Optical Characterization of CdTe/CdMgTe Quantum Wells Containing Single (Sub)Monolayers MnTe

W. Heiss, G. Prechtl, W. Jantsch

Microelectronics Institute, Johannes Kepler University Linz, A-4040 Linz, Austria

We perform photoluminescence (PL), polarization dependent PL excitation, and magneto-optical Kerr rotation experiments on II-VI quantum wells containing ultra narrow MnTe layers. From a comparison of the change of the optical transition energies upon rapid thermal annealing with model calculations we determine the actual width of the MnTe barriers. We find the barrier width to be very close to its nominal values. In addition, for a single monolayer MnTe we detect the antiferromagnetic-paramagnetic phase transition at a critical temperature of 50 K by a kink in the temperature dependence of the inverse Zeeman splitting. Furthermore, we have used the Zeeman splitting induced by a quarter monolayer MnTe inserted in a nonmagnetic quantum well at various positions to map the probability density of free holes in growth direction.

1. Introduction

Zincblende (ZB) MnTe is a prototype of a fcc Heisenberg system with strongly dominating antiferromagnetic nearest-neighbor interactions. While bulk grown crystals of MnTe exhibit the hexagonal NiAs crystal structure [1], by non-equilibrium growth techniques like molecular beam epitaxy (MBE) single crystals of MnTe can be synthesized also in the ZB phase [2]. In previous works, mainly epilayers of ZB MnTe [3], [4] and superlattices containing MnTe layers with a thickness of *several* monolayers (MLs) [4], [5] were investigated. Recently, new heterostructures have been developed in which fractional MLs of magnetic ions are introduced digitally within a semiconductor quantum well [6]. These structures are of special interest due to the possibility to tailor the spin splitting in addition to the electronic eigenstates [6]. Therefore, we investigate the magneto-optical properties of single ZB MnTe layers with a thickness corresponding to a coverage of *one*-or *one half* or *one quarter* of a ML, embedded in non-magnetic quantum wells. In particular, we investigate (a) the migration of the Mn ions in such structures, leading to a broadening of the MnTe barriers [7], and (b), the antiferromagnetic – paramagnetic phase transition of a single monolayer MnTe [8]. Furthermore, we apply a quarter ML MnTe to map out the probability density of free carriers in a square quantum well [9].

2. Samples and Experimental Details

Our samples are CdTe quantum wells with various width embedded between $Cd_{1-x}Mg_xTe$ barriers. All samples contain a single MnTe (sub)monolayer at various positions. The growth of the samples was performed at the Institute of Physics, Polish

Academy of Sciences in Warsaw (Poland). The samples are grown by molecular beam epitaxy (MBE) either on (001)-oriented $Cd_{0.96}Zn_{0.04}Te$ substrates ore on GaAs substrates. The growth rate was chosen to be very slow (8 s/ML) in order to control the Mn incorporation precisely and to get long enough growth times as compared to the opening and closing times of the shutters. Each interface was smoothed by performing growth interruptions under Te excess. The two-dimensional growth was monitored by RHEED oscillations.

Similar as in diluted magnetic semiconductors, in our quantum well samples, the exciton spin (Zeeman) splitting is strongly enhanced by spin-spin exchange interactions between the d electrons of the Mn^{2+} ions and the s-like conduction-band and p-like valence band electrons in the quantum well. With applied field, all e_1-hh_1 transitions, one with positive (σ^+) and the other with negative helicity (σ^-), can be observed. The energy difference between these transitions ΔE is directly proportional to the magnetization M [10]. Therefore, M can be measured directly by magneto optical spectroscopy.

All measurements were performed in Faraday configuration. We performed polarization dependent photoluminescence (PL) and PL excitation (PLE) experiments in magnetic fields up to 6 T. Furthermore, the magneto-optical Kerr rotation was measured by the use of a photoelastic modulator. For excitation a tunable Coherent CR 599 dye laser was used, operating in the wavelength range between 620 nm and 800 nm.

3. Results and Discussions

To estimate the actual barrier width of a nominal 1 ML thick MnTe barrier we have performed rapid thermal annealing experiments. The annealing step results in diffusion of the Mn out of the barriers. For small Mn diffusion lengths, the PL transition shifts to the blue due to the broadening of the MnTe barrier while for larger diffusion lengths the PL shifts to the red, because the barrier height decrease. Furthermore, the Zeeman splitting increases significantly upon annealing, since the number of antiferromagnetically coupled Mn ions in the barrier decreases while the number of paramagnetic background ions in the quantum well increases. In particular, in a quantum well sample containing a single ML MnTe we find a 14 meV blue shift of the PL transition upon annealing and an increase of the spin splitting from 23 meV to 60 meV at 6 T. In contrast, for a similar sample containing a MnTe barrier with a nominal width equivalent to one half ML, we find a 3 meV red shift upon 15 s rapid thermal annealing at 440 °C. From a comparison of these experimental observations with band structure calculations assuming Gaussian shaped MnTe barriers we obtain the actual barrier width before and after annealing. The width of the Mn distribution after annealing corresponds to a diffusion length of 1.5 nm. From this value a diffusion coefficient of 1.5×10^{-15} cm^2/s can be obtained, in good agreement with results given in the literature for the same annealing conditions [11]. Before annealing, the deduced width of the Mn distribution corresponds to a barrier width of 1.25 MLs. In addition, an absolute upper limit for this value of 1.65 MLs can be obtained.

For a quantum well sample containing a MnTe barrier with the nominal thickness of one ML we have studied the temperature dependence of the inverse Zeeman splitting $1/\Delta E$ in detail. At low temperatures, $1/\Delta E$ increases linearly with increasing T. At a critical temperature of 50 K a kink is observed and for higher temperatures $1/\Delta E$ rises linearly again, but with a smaller gradient. The temperature dependence of the Zeeman splitting

can be fitted by the use of a Brillouin function and two phenomenological quantities, the effective manganese concentration x_e and an antiferromagnetic temperature T_{AF}. Up to 50 K, a good fit of the temperature dependence of $1/\Delta E$ can be obtained using a constant value for $T_{AF} = 23$ K and $x_e = 0.9$ %. For higher temperatures, above the kink, a good fit can be obtained only by increasing both of these parameters. This sudden increase of x_e at $T_{critical} = 50$ K indicates a phase transition of the antiferromagnetically coupled magnetic ions in the MnTe ML. The value of x_e obtained for the temperature range below $T_{critical}$ is much smaller than the value obtained by averaging the total number of magnetic ions present in a single MnTe ML over the whole CdTe quantum well ($x_{av} = 6$ %). This indicates that the enhanced Zeeman splitting observed below $T_{critical}$ is caused mainly by the isolated Mn ions migrated from the antiferromagnetically coupled MnTe layer into the CdTe quantum well by diffusion.

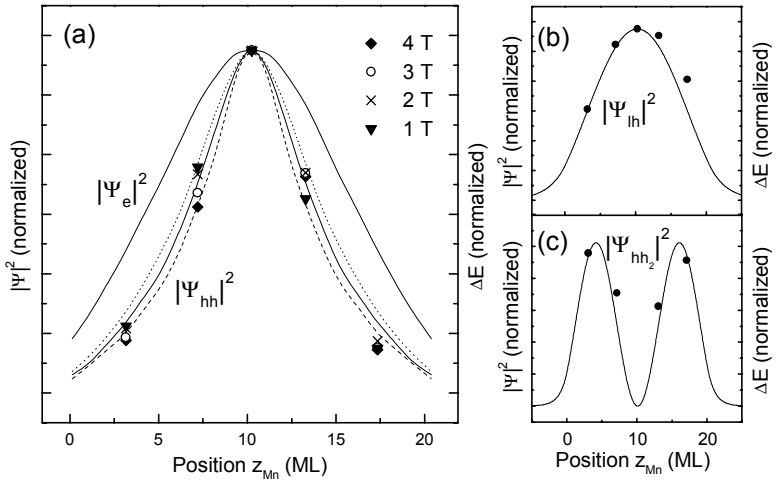

Fig. 1: (a) Probability density (PD) of the heavy hole ground state calculated for various valence band offsets (0.25 (dashed), 0.33 (solid) and 0.45 (dotted)) and of the electron ground state (valence band offset 0.33). The symbols display the experimentally observed interband Zeeman splitting of the e_1-hh_1 transition. (b) PD of the light hole ground state compared with the Zeeman splitting of the e_1-lh_1 transition. (c) As (b) but for the first excited heavy hole state hh_1.

Furthermore, we have investigated the Zeeman splitting in a series of five 20 ML wide quantum well structures (S1 to S5), where each sample contains a narrow MnTe barrier with a Mn content equivalent to ¼ ML coverage. In sample S1 the MnTe layer is inserted after the 3rd ML of CdTe, while in S2 to S5 the magnetic probes are embedded after the 7th, 10th, 13th, and 17th ML of CdTe, respectively. Assuming the sp-d exchange interaction to be strongly localized, it can be shown that the Zeeman splitting in this set of samples is proportional to the probability density of free carriers as function on the position of the magnetic probe in the well [9]. This is demonstrated in Fig. 1, where the probability density of the electrons and the holes calculated for different valence band offsets is compared to the experimental Zeeman splitting. This comparison shows that the measured values correspond to the probability density of the holes and not of the electrons, which is in contrast to all previous experiments [12] – [14]. Furthermore, Fig. 1 shows the probability density of the light holes in (b) and of the first excited heavy hole state in (c), in good agreement with the experimental data.

4. Summary

We characterize CdTe/CdMgTe quantum wells containing fractional MnTe monolayers by magneto-optical spectroscopy. In particular, we perform photoluminescence (PL), polarization dependent PL excitation, and magneto-optical Kerr rotation experiments. By comparing the change of the PL transition energies upon rapid thermal annealing with results of band structure calculations we determine the actual width of the MnTe barriers. We find for a nominal one monolayer thick MnTe barrier a broadening of the barrier width by Mn migration of only 0.25 monolayers. In this sample the antiferromagnetic-paramagnetic phase transition is detected by optical spectroscopy at a critical temperature of 50 K. In addition, we have demonstrated that the Zeeman splitting induced by a quarter monolayer MnTe inserted in a nonmagnetic quantum well at various positions can be used to map the probability density of free holes in growth direction.

References

[1] J.W. Allen, G. Lucovsky, and J.C. Mikkelsen, Jr., *Solid State Commun.* **24**, 367 (1977)

[2] S.M. Drubin, J. Han, Sungki O, M. Kobayashi, D.R. Menke, R.L. Gunshor, Q. Fu, N. Pelekanos, A.V. Nurmikko, D. Li, J. Gonsalves, and N. Otsuka, *Appl. Phys. Lett.* **55**, 2087 (1989)

[3] K. Ando, K. Takahashi, T. Okuda, and M. Umehara, *Phys. Rev.* **B 46**, 12289 (1992)

[4] T.M. Giebultowicz, P. Klosowski, N. Samarth, H. Luo, J.K. Furdyna, and J.J. Rhyne, *Phys. Rev.* **B 48**, 12817 (1993)

[5] M. Pohlt, W. Herbst, H. Pascher, W. Faschinger, and G. Bauer, *Phys. Rev.* **B 57**, 9988 (1998)

[6] S. A. Crooker, D. A. Tulchinsky, J. Levy, D. D. Awshalom, R. Garcia, N. Samarth, *Phys. Rev. Lett.* **75**, 505 (1995)

[7] G. Prechtl, W. Heiss, S. Mackowski, A. Bonanni, G. Karczewski, H. Sitter, W. Jantsch, *Sem. Sci. Technol.* (in print)

[8] G. Prechtl, W. Heiss, A. Bonanni, H. Sitter, W. Jantsch, S. Mackowski, G. Karczewski, *Physica E* (in print)

[9] G. Prechtl, W. Heiss, A. Bonanni, W. Jantsch, S. Mackowski, E. Janik, G. Karczewski, *Phys. Rev. B* (in print)

[10] D. U. Bartholomew, J. K. Furdyna, A. K. Ramdas, *Phys. Rev. B*. **43**, 6943 (1986)

[11] D. Tönnies, G. Bacher, A. Forchel, A. Waag, G. Landwehr, *Appl. Phys. Lett.* **64**, 766 (1994)

[12] P. H. Beton, J. Wang, N. Mori, L. Eaves, P. C. Main, T. J. Foster, M. Henini, *Phys. Rev. Lett.* **75**, 1996 (1995)

[13] J. Y. Marzin, J. M. Gerard, *Phys. Rev. Lett.* **62**, 2172 (1989)

[14] G. Salis, B. Graf, K. Ensslin, K. Campman, K. Maranowski, A. C. Gossard, *Phys. Rev. Lett.* **79**, 5106 (1997)