Epitaxial growth and Photoluminescence Excitation spectroscopy of CdSe Quantum Dots in (Zn,Cd)Se barrier

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Abstract
Design, epitaxial growth, and resonant spectroscopy of CdSe Quantum Dots (QDs) embedded in an innovative (Zn,Cd)Se barrier are presented. The (Zn,Cd)Se barrier enables shifting of QDs energy emission down to 1.87 eV, that is below the energy of Mn$^{2+}$ ions internal transition (2.1 eV). This opens a perspective for implementation of epitaxial CdSe QDs doped with several Mn ions as, e. g., the light sources in high quantum yield magnetooptical devices. Polarization resolved Photoluminescence Excitation measurements of individual QDs reveal sharp (Γ < 150 µeV) maxima and transfer of optical polarization to QD confining charged exciton state with efficiency attaining 26%. The QD doping with single Mn$^{2+}$ ions is achieved.

Keywords: quantum dot, exciton, Auger recombination, Photoluminescence Excitation, CdSe

1. Introduction
Selenium based Quantum Dots (QD) act as temperature robust non-classical light sources [1], what makes them excellent candidates for applications in optoelectronic and spintronic devices [2, 3]. In particular, appealing properties and functions have been predicted and demonstrated for CdSe QDs doped with either a single [4, 5] or a density [6, 7, 8] of magnetic ions, like Mn$^{2+}$. The development of advanced devices involving Mn-doped CdSe QDs and nanostructures is impeded, however, by Auger-type, non-radiative carrier recombination introduced by the magnetic dopants [9, 10, 11]. In the case of typical Mn-doped CdSe/ZnSe QDs, the QDs emission energy (~2.3 eV) exceeds the energy of the Mn$^{2+}$ intraionic $^6A_1 \rightarrow ^4T_{1(2)}$ transition (2.1 eV). As a result, the non-radiative channel of exciton decay related to the ions is efficient enough to limit significantly or quench completely the excitonic emission [6, 7, 8, 12, 13]. The excitonic emission can be, however, recovered by shifting the QD levels below the energy of the Mn$^{2+}$ ion transition. In the case of colloidal CdSe QDs such shifting has been achieved by increasing the nanocrystal size leading to a reduction of the exciton confinement energy [7] or by a coupling of the QDs to a graphene sheet [14]. A respective shifting method has been still missing, however, for epitaxial CdSe QDs.

Here, we design and demonstrate epitaxially grown CdSe QDs with energy of confined levels lowered with respect to a typical case of CdSe/ZnSe QDs by lowering of the barrier energy. We implement (Zn,Cd)Se barrier exploiting the fact that (Zn,Cd)Se band gap decreases with increasing Cd content [15, 16]. We show that for Cd content attaining 30 %, the CdSe QDs transitions are shifted down to 1.87 eV, that is far below 2.1 eV. This is beneficial for high quantum yield magnetooptical devices involving CdSe/(Zn,Cd)Se QDs doped with several Mn$^{2+}$ ions.

Due to a shortage of tunable and stable excitation sources operating in 590 nm - 620 nm spectral range, Photoluminescence Excitation studies of the selenium based QDs have been primarily limited to ensembles of 7, 17, 18 or individual 19 colloidal type CdSe QDs so far.

We demonstrate an optical addressing of individual CdSe QDs using a quasi-resonant excitation [20, 21, 22] with a standard Rhodamine 590 laser. We find an efficient transfer of the optical polarization for a charged exciton confined in the QD. We incorporate single Mn$^{2+}$ ions to the QDs by a low density doping at the sample growth stage. When combined with a possibility of resonant excitation, it makes the studied QDs promising for efficient optical orientation of a single magnetic ions spin [4, 5, 23]. Moreover, the emission range of the presented QDs provides a perspective for their implementation as temperature robust light sources and as building blocks of lasers operating in the short wavelength transmission window of plastic optical fibers [24].

2. Samples and experiment
A sample series comprises of twelve structures grown by Molecular Beam Epitaxy (MBE) on a GaAs (100) substrate followed by a 1.1 µm thick ZnSe buffer and 1 µm thick (Zn,Cd)Se barrier layer. The CdSe QDs are formed...
out of 2-3 monolayers of CdSe deposited in atomic layer epitaxy mode, 100 nm below the sample surface. The cadmium content in the barrier layer $x_{\text{Cd}}$ varies from 0% to 30%, depending on a sample from the series. Some of the samples are doped with a very low density of Mn$^{2+}$ ions in the QDs layer.

The samples undergo a characterization by $\mu$-Photoluminescence ($\mu$-PL) at $T = 10$ K excited non-resonantly at $E_{\text{exc}} = 3.06$ eV. Micro-Photoluminescence Excitation (PLE) experiment at $T = 1.8$ K employs a continuous wave dye (Rhodamine-590) laser tunable between 2.21 eV and 2.06 eV (560 nm-600 nm) as an excitation source. The excitation power is live-stabilized by a noise eater. The excitation beam is focused by a microscope objective down to 1 $\mu$m or 0.5 $\mu$m, in the case of $\mu$-PL characterization and PLE studies, respectively. Magnetic field of up to 10 T is applied in Faraday configuration ($\vec{k} \parallel \vec{B}$). The polarization resolved signal is detected using a Peltier-cooled CCD camera coupled to a 0.5 m grating spectrometer (100 $\mu$eV of overall spectral setup resolution).

3. **Tuning of CdSe/(Zn,Cd)Se Quantum Dots emission energy**

A typical emission pattern of a sample from the studied series is shown in Figure 1a). Here, the QDs emission is seen as a band in range of 2.04-2.08 eV. Much weaker (Zn,Cd)Se barrier and ZnSe buffer emission is centered at 2.49 eV at 2.79 eV, respectively. In the sample set, the (Zn,Cd)Se barrier emission energy varies from 2.46 eV to 2.79 eV (the latter corresponding to the pure ZnSe case), while the central energy of the QDs ensemble emission band covers the range from 1.87 eV to 2.38 eV, respectively. In Figure 1b) both, QDs and barrier, emission energies are plotted against one another.

For a quantum box, the lower potential of the confining barrier, the lower energies of the confined levels. For the case of QDs, the smaller band gap of a barrier and/or a QD material, the smaller energy of optical transitions of the QD confined excitons. Expected decrease of the QD emission energy with the decreasing barrier energy is indeed evidenced in Figure 1b). Despite of this clear trend, a scatter of QDs emission energies is present in Figure 1b). It reflects differences in the QD layer growth parameters (e. g., an amount of CdSe deposited or deposition temperature) in the case of a different samples. Thus, the Figure 1b) confirms that decreasing of the (Zn,Cd)Se barrier energy by the increase of Cd content provides an efficient way of tuning of the CdSe QDs energy. The lowest (Zn,Cd)Se achieved barrier energy in the sample set is equal to 2.46 eV, what determines the largest obtained $x_{\text{Cd}}$ in the barrier to 30% ± 3%, as intended on the sample growth stage. A further increasing of the Cd content in the barrier would result in a further lowering of the QD energies. We estimate that $x_{\text{Cd}} = 80\%$ would still ensure at least single QD confined level.

The Fig. 1b) clearly shows that, in particular, decreased barrier energy enables CdSe QDs with energy of emission lower then the Mn$^{2+}$ ion internal transition energy. The resulting hindering of the non-radiative recombination related to Mn$^{2+}$ ions is highly advantageous for studies and implementation of Mn-doped CdSe QDs [1]. Moreover, the QDs emitting in yellow spectral range (2.05 eV - 2.15 eV) are promising for implementation in lasers operating in the short wavelength transmission window of plastic optical fibers [24].

4. **Quasi-resonantly excited individual CdSe/(Zn,Cd)Se Quantum Dots**

In most of the studied samples, in the low energy tail of the QD emission band there are spectrally resolved emission lines attributed to individual QDs. Resonant or quasi-resonant excitation enables further limiting of the number of excited QDs and, in consequence, optical accessing of individual QDs. It opens a perspective for coherent manipulation of QD confined excitonic states [25, 21], or studies...
of spin memory effects on electron or ion confined to the QD.

Figure 2 presents the result of the PLE measurement on a selected CdSe/(Zn,Cd)Se QD (QD1). A map composed of emission spectra of the QD1 acquired for excitation energies increasing consecutively with a step of 72 meV in a range from 2107 meV to 2127 meV is shown in Figure 2a). A set of sharp (FWHM down to 0.15 meV) maxima is evidenced in the PLE map in Fig. 2a) for each excitonic transition of the QD1. The emission spectra of QD1 excited quasi-resonantly (at 2110.8 meV) and out of resonance (at 2107.5 meV) are displayed in Figure 2b). Much larger intensity of the emission for the quasi-resonant excitation case. as indicated by dashed lines for resonant (E_{exc} = 2110.8 meV) and non-resonant (E_{exc} = 2107.5 meV) excitation case.

Figure 2: (a) Photoluminescence Excitation spectra of a selected CdSe/(Zn,Cd)Se QD (QD1) at 2105-2127 meV excitation energy range. (b) μ-PL spectra of the QD1 at cross-sections of the panel a), as indicated by dashed lines for resonant (E_{exc} = 2110.8 meV) and non-resonant (E_{exc} = 2107.5 meV) excitation case.

The QD1 transitions are identified by measurements of anisotropy of linear polarization of emission (see Fig. 3a)) as a recombination of neutral exciton (X), biexciton (XX) and charged exciton (X^-). Exciton anisotropy splitting is determined to 80 μeV. The negative sign of the charged exciton is determined in PLE experiment, as described below. Energy difference between X and XX transitions is equal to 19.4 meV, the value that is common for the QDs in the studied set of samples and generally quite typical for CdSe QDs [28, 33]. The variation of transition energy of the QD1 with magnetic field applied in Faraday configuration (see Fig. 3b)) yields excitonic g-factor equal to 1.2, slightly smaller than for typical CdSe/ZnSe QDs [28, 20], and diamagnetic shift γ = 1.2 μeV/T^2 comparable to the one observed previously [29].

As it is seen in Figure 2b), no difference in resonance energy for X^- and neutral excitons is found, in contrary to the case of previously studied (In,Ga)As QDs [30]. Instead, the resonance energies are common for the X, X^- and XX transitions of QD1. A similar conduct has been observed previously in the case of CdTe/ZnTe QDs [31, 26], where it has been interpreted as a result of coupling between two neighboring QDs: the one at higher energy absorbing the excitation, and the emitting one at lower energy, to which the excitation is transferred. In the case of the presently investigated QDs, however, the absorbing state does not exhibit optical properties expected for an absorbing QD observed previously [31], like anisotropy exchange splitting with two orthogonal polarization axes and linear dependence of energy on the magnetic field due to the Zeeman effect (not shown). Energy distance between PLE maxima and QD1 transitions does not match to any multiple of LO-phonon energy in CdSe (26.5 meV), what excludes also the possibility of excitation via LO-phonon [28, 21, 22].

In order to investigate a transfer of exciton spin polarization between the absorbing and the emitting state, we perform polarization resolved PLE measurements. We
The heavy-hole spin relaxation rate is much faster than the emitting singlet state. Within such interpretation, the exciton, being the intermediate state of the transfer to heavy hole spin flip-flop process in triplet state of charged polarization transfer [33, 34, 2, 31, 5] results from electron-in Refs. [33, 34], the mechanism of such efficient, negative QDs is -26.1%. As it has been described in detail, e.g., for X, X
it is confined to the QD. The polarization transfer [32]. As a result, transitions exhibit a negligible transfer of polarization. It is expected, since exchange interaction in the anisotropic QDs are indicated in respective panels in Fig. 4. As it is seen, the X and XX transitions of QD1 obtained in that way are presented in Fig. 4.

In order to quantify the amount of exciton spin polarization conserved in the transfer process, we introduce degree of the polarization transfer $P$. The $P$ is defined for polarization of the excitation $\sigma^+$ as:

$$P_{\sigma^+} = \frac{I_{\sigma^+/-\sigma^+} - I_{\sigma^+/-\sigma^-}}{I_{\sigma^+/-\sigma^+} + I_{\sigma^+/-\sigma^-}}.$$

where $I_{\sigma^+/-\sigma^-}$ denotes the intensity of exciton transition excited in $\sigma^+$ and detected in $\sigma^-$ polarization. In analogy, the $P_{\sigma^-}$ is defined as $P_{\sigma^-} = (I_{\sigma^-/-\sigma^+} - I_{\sigma^-/-\sigma^+})/(I_{\sigma^-/-\sigma^+} + I_{\sigma^-/-\sigma^+})$. The values of $P_{\sigma^+}$ and $P_{\sigma^-}$ obtained for X, X’ and XX transitions of QD1 are indicated in respective panels in Fig. 4. As it is seen, the X and XX transitions exhibit a negligible transfer of polarization. It is expected, since exchange interaction in the anisotropic QD mixes spin-up and spin-down exciton states, which leads to a linearly polarized eigenstates of exciton. As a result, the initial polarization of the neutral exciton is erased after it is confined to the QD.

The Fig. 4 indicates, however, that in the case of the X’ transition the cross-polarized emission is favored with respect to the co-polarized one. The $P_{\sigma^+}$ and $P_{\sigma^-}$ attain the values of -15.6% and -18.3%, respectively. The maximum value of $P$ obtained for the X’ in the studied QDs is -26.1%. As it has been described in detail, e.g., in Refs. [33, 34], the mechanism of such efficient, negative polarization transfer [33, 34, 2, 31] results from electron-heavy hole spin flip-flop process in triplet state of charged exciton, being the intermediate state of the transfer to the emitting singlet state. Within such interpretation, the heavy-hole spin relaxation rate is much faster than the one of electron, and the electron is a majority carrier in the charged exciton (X’) complex. QD a

The QD embedding the single Mn$^{2+}$ ion exhibits a characteristic six-fold splitting of the exciton emission line [4, 33, 26]. An example PLE spectrum along with respective $\mu$-PL spectra providing the evidence for a quasi-resonant optical addressing of QDs that contain the single Mn$^{2+}$ ion are shown in Fig. 5. Similar emission patterns in the case of the line sets observed at energies of 2043 meV, 2047 meV, 2057 meV suggest that they are related to excitonic complexes (e.g., X, X’ and XX) coming from the same QD. However, for QDs without the Mn$^{2+}$ ion in the studied samples, resonance energies are common for all excitonic complexes confined in the same QD (see the example case of the QD1 in Fig. 4). Thus, the Fig. 5 with different resonance energy for each of the four transitions, would suggest that four QDs embedding the Mn$^{2+}$ ion contribute to the PLE and $\mu$-PL spectra.

Presence of four QDs with the single Mn$^{2+}$ ion under the 0.5 $\mu$m excitation spot would indicate the average planar density of such QDs equal to $50/\mu$m$^2$. Such value is at least two orders of magnitude higher than the estimated one. In any case, the locally increased density of QDs embedding individual Mn ions has been observed for several regions within the sample. The inhomogeneity of spatial distribution of such QDs suggests a positive correlation between Mn ions spatial positions within the QD layer.
arising at the sample growth stage, in particular during self-assembled QD formation. It may result from a kind of aggregation of the Mn$^{2+}$ ions prior to their adsorption on the sample surface during the MBE growth. More detailed studies are required in order to confirm and to determine the driving mechanism here.

5. Conclusions

Fabrication and properties of resonantly excited emission of CdSe QDs embedded in (Zn,Cd)Se barrier are presented. The (Zn,Cd)Se barrier enables lowering of energy of QD confined levels (down to 1.87 eV) with respect to a typical CdSe/ZnSe QDs (2.32 eV). This should facilitate transfer of polarization in the studied QDs is valuable for the field of quantum information processing, where stored spins may play a role of quantum memories or qubits controlled and measured optically [53, 33, 27].

Acknowledgments

This work was supported by the Polish National Research Center (NCN) projects under numbers UMO-2013/10/E/ST3/00215, DEC-2013/09/B/ST3/02603, DEC-2011/02/A/ST3/00131, and MNISW project Inven tus Plus IP2014 034573.

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