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Magnetooptical studies in CdSe/(Zn,Mn)Se semimagnetic nanostructures

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Recently diluted magnetic semiconductor (DMS) heterostructures have received increasing attention due to the potential optoelectronic and magnetoelectronic applications based on spin-polarized transport in semiconductors [1]. In this context, the studies of electronic spin behavior in 0D structures with a discretized electronic system are of particular interest. An excellent opportunity for studying the spin-related phenomena in DMS nanostructures is provided by recently developed molecular beam epitaxy (MBE) technique allowing growth of Zn(Cd)Se-based quantum structures with varying dimensionality, from quantum wells (QWs) to quantum disks and dots [2–5]. To produce the zero-dimensional (0D) DMS structure one can incorporate Mn ions either into the quantum objects (disks or dots) or into their surroundings. The preliminary data on magnetooptics in the quantum dot (QD) structures of this type have recently been reported by Kim *et al.* [6].

In this paper we report on magneto-PL studies of CdSe/(Zn,Mn)Se nanostructures, focusing on the effects of Zeeman splitting and spin polarization of strongly localized 0D excitons. Three samples were grown for this purpose by MBE pseudomorphically on (001) GaAs substrates. The structures are non-magnetic CdSe single layers of nominal thickness w, embedded in the center of a 20 nm thick DMS layer of $Zn_{0.89}Mn_{0.11}Se$. The whole structure is surrounded by thick ZnBeSe barriers lattice-matched to GaAs. The CdSe insertion was grown using migration enhanced epitaxy (MEE) technique [2], whereas conventional MBE was used to fabricate all other layers of the samples. A, B and C samples differ only in the CdSe nominal thickness, with w being equal to 0.4, 0.8 and 1.9 monolayer (ML), respectively. According to the data obtained previously for CdSe/ZnSe (see e.g. [4, 5]), the intrinsic morphology of the insertion depends drastically on w, varying from the relatively homogeneous ZnCdSe graded-gap QW ($w < 0.6–0.8$ ML) with the average thickness of 4–6 ML to laterally inhomogeneous layers containing flat ZnCdSe islands with enlarged Cd content at larger w. The islands arise, most probably, due to strain-driven decomposition of the alloyed layer with large Cd content. Electronically they can be regarded as quantum disks providing 3D confinement for excitons [3]. Therefore, neglecting the possible effect of Mn atoms in the barriers on the ZnCdSe islands formation, one can assume that sample A represents a 2D QW exciton. Sample B is expected to present a QW system as well, perhaps suffering larger interface disordering. Sample C is an example of the inhomogeneous ensemble of 0D excitons localized in the ZnCdSe islands of different sizes and compositions. Magneto-PL measurements were carried out in Faraday geometry in a He cryostat with a split-coil superconducting magnet at 1.8 K, using a linearly-polarized emission of a cw Ar^+ laser at 364 nm. The PL signal was dispersed by a double-Spex monochromator and detected by a GaAs photomultiplier tube. Either

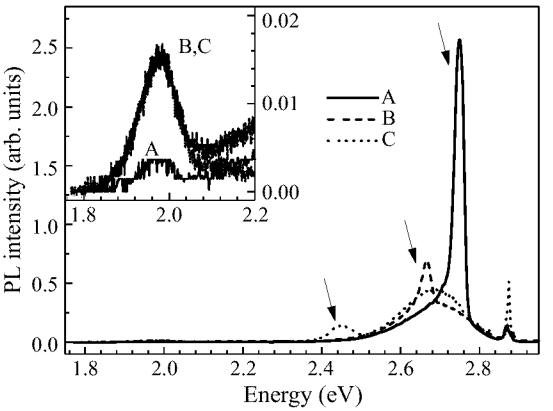

Fig. 1. PL spectra of samples A, B, and C at zero magnetic field.

the σ^+ or σ^- component of the PL was selected by a quarter-λ plate coupled with a linear polarizer.

Figure 1 demonstrates zero-field PL spectra of the samples A, B, and C. An increase in w causes a red shift of the PL peak attributed to the emission of localized heavy-hole excitons in a CdSe insertion (the peaks are pointed in Fig. 1 by arrows). Another effect is the peak broadening, which reflects enhancement of the layer structural disordering in good agreement with the data on similar nonmagnetic CdSe/ZnSe structures [2]. Three additional emission bands are visible for all the samples at 2.875 eV, 2.69 eV, and 2.0 eV, being attributed to the near-band-edge excitonic emission in ZnBeSe claddings, DAP recombination in the barriers, and intra-shell transitions within Mn ions, respectively. Note that only position of the peaks related to the CdSe insertions depends on w. Furthermore, this peak is the only feature in the spectra, depending on a magnetic field. Particularly, an increase in a magnetic field results in (i) Zeeman shift of the PL band, (ii) rapid increase in intensity of the σ^+ component, and (iii) large enhancement of the integral PL intensity. The latter observation was also reported by Kim *et al.* [6], who observed a field-induced increase in the integral intensity of exitonic emission from CdSe/(Zn,Mn)Se nanostructures, accompanied by quenching of the PL related to Mn inter-shell transitions. Owing to this observation, the authors of [6] explained this effect in terms of an efficient transfer of excitation between the excitons and the Mn ions, which is suppressed by a magnetic field. In our samples, we do not observe any field-induced changes of the Mn-related PL and, hence, the observed enhancement of the excitonic PL intensity implies some other mechanism of non-radiative recombination, quenched by a magnetic field. As it is seen from Fig. 2, the absolute value of the PL intensity decreases with w for any field in the range studied. The w-dependent behavior results probably from Mn ions clustering [7], which can be especially enhanced in the vicinity of the laterally inhomogeneous CdSe insertions. In this case, the centers of nonradiative recombination can be formed due to the locally enhanced concentration of Mn ions, with the cluster density and distribution depending on w.

Figure 3 illustrates the Zeeman shifts of the PL peak maximum for all three samples. For samples A and B the dependencies are alike and generally consistent with the giant Zeeman splitting effect observed in DMS QWs (see e.g. [8]). For sample C the dependence is almost twice weaker and the curve shape is more complicated. It is well known that the magnitude of Zeeman splitting in the structures containing both DMS and nonmagnetic

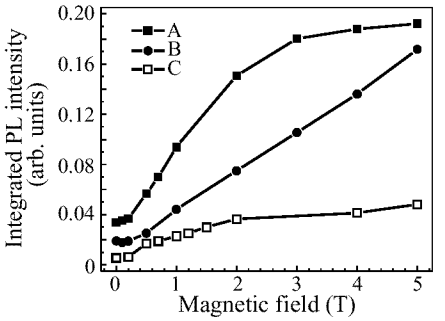

Fig. 2. Integrated PL intensity versus magnetic field.

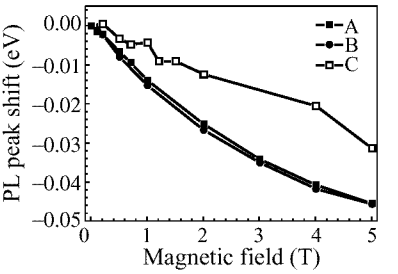

Fig. 3. PL peak shift versus magnetic field.

layers depends on many factors. Particularly, the Zeeman splitting of a given state reflects its weighted probability distribution over the two media, which can be a function of the magnetic-field-dependent band offsets in the heterostructure. Another important factor is the structure of an interface between the DMS and non-DMS materials, because manganese ions near to the interface possess the larger magnetism relative to those in bulk material [7]. In regard to the QW structures, an increase in w is expected to decrease Zeeman splitting due to simultaneous action of both the factors. Really, the larger content of Cd results in the deeper wells for confined carriers and, hence, in smaller penetration of the carrier wave function into the DMS barriers. Simultaneously, the probability to find an electron near the interface is smaller for a deeper QW, which should decrease the interface contribution.

However, very small difference between the curves in Fig. 3 for the QW samples (A and B) indicates negligible effect of w on the relative probability to find electron within the regions with higher effective concentration of Mn ions. Most probably, this results from the effect of Zn(Cd)-Mn interdiffusion during the structure growth, which causes unintentional incorporation of Mn into the thin layer of otherwise non-DMS CdZnSe alloy. To contribute to understanding of the different behavior of the sample C, we plot in Figs. 4(a) and (b) (samples A and C, respectively) the spectral contours of the PL intensity and circular polarization rate for two values of a magnetic field. For the sample A, the PL line shape weakly depends on a magnetic field. The respective polarization-rate contour follows the PL line when the latter is shifted by a magnetic field. The shape of the contour depends on the field rather weakly as well. The difference concerns mainly the region of two visible LO-phonon replica of the PL band. In contrast to that, the PL line of the sample C changes its shape noticeably with a magnetic field increase. Furthermore, the respective polarization-rate contour does not follow the PL line, but rather changes its shape, expanding to the red side with a field increase. These observations for the sample C can be explained, considering inhomogeneous nature of the PL line involving responses from 0D excitons localized in different cites. However, they hardly can be attributed only to the energy dispersion among the sites, because no large field-induced changes of the contour shape are expected in this case. The analyses of the inhomogeneous contours at different fields indicate that the observed behavior is rather related to the dispersion of the excitons magnetic properties, particularly of their Zeeman splittings, which assumes inhomogeneous distribution of Mn ions in the vicinity of the CdSe-based islands.

In conclusion, the performed magneto-PL measurements exhibit formation of 0D excitons in CdSe/(Zn,Mn)Se nanostructures. The excitonic states forming the inhomogeneously broadened emission band differ both in their energy (due to fluctuations in the sizes and compositions among the localization sites) and in magnetic properties (most probably,

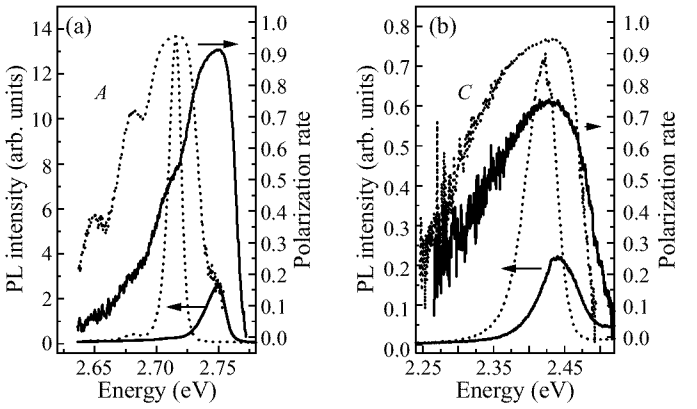

Fig. 4. PL spectra and circular-polarization-rate contours at 0.5 T (solid curves) and 4 T (dot curves) in the samples A and C (figures (a) and (b), respectively).

due to the effect of Mn ions clustering in the nearby regions). The structure potential for spin-related optoelectronic applications is demonstrated, owing to almost complete circular polarization of the emitted light within the wide spectral range (~100 meV) at moderate magnetic fields.

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