

## Diffusion of Carriers Induced by Exchange Interaction with Magnetic-Ion System in (Zn, Mn)Se/(Zn, Be)Se Quantum Wells

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We studied the diffusion of carriers in dilute magnetic semiconductors for conditions when an additional force caused by a spatially inhomogeneous exchange potential is active. The spatially inhomogeneous exchange potential arises from a creation of photocarriers in the  $\text{Zn}_{0.988}\text{Mn}_{0.012}\text{Se}/\text{Zn}_{0.94}\text{Be}_{0.06}\text{Se}$  quantum well. The photocarriers enhance the spin temperature of the Mn ions by a spin-flip scattering with the magnetic ions, which results in a reduction of the giant Zeeman splitting. The gaussian like intensity profile of the exciting laser beam causes a locally varying exchange potential for the carriers. A strong influence of the gradient in the exchange potential on the diffusion of the carriers was found. The diffusion of the carriers was examined by a spatially resolved photoluminescence technique.

**Introduction** The strong sp–d exchange interaction of the carriers with the localized Mn electrons in diluted magnetic semiconductors (DMSs) results in the well-known giant Zeeman splitting of the spin sublevels of the valence and conduction band. For small Mn concentrations the creation of photocarriers causes a reduction of the Zeeman splitting, due to an elevated spin temperature of the magnetic ions. In detail the heating of the Mn system has been studied in Refs. [1, 2]. In short the mechanism can be explained with the inset of Fig. 1c. The photocarriers transfer energy to the magnetic ion system by a spin-flip scattering. The excess energy of the magnetic ion system can be transferred to the phonon system via a spin-lattice relaxation. The efficiency of this channel depends on the spin-lattice relaxation time  $\tau_{\text{SL}}$ , which is a strong function of Mn content  $x$  [3]. For small Mn concentrations of about  $x = 0.01$   $\tau_{\text{SL}}$  is very long (about 100  $\mu\text{s}$ ) and as a result the temperature of the Mn system exceeds the lattice temperature under steady state photoexcitation. In external magnetic fields a local laser illumination causes a locally varying exchange potential (see scheme in Fig. 3). The additional force originating from the gradient of the exchange potential is independent of the charge state of the particles and acts on electrons, holes and neutral excitons. We found experimentally that the gradient of the exchange potential enforces the diffusion of the carriers.

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**Experimental Details** The studied  $\text{Zn}_{0.988}\text{Mn}_{0.012}\text{Se}/\text{Zn}_{0.94}\text{Be}_{0.06}\text{Se}$  multiple-quantum-well (MQW) structure was fabricated by molecular beam epitaxy. It consists of five periods of 200 Å thick  $\text{Zn}_{0.94}\text{Be}_{0.06}\text{Se}$  barriers with an energy gap of 2.97 eV and 100 Å thick  $\text{Zn}_{0.988}\text{Mn}_{0.012}\text{Se}$  wells. The spatially resolved photoluminescence measurements were performed in liquid helium at a temperature of 4.2 K [5]. Magnetic fields up to 6 T were applied parallel to the growth axis of the structure and to the direction of the collected light (Faraday geometry). An Ar-ion laser operating at the wavelength of 363.8 nm was used for excitation of photoluminescence (PL). The laser was focused onto the sample surface by a 40× microscope objective. The luminescence was collected by the same objective and imaged on the slit of a monochromator. The signal detected by a CCD camera was spectrally resolved along the horizontal axis, and spatially resolved along the vertical axis [5]. The spatial resolution of the setup is limited by 0.7 μm.

**Experimental Results and Discussion** Figure 1a shows photoluminescence spectra of the sample for different magnetic fields at 1.6 K. The spectra were measured under very low photoexcitation density of  $P = 0.016 \text{ W/cm}^2$  and without microscope objective. At  $B = 0 \text{ T}$  the spectrum consists of two lines. The line at 2.8154 eV corresponds to the heavy-hole exciton (X). Another line at 2.8105 eV is due to the recombination of the negatively charged exciton ( $X^-$ , trion), which is a complex of two electrons bound to

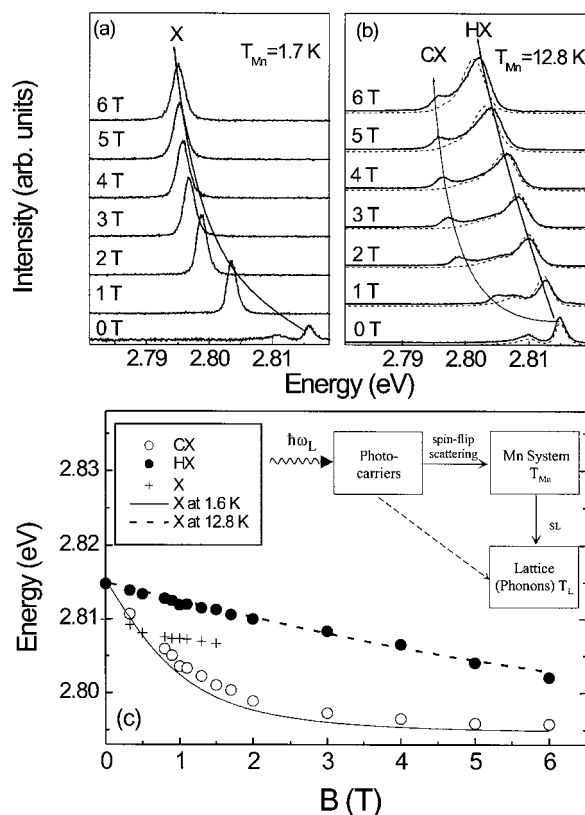


Fig. 1. a) PL spectra with  $P = 0.016 \text{ W/cm}^2$  at  $T = 1.6 \text{ K}$ . b) Spectra with  $P = 3.8 \text{ W/cm}^2$  (dashed lines) measured with defocused laser and mask, and with an excitation density which induces comparable heating of the Mn system (solid lines) measured with focused laser. Exciton lines associated with crystal sites with “hot” and “cold” Mn system are labeled as HX and CX, respectively. c) Peak energy of the lines in panel (b) as a function of magnetic field. The inset shows the energy reservoirs and relaxation processes which participate in the Mn heating

one hole. In external magnetic fields the X line shifts to lower energies due to the sp–d exchange interaction between the carriers and the localized electrons of the Mn ions. The temperature of the Mn spin system can be derived by fitting the energy shift of the X line [1, 2]. For such a low excitation density as  $0.016 \text{ W/cm}^2$  the Mn system is not heated over the lattice temperature. A fitting of the energy shift yields a value of  $T_{\text{Mn}} = 1.7 \text{ K}$ , which is very close to the bath temperature of  $1.6 \text{ K}$ . The trion line is suppressed with growing magnetic fields (for details see Ref. [4]). Dashed lines in Fig. 1b are PL spectra of the sample measured at a higher excitation density of  $3.8 \text{ W/cm}^2$ . The sample was covered by a mask with a  $1 \text{ mm}$  pinhole. The exciting laser spot was chosen larger than the pinhole size to provide a spatially homogeneous excitation. At zero magnetic field the spectrum coincides with the spectrum measured under low excitation density. However, it can clearly be seen that the energy shift of the exciton is reduced in magnetic fields. The reduction of the Zeeman splitting can be explained by an elevated spin temperature of magnetic ions. A fitting of the energy shift of the exciton yields a value of  $T_{\text{Mn}} = 12.8 \text{ K}$ . We stress here that at these experimental conditions the lattice temperature of the sample immersed in the pumped He is not expected to deviate strongly from  $1.6 \text{ K}$ . Solid lines in Fig. 1b represent spectra of the sample measured with focused laser beam and without mask, using an excitation density which induces comparable heating of the magnetic ion system. In magnetic fields a third line, labeled by CX, appears in these spectra in addition to the exciton and the trion line in the corresponding spectra measured with mask. Figure 1c shows the energy of the three lines in dependence of magnetic field. The energy of the additional line CX follows the energy shift of the exciton at a temperature of  $1.6 \text{ K}$ . From the fact that the spectra, represented by solid lines are measured without mask, we conclude that this line originates from a recombination of carriers at the edge of the laser spot. If we take into account that the intensity profile of the laser has a gaussian like shape, we can conclude that the heating of the magnetic ion system decreases at the edge of the laser spot, because it depends strongly on the laser intensity. But for a smooth decrease of the exciton intensity at the edge of the laser spot, we would expect a low energy tail of the exciton line rather than a sharp additional line of such high intensity. A possible explanation for the sharp line could be that the carriers, excited at the edge of the laser spot are driven into regions with a lower Mn temperature, because of the gradient of the sp–d exchange potential induced by the profile of the laser. The inset of Fig. 3 illustrates schematically the formation of such a gradient.

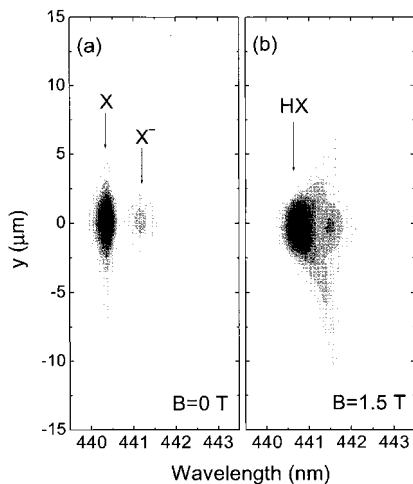


Figure 2 shows spatially resolved PL spectra of the sample for  $B = 0 \text{ T}$  and  $1.5 \text{ T}$ , in which the intensity is given as a contour plot in de-

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Fig. 2. Spatially resolved PL spectra at  $B = 0 \text{ T}$  and  $B = 1.5 \text{ T}$  at a temperature of  $T = 4.2 \text{ K}$

pendence of the wavelength and of the position  $y$  on the sample. At zero magnetic field two lines are clearly seen, corresponding to the exciton and the trion. The full-width-at-half-maximum of the exciting laser spot is  $\Delta_L = 1 \mu\text{m}$ . The larger width of the luminescence spot of  $\Delta_{\text{PL}} = 3.8 \mu\text{m}$  results from the diffusion of the carriers. The spectrum at  $B = 1.5 \text{ T}$  shows also two peaks due to the recombination of the exciton and the trion, respectively. The spectrum was measured at a high excitation density of  $P = 250 \text{ kW/cm}^2$ . As a result the temperature of the Mn spin system is elevated under the laser spot, i.e. the luminescence is shifted to shorter wavelengths under the laser spot. For increasing  $y$ , however, the Mn temperature decreases and as a result the energy of the  $X$  shifts to smaller values. The trion formation is suppressed outside of the laser spot, because of the increasing Zeeman splitting of the electrons.

The effect of the gradient in the exchange potential on the carrier diffusion is illustrated in Fig. 3, which shows the spectrally integrated PL intensity in dependence of the position  $y$  on the sample. For zero magnetic field no additional force is present (circles). For  $B = 5 \text{ T}$  (triangles) the width of the luminescence is enhanced compared to the case of  $B = 0 \text{ T}$ . This differs from the case of nonmagnetic semiconductors, where the diffusion coefficient decreases with growing magnetic fields [5]. The intensity profile of the exciting laser is represented by squares. It can be fitted reasonably well with a gaussian with  $\Delta_L = 1 \mu\text{m}$  (dotted line). Experiments with low excitation densities, for which no heating of the Mn system is expected show no dependence of the PL spot width on the magnetic field. From this fact we conclude that the enhancement in the PL spot width can be assigned to the locally varying sp-d exchange potential.

In order to quantify the influence of the gradient of the exchange potential, the dependences from Fig. 3 were fitted with a simple model in which the exchange potential is assumed to have gaussian like profile, i.e.  $V_{\text{ex}} = V_0 \exp[-(x^2 + y^2)/2\Delta_{\text{PL}}^2]$ . The diffusion of the carriers including the current induced by the spatially varying exchange potential can be described by the equation

$$\frac{\partial n}{\partial t} = \mu \nabla \left( n \nabla \frac{V_{\text{ex}}}{e} \right) + D \Delta n - \frac{n}{\tau} + G = 0,$$

where  $n$  is the carrier density, which is assumed to be proportional to the luminescence intensity,  $\tau$  is the carrier lifetime,  $\mu = De/k_B T$  is the mobility and  $G$  is the generation rate

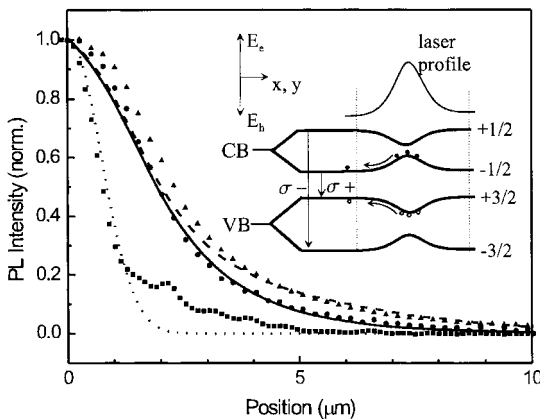


Fig. 3. Spectrally integrated PL intensity at  $B = 0$  (circles) and  $B = 5 \text{ T}$  (triangles) in dependence of the position on the sample. The intensity profile of the exciting laser is shown by squares. Solid and dashed lines represent fittings with the model described in the text. The inset illustrates the formation of the gradient in the sp-d exchange interaction

of the photocarriers. The generation rate is considered by  $G = G_0 \exp [-(x^2 + y^2)/2\Delta_L^2]$ , i.e. a gaussian-like laser profile. The results of the numerical solution of the diffusion equation using supposed lifetime of  $\tau = 100$  ps, which is a typical decay time for exciton PL in ZnSe-based QW are represented by lines in Fig. 3. For zero magnetic field no additional exchange potential is present ( $V_{\text{ex}} = 0$ ) and the experimental data can be described reasonably well using a diffusion coefficient of  $D = 400$  cm<sup>2</sup>/s. In order to fit the PL spot profile at  $B = 5$  T with the diffusion equation the value of the diffusion coefficient was fixed to  $D = 400$  cm<sup>2</sup>/s, as derived for  $B = 0$ , and the value of maximum difference of exchange potential  $V_0$  was varied. The best agreement of experimental data with the simulations could be achieved for  $V_0 = 2$  meV (dashed line), which is in good qualitative agreement with the value of  $V_0$  derived experimentally from the shift of the exciton line of  $V_0 = 7$  meV. Compared to literature data of the diffusion coefficients in bulk II–VI semiconductors the absolute value of  $D$  is about one order of magnitude too large [6]. A possible explanation for this could be that we do not excite excitons resonantly, but we excite carriers with a big excess energy above the band gap. Non-resonant excitation results in a broadening of the PL spot profile [5].

In conclusion we found a possibility to enhance the diffusion of carriers in DMS heterostructures by an additional force. The force which acts on the carriers is generated by a creation of photocarriers which causes a local heating of the Mn-ion system, which results in a gradient of the sp–d exchange potential.

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## References

- [1] B. KÖNIG, I.A. MERKULOV, W. OSSAU, S.M. RYABCHENKO, M. KUTROWSKI, T. WOITOWICZ, G. KARCEWSKI, and J. KOSSUT, *Phys. Rev. B* **61**, 16870 (2000).
- [2] D. KELLER, D.R. YAKOVLEV, B. KÖNIG, W. OSSAU, TH. GRUBER, A. WAAG, and L.W. MOLENKAMP, to be published in *Phys. Rev. B* **64** (2002).
- [3] A.V. SCHERBAKOV, A.V. AKIMOV, D.R. YAKOVLEV, W. OSSAU, G. LANDWEHR, T. WOITOWICZ, G. KARCEWSKI, and J. KOSSUT, *Phys. Rev. B* **62**, R10641 (2000).
- [4] B. KÖNIG, U. ZEHNDER, D.R. YAKOVLEV, W. OSSAU, T. GERHARD, M. KEIM, A. WAAG, and G. LANDWEHR, *Phys. Rev. B* **60**, 2653 (1999).
- [5] F. PULIZZI, W.H.A. THIJSSSEN, P.C.M. CHRISTIANEN, and J.C. MAAN, *Physica B* **298**, 441 (2001)
- [6] J. ERLAND, B.S. RAZBIRIN, K.H. PANTKE, V.G. LYSENKO, and J.M. HVAM, *Phys. Rev. B* **47**, 3582 (1993).

