Presented: 23rd International Conference on the Physics of Semiconductors; Berlin, Germany; July 21-26, 1996

Published: in "The Physics of Semiconductors", edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996), pp. 393-396

II. F. 3

# **OPTICALLY STUDIED SPIN RELAXATION PROCESSES IN CdMnTe**

M. GODLEWSKI<sup>a</sup>, A. WITTLIN<sup>a,d</sup> and R.R. GAłĄZKA<sup>a</sup> <sup>a</sup>Institute of Physics, Polish Acad. of Sci., 02-668 Warsaw, AL. Lotników 32/46, Poland

B. MONEMAR<sup>b</sup>

<sup>b</sup>Dept. of Physics and Meas. Technol., Linköping University, Linköping, Sweden

T. GREGORKIEWICZ<sup>c</sup> and C.A.J. AMMERLAAN<sup>c</sup> <sup>c</sup>Van der Waals-Zeeman Lab., Univ. of Amsterdam, Amsterdam, The Netherlands

P.H.M. VAN LOOSDRECHT<sup>d</sup> and J.A.A.J. PERENBOOM<sup>d</sup> <sup>d</sup>High Magnetic Field Lab., Nijmegen University, Nijmegen, The Netherlands

Mechanisms of optical detection of magnetic resonance in CdMnTe are discussed. We show that magnetization of CdMnTe samples is "macroscopically" reduced at the  $Mn^{2+}$  magnetic resonance conditions. An efficient spin cross-relaxation between Mn subsystem and free holes explains the magnitude and sign of the observed ODMR signal.

In CdMnTe a strong magnetic interaction between subsystems of localized spins of  $Mn^{2+}$  (3d<sup>5</sup>) ions and spins/magnetic moments of free or bound carriers leads to a "giant" Zeeman splitting of free and bound excitons and to a formation of magnetic polarons [1,2]. In the present work we discuss the mechanisms of optical detection of magnetic resonance of  $Mn^{2+}$  ions in bulk CdMnTe samples with 9.5 % and 0.7 % Mn fraction. Based on the results of photoluminescence (PL), magneto-luminescence, optically detected magnetic resonance (ODMR) and optically detected cyclotron resonance (ODCR) studies we show that the PL changes at the  $Mn^{2+}$  magnetic resonance cannot be related to the macroscopic decrease of a sample magnetization at the resonance conditions only. The present study indicates that spin systems of  $Mn^{2+}$  ions and of free carriers (mostly of holes, due to a stronger exchange coupling with Mn spins) are in a thermal contact due to efficient spin cross-relaxation transitions. Cross-relaxation between  $Mn^{2+}$  ions and free carriers is much more efficient than the spin-lattice relaxation.

The first experiments of optically detected magnetic resonance of  $Mn^{2+}$  ions in CdMnTe have been performed by Komarov et al. [3]. The observed changes of optical properties (Faraday rotation) have been related to a decrease of a sample magnetization at the magnetic resonance conditions. Such mechanism of the

393

#### 394 M. Godlewski, A. Wittlin et al.

ODMR detection is also confirmed by the present ODMR study. We have observed a shift towards higher energy of a spectral position of free exciton (FE) (by 52 cm<sup>-1</sup> in a Q-band ODMR experiment for the 9.5 % Mn sample and for 200 mW of microwave power) and acceptor bound exciton (ABE) PL transitions occurring at the Mn<sup>2+</sup> magnetic resonance conditions [4]. The magnitude of magnetization changes induced by the magnetic resonance could be estimated by comparing PL shift observed in the ODMR and in the magneto-optical experiments. It is about 60 % for the 9.5 % Mn CdMnTe sample and the Q-band ODMR study (200 mW microwave power). This is an extremely large effect resulting in macroscopical changes of optical and magnetic properties of the sample.

The ABE PL emission is deactivated for an increased magnetic field and the FE PL is enhanced. This is a consequence of a much larger Zeeman effect for FE excitons, for which magnetic momenta of an electron and hole can freely adjust to the direction of an external magnetic field. For ABEs magnetic momenta of two holes are antiparallel, resulting in a much smaller Zeeman splitting than that observed for FEs [2]. The difference of Zeeman splitting for the FE and ABE PL transitions allows to estimate an efficiency of Auger-type nonradiative recombination in CdMnTe.



Figure 1: Optically detected magnetic resonance of  $Mn^{2+}$  ions (structure at about 0.33 T) in two CdMnTe samples studied. In each scan the excitation was cut off for a moment (structure at about 0.1 T) which allowed us to evaluate the magnitude of the change of the PL signal at the magnetic resonance conditions.

Auger-type recombination processes (energy of recombining electron-hole pair is transferred to a third carrier bound at the same center (electron for donor bound exciton (DBE) and hole for ABE), which is ionized into a continuum of conduction (electron) or valence (hole) band states) are responsible for an efficient

### II. F. 3 Optically Studied Spin Relaxation Processes in CdMnTe 395

nonradiative recombination of bound excitons in GaP [5,6]. The efficiency of Auger transitions for DBE and ABE excitons in direct band gap II-VI compounds is not well known. If such transitions are efficient in CdMnTe, magnetic resonance of  $Mn^{2+}$  ions, which decreases magnetization of the sample, should lead to a decrease of the PL intensity in the excitonic part of the emission spectrum. This happens since radiative FE transitions are deactivated and are partly replaced by ABE transitions, with competing Auger-type nonradiative transitions, when the sample magnetization is reduced at the  $Mn^{2+}$  magnetic resonance conditions. In Fig. 1 we show the results of the respective ODMR experiments for the two CdMnTe samples studied.



Figure 2: PL and spectral dependencies of the ODCR and ODMR signals for the 0.7 % of Mn CdMnTe sample and X-band microwave experiment.

The observed decrease of the overall intensity of the "edge" PL is very large for the 9.5 % Mn sample. The excitonic PL is quenched by about 40-50 % and is smaller at the  $Mn^{2+}$  magnetic resonance than that observed at the 0 T magnetic field. The latter means that the observed effect can not be related to the decrease of the sample magnetization only. Its magnitude is too large to be entirely explained by an efficient Auger recombination for ABEs. We have performed the additional ODCR and ODMR investigations to explain the observed magnitude of the ODMR signal (Fig. 2). By setting conditions for electron cyclotron resonance (ECR) we have observed that PL of FE and ABE excitons and of donor-acceptor pairs (DAP) is deactivated. This is a consequence of the reduced rate of excitons formation and 396 M. Godlewski, A. Wittlin et al.

of their dissociation (also for shallow donors) by impact with microwave heated free electrons at the ECR conditions [7]. Fig. 2 shows that the ABE PL is reduced in the intensity at the  $Mn^{2+}$  magnetic resonance conditions. This at first seems to contradict the proposed above mechanism of the ODMR detection. The ABE PL should be enhanced when the sample magnetization is reduced. From shifts of PL positions we know that such reduction of the sample magnetization takes place. To explain this apparent contradiction we propose that  $Mn^{2+}$  spin subsystem is in a thermal contact with magnetic momenta of free carriers. There are several experimental indications that a spin-lattice coupling is too weak in CdMnTe to account for the observed dynamics of  $Mn^{2+}$  spins [8]. We propose that effective temperature of hole subsystem is increased at the  $Mn^{2+}$  magnetic resonance conditions, which results in a reduced rate of ABEs formation. Mn spin system relaxes by efficient spin cross-relaxation transitions increasing an effective spin temperature of free carriers (mostly holes). The process is more efficient for holes due to a large p-d exchange coupling in CdMnTe [1,2].

Concluding we relate the ODMR detection in our samples to a macroscopical decrease of the sample magnetization (shift of the spectral position of excitonic PLs) and to efficient spin cross-relaxation between  $Mn^{2+}$  ions and free holes.

## Acknowledgments

This work was partly financed by grant no. 2 P03B 191 08 of Polish State Committee for Scientific Research

### References

- 1. J.K. Furdyna, J. Appl. Phys. 64, R29 (1988).
- J.A. Gaj, in Diluted Magnetic Semiconductors, eds. J.K. Furdyna and J. Kossut, (Academic, London, 1988) Semiconductors and Semimetals Vol. 25, p. 275.
- 3. A.V. Komarov, S.M. Ryabchenko, O.V. Terletskii, I.I. Zheru and R.D. Ivanchuk, Sov. Phys. JETP 46, 318 (1977).
- 4. S.J.C.H.M. van Gisbergen, M. Godlewski, R.R. Galazka, T. Gregorkiewicz, C.A.J. Ammerlaan and Nguyen The Khoi, *Phys. Rev.* **B48**, 11767 (1993).
- 5. D.F. Nelson, J.D. Cuthbert, P.J. Dean and D.G. Thomas, *Phys. Rev. Lett.* 17, 1262 (1966).
- 6. P.J. Dean, R.A. Faulkner, S. Kimura and M. Ilegems, *Phys. Rev.* B4, 1926 (1971).
- 7. M. Godlewski, W.M. Chen and B. Monemar, CRC Critical Reviews in Solid State and Material Sciences 19, 241 (1994).
- 8. T. Dietl, P. Peyla, W. Grieshaber and Y. Merle d'Aubigne, *Phys. Rev. Lett.* 74, 474 (1995).