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Homogeneous linewidth of excitons in semimagnetic CdTe/Cd$_{1-x}$Mn$_x$Te multiple quantum wells

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We report on a detailed experimental study of the homogeneous linewidth of heavy-hole excitons in semimagnetic CdTe/Cd$_{1-x}$Mn$_x$Te multiple quantum wells. Transient four-wave-mixing experiments are used to obtain the homogeneous linewidth by measuring the dephasing time of the excitons. We find a dephasing time of 2.2 ps corresponding to a homogeneous linewidth of 0.6 meV at low temperature and moderate exciton density. The homogeneous linewidth increases linearly with both exciton density and lattice temperature. The contributions of exciton–acoustic-phonon scattering, exciton-exciton interaction, and disorder to the homogeneous linewidth are determined.

Optical spectroscopy of excitons in II-VI semiconductor quantum wells (QW's) has attracted increasing interest[1-6] recently due to the technological progress in the growth of the respective heterostructures and their possible applications. In addition, a comparison of the optical properties of quantum wells based on wide-gap II-VI semiconductors with III-V quantum wells is of fundamental interest. For example, the question arises whether at room temperature in II-VI heterostructures lasing takes place on excitonic rather than on free carrier transitions.[7]

Insight into the exciton dynamics is provided by time-resolved spectroscopy. In particular, transient four-wave-mixing (FWM) experiments allow a direct measure of the exciton dephasing time T_2, and thus a determination of the homogeneous linewidth by $\Gamma_{\text{hom}}=2\hbar/T_2$, even in the case of a strongly inhomogeneously broadened optical transition. The dephasing time T_2 and thus the homogeneous linewidth Γ_{hom} of excitons is determined by elastic and inelastic scattering processes with phonons, excitons and crystal defects, such as impurities, interface roughness, and alloy fluctuations. Therefore, a detailed study of Γ_{hom} provides information about the various scattering processes and offers the possibility to determine quantitatively the various contributions, as has been demonstrated for III-V semiconductor bulk materials[8] and heterostructures.[9,10] However, for II-VI semiconductor heterostructures the knowledge about these interactions is less elaborate and particularly for semimagnetic II-VI heterostructures a detailed experimental analysis of the homogeneous linewidth of excitons and its dependence on crystal parameters is still lacking. In this paper, we present a detailed experimental study of the homogeneous linewidth of heavy-hole excitons in semimagnetic CdTe/Cd$_{1-x}$Mn$_x$Te quantum-well structures. We perform transient four-wave-mixing experiments in a two-pulse self-diffraction configuration in a reflection geometry. The homogeneous linewidth of the exciton transition increases linearly with both increasing temperature and exciton density. On the basis of our data we estimate the coupling strengths for exciton–acoustic-phonon and exciton-exciton scattering and compare these values to those reported for GaAs/Al$_x$Ga$_{1-x}$As QW's.[9,10]

We have studied a CdTe/Cd$_{0.86}$Mn$_{0.14}$Te multiple-quantum-well (MQW) structure consisting of 50 periods of 85-Å-thick CdTe wells and 95-Å-thick Cd$_{0.86}$Mn$_{0.14}$Te barrier layers. In accordance with previous findings,[11] magnetic polaron formation due to the exchange interaction between excitons in the well and the manganese ions situated in the semimagnetic barriers can be neglected in these rather thick quantum wells with low Mn content in the barrier layer. Nevertheless, scattering between the exciton spin and the spin of the manganese ions may have to be taken into account as a possible scattering mechanism, giving an additional contribution to Γ_{hom}.

The structure was grown by molecular-beam epitaxy on (100)-oriented CdTe substrates with a 0.2-μm buffer layer of CdTe. The thickness of the MQW structure is well below the critical thickness for strain relaxation, providing excellent structural characteristics, as determined by x-ray analysis and optical methods. Optical characterization by means of photoluminescence-excitation (PLE) spectroscopy and reflectivity reveals the following features: in the reflection spectrum (see inset of Fig. 1) the resonances of the heavy- and light-hole excitons of the first ($n=1$) quantum confinement subband can be clearly resolved at 1.621 eV (hh) and 1.634 eV (lh). The energy splitting between the heavy- and light-hole excitons is 13 meV. The full width at half maximum of the 1*s* state of the heavy-hole exciton line in the PLE spectrum is found to be 4.7 meV and is attributed to inhomogeneous broadening due to well width fluctuations and alloy disorder.[12] Therefore, we consider the excitons to be free, i.e., not bound to impurities, but possibly localized by potential fluctuations induced by interface roughness and alloy disorder.

We used a tunable mode locked Ti-sapphire laser as ex-

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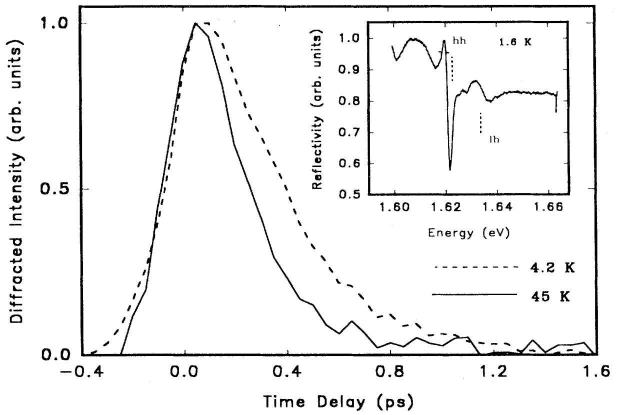

FIG. 1. Diffracted intensity from a CdTe/Cd$_{0.86}$Mn$_{0.14}$Te MQW structure ($L_z = 85$ Å; $L_B = 95$ Å) as a function of the delay time τ between the two exciting laser pulses for temperatures of 4.2 and 45 K. An exponential fit to the data yields dephasing times of 1.38 ps and 840 fs, respectively. The inset shows the reflection spectrum for the structure under study. The resonant energies of the light- and heavy-hole excitons are at 1.634 and 1.621 eV, respectively.

citation source for the FWM experiments. This laser system produces a train of 110-fs pulses (spectral width 22 meV) at a repetition rate of 76 MHz and was tuned slightly below the heavy-hole exciton transition in order to excite only the heavy-hole exciton. The sample was held in a temperature-variable helium flow cryostat. The experiments were performed in the temperature range from 4.2 to 45 K and in the exciton density range of 3.4×10^{11} to 5.5×10^{12} cm^{-2}. The exciton density was changed by varying the intensity of the exciting laser beams and estimated from the excitation power density assuming an absorption coefficient at the exciton resonance of 2×10^5 cm^{-1}.[13]

The T_2 times were measured in the two-pulse self-diffraction four-wave-mixing configuration in the reflection geometry,[14] allowing FWM experiments on structures with opaque substrates. In the two-pulse self-diffraction experiment the interaction of two exciting laser pulses having wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, separated by a time delay τ generates a nonlinear third-order polarization which gives rise to an electromagnetic field coherently emitted into the phase matched direction $2\mathbf{k}_2$-$\mathbf{k}_1$. However, for thin layers a nonlinear signal is also emitted in the backward direction, since in this particular case, besides energy, only the resulting wave vector parallel to the layer has to be conserved.[14] This nonlinear signal is detected by a slow photomultiplier, i.e., time integrated, as a function of the time delay τ. Its decay yields directly the dephasing time T_2: in the case of an exponential decay T_2 equals four times the decay constant of the FWM signal τ_{FWM} ($T_2 = 4\tau_{\mathrm{FWM}}$) for inhomogeneously broadened exciton transitions,[15] as is the case in the quantum wells under study.

Figure 1 shows two transient FWM signals measured at two different temperatures of 4.2 and 45 K and an exciton density of 1×10^{12} cm^{-2}. The decay times of the FWM signals, obtained by an exponential fit to the data, correspond to dephasing times T_2 of 1.38 ps and 840 fs, respectively. The decrease of the dephasing time in the temperature range between 4.2 to 45 K corresponds to a linear increase of Γ_{hom} from 0.95 to 1.56 meV, as shown in Fig. 2. The linear increase implies that acoustic-phonon scattering is the dominant temperature-dependent dephasing mechanism at temperatures below 45 K,[16] which can be described by[9,17]

$$\Gamma_{\mathrm{hom}}(T) = \Gamma_{\mathrm{hom}}(T=0) + \gamma_{\mathrm{ph}} T \ . \qquad (1)$$

In this formula γ_{ph} denotes the exciton–acoustic-phonon coupling strength whereas $\Gamma_{\mathrm{hom}}(T=0)$ accounts for exciton-exciton interaction and scattering of excitons by impurities, interface roughness, and alloy fluctuations.

Equation (1) is based on the assumption that the contribution of acoustic-phonon scattering to the homogeneous linewidth of optically excited excitons, i.e., excitons with wave vectors $K \simeq 0$, is solely determined by the absorption of acoustic phonons, i.e., anti-Stokes scattering. Thus, it depends on the phonon occupation number n_q, given by the Bose-Einstein statistics. For phonons with a given wave vector the occupation number increases at low temperatures approximately linearly with temperature, causing the observed linear dependence of Γ_{hom}.

From a linear regression to our data we find a coupling strength of $\gamma_{\mathrm{ph}} = (13.7\pm1.8)$ μeV/K and $\Gamma_{\mathrm{hom}}(T=0)$ is found to be 0.89 ± 0.05 meV. The obtained value for γ_{ph} is by a factor of 3 larger than the one reported by Kuhl *et al.* for a 135-Å-thick GaAs/Al$_x$Ga$_{1-x}$As QW.[9,18] Thus, we have to conclude that exciton–acoustic-phonon scattering is weaker in GaAs/Al$_x$Ga$_{1-x}$As QW's as compared to CdTe/Cd$_{1-x}$Mn$_x$Te heterostructures.[19] Possible contributions due to the exchange interaction between the exciton spin and the spin of the manganese ions will be discussed later.

We now turn to the discussion of the value of the

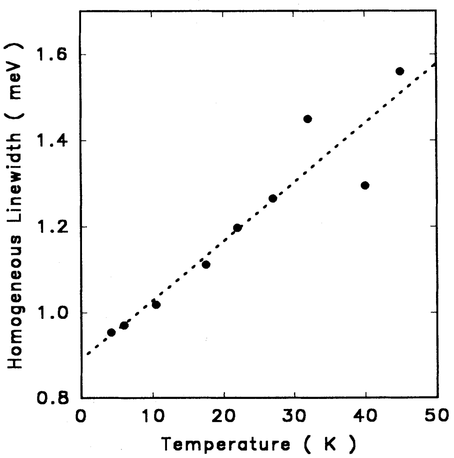

FIG. 2. Homogeneous linewidth of a CdTe/Cd$_{0.86}$Mn$_{0.14}$Te MQW structure as a function of the crystal temperature at a constant exciton density of 1×10^{12} cm^{-2} The dashed line represents a linear regression according to Eq. (1), which allows us to determine the coupling strength for exciton–acoustic-phonon scattering $\gamma_{\mathrm{ph}} = 13.7$ μeV/K.

homogeneous linewidth extrapolated for zero temperature, $\Gamma_{\text{hom}}(T=0)$. As considered above, exciton-exciton interaction may contribute to $\Gamma_{\text{hom}}(T=0)$ in addition to scattering by defects, impurities, alloy disorder, and interface roughness. We have determined the contribution of exciton-exciton scattering by measuring the dependence of Γ_{hom} on the excitation density at a constant temperature of 4.2 K. The experimentally obtained data are plotted in Fig. 3. The homogeneous linewidth increases linearly with density from 0.6 to 2.4 meV in the range studied. Note, that for densities higher than 5×10^{12} cm^{-2} the decay of the FWM signal is limited by the laser pulse width.

We attribute the linear increase of the homogeneous linewidth with exciton density to exciton-exciton scattering in accordance with FWM studies on other materials.[10] Exciton-free carrier scattering, which has been shown to be more efficient than exciton-exciton scattering,[10] can be excluded since the center laser frequency is tuned below the exciton resonance and thus no free carriers are excited optically. Furthermore, thermal dissociation of excitons is negligible at $T=4.2$ K.

The contribution of exciton-exciton scattering to the homogeneous linewidth can be approximately described by[20]

$$\Gamma_{\text{hom}}(n_x)=\Gamma_{\text{hom}}(n_x=0)+\gamma_{xx}a_B^2E_Bn_x\ , \qquad (2)$$

where a_B is the exciton Bohr radius (45 Å in our particular structure) and E_B the exciton binding energy (20 meV in the structure under study). γ_{xx} is a dimensionless parameter and gives a measure of the exciton-exciton interaction strength. $\Gamma_{\text{hom}}(n_x=0)$ is the density-independent contribution to the homogeneous linewidth which includes all residual interactions of the excitons with acoustic phonons, impurities, interfaces, and alloy fluctuations. According to Eq. (2) we obtain $\gamma_{xx}=0.16\pm0.01$ and $\Gamma_{\text{hom}}(n_x=0)=0.35\pm0.05$ meV at 4.2 K from the data shown in Fig. 3. The value for γ_{xx} is smaller by a factor of 9 compared to those reported by Honold *et al.*[10] for a 120-Å-thick GaAs single quantum well at 2 K. Since the differences in exciton parameters (a_B,E_B) are already taken into account in Eq. (2), we expect γ_{xx} to be similar in different materials. Slight differences can be partly attributed to the different screening of the Coulomb potential between excitons, i.e., due to the different dielectric constants. In addition, the degree of localization will also affect γ_{xx} (for localized excitons a smaller γ_{xx} is expected). However, according to Takagahara,[17] localization of excitons will also modify the temperature dependence of exciton–acoustic-phonon interaction, resulting in an exponential temperature dependence instead of the linear variation as given by Eq. (1). This, however, is not found (see Fig. 2), similar to results obtained by Stanley in $\text{Cd}_x\text{Zn}_{1-x}\text{Te/ZnTe}$ MQW's.[21] In these samples, which clearly exhibit the effects of exciton localization, the low-temperature homogeneous linewidth also depends linearly on temperature. Nevertheless, this point is not yet fully understood at present and further work is needed to explore the role of exciton-exciton scattering in the case of localized excitons.

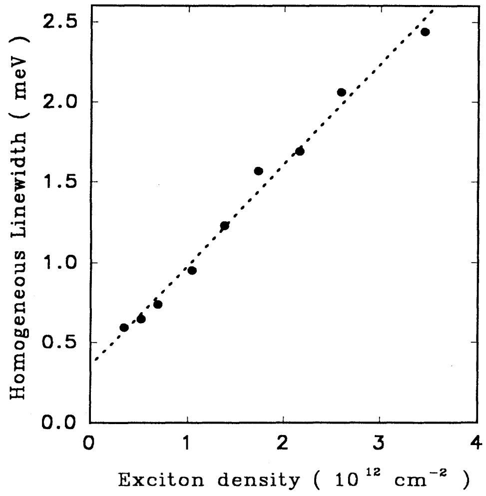

FIG. 3. Homogeneous linewidth Γ_{hom} of a $\text{CdTe/Cd}_{0.86}\text{Mn}_{0.14}\text{Te}$ MQW structure vs exciton density in the range of 3.4×10^{11} to 3.4×10^{12} cm^{-2}, measured at 4.2 K. Also shown is the result of a linear regression according to Eq. (2) (dashed line) which yields the parameter for the exciton-exciton interaction $\gamma_{xx}=0.16$.

Another set of measurements on $\text{Cd}_x\text{Zn}_{1-x}\text{Te/ZnTe}$ MQW's studied the excitonic absorption linewidth.[22] This work focused on sufficiently high temperatures that the exciton absorption linewidth is homogeneously broadened by LO-phonon scattering. At low temperatures inhomogeneous broadening dominated the linewidth, as was the case for the temperatures used in Ref. 21 and is the case for the results presented here. The presence of inhomogeneous broadening means that the simple linear spectroscopic technique employed in Ref. 22 cannot extract the homogeneous linewidth at these low temperatures. Furthermore, the use of alloy well material in both of these earlier studies complicates any comparison to the results presented here.

Since we have measured both the temperature and density dependence of the homogeneous linewidth, we now are able to extrapolate the exciton homogeneous linewidth for zero temperature and zero density $\Gamma_{\text{hom}}(T=0;n_x=0)$. This value then accounts for all residual scattering processes like interface roughness, alloy fluctuation, and impurity scattering. Furthermore, spin-spin interaction (in the case of a temperature-independent process) may also contribute to this value. By extrapolating the observed data we find $\Gamma_{\text{hom}}(T=0;n_x=0)$ to be about 0.28 meV, which is about a factor of 2.5 larger than the value reported for GaAs single quantum wells.[9] Spin scattering may contribute to this; however, further careful study is required to determine to what extent. The primary contribution to the difference in values is probably due to the different structural quality.

The unique feature of the samples studied here is the semimagnetic nature of the barrier material. The exchange interaction between the manganese spins in the barrier and the exciton spin may lead to new phase relaxation mechanisms. Because magnetic polaron formation induces additional localization of the excitons, it will result in excitonic dephasing. However, as mentioned earlier, magnetic polarons are not expected to form in these samples due to the low Mn content and wide wells. Indeed, any interaction between the exciton and collective magnetic excitations in the barrier should result in an additional temperature-dependent dephasing. The temperature dependence observed in our samples is consistent with acoustic-phonon scattering, suggesting that any interaction with magnetic excitations is not important. However, this does not exclude dephasing due to spin-spin interactions between the exciton and single Mn ions. Such spin-spin scattering is expected to be relatively independent of temperature, and may contribute to the extrapolated zero-density, zero-temperature scattering. Definite identification of such new scattering mechanisms due to the semimagnetic barriers is quite difficult. Additional studies in this direction, examining the effects of Mn content, well width, and external magnetic fields on the excitonic dephasing as well as comparison to similar nonmagnetic structures, are currently underway. In turn, a detailed understanding of how the magnetic nature of the barriers influences the excitonic dephasing may yield insight into the dynamics of free spin and spin-correlated systems, realized in dilute magnetic semiconductors.

In summary, we have studied the broadening mechanisms contributing to the homogeneous linewidth of heavy-hole excitons in semimagnetic $CdTe/Cd_{1-x}Mn_xTe$ multiple quantum wells. The observed linear increase of Γ_{hom} with both increasing temperature and exciton density permits the determination of the relevant contributions to the dephasing. In particular, we obtain $\gamma_{xx}=0.16$ for the exciton-exciton interaction strength, $\gamma_{ph}=13.7$ μeV/K for the exciton–acoustic-phonon coupling constant and 0.28 meV as the contribution from all residual temperature and density-independent scattering processes. A comparison with $GaAs/Al_xGa_{1-x}As$ quantum wells leads to the conclusion that the structural properties (alloy disorder, interface roughness, defect density, etc.) or our II-VI quantum-well structures are comparable.

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[19] This, unfortunately, is in contrast to our recent conclusions based on the results for the temperature dependence of the radiative exciton lifetime (Ref. 3). However, opposite to the lifetime data, the FWM experiments provide a very direct determination of the homogeneous linewidth and thus the acoustic-phonon coupling strength. We therefore consider the present data as most reliable. In addition, the recombination lifetimes were obtained under nonresonant excitation conditions while in FWM experiments the excitation is resonant. Nevertheless, further work is definitely needed to clarify this point.

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