Exciton Tunneling Revealed by Magnetically Tuned Interwell Coupling in Semiconductor Double Quantum Wells

I. Lawrence,[1,2] S. Haacke,[3,*] H. Mariette,[1] W. W. Rühle,[2] H. Ulmer-Tuffigo,[1] J. Cibert,[1] and G. Feuillet[1]

[1]*Commissariat à l'Energie Atomique–Centre National de la Recherche Scientifique joint group "Microstructures de Semiconducteurs II-VI" Department de Recherche Fondamentale sur la Matière Condensée and Laboratoire de Spectrométrie Physique, Université J. Fourier, 17 Rue des Martyrs, 38054 Grenoble Cedex 9, France*

[2]*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Germany*

[3]*Grenoble High Magnetic Field Laboratory, Max-Planck-Institut für Festkörperforschung, Centre Nationale de la Recherche Scientifique, 38042 Grenoble, France*

(Received 14 February 1994)

We have found strong evidence for tunneling of excitons as an entity in CdTe/CdMnTe and CdTe/CdZnTe asymmetric double quantum wells (QWs). An external magnetic field changes the coupling between the two QWs by allowing resonances to occur between excitonic states. The tunneling dynamics are investigated by time-resolved and steady-state photoluminescence spectroscopies under magnetic field. Very efficient tunneling is found when the transfer of a spatially direct exciton is possible, either with the emission of LO phonons or via the resonance with the $2s$ state of the low energy QW.

PACS numbers: 78.66.Hf, 71.35.+z, 78.47.+p

Tunneling of carriers from one quantum well (QW) to an adjacent one through a thin potential barrier in semiconductor heterostructures is an area of active research because of its basic quantum mechanical aspects [1] and its importance for tunneling devices [2]. A basic question for the transfer of an optical excitation is: Do the carriers—electrons or holes—tunnel independently, or is it a bound electron-hole pair, that is an exciton, which transits between the wells? In a first approach, experimental results on transfer mechanisms between adjacent QWs made with III-V semiconductors were interpreted as a tunneling of free electrons (free holes) whose energy levels were brought into resonance by an electric field [3,4]. More detailed results have demonstrated that the correct description is a transfer from a direct to an indirect exciton: in this mechanism the electron effectively transfers from a bound state within a direct exciton (electron and hole wave functions localized in the same well), to a bound state within an indirect exciton (electron and hole wave functions localized in different wells) [5–8], whereas "the hole only participates as a spectator" [6]; all these studies concluded then, that although excitonic effects in carrier tunneling must be considered, the *simultaneous* transfer of electron and holes between direct excitons was *not* the dominant mechanism.

In II-VI-based heterostructures the tunneling properties must be much more affected by exciton effects. The exciton binding energies are much larger than in III-V heterostructures and they can be of the same order as, or even larger than, the hole confinement potential [9]; for example, quasi-two-dimensional exciton effects are of great relevance for the origin of optical gain and laser emission in wide-gap II-VI QW structures [10]. In this Letter we present unequivocal evidence for direct exciton tunneling in asymmetric double QWs (ADQWs), i.e., the tunneling, at the same time, of an electron and a hole correlated by the Coulomb interaction. Resonances between excitons were induced by a magnetic field in two different types of II-VI ADQWs, CdTe/CdMnTe and CdTe/CdZnTe, using the enhanced Zeeman splitting of an exciton in the first type of system, and the strong diamagnetic shift of excited exciton states in the second type.

The semimagnetic semiconductor CdMnTe is known to show a giant Zeeman effect, 2 orders of magnitude larger than in nonmagnetic CdTe, resulting from an exchange interaction between the free carriers and the Mn^{2+} ions [11]. In CdTe/CdMnTe ADQWs, where the wider QW contains Mn [Fig. 1(a)], the magnetic field B allows therefore a continuous tuning of the potential of the magnetic QW with respect to that of the nonmagnetic QW [12]. Then the magnetic field affects mainly the $1s$ excitonic state of the CdMnTe QW and changes thereby the energy separation ΔE between the exciton ground states of both QWs, which modifies the coupling between them [Figs. 1(b) and 1(c)]. In the nonmagnetic CdTe/CdZnTe ADQW [Fig. 1(a)], the magnetic field affects mainly the excited states of excitons $(2s, 3s, \ldots)$ due to their smaller binding energy as compared to the $1s$ states. This property allows us to bring the excited $(2s, 3s, \ldots)$ excitonic states of the low energy QW into resonance with the fundamental excitonic state of the high energy QW [Figs. 1(b) and 1(c)].

In the two systems we have made use of the specific field dependences of excitonic transitions to demonstrate a new effect: the transfer of the $1s$ exciton of the high energy QW *as a whole*. It is observed when it occurs via the $2s$ exciton state of the adjacent QW or directly to the $1s$ state of this QW with the emission of an LO phonon. Photoluminescence excitation (PLE) and time-

0031-9007/94/73(15)/2131(4)$06.00

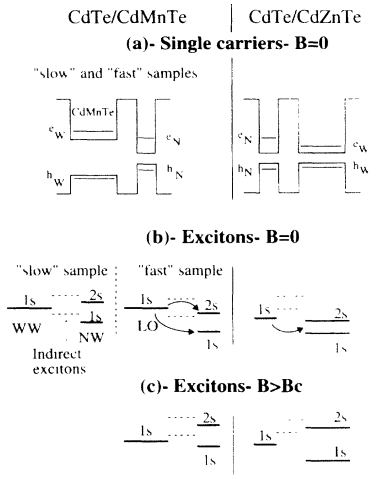

FIG. 1. Two CdTe/CdMnTe samples ("fast" and "slow," differing in the composition of the CdMnTe QW, left column) and one CdTe/CdZnTe QW (right column) are discussed in the text. (a) Schematic representation of the carrier energy levels in the conduction and valence band potential wells in the two systems studied. (b), (c) show direct exciton states (full levels) and indirect exciton states (dotted levels) in the two systems. At $B = 0$, high efficiency transfer between the two QWs is possible through emission of LO phonon or via the resonance with the $2s$ state [curved arrows in (b)]. At a magnetic field above a critical value B_c [(c)] these transfers are inhibited.

resolved photoluminescence (TRPL) under magnetic field are employed as complementary methods for the study of, respectively, the relative energy positions of all excitons and the dynamical behavior associated with this magnetic field dependent energy configuration. Changes in the transfer efficiency are observed as drastic variations of the PL decay time of the $1s$ exciton in the high energy QW.

The CdTe/CdZnTe and CdTe/CdMnTe heterostructures [Fig. 1(a)] are grown by molecular beam epitaxy on (100) CdZnTe substrates. They consist of two QWs on a 2 μm buffer layer of the same composition as the barriers: $x_{Zn} = 13\%$ (for CdTe/CdZnTe) and $x_{Mn} = 12\%$ (for CdTe/CdMnTe). In the CdTe/CdZnTe sample the narrow well (NW, $L_N = 40$ Å) and the wide well (WW, $L_W = 74$ Å) are separated by a barrier of thickness $L_B = 109$ Å. In the CdTe/CdMnTe samples the thickness of the CdTe NWs, the CdMnTe WWs, and the tunneling barriers are, respectively, $L_N = 50$ Å, $L_W = 143$ Å, and $L_B = 50$ Å. In these structures the two samples differ only by the Mn concentration in the WW which is $x_W = 3.7\%$ and $x_W = 4.5\%$, respectively. The sample parameters are determined precisely as described in Ref. [12]. Note that in CdTe/CdMnTe ADQWs the recombination in the CdMnTe WW occurs at higher energy than the recombination in the CdTe NW due to the larger band gap of CdMnTe with respect to CdTe, in contrast to the CdTe/CdZnTe system where both QWs are CdTe layers [13] [see Fig. 1(a)]. A common feature of these two systems is that only one electron and one heavy hole level are present in the low energy QW. Also, due to the strain experienced by the QWs, light hole states need not be considered, because they are at considerably higher energy than heavy hole states (for CdTe/CdMnTe) or even are confined in the barrier (for CdTe/CdZnTe).

The TRPL data were obtained by a two dimensional synchroscan streak camera. A typical lower limit for the measured PL decay time was 7 ps. A tunabled, mode-locked Ti:sapphire laser pumped by an Ar^+ laser was used as an excitation source. The density of excitons created was low enough (10^{10} cm^{-2} per pulse) to avoid space charge effects and to ensure an excitonic regime. The magnetic field is applied in the Faraday configuration along the growth direction.

The time-integrated photoluminescence spectra at 0 T of CdTe/CdMnTe ADQWs are depicted in Fig. 2. The emission energies of the fundamental excitons confined in the CdTe QWs ($1s$-NW) are around 1.647 eV for both samples. The transition related to the fundamental exciton confined in the CdMnTe QW ($1s$-WW) is found at 1.662 and 1.674 eV depending on x_W. The shoulders or lines labeled Y on the low energy side of these transitions are attributed to extrinsic excitons. The PL decay times of the high energy $1s$-WW excitons, τ_W, are strikingly different between the two samples (inset in Fig. 2). A short decay time, $\tau_W = 25$ ps, is observed for the sample with the largest x_W (called hereafter the "fast sample") whereas a longer one, $\tau_W = 100$ ps, is measured for the other sample (called the "slow sample"). Tunneling times τ_T are deduced from these values of τ_W from the relation $1/\tau_W = 1/\tau_R + 1/\tau_T$, where the intrinsic recombination time $\tau_R = 155 \pm 5$ ps was measured at 0 T on single CdMnTe QWs of the same width (the value of the recombination time τ_R is reproducible and the recombination is of radiative origin [14]). The tunneling times obtained are then $\tau_T = 30$ ps and $\tau_T = 280 \pm 20$ ps for the fast and slow samples, respectively.

As a consequence of the different Mn concentrations x_W in both samples, the barrier transparencies which govern the nonresonant single carrier tunneling process [3] are slightly different. However, this difference is too small to explain a 1 order of magnitude difference in the transfer times. Rather, the important point is that different concentrations x_W introduce a different energy separation between the fundamental excitons, $\Delta E = (1s\text{-WW}) - (1s\text{-NW})$. The dynamical behavior of these two samples suggests that the transfer is less efficient when ΔE is lower than a $\hbar\omega_{LO}$ phonon energy. Therefore a series of time-resolved experiments in magnetic fields up to 10 T were performed in order to tune the exciton energy levels, i.e., ΔE, and to observe the changes of the transfer efficiency.

Figure 3 shows for the fast sample the magnetic field dependence of ΔE obtained by steady state measurements (left scale) together with the measured decay time τ_W (right scale) of the low energy component of the $1s$-WW exciton luminescence ($\sigma+$ polarization). Thanks to the strong field dependence of the $1s$-WW transition, ΔE

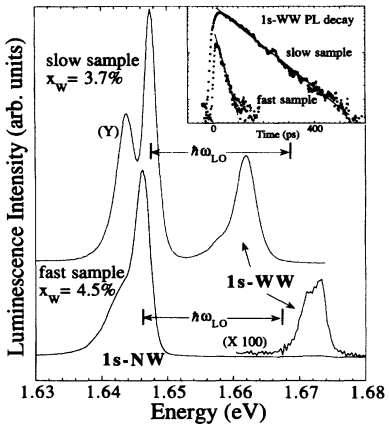

FIG. 2. Time integrated PL spectra and PL decays (inset) of the two CdTe/CdMnTe samples at 0 T and 5 K.

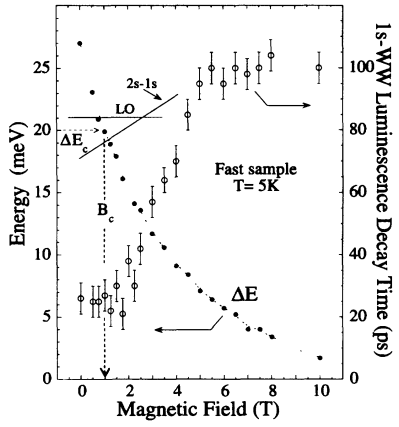

FIG 3. Left scale: energy difference ΔE (black dots) measured between the two excitonic transitions $1s$-WW and $1s$-NW as a function of B. Right scale: variation of the PL decay time τ_W of the high energy CdMnTe QW (white dots). The decay time τ_W starts to rise at a critical field B_c for which ΔE is approximately equal to the LO phonon energy and to the energy difference between the $2s$- and $1s$-NW excitons ($\Delta E = \Delta E_c$).

decreases from 27.5 meV at 0 T to 1.5 meV at 10 T. A strong increase of τ_W is obtained simultaneously, reaching a value of 100 ps for $B > 6$ T. The rise of τ_W starts for $B > B_c$ ($B_c \approx 1$ T) corresponding to a critical value of ΔE, ΔE_c, approximately equal to the longitudinal optical phonon energy ($\hbar\omega_{LO} = 21.3$ meV), but also to the energy difference ($2s$-NW)$-$ ($1s$-NW) (Fig. 3). Such an increase is not observed either for the decay measured on the slow sample or for the decay of a CdMnTe single QW sample. In such samples the experimental decay time (not shown here) presents only weak dependence on the magnetic field. So the strong increase of τ_W with B observed in the fast sample cannot be attributed to a variation of the recombination time τ_R, but mainly to a variation on the tunneling time. Moreover, in the present sample, the effect of the magnetic field never compensates the initial confining potentials for electrons or heavy holes of either spin state. Thus no change in the exciton dynamics due to a variation of spin relaxation phenomena is expected [15]. The clear link observed between the excitonic energy difference ΔE and the lifetime τ_W as B varies suggests that an exciton transfer governs the dynamics of these samples. A complete PLE study under magnetic field confirms that the resonance of $1s$-WW and $2s$-NW and that of $1s$-WW and ($1s$-NW) $+ \hbar\omega_{LO}$ do indeed occur at field values close to $B_c = 1$ T. Thus, the transfer of the $1s$-WW exciton to the $1s$-NW exciton is enhanced when it can occur via the emission of an optical phonon or via the $2s$-NW exciton state. The relative importance of the two identified mechanisms could be determined by measurements at higher temperatures, which would weaken the giant Zeeman effect in CdMnTe without affecting significantly the energy shift of the $2s$-NW exciton.

The latter process, transfer through the $2s$ excitons, is demonstrated clearly in the CdTe/CdZnTe ADQW. The excitonic transition energies of the CdTe/CdZnTe sample under magnetic field as determined by PLE spectra are displayed in Fig. 4(a). The $2s$-WW transition is identified by its strong linear field dependence at high fields (1.3 meV/T for $B \geq 6$ T), which is due to a smaller binding of $2s$ states as compared with $1s$ states. The other weak transition is assigned to the $e_W h_N$ indirect exciton. All these assignments are confirmed by the good agreement obtained with a complete calculation of the exciton transition energies [16]. We should point out that the extrapolation of the field variation of the $e_W h_N$ energy down to 0 T shows $e_W h_N$ at higher energy than the $1s$-NW so that these two transitions never cross. We will hence focus on the fact that the $1s$-NW and $2s$-WW exciton states have the same energy at $B_c \approx 5$ T. The PL transients of both the NW and the WW have been studied for magnetic fields up to 14 T. The effect on the high energy NW PL decay time τ_N is depicted in Fig. 4(b). Starting with a zero field decay time of $\tau_N(B = 0) = 28 \pm 3$ ps we observe a decrease down to $\tau_N = 18 \pm 5$ ps for $B = 6$ T. For $B \geq 6$ T, τ_N increases drastically up to $\tau_N = 60 \pm 5$ ps and then remains constant. The dependence of the low energy WW PL decay time on the magnetic field was also recorded; these values were used as typical intrinsic recombination times τ_R in these QWs to deduce the transfer times τ_T as a function of B from the observed τ_N according to $1/\tau_N = 1/\tau_R + 1/\tau_T$. The transfer times τ_T exhibit a field dependence slightly more pronounced than τ_N, reaching a value of $\tau_T = 80$ ps for $B \geq 10$ T. The magnetic field dependence of τ_N shows unambiguously that the two QWs become much less coupled for $B \geq 6$ T. Thus the transfer of the $1s$-NW exciton via the $2s$-WW state is identified as the relevant process for the coupling of the two QWs, given the crossing of the $2s$-WW and the $1s$-NW exciton states at $B_c \approx 5$ T.

Our interpretation of the experimental results based upon direct exciton tunneling in these II-VI systems is strongly supported by calculations both of confined car-

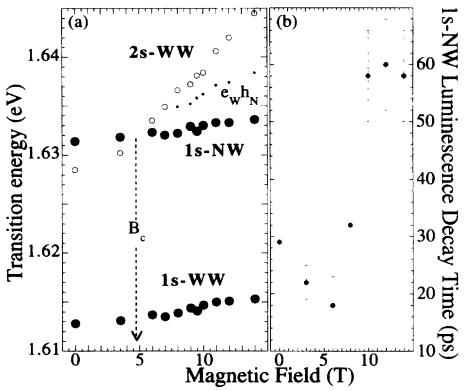

FIG. 4. (a) Magnetic field dependence of the exciton transitions in the CdTe/CdZnTe ADQW, recorded in PLE under σ excitation and detected on the low energy σ component of $1s$-WW at $T = 2$ K (the size of the symbols reflects the transition intensity). For $B = B_c$ the $1s$-NW and the $2s$-WW exciton levels are in resonance. (b) PL decay time τ_N of the high energy CdTe QW showing a minimum at B_c.

rier energies and of exciton energies (direct and indirect excitons), the results of which can be summarized as follows. Let us first recall that, in CdTe/CdMnTe samples, the magnetic field affects mainly the hole levels and consequently the exciton levels but hardly the electron levels [11]. From the calculation, no resonances between electron states are found [4,17], but only resonances involving holes (or excitons) are possible. However, the consideration of hole levels [Fig. 1(a)] and of the corresponding indirect excitons [Fig. 1(b)] cannot explain the different dynamical behaviors of the fast and the slow CdTe/CdMnTe samples of 0 T, since the calculation gives similar configurations for both samples. For CdTe/CdZnTe the transfer via an indirect exciton is energetically forbidden [Fig. 1(b)]. In summary, only transfer between direct exciton states consistently explains the behavior of all samples (Fig. 1).

The two dominant mechanisms discussed above, namely the LO phonon assisted excitonic transfer and the transfer via a $2s$ exciton state, should induce a sharp increase of τ_W as soon as $\Delta E < \Delta E_c$. Nevertheless, for CdTe/CdMnTe, τ_W keeps increasing as ΔE decreases further. This suggests that other transfer mechanisms are still efficient for $B > B_c$ (transfer by acoustical phonons or impurity scattering) and that these transfers are successively suppressed as ΔE decreases. Note that for higher fields ($B > 8$ T), when the PL lines of the two $1s$ excitonic transitions are superimposed ($\Delta E < 5$ meV), τ_W reaches a value of 100 ps close to the intrinsic decay time observed at 10 T for a single QW of either CdTe or CdMnTe ($\tau_R = 110 \pm 10$ ps). This experimental result suggests that a thermal equilibrium between the $1s$ direct excitons of the two QWs is effectively established thanks to elastic processes, when ΔE is smaller than the full width at half maximum of the excitonic lines. This behavior is in agreement with our interpretation for which no decrease of the observed decay time is expected when resonance in energy occurs between the two $1s$ direct exciton transitions.

For the CdTe/CdZnTe ADQW, the value of τ_N obtained for $B \geq 10$ T, when the fast tunneling via the $2s$ state is inhibited, never reaches the PL decay time of an isolated CdTe QW having the same width. This indicates that the QWs are still partially coupled at high magnetic fields. In this case the transfer of direct $1s$-NW excitons under emission of LO phonons into the Y extrinsic state of the WW is still possible.

In conclusion, the transfer dynamics between two adjacent QWs have been tuned by an external magnetic field. Our results demonstrate that tunneling processes involving the exciton as a whole have to be considered in order to understand the strong variations of the PL decay times obtained for CdTe/CdMnTe and CdTe/CdZnTe ADQWs. Very efficient tunneling is observed for magnetic field values inducing an excitonic configuration where the transfer of spatially direct excitons between the two wells is possible either with the emission of an LO phonon or via the resonance with the $2s$ state of the low energy QW exciton.

We are grateful to A. Heberle, N. Magnea, M. Oestreich, and N. Pelekanos for their collaboration, and to K. Rother and H. Klann for their technical assistance during the TRPL measurements.

*Present address: Institute of Micro- and Optoelectronics, Federal Institute of Technology, CH-1015 Lausanne, Switzerland.

[1] S. Lury, Solid State Commun. **65**, 787 (1988).
[2] F. Capasso *et al.*, IEEE J. Quantum Electron. **22**, 1853 (1986).
[3] M. Nido *et al.*, Appl. Phys. Lett. **56**, 355 (1990).
[4] D. Y. Oberli *et al.*, Appl. Phys. Lett. **56**, 1239 (1990).
[5] G. Bastard *et al.*, J. Lumin. **44**, 247 (1989).
[6] R. Ferreira *et al.*, Phys. Rev. B **45**, 11 782 (1992); Surf. Sci. **229**, 192 (1990).
[7] A. M. Fox *et al.*, Phys. Rev. B **42**, 1841 (1990).
[8] A. P. Heberle *et al.*, Semicond. Sci. Technol. B **7**, 421 (1992); Phys. Status Solidi (b) **173**, 381 (1992).
[9] N. T. Pelekanos *et al.*, Phys. Rev. B **45**, 6037 (1992); H. Mariette *et al.*, Phys. Scr. **T39**, 204 (1991).
[10] J. Ding *et al.*, Phys. Rev. Lett. **69**, 1707 (1992); A. Nurmikko and R. Gunshor, in *Proceedings of the Trieste Semiconductor Symposium* [Physica (Amsterdam) **185B**, 16 (1993)].
[11] J. K. Furdyna, J. Appl Phys. **64**, R29 (1988), and references therein.
[12] I. Lawrence *et al.*, Superlattices Microstructures **12**, 119 (1993).
[13] S. Haacke *et al.*, Phys. Rev. B **47**, 16 643 (1993); J. Cryst. Growth **138**, 831 (1994).
[14] I. Lawrence *et al.*, J. Phys. IV (France) **3**, C5-405 (1993).
[15] J. F. Smyth *et al.*, Phys. Rev. Lett. **71**, 601 (1993).
[16] P. Peyla *et al.*, in *Proceedings of the 21th ICPS, Beijing, China, 1992* (World Scientific, Singapore, 1993).
[17] B. Deveaud *et al.*, Europhys. Lett. **11**, 367 (1990).