectrum. The signal is heterodyne-detected with a phase-stabilized reference and is spectrally resolved with a resolution of 17 μeV.

Figure 1b shows the experimental setup [6]. A mode-locked Ti/sapphire laser produces 100-fs pulses at a repetition rate of 76 MHz. Each pulse is split into four with interferometric delay control and the pulses are incident on the sample in the box geometry. The pulses are centered spectrally over the QD resonance and are broad enough to excite both the QD and QW excitons, but not the continuum. The excitation beams overlap on the sample with a spot diameter of 80 μm, enclosing ≈10^5 QDs. The pulses are co-linearly polarized and the average beam power incident on the sample is set to 1.0 mW (1×10^{12} photons pulse^{-1} cm^{-2}). The emission axis of a 2D spectrum is constructed by spectrally resolving the signal, while the absorption axis is obtained by Fourier transforming the signal with respect to τ. The absorption axis is $-\hbar\omega_\tau$ in these experiments because the first pulse is conjugated.

Figure 2 shows 2D amplitude spectra at a temperature of 50 K for $T =$ (Fig. 2a) 200 fs, (Fig. 2b) 10 ps, and (Fig. 2c) 20 ps. The QW exciton and biexciton resonances are peaked at an absorption energy of 1648 meV and the biexciton is red-shifted along the emission energy axis by the biexciton binding energy. The QD ensemble is red-shifted by 9 meV from the QW and has an asymmetric inhomogeneous full-width at half-maximum of ≈2 meV. The homogeneous and inhomogeneous linewidths, labeled in Fig. 2b, are measured along the cross-diagonal and diagonal (dashed line) of the spectrum, respectively. A relaxation peak (RP) is observed at long T at the absorption energy of the QW and emission energy of the QDs. The spectra are normalized to the QW peak and truncated in order to highlight the RP.

3 Results and discussion

3.1 Homogeneous linewidth thermal broadening The homogeneous linewidth, γ, is related to the population decay rate, Γ, and the pure dephasing rate, γ^*, by $\gamma = (\Gamma/2) + \gamma^*$. We measure the homogeneous linewidth by fitting a cross-diagonal slice of a 2D amplitude spectrum to a $\sqrt{\text{Lorentzian}}$ [7]. The homogeneous linewidth measured at line center of the inhomogeneous distribution for $T = 200$ fs is shown in Fig. 3a as a function of

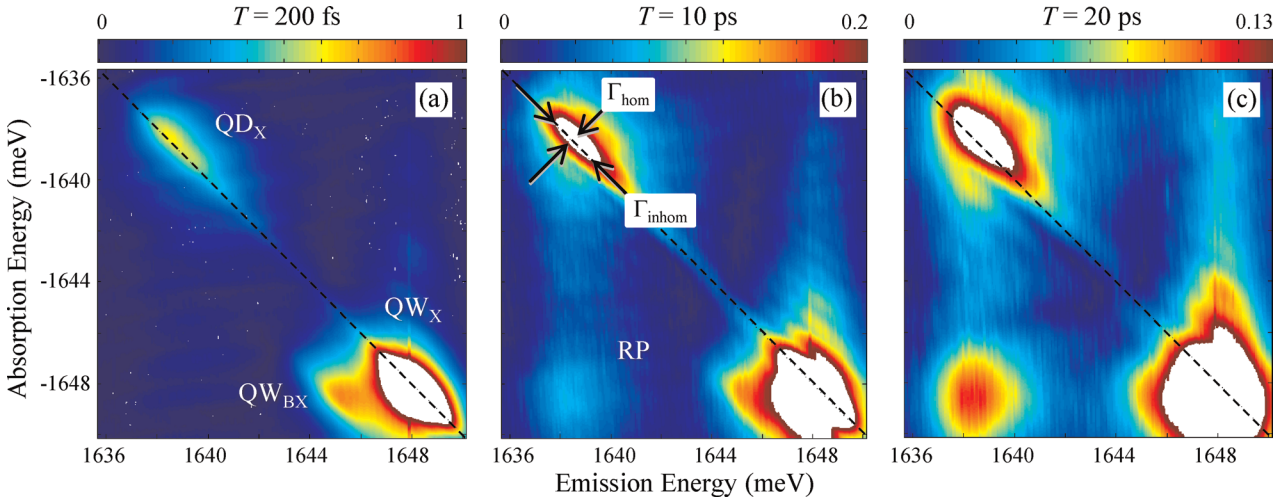

Figure 2 (online color at: www.pss-b.com) The QW exciton (QW_X), QW biexciton (QW_{BX}), QD exciton ensemble (QD_X), and QW → QD RP are shown on the 2D amplitude spectra at a temperature of 50 K and T = (a) 200 fs, (b) 10 ps, and (c) 20 ps. The homogeneous and inhomogeneous linewidths are measured along the cross-diagonal and diagonal lines, respectively. The spectra are normalized to the QW_X peak and truncated to emphasize the RP.

temperature. Thermal broadening of exciton linewidths in QDs has been modeled using an activation term + offset, which describes exciton population activation to a higher-lying state by one-phonon absorption [2]. We fit this model to the data (solid line in Fig. 3a) and extract an activation energy of $E_{12} = 4.4$ meV and an offset of $\gamma_1^* = 0.11$ meV. The offset is due to a combination of radiative broadening and excitation-induced dephasing, and we measure a low temperature and extrapolated zero excitation-density value of 30.6 μeV. Filtering the excitation spectrum such that neither QW excitons nor carriers are created has no apparent effect on the measured linewidths. While PLE spectra reveal

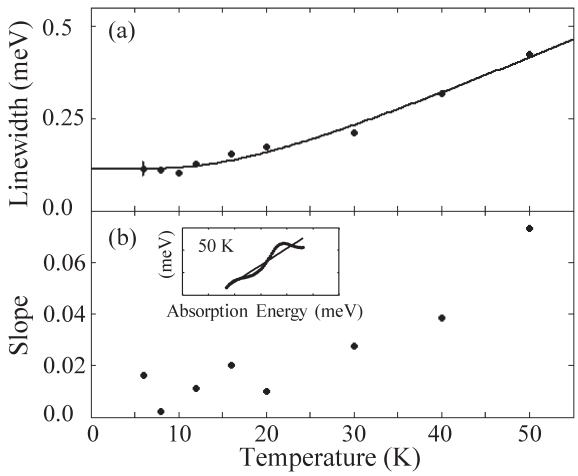

Figure 3 (a) Thermal broadening of the homogeneous linewidth for T = 200 fs (points), with a representative error bar obtained from repeating the measurement at 6 K. The line shows the phonon activation term + offset. (b) The slope of a linear fit to the QD homogeneous linewidth measured across the inhomogeneous distribution. Inset: Homogeneous linewidth as a function of absorption energy at 50 K and T = 200 fs (points) and linear fit (line).

energy levels with a separation similar to our value of E_{12}, we eliminate population activation as a possible thermal broadening mechanism because we do not observe an activation peak in the 2D spectra at long T [8]. If the thermal broadening was due to activation, an additional peak would be observed blue-shifted from the QD line center by 4.4 meV along the emission axis. Development of a peak at the QD absorption and QW emission energies indicates QD → QW activation; however this process occurs in a time over an order of magnitude larger than the QD dephasing time and is not expected to contribute to the QD homogeneous linewidth thermal broadening.

The absence of an activation peak indicates that thermal broadening is a result of population decay and pure dephasing. Previous studies show that population decay contributes only weakly to the thermal broadening [3] and that pure dephasing is the dominant broadening mechanism. Calculations based on the Huang–Rhys theory predict that for excitons weakly confined in GaAs QDs, exciton–phonon coupling can drive *virtual* transitions from the ground state → excited state → ground state that significantly contribute to the exciton dephasing with no effect on the population [3, 9]. These calculations support our observation of an activation-like thermal broadening without the observation of an activation peak in the 2D spectra [8]. Our observations are direct evidence that elastic exciton–phonon scattering dominates the thermal broadening of the homogeneous linewidth.

We measure the homogeneous linewidth across the inhomogeneous distribution and perform a linear fit to the measured widths. Oscillations are due to time-truncation artifacts of the TFWM signal. As the energy increases across the distribution, the QD size decreases. The slope of the fitted line increases with temperature above 25 K, shown in Fig. 3b. An increase in the slope with temperature indicates that smaller QDs experience greater thermal broadening.

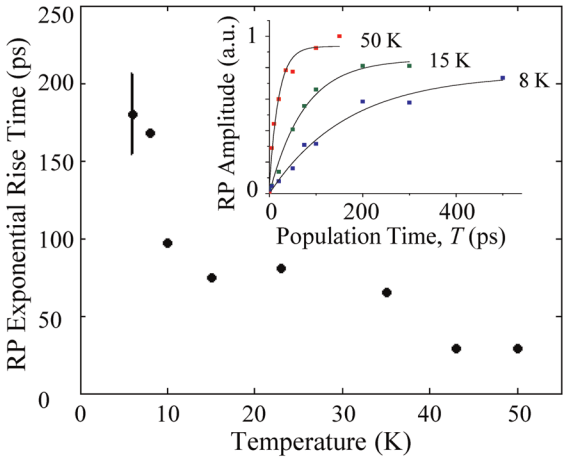

Figure 4 (online color at: www.pss-b.com) QW → QD RP exponential rise time as a function of temperature. Inset: RP amplitude (points) and exponential fit (line) as a function of population time T for temperatures of 8, 15, and 50 K.

This result is in agreement with the model in Ref. [9], which predicts stronger exciton–phonon coupling for smaller QDs.

3.2 QW → QD exciton relaxation While photoluminescence spectroscopies can be used to study transfer phenomena, the non-resonant generation of free carriers strongly influences the exciton dynamics. In order to provide a clear picture of exciton transfer dynamics, we record 2D spectra as a function of temperature and population time T and observe incoherent relaxation of QW excitons to QD states. Because the QW and QD states are independent systems, symmetric cross peaks above and below the diagonal indicating Raman coherences [10] and ground state bleaching are absent. The signal phase does not evolve during T for incoherent processes; therefore energy relaxation dynamics occurring during T will be visible as additional peaks in the 2D spectra, shown in Fig. 2b and c. The RP grows in amplitude with increasing T and lattice temperature, consistent with an energy relaxation process. The RP amplitude is fit to an asymptotically rising exponential for a given temperature, shown in the inset of Fig. 4. The extracted rise time, shown in Fig. 4, decreases from 180 ps at 6 K to 30 ps at 50 K. In this relaxation process, QW excitons are scattered by the temperature-dependent phonon population and subsequently localize in QD states while emitting acoustic phonons to conserve energy and momentum.

4 Conclusion In summary, we use optical 2DFTS to study homogeneous linewidth thermal broadening and energy relaxation of an ensemble of interfacial GaAs QDs. By unfolding spectra onto 2D, we are able to measure the size-dependent homogeneous linewidth of the QD ensemble. The homogeneous linewidth increases nonlinearly with temperature and this behavior is modeled with an activation term + offset. Combining the nonlinear temperature dependence with the absence of an activation peak in the 2D spectra reveals that elastic exciton–phonon coupling is the dominant thermal broadening mechanism. The homogeneous linewidth is measured across the inhomogeneous distribution and the thermal broadening is stronger for smaller QDs. We observe incoherent exciton population relaxation from QW to QD states. The relaxation rate increases with population time and temperature, indicating that QW excitons relax to QD states via exciton–phonon scattering.

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