Proceedings of the XXXII International School of Semiconducting Compounds, Jaszowiec 2003

Microwave-Induced Delocalization of Excitons in Ternary Compounds of II–VI and III–V Semiconductors

V.Yu. Ivanov[a], M. Godlewski[a,b], A. Khachapuridze[a], S. Yatsunenko[a], T. Wojtowicz[a], G. Karczewski[a], J.P. Bergman[c], B. Monemar[c], T. Shamirzaev[d], K. Zhuravlev[d], K. Leonardi[e] and D. Hommel[e]

[a]Institute of Physics, Polish Academy of Sciences
al. Lotników 32/46, 02-668 Warsaw, Poland
[b]College of Science, Cardinal S. Wyszyński University, Warsaw, Poland
[c]Dept. of Physics and Meas. Techn., Linköping University
581 83 Linköping, Sweden
[d]Institute of Semiconductor Physics, SB RAN, Novosibirsk 630090, Russia
[e]Institute of Solid State Physics, Bremen University
28334 Bremen, Germany

In this work we employ technique of optically detected cyclotron resonance for evaluation of the role of localization processes in CdTe/CdMnTe and CdMnTe/CdMgTe quantum well structures. From microwave-induced changes of excitonic emissions we evaluate magnitude of potential fluctuations (Stokes shift), correlate optically detected cyclotron resonance results with the results of time-resolved experiments and discuss nature of recombination processes in the limit of a strong localization.

PACS numbers: 71.35.Ji, 72.25.Rb, 76.70.Hb, 78.55.Et

1. Introduction

It was demonstrated by several groups that at low temperatures excitons can be site localized in quantum well (QW) structures of II–VI and III–V compounds. By site localization we mean here trapping of excitons (carriers) by potential fluctuations present in a QW plane. Then excitons are localized in a given region and cannot move freely in a QW. Such site localization significantly affects the properties of excitons. Increase in exciton's lifetime, photoluminescence (PL) line broadening, weaker exciton–phonon coupling were related to the localization effects, as discussed in Ref. [1] for CdTe/CdMnTe and CdMnTe/CdMgTe QW structures. Definitely, the most striking localization-related observation is efficient radiative recombination in heavily defected (high density of dislocations) GaN-based QW structures [2].

In this work we employ technique of optically detected cyclotron resonance (ODCR) for evaluation of strength of localization. Modifications of PL spectra at cyclotron resonance (CR) conditions in selected low-dimensional structures and thin films of II–VI and III–V semiconductors are first discussed. Then we relate S-like shape of temperature dependence of PL line width and an anomalous temperature dependence of PL energy to strong localization effects in QW structures of CdTe/CdMnTe and CdMnTe/CdMgTe. Finally, we show that not only excitons but also free carriers can be localized in QW structures showing strong potential fluctuations.

2. Experimental

ODCR investigations were performed with 10, 36, and 60 GHz microwave systems, the latter two developed by us in the Institute of Physics, Warsaw, with a microwave cavity mounted in a split-coil Spectromag 6000 magnet of the Oxford Instruments. We used argon laser (2.41 eV to 3.64 eV excitation), semiconductor laser diode (1.92 eV excitation) or 2nd harmonics of YAG:Nd diode-pumped solid state laser (2.34 eV excitation) for the PL excitation. ODCR signals were detected via relevant PL changes with either photomultiplier or CCD camera. Changes in intensity or a shift of the PL spectral position were measured synchronously with on-off modulated microwave power. The experiments were performed on a range of II–VI (CdTe/CdMnTe, CdMnTe/CdMgTe, ZnCdSe/ZnSe) and III–V (GaAs/AlGaAs) QW structures and in thin films, supplied by the co-authors. PL, magneto-PL, and time-resolved investigations were also performed using conventional setups. The results for the two first types of structures will be discussed in detail.

3. Principles of the ODCR method

The mechanisms responsible for the optical detection of cyclotron resonance in bulk samples are related to carrier heating at CR conditions [3, 4]. In the case of dumped CR signals carriers are heated at nonresonant conditions. Then the method should be referred to as microwave-induced free carrier heating. In both cases (resonant or nonresonant conditions) carrier heating can influence the rate of their trapping, the rate of exciton formation, or can lead to dissociation of bound excitons and ionization of shallow impurities. The latter two effects are caused by impact with microwave-heated hot carriers (see Ref. [3] and references given therein).

At low temperatures and for low-dimensional structures with rough interfaces, interaction of hot carriers (heated by microwaves) can result in carrier/ exciton delocalization, as is discussed here. Then, the CR signal is detected via a shift in a spectral position of the relevant PL emission, as shown in Fig. 1. The up-in-energy shift of the PL is shown for a multiple ZnCdSe/ZnSe QW structure with 12% Cd fraction in the QWs. We show the PL response to the CR conditions for two ZnCdSe QWs, 2.2 nm and 4.3 nm wide. For a narrow QW PL shifts towards a higher energy once microwave power is applied. The magnitude of this shift allows us to estimate the Stokes shift, as shown in Fig. 1, equivalent to the one estimated by us from the comparison of the PL and PL excitation (PLE) spectra. We detected a similar response for a high quality ternary GaAlAs alloy, showing a dominant free excitonic (FE) PL and only weak donor bound exciton (DBE) PL. DBE PL is reduced (by about 1%) in intensity, whereas FE PL shows a very small up-in-energy shift, which could not be resolved in the PLE study.

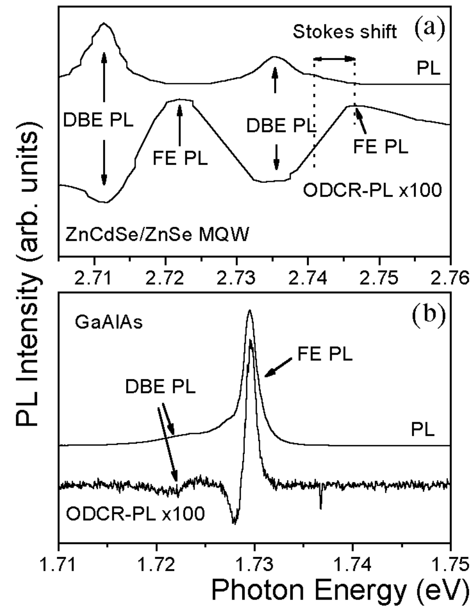

Fig. 1. PL and response of the PL emission to the cyclotron resonance conditions, observed for (a) ZnCdSe/ZnSe MQW structure and for (b) GaAlAs epilayer. DBE PL emissions are deactivated and localized excitons are replaced with free excitons. The observed microwave-induced shift between localized and FE excitons estimates the Stokes shift.

For the 4.3 nm wide ZnCdSe QW ODCR proved that a low temperature PL is dominated by a radiative decay of donor bound excitons. DBE PL is quenched and free excitonic PL is enhanced at CR conditions, as can be seen in Fig. 1. We observed very different response to microwaves in ZnCdSe/ZnSe structures showing stronger localization effects. DBE PL was enhanced once excitons were delocalized. Apparently DBE formation was deactivated once excitons are trapped in a QW plane.

To confirm the latter conclusion we performed ODCR and PLE investigations for a series of ZnCdSe/ZnSe QW structures with a different strength of localization. Properties of these structures were discussed in Ref. [5]. By selecting excitation conditions we could excite either free (localized) or DBE PL. At low temperatures selectivity of the excitation was 100%, confirming that the link between two excitonic transitions is deactivated by exciton localization. This link is retained at increased temperatures or by applying microwave power. With increasing temperature the selectivity of the PLE decreases fast and is not observed once excitons are thermally delocalized.

4. Excitons in the limit of a strong localization

Once we established possible responses of the QW PL to the applied microwave power, we used the ODCR to explain anomalous PL properties observed by us for a series of CdTe/CdMnTe structures with a varying Mn fraction (10, 30, 51, and 68%)) in 50 nm wide CdMnTe barriers and with 2, 4, 6, and 10 nm wide CdTe QWs. In this work we discuss the results of the ODCR study for the 10 nm wide QW in the CdTe/CdMnTe structure with 68% Mn fraction. For 10 nm wide QW penetration of an exciton wave function to the barrier regions is minimized. Thus we could avoid complications related to a magnetic polaron formation.

4.1. Origin of anomalous temperature dependence of QW PL

PL for the 10 nm wide QW shows several anomalous properties. PL decay time is not only significantly longer than that expected for free excitons, but is also energy dependent, i.e., PL decay time changes within the whole PL line width [1]. PL decay time is the shortest at high-energy wing of the PL and is the longest at low-energy wing of the PL. In addition, S-like temperature dependence of the PL line width, similar to that reported for quantum dot systems [6, 7], was observed. PL line first narrows at increased temperatures, then its width comes back to the one observed at liquid helium temperature. Only for temperatures above 80 K it shows a linear dependence on temperature, as expected for free excitons interacting with phonons. Anomalous temperature dependence of the PL energy is also observed. Energy of the PL first increases with increasing temperature and only for further increased temperatures starts to follow temperature-induced changes of the band gap energy.

For quantum dots (QDs) S-like temperature dependences of the PL line width and PL energy were explained by a temperature-induced exciton migration among dots of various sizes [6, 7]. By analogy, we tentatively related PL properties for the system studied to a high density of localization sites in a QW plane and to a very slow migration (tunneling) of excitons among these sites [8]. Such migration must be slow, as compared to exciton decay time. Then excitons do not thermalize to the sites of the lowest energy (the largest localization strength) during their lifetime. PL line is inhomogeneously broadened and shows energy dependence of the lifetime. Increase in temperature can first help excitons to redistribute among localization sites — at low temperature mostly to sites of the lowest energy, and at increased — among all localization sites, including those with the highest energy (the weakest localization).

To verify this model, and explain why S-like dependence of the PL line width is not accompanied by a similar dependence of the PL energy, we applied the ODCR study. We found that redistribution of excitons can also be promoted by application of microwaves at CR conditions, as shown in the inset of Fig. 2. Scattering of microwave-heated hot carriers at localized excitons helps them to redistribute to sites of a lower energy (low microwave power) or a higher energy (high microwave power). Microwave-induced PL shifts occur within the PL line width. We observe this in the ODCR study as a derivative-like response of the 10 nm QW PL to the applied microwave power. Positive signal means that PL at a given energy is enhanced. Negative signal means that PL is deactivated.

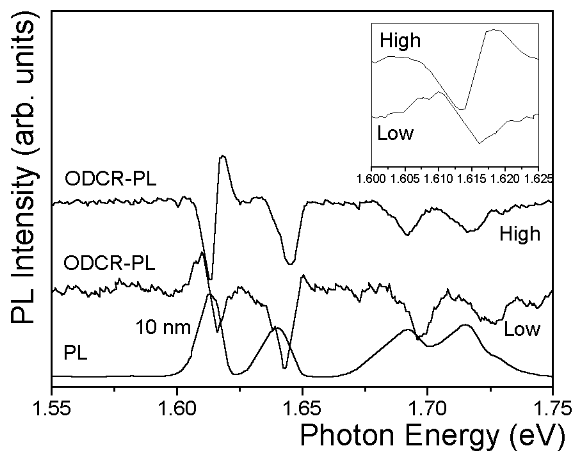

Fig. 2. Photoluminescence and response of the QW PL to the cyclotron resonance conditions (ODCR-PL spectrum), observed for the 2, 4, 6, and 10 nm wide CdTe QWs in CdTe/CdMnTe structure with 68% Mn fraction in the barriers. ODCR-PL spectra for low (1 mW) and high (200 mW) microwave power are shown. In the inset we show in more detail ODCR-PL spectrum observed for the 10 nm QW.

Microwave induced PL changes are fairly small and are of about 1% of a signal amplitude. AC detection in phase with on-off modulated microwaves was necessary to measure ODCR-PL spectra.

The microwave induced PL shifts are of about 8 meV (taken as a difference between peaks in the ODCR-PL spectra shown in the inset of the Fig. 2), which estimates strength of potential fluctuations present in a QW plane. This value is comparable to a Stokes shift between PL and PLE spectra, measured for this QW. The estimated localization energy is relatively large. Thus we did not observe microwave-induced delocalization of excitons but mostly their redistribution among localization sites. Exciton migration among localization sites is very small, as we confirmed by time-resolved PL and also a small magnitude of microwave-induced PL shifts. This most likely explains why we do not observe S-like shift of the PL energy.

Figure 2 also shows that scattering of hot carriers at excitons can help them to escape from narrower QWs, observed in the ODCR-PL as a weak quenching of the PL from 2, 4, and 6 nm wide QWs. For structures with large Mn fraction in the barriers the effects were relatively small. For those with 10% Mn fraction the effect was much stronger. For this structure the PLE experiments showed a surprisingly efficient inter-QW transfer. The process was fast deactivated with increasing temperature and also at increased magnetic field. Thus, it cannot be explained by over the barrier carrier migration. Mechanism of inter-well migration remains unknown.

4.2. Mechanism of localization

Surprisingly, the scenario of localization often remains unknown. In an ideal case (smooth interfaces, weak localization) photogenerated and inequilibrium free carriers dissipate their excess energy by a multiphonon emission. First, a cascade of LO phonons is emitted, followed by emission of rest of energy by acoustic phonons. Then, free excitons are formed or free carriers are trapped at impurity sites. Free excitons decay radiatively or some of them can be bound at localization sites.

Definitely, such scenario cannot be used for structures showing strong localization effects. For these structures thermalization of energy by hot carriers (excitons) is significantly slower. Then, recombination of unthermalized carriers (excitons) can be observed. The most striking consequence was observation of the so-called hot PL and of much slower PL decay times [1].

The unsolved question is if carriers can be localized in a QW plane before being trapped to form excitons. We applied the ODCR study to answer this question. CR signals were detected via the change in intensity of the PL of localized excitons, with the PL detection set at different energies within the PL line width. The idea of the experiment is very simple. We showed that shift of the PL in the ODCR-PL spectra can be explained by delocalization of excitons caused by scattering of microwave-heated hot carriers. If carriers interacting with excitons are free then identical CR signals should be detected via QW PL, when detection is set at different PL energies. If carriers interacting with localized excitons are also site localized, then resonant signals should be observed at different fields, reflecting carrier localization.

The ODCR results shown in Fig. 3 were taken for a single 8 nm wide QW in CdMnTe/CdMgTe structure with 1% Mn fraction in the well and 20% Mg fraction in the barriers. For the 10 nm QW in the CdTe/CdMnTe (68% of Mn) structure electron and heavy hole CR detected via QW PL were too broad and too weak to perform a similar study.

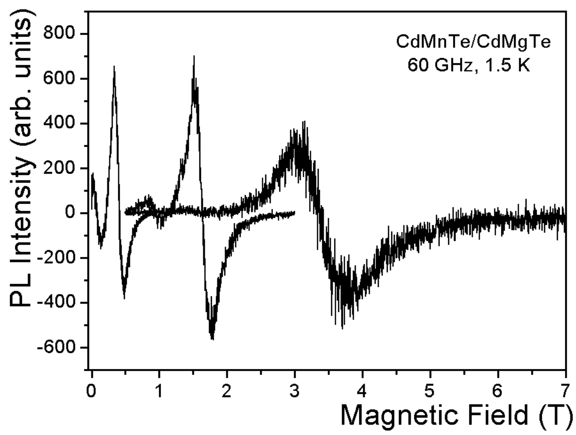

Fig. 3. Cyclotron resonance signals of electrons detected at high-energy (signal in the lowest field), at maximum, and at low-energy wing (signal in the highest field) of the PL emission of localized excitons in 8 nm wide QW in CdMnTe/CdMgTe (1% of Mn in the QW, 20% of Mg in the barriers) structure, showing strong localization effects.

We attribute the observed resonance signal (see Fig. 3) to CR of electrons. When detected at high-energy wing of the PL the calculated effective mass is $0.098m_0$, which is close to the electron effective mass in CdTe. However, the CR signal shifts to higher magnetic fields for PL detection shifted to lower energies within the PL line width. Moreover, the signal significantly broadens.

For a magnetic QW two effects contribute to the observed exciton localization — potential fluctuation caused by fluctuations of a QW width and also composition fluctuations. Moreover, for localized excitons a magnetic polaron effect is expected. We have shown recently that scattering of free carriers at Mn localized spins can increase an effective spin temperature and thus decrease sample magnetization [9]. Then PL of a magnetic QW should shift up-in-energy. In our experiment PL is shifted down-in-energy at CR conditions, indicating a dominant role of exciton redistribution among localization sites, rather than of magnetic polaron effects.

Shifting detection energy to a low energy wing of the PL, i.e., increasing contribution of strongly localized excitons to the PL, we observed that CR resonance signals also shift. This observation indicates that electrons interacting with localized excitons are also localized, i.e., not only excitons but also carriers can be site-localized in a QW plane.

5. Conclusions

We demonstrate that the ODCR study can yield detailed information on exciton properties in low-dimensional structures. Some anomalous temperature dependences of excitonic PL are related to strong localization effects. Using the ODCR we also show that localization limits formation rate of bound excitons by trapping at donor site of free excitons, and that not only excitons, but also carriers can be localized in a QW plane in structures showing strong localization effects.

Acknowledgment

This work was partly supported by grant no. 5 P03B 007 20 of the State Committee for Scientific Research (Poland) granted for years 2001–2003.

References

[1] M. Godlewski, *Mater. Sci. Forum, Solid State Phys. Eng.* **297/298,** 197 (1999).

[2] S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Matushita, T. Mukai, *MRS Internet J. Nitride Semicond. Res.* **4S1,** G1.1 (1999).

[3] M. Godlewski, W.M. Chen, B. Monemar, *CRC Crit. Rev. Solid State Mater. Sci.* **19,** 241 (1994).

[4] M. Godlewski, *Opt. Appl.* **30,** 463 (2000).

[5] M. Godlewski, J.P. Bergman, B. Monemar, E. Kurtz, D. Hommel, *Appl. Phys. Lett.* **69,** 2843 (1996).

[6] A.E. Zhukov, V.M. Ustinov, A.Yu. Egorov, A.R. Kovsh, A.F. Tsatsulnikov, N.N. Ledentsov, S.V. Zaitsev, N.Yu. Gordeev, P.S. Kopev, Z.I. Alferov, *Jpn. J. Appl. Phys.* **36,** 4216 (1997).

[7] G. Karczewski, S. Mackowski, M. Kutrowski, T. Wojtowicz, J. Kossut, *Appl. Phys. Lett.* **74,** 3011 (1999).

[8] M. Godlewski, V.Yu. Ivanov, A. Khachapuridze, R. Narkowicz, P.J. Bergman, B. Monemar, *Mater. Sci. Forum* **384/385,** 19 (2002).

[9] M. Godlewski, V.Yu. Ivanov, A. Khachapuridze, S. Yatsunenko, *Phys. Status Solidi B* **229,** 533 (2002).