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Photoluminescence and magneto-optical properties of multilayered type-II ZnTe/ZnSe quantum dots

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ABSTRACT

Multilayered Zn–Se–Te structures grown by migration enhanced epitaxy are studied by temperature- and excitation-dependent photoluminescence (PL) as well as magneto-PL. The PL consists of two bands: a blue band, overlaid with band edge sharp lines, dominant at low temperatures and high excitation, and a green band, which appears at elevated temperature and low excitation. Upon varying excitation intensity by four orders of magnitude, the green band peak energy shifts by ∼60 meV, indicating recombination of excitons in type-II quantum dots (QDs); no significant shift is observed for the blue band. Therefore, the green emission is attributed to ZnTe/ZnSe type-II QDs, which co-exist with isoelectronic centers, responsible for the blue and band edge emissions. The existence of type-II ZnTe/ZnSe QDs is further confirmed by magneto-PL, for which the observed oscillations in the PL intensity as a function of magnetic field is explained in terms of the optical Aharonov–Bohm effect.

1. Introduction

Over the past decades, there have been intensive studies on the properties of quantum dots (QDs) for their device fabrication potential and interesting physical properties. Until recently, most of the studies on self-assembled quantum dots were carried out on the so-called type-I QDs, whereas only a few reports were found on type-II QDs. We note that recently interest in type-II QD systems has

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increased significantly [1–5]. Among many systems, the ZnTe/ZnSe QDs appear interesting and useful since it is predicted that the band alignment of ZnTe and ZnSe is type-II with a relatively large valence band offset between 0.8 and 1.0 eV [6] as well as substantial conduction bad offset. Moreover, the photoluminescence (PL) of bulk $ZnSe_{1-x}Te_x$ alloys is well studied by now [7–9], which allows one to separate ZnTe/ZnSe QD PL from that of Te-related isoelectronic centers (IC), which in general, co-exist with QDs [4,10].

Previously, Gu et al. [10] showed that ZnTe/ZnSe QDs form in multilayered Zn-Se-Te system grown by combination of molecular beam epitaxy (MBE) and its variation known as migration enhanced epitaxy (MEE) when three contiguous MEE deposition cycles of Zn-Te-Zn were sandwiched between ZnSe barriers. The samples grown with one such cycle did not show evidence for type-II ZnTe/ZnSe QDs [10,11], and the emission from such samples was always attributed to isoelectronic bound excitons (IBEs) [11]. For sample details, see Refs. [12,13].

Here we show via detailed temperature and excitation intensity PL studies, including magneto-optical experiments, that such QDs, albeit relatively small and low density, are also formed in the Zn-Se-Te multilayers grown with only one deposition cycle of Zn-Te-Zn. Henceforth, to ensure continuity in notations [10], we denote samples grown with one and three Zn-Te-Zn cycles as δ-ZnSe:Te and δ^3-ZnSe:Te, respectively. We also discuss the nature of the band edge excitons, which reveal themselves via very sharp emission lines.

2. Experimental

The low temperature and temperature-dependent PL measurements were carried out using a closed cycle refrigerating system. The 325 nm emission line from a He–Cd laser was used as the excitation source, and the excitation intensity can be varied over four orders of magnitude with the aid of the neutral density filters mounted on a dual-wheel holder in front of the laser. The emitted light was dispersed through a 3/4 m monochromator, and was then detected with a thermoelectrically cooled GaAs photomultiplier tube coupled to a photon counter. Magneto-PL was performed at the National High Magnetic Field Laboratory (NHMFL). The magneto-PL was recorded at 4.2 K in the Faraday configuration with magnetic field up to 25 T.

3. Results and discussion

Fig. 1(a) shows the low temperature PL on a δ-ZnSe:Te samples at zero magnetic field. The PL is a broad blue band overlaid with the sharp lines in the band edge region (see inset). Intensity-dependent PL at $T = 10$ K did not show shift in the peak energy of either the whole band or the sharp lines. In Fig. 2 we show in high resolution band edge PL for another sample to indicate the presence of the peak previously attributed to a free exciton in the barriers [11]. We note that the sharp lines are observed only in Zn-Se-Te with very low Te fraction [9,11]; samples, which have relatively high Te fraction show shoulders instead of clear PL lines [8]. These lines are tentatively attributed to excitons bound to pairs of Te atoms (Te_2) [9,11], and are shown to be single photon emitters [9].

When the temperature is increased above 40 K another broad peak starts to appear at the lower energy side of the dominant peak, and becomes more pronounced with a further increase in the temperature. As an example, in Fig. 2, we show the PL for one of the samples at $T = 70$ K taken at various excitation intensities. We fit the broad bands with two Gaussians centered at ~2.66 eV ("blue" band) and at ~2.47–2.52 eV ("green" band). For the energy shift of the latter band is obvious, we plot the peak energy positions of both bands as a function of the excitation intensity in Fig. 3. It is found that as the excitation intensity increases, over four orders of magnitude, the peak energy of the green band exhibits a large, ~60 meV, shift to higher energies, whereas the blue band barely shifts (~6 meV). This suggests different origins of the green and blue bands.

For the blue emission, which exhibits very little shift, a plausible explanation is the recombination of IC bound excitons formed by, nearest or non-nearest Te_2 [9,11,12]. The PL due to these IBEs have been widely reported for dilute bulk $ZnSe_{1-x}Te_x$ ($x \leq 4\%$) alloys and even for ZnTe/ZnSe quantum

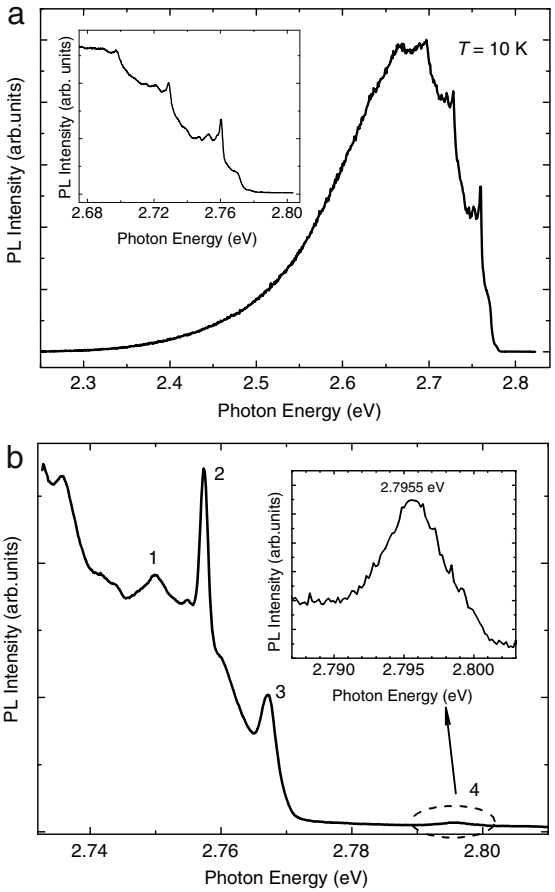

Fig. 1. Low temperature PL of δ-ZnSe:Te samples: (a) full spectrum PL with inset showing near band edge region; (b) a band edge PL region in detail with the inset showing the free exciton peak.

wells [14] with no reported intensity shifts. Moreover, in our previous low temperature work on δ^3-ZnSe:Te samples [10], a blue band did not shift with increasing excitation intensity either. The small shift observed here is probably due to overlap with the green band.

As to the origin of the green band, we note that a strong blue shift of emission spectra with increasing excitation intensity has been reported previously for various type-II quantum structures, including ZnTe/ZnSe QDs, and a band bending model at the interface has been proposed to explain this behavior [10,15–18]. Thus, we conclude that the green band is due, at least partially, to recombination of excitons within type-II quantum structures, and QDs in particular. We note that for our material structure, due to the large valence band offset between ZnTe and ZnSe, the holes are strongly confined in ZnTe QDs, while electrons locate in ZnSe barriers.

Therefore, it apparent that the δ-ZnSe:Te samples, even though grown with minimal amounts of Te, also contain both type-II QDs and ICs. Furthermore, the absence of the peak energy shift at low temperature suggests relatively low density of ZnTe/ZnSe QDs formed in the δ-ZnSe:Te sample, which is plausible due to low concentration of Te. It must be noted that the PL of the δ^3-ZnSe:Te sample is dominated by the QD-related emission even at 10 K, indicating high density of ZnTe/ZnSe QDs [2,10].

To further investigate the structures we performed magneto-PL measurements. In Fig. 4 we plot the peak position of the various sharp lines (as marked in Fig. 1(b)) as functions of the magnetic field up to 25 T. The peak at $\sim$2.798 eV exhibits relatively large diamagnetic shift, whereas the sharp peaks

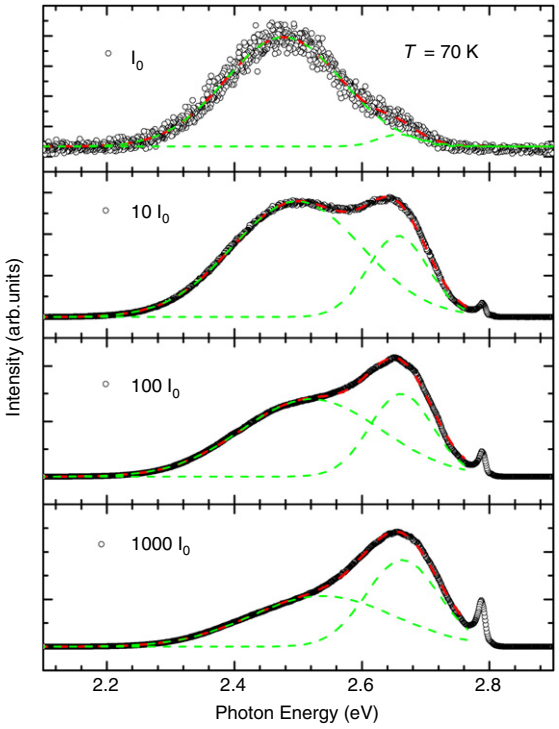

Fig. 2. Intensity-dependent PL of a δ-ZnSe:Te sample measured at 70 K. I_0 is the minimum intensity (in arbitrary units) of the experiment. Dashed (and dotted) lines are the Gaussian fits of two broad bands: centered at ∼2.66 eV ("blue" band) and at ∼2.47–2.52 eV ("green" band).

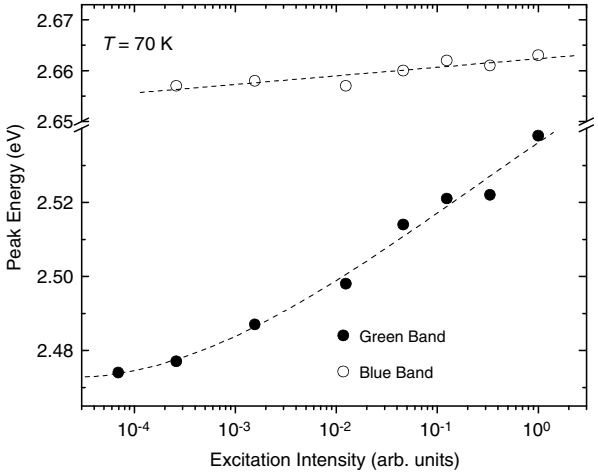

Fig. 3. Peak energy position of the blue and green bands as functions of excitation intensity as obtained via the two Gaussian fittings as shown in Fig. 2. The dashed lines are for eye guidance only.

barely show any shift. This strongly supports our conclusions that the sharp peaks are due to IBEs, for which hole is strongly confined and exciton wavefunctions is localized at the defect (this is consistent with the observed multiple phonon replicas, with Huang–Rhys factor, $S > 1$). The most significant

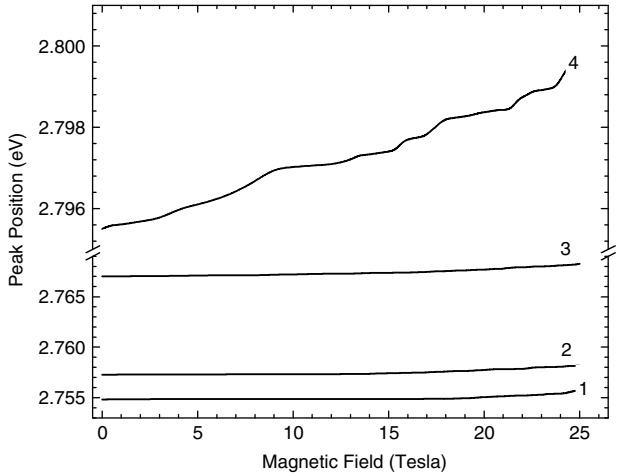

Fig. 4. Magnetic field dependence of the peak position for the band edge emission lines as denoted in Fig. 1(b). This indicates a strongly localized nature of the centers responsible for the emission lines 1–3.

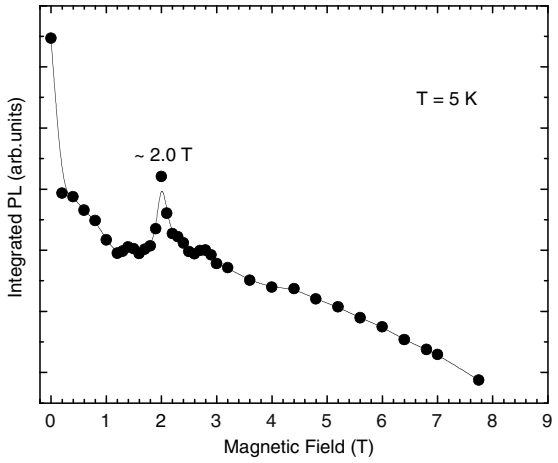

Fig. 5. The integrated PL intensity of a δ-ZnSe:Te sample as a function of magnetic field, B. The peak at $\sim$2.0 T corresponds to the electronic transition to non-zero orbital momentum. The overall decrease in intensity is attributed to the magnetic field induced carrier localization.

observation in magneto-PL, however, is the oscillation (with the peak at $B_0 \approx 2.0$ T) in the integrated PL intensity of the δ-ZnSe:Te samples as a function of the magnetic field B (Fig. 5). Despite overall decrease in the intensity due to localization effects, the oscillatory behavior of the intensity indicates a transition between bright and dark excitonic states with increasing magnetic field with B_0 being the field for which electron's orbital momentum, l_e, changes from zero to $l_e = -1$. Such behavior has been predicted previously and attributed to the optical Aharonov–Bohm (AB) effect for type-II QDs of cylindrical symmetry as well as quantum rings (see e.g. Refs. [19–24] and references there in).

This behavior is similar to observations for the δ^3-ZnSe:Te samples [2,25]. The value of the magnetic field B_0 allows one to estimate the lateral size of the dots—the higher the field the smaller the dots. The highest value for B_0 reported in Ref. [2] was 1.79 T, which suggests that the QDs formed in the δ-ZnSe:Te sample are indeed smaller. Assuming that the model developed in Ref. [2] is valid for our multilayered systems we estimate the radius of the quantum dots to be less than 9 nm.

4. Conclusions

We studied the properties of multiple multilayered Zn-Se-Te systems grown with a single deposition cycle of Zn-Te-Zn (δ-ZnSe:Te) sandwiched between ZnSe barriers. Temperature- and excitation-dependent PL as well as magneto-PL measurements confirmed the formation of ZnTe/ZnSe quantum dots that co-exist with isoelectronic centers. The density of these dots is low, as compared to a typical sample grown with three consecutive deposition Zn-Te-Zn cycles (δ^3-ZnSe:Te sample). The average size of QDs is also smaller. The sharp lines observed in the samples exhibit small diamagnetic shift due to strong exciton localization. The oscillations in the magneto-PL intensity are explained by the Aharonov–Bohm effect.

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