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Homogeneous linewidths of excitons in CdTe/(Cd,Zn)Te single quantum wells

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The homogeneous linewidth Γ_h of the lowest exciton state in CdTe/(Cd,Zn)Te single quantum wells (QW's) has been analyzed as a function of QW thickness, temperature, and exciton density. The zero-temperature and density limit of Γ_h suggests the presence of an ordering mechanism that decreases the effective interface roughness for the narrower QW's in the CdTe/(Cd,Zn)Te system. The exciton–acoustic-phonon and the exciton-exciton scattering are of comparable strength as in GaAs QW's, indicating the absence of strong localization effects, present in other II-VI QW systems.

The II-VI quantum-well (QW) systems are currently attracting general interest, mainly due to their potential as short wavelength light emitters.[1,2] They are typically characterized by enhanced excitonic features with profound consequences on both their optical properties and potential device applications. Recently, in a ZnSe QW, excitons with about 40 meV binding energy[3] were found to play a major role in the stimulated emission process.[4]

Here, we present a comprehensive study of exciton dephasing in CdTe/(Cd,Zn)Te QW's as a function of QW thickness L, temperature, and excitation density utilizing degenerate-four-wave-mixing (DFWM). The loss of coherence is characterized by the dephasing time T_2, which in turn defines the homogeneous linewidth of the excitonic transition as $\Gamma_h = 2\hbar/T_2$.

For our studies, we used a series of single CdTe/$Cd_{1-x}Zn_xTe$ QW's with $x = 16-18$ % and L between 18 and 112 Å. All samples were grown by molecular beam epitaxy (MBE) on (100) $Cd_{0.96}Zn_{0.04}Te$ substrates. Details of the growth conditions are published elsewhere.[5] The samples were characterized by cw photoluminescence (PL) and reflectivity measurements. In comparison to previously studied II-VI QW's,[6–8] the CdTe/(Cd,Zn)Te system is distinguished by narrow exciton linewidths and small Stokes shifts. For selective, resonant excitation of the e_1h_1 exciton in the DFWM experiments, we reduced the bandwidth of 120-fs pulses from a mode-locked Ti:sapphire laser operating at 76 MHz to 4 meV (corresponding to 0.8 ps) by external spectral filtering. For incident photon wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, the time-integrated DFWM signal was detected in the self-diffraction backward reflection geometry $\mathbf{k}_s = 2\mathbf{k}_2 - \mathbf{k}_1$.

Typical PL and reflectivity spectra at $T = 2$ K are shown in the inset of Fig. 1 for the 18-Å QW sample. Both traces are dominated by the e_1h_1 transition. From a fitting analysis of these spectra taking into account the homogeneous linewidth determined by the DFWM experiments, we obtain the inhomogeneous exciton linewidth $\Gamma_{\rm inh}$ and the Stokes shift S plotted in Fig. 1 as a function of QW thickness L. For all L, both quantities remain remarkably small ($1 \leq \Gamma_{\rm inh} \leq 2$ meV and $0.1 \leq S \leq 1$ meV). This implies that the interface roughness in CdTe/(Cd,Zn)Te is different than in GaAs/(Ga,Al)As. In GaAs QW's, the formation of monolayer (ML) islands of lateral size comparable to the exciton Bohr radius a_B is the most common interface model used to account for $\Gamma_{\rm inh}$. This model obviously fails in the present case. The expected $\Gamma_{\rm inh}$ originating from fluctuations by ± 1 ML in the 18-Å QW is 4 meV, while the experimental $\Gamma_{\rm inh} = 1.3$ meV. We interpret the small $\Gamma_{\rm inh}$ and S values as a clear indication for the formation of pseudosmooth CdTe/(Cd,Zn)Te heterointerfaces, i.e., ML roughness on a length scale much smaller than a_B ($a_B \leq 70$ Å in our system). In this case, the roughness is averaged out by the exciton wave function, leading to narrower linewidths.[9]

The most striking information of Fig. 1 is the narrowing of $\Gamma_{\rm inh}$ and decrease of S observed for decreasing QW width. The formation of pseudosmooth interfaces alone cannot account for this behavior. For a fixed interface morphology,

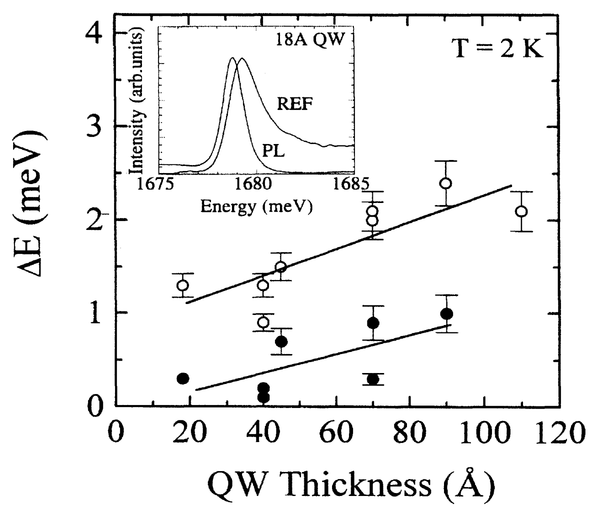

FIG. 1. PL linewidths (open circles) and Stokes shifts (closed circles) in CdTe/(Cd,Zn)Te single QW's at $T = 2$ K, as a function of QW thickness. In the inset, the PL and reflectivity (REF) spectra for the 18-Å QW are shown.

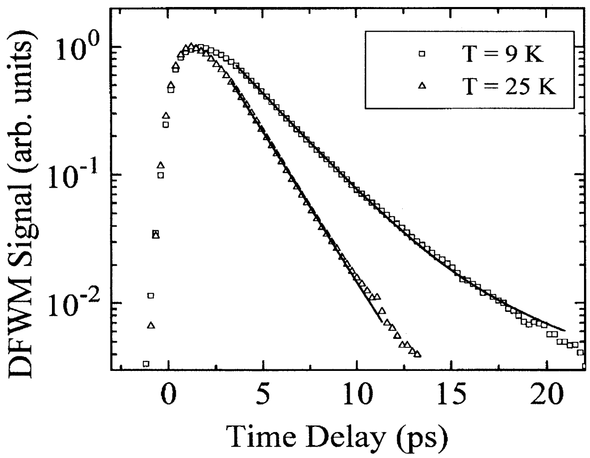

FIG. 2. Diffracted intensity as a function of time delay, for a CdTe/(Cd,Zn)Te 18-Å single QW, at $T=9$ K (open squares) and 25 K (open triangles). The solid lines are theoretical fits. The 9-K curve is fitted by a biexponential function and the 25-K one by a single exponential.

one still expects an increasing $\Gamma_{\rm inh}$ with decreasing L.[9] One possible explanation is that the two QW heterointerfaces are increasingly correlated as the QW thickness decreases. The idea here is that, for sufficiently thin QW's, the growth of the QW layer occurs in a way that preserves the roughness topography of the first heterointerface and results in a reduced average thickness fluctuation along the QW plane. Such a behavior has already been detected in metallic heterostructures,[10] but to our knowledge never in semiconductors.

In Fig. 2, we show the DFWM signal versus time delay for the 18-Å QW sample, for 9 K and 25 K. In the 9-K curve, we distinguish a biexponential decay with a fast ($t_D=2.8$ ps) and a slow ($t_D=11.5$ ps) decay time component. We attribute the fast decay to free excitons and the slow to a small population of weakly localized excitons.[11] This interpretation is supported by the disappearence of the slow decay at 25 K. The presence of weak localization is further confirmed by the small Stokes shift of the PL line. The contribution of weakly localized excitons to the DFWM signal amounts to $\approx$ 2%. In the rest of the paper, we only consider the fast time decay. The signal decay time t_D is much smaller than $2\hbar/\Gamma_{\rm inh}$, confirming that the e_1h_1 transition in these samples is inhomogeneously broadened. In this case, $T_2=4t_D$ (Ref. 12) and $\Gamma_h=\hbar/2t_D$.

We investigated the contribution of the exciton–acoustic-phonon interaction to Γ_h, by measuring Γ_h as a function of temperature T, in the range 10–40 K for a constant exciton density of $n_X\approx5\times10^9$ cm^{-2}. The plot of Γ_h versus T for the 18-Å QW in Fig. 3(a) displays a linear increase of Γ_h in the range 9–30 K,

$$\Gamma_h(T,n_X)=\Gamma_h(T=0,n_X)+\gamma_{XP}T. \quad (1)$$

This relation observed for all samples is due to anti-Stokes scattering of excitons by acoustic phonons. γ_{XP} is a measure of the exciton–acoustic-phonon coupling strength. $\Gamma_h(T=0,n_X)$ is the low temperature limit of Γ_h and contains contributions due to collisions with other excitons, scattering

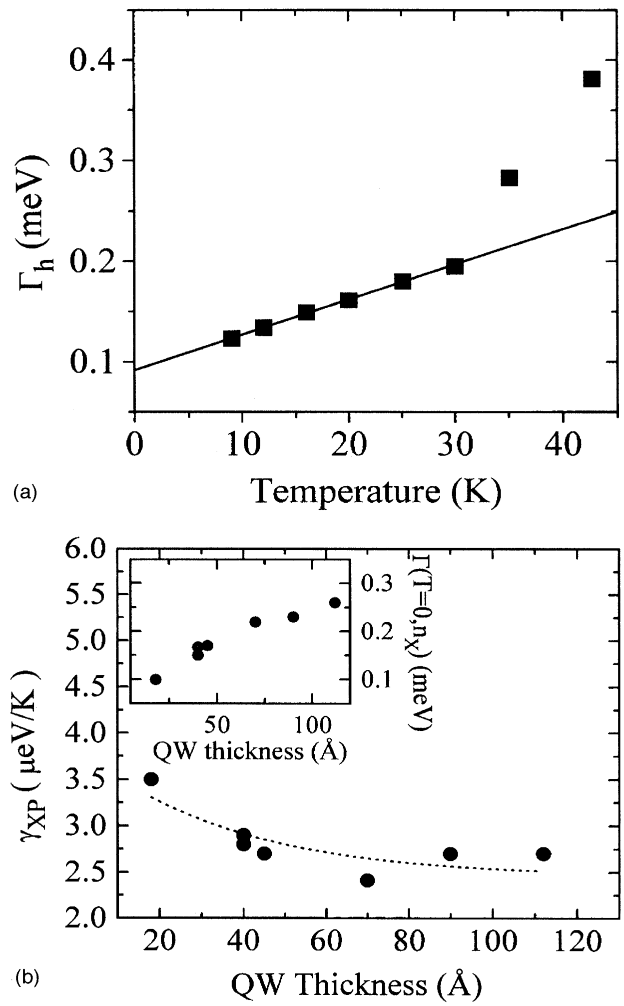

FIG. 3. (a) Homogeneous linewidth Γ_h of the 18-Å QW as a function of temperature. The solid line is a linear fit. (b) Exciton–acoustic-phonon coupling strength γ_{XP} vs QW thickness. The dashed line is to guide the eye. The inset shows the zero-temperature limit of Γ_h as a function of QW thickness. The incident exciton density was kept constant $n_X=5\times10^9$ cm^{-2}.

by crystal imperfections, or radiative recombination. For $T\geqslant30$ K, Γ_h increases superlinearly owing to the onset of exciton–LO-phonon scattering, occurring at lower temperatures in CdTe as compared to GaAs, due to the smaller LO-phonon energy $\hbar\omega_{\rm LO}$ and stronger exciton–LO-phonon coupling. We can estimate the contribution of this process to Γ_h by

$$\Delta\Gamma=\frac{\Gamma_{\rm LO}}{[\exp(\hbar\omega_{\rm LO}/k_BT)-1]}, \quad (2)$$

where $\Gamma_{\rm LO}$ is a constant representing the strength of the exciton–LO-phonon coupling. For $\Gamma_{\rm LO}\approx$ 30 meV (Ref. 13) and $\hbar\omega_{\rm LO}=21$ meV, we estimate $\Delta\Gamma$ = 0.07 meV at $T=40$ K, in reasonable agreement with our data.

Figure 3(b) shows γ_{XP} and $\Gamma_h(T=0,n_x)$ (inset) as a function of QW thickness. In the range between 70 Å and 112 Å, no significant variation of γ_{XP} about an average value of 2.5

μeV/K occurs. Below $L=70$ Å γ_{XP} slightly increases with decreasing L. In order to stress the significance of the experimental result, it should be mentioned that the 18-Å and 45-Å well and also one 40-Å and the 70-Å well were grown during the same MBE run on a single substrate. These wells, expected to have the same high sample quality, exhibit a pronounced well width dependence of both γ_{XP} and $\Gamma_h(T=0,n_x)$. An increasing exciton–acoustic-phonon coupling with increasing two-dimensional (2D) confinement has been theoretically predicted.[14,15] The experiment reveals, however, a much weaker effect than the $1/L$ dependence calculated in Ref. 15. Moreover, this effect seems to play a role only in the 2D limit and becomes negligible for $L \geqslant a_B$.

The γ_{XP} are five times smaller than the value measured on CdTe/(Cd,Mn)Te QW's (Ref. 7) and comparable to the lower values available for GaAs QW's.[16,17] The previous assertion that the exciton–acoustic-phonon interaction is enhanced in CdTe QW's as compared to GaAs QW's (Ref. 7) is not supported by our results. Probably, part of the strong temperature dependence of the exciton dephasing rate observed in the CdTe/(Cd,Mn)Te QW's originates from a temperature dependent exchange interaction between the exciton spin and the spin of the Mn ions.

The role of exciton-exciton interaction as a dephasing mechanism can be explored by intensity dependent DFWM. In Fig. 4(a), we plot Γ_h versus exciton density n_X for the 18-Å sample, at $T=9$ K. For low n_X, Γ_h is expected to have a linear dependence on n_X,

$$\Gamma_h(n_X,T)=\Gamma_h(n_X=0,T)+\gamma_{XX}a_B^2E_Bn_X, \quad (3)$$

where γ_{XX} represents the strength of the exciton-exciton interaction and E_B the exciton binding energy. We verified the linear dependence in all samples. In order to evaluate γ_{XX}, we take that, as the QW width decreases from 112 down to 18 Å, a_B decreases from 65 to 58 Å, E_B increases from 16 to 18 meV, and the exciton oscillator strength increases by 30% over a $f_X=1.7\times10^{-3}$ Å^{-2} value, measured in a 100-Å QW.[18] These values are consistent with experimental data for E_B (Ref. 13) as well as variational calculations.[19] It should be noticed that the confinement effects in the CdTe QW's are relatively small as compared to the GaAs/(Al,Ga)As QW's because of the shallow potential barriers (80 meV for electrons and 25 meV for the heavy holes).

Figure 4(b) shows an appreciable reduction of γ_{XX} for the thinner QW. The origin of this reduction remains at present unclear. It could be tentatively attributed to an increasing exciton localization in the narrower wells. This interpretation, however, is not supported by the following considerations. First, the results of Fig. 1 give no evidence of increasing localization with decreasing L, rather $\Gamma_{\rm inh}$ and S, usual measures of localization, seem to decrease. Second, Γ_h doubles for rather small exciton densities of $n_X\approx10^{10}$ cm^{-2} [Fig. 4(a)] corresponding to an average interparticle distance of $\approx 20a_B$. For an efficient exciton-exciton interaction at such a large interparticle distance ($\geqslant 1000$ Å), the excitons ought to be at least quasidelocalized. Furthermore, we note that the γ_{XX} are comparable to those in GaAs QW's. For example, in a 120-Å GaAs/(Ga,Al)As QW, $\gamma_{XX}=1.5$.[16] The slightly larger value of $\gamma_{XX}=2.5$ observed here for a 60-Å well (this thickness yields the same ratio of

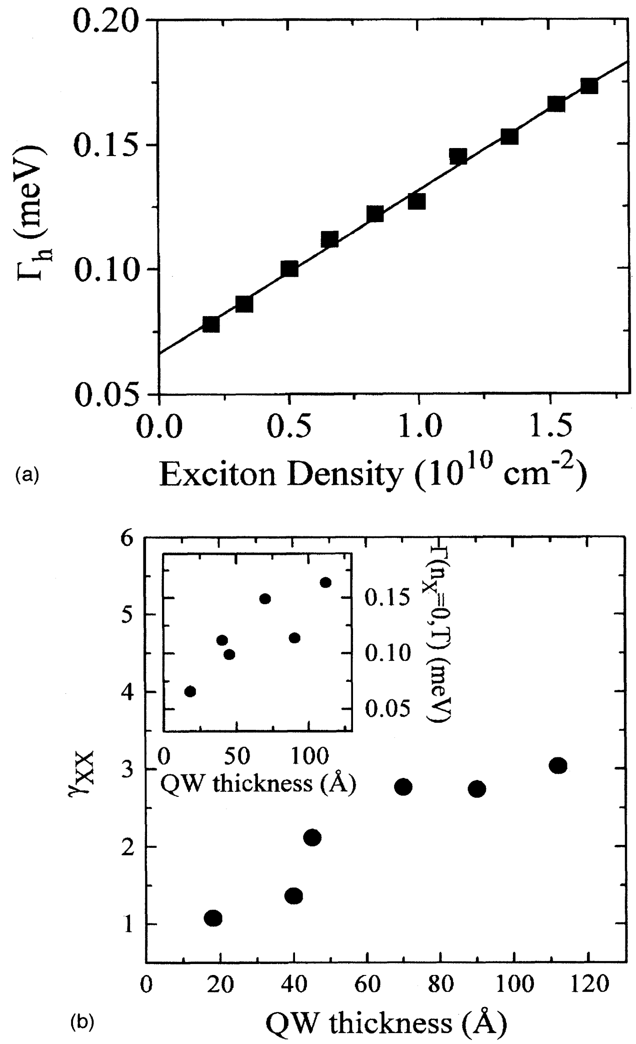

FIG. 4. (a) Homogeneous linewidth Γ_h of the 18-Å QW as a function of exciton density, at $T=9$ K. The solid line is a linear fit. (b) Exciton-exciton interaction strength γ_{XX} vs QW thickness. The inset shows the zero-density limit of Γ_h as a function of QW thickness, at $T=9$ K.

L/a_B) is probably explainable by the weaker screening associated with the smaller dielectric constant. On the other hand, γ_{XX} was found to be about 20 times smaller in a CdTe/(Cd,Mn)Te QW sample, where efficient localization occurred.[7]

In the inset of Fig. 4(b), we show $\Gamma_h(n_X=0,T)$ for the various QW thicknesses. A clear tendency of smaller $\Gamma_h(n_X=0,T)$ is observed for decreasing L. To magnify this observation, we plot in Fig. 5 $\Gamma_h(n_X=0,T=0)$ versus QW width, after subtraction of the finite contribution of exciton–acoustic-phonon interaction at $T=9$ K to Γ_h. We observe a strong decrease of $\Gamma_h(n_X=0,T=0)$ for smaller L. In interpreting these results, we exclude any localization effects based on the same arguments presented in the previous paragraph. We also exclude the possibility that $\Gamma_h(n_X=0,T=0)$ is dominated by radiative recombination because the recombination rate, being proportional to the exciton oscillator strength,[20] is expected to increase for the narrower QW's,

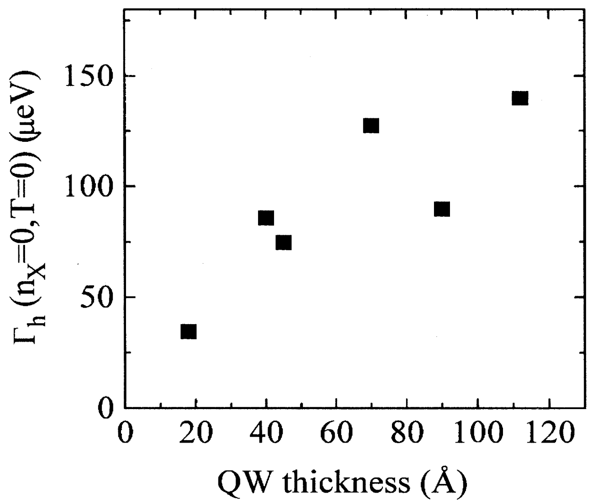

FIG. 5. Homogeneous linewidth Γ_h, at zero temperature and exciton density, as a function of QW thickness.

opposite to the behavior shown in Fig. 5.[21] We also eliminate impurity scattering since our samples are nominally undoped, with a residual dopant concentration $\leq 10^{10}$ cm^{-2}. Since the ionized impurity concentration below 40 K remains less than 10^8 cm^{-2}, exciton-impurity scattering should be negligible. Moreover, the decrease of Γ_h for the narrower QW's for which the excitons increasingly penetrate into barriers, implies that small barrier alloy fluctuation have a negligible contribution to Γ_h. This is further supported by the weak dependence of the e_1h_1 confinement energy on the Zn content.

These considerations allow us to conclude that $\Gamma_h(n_X=0,T=0)$ is dominated by scattering on interface roughness. At first sight, the results of Fig. 5 are puzzling, because one intuitively expects that the role of interface roughness is enhanced in narrow QW's, leading to more dephasing events and larger Γ_h. This tendency has been confirmed in GaAs QW's, where both Γ_h and Γ_{inh} increase with decreasing QW width. The opposite, however, occurs in the CdTe/(Cd,Zn)Te QW's. We correlate the observed reduction of Γ_h for the narrower CdTe/(Cd,Zn)Te QW's with the parallel reduction of Γ_{inh}, as seen in Fig. 1. Both results are a manifestation of increasing order in the narrower QW's, reducing the effective interface roughness.

In summary, we analyzed the main contributions to the homogeneous linewidth of CdTe/(Cd,Zn)Te single QW excitons as a function of QW thickness. The exciton–acoustic-phonon scattering rate is independent of QW thickness in the range 70–112 Å, and shows a significant increase with decreasing well width between 70 and 18 Å in accordance with theoretical expectations. On the other hand, the strength of exciton-exciton interaction shows an appreciable decrease with decreasing well width below 70 Å. Both scattering processes are of comparable strength as in GaAs QW's, indicating the absence of strong localization effects present in other II-VI QW systems. The zero-temperature and density limit of Γ_h mainly due to scattering by interface roughness shows a remarkable decrease with decreasing QW width. Associating this effect with a parallel decrease of the inhomogeneous linewidth in the narrower QW's we suggest that an increasing interface ordering occurs for the thinner QW's in this material.

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[21] In GaAs QW's, the calculated radiative recombination time τ_0 for a 100-Å GaAs/(Ga,Al)As QW of $\approx$25 ps is much longer than other dephasing channels. However, the oscillator strength of a 100-Å CdTe/(Cd,Zn)Te QW is three times larger than in a 100-Å GaAs QW. Hence, we would expect radiative recombination to be detectable in coherent DFWM experiments, which is not the case.