

Optical Properties of Heavily Doped n-type CdSe Quantum Dots for Intersubband Device Applications

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ABSTRACT

In this research, interband and intersubband optical properties of heavily doped n-type CdSe quantum dots were investigated by temperature dependent photoluminescence (PL) spectroscopy, picosecond time-resolved PL spectroscopy and Fourier transform infrared (FTIR) spectroscopy. Two doped and one undoped CdSe quantum dot samples with multiple QD layers were grown over ZnCdMgSe barrier layers on InP (001) substrate by molecular beam epitaxy. Heavy doping leads to decreasing of activation energy of nonradiative recombination centers, however, does not affect the luminescence efficiency of doped quantum dots. Time resolved PL experiments show that the PL decay times of the doped samples have weak dependence on dot size and are much longer than that of the undoped sample. The two doped CdSe QD samples show strong Intersubband IR absorption that peaked at 2.54 μm , 2.69 μm and 3.51 μm .

INTRODUCTION

Semiconductor quantum dots (QDs) have been extensively investigated for optical and optoelectronic devices due to their unique advantages over bulk and quantum well materials. One of the important applications is quantum dot infrared photodetectors (QDIPs) where intersubband (ISB) transition is applied. The expected advantages of QDIPs are their inherent sensitivity to normal-incidence radiation, high operating temperature, and low dark current. QDIP becomes a promising candidate as an alternative technology aiming to replace quantum well and HgCdTe infrared detectors. In(Ga)As/GaAs quantum dots grown on GaAs substrate [1], being probably the most explored quantum dot material system, are typically used in QDIPs. In this study, a different material system, CdSe QDs on ZnCdMgSe barriers, is introduced and experimentally tested for intersubband infrared device applications. Both interband and intersubband optical properties of heavily doped n-type CdSe QDs have been investigated. Intersubband absorption of these CdSe quantum dots was successfully observed in mid-infrared range.

EXPERIMENTAL DETAILS

Three CdSe quantum dot samples, A, B and C, were grown by molecular beam epitaxy (MBE) on (001) semi-insulating InP substrates in a dual-chamber Riber 2300P MBE system. The CdSe QDs were self-assembled over ZnCdMgSe materials that are lattice-matched to InP substrates. The CdSe QD/ZnCdMgSe structure was repeatedly grown for 50 periods for each

sample. In all the three samples, the ZnCdMgSe barrier layer thickness is kept at 18 nm. The band gap of the ZnCdMgSe barrier is about 3.0 eV at 77 K as determined from separate calibration layers. Samples A and B have identical structures except that sample A is undoped while in sample B the CdSe QDs layers were doped with Cl (using ZnCl_2 as dopant). The nominal CdSe thickness deposited for samples A and B is 1.56 nm, correspond to 5.2 monolayers (MLs). For sample C, the CdSe QDs layers with a nominal deposited thickness of 6.9 MLs were also doped with Cl. The actual dot sizes are unknown. Inferred from the atomic force microscopy measurement results on uncapped QDs, the height of the dots is in the range between 2.5 and 5.5 nm while the lateral size is in between 30 and 60 nm. If we assume that the Cl doping efficiency in CdSe is the same as that in ZnCdSe, the 2-dimensional carrier density in samples B and C is about $3 \times 10^{11} \text{ cm}^{-2}$ and $4 \times 10^{11} \text{ cm}^{-2}$, respectively. Each sample is capped with 25-nm ZnCdSe layers.

The samples were mounted into a low temperature cryostat. The sample temperature varied from 77 K to 295 K. CW photoluminescence was excited at different temperatures by a 325 nm He–Cd laser and measured using a 0.3 m SPEX 1680-B spectrometer connected to a photomultiplier. For time-resolved photoluminescence experiments, the second harmonic radiation at 400 nm obtained from a mode-locked tunable Ti-Sapphire laser (Spectra Physics Tsunami) was used as excitation source. The laser pulses have a pulse width of 100-fs and a repetition rate of 82 MHz. The power of the 400 nm second harmonic radiation was measured to be 3 mW with 5% fluctuation. The time evolution of the luminescence was recorded in 2000 picoseconds by a streak camera (Hamamatsu Model C5680) with a typical temporal resolution of 10 ps. Intersubband absorption measurements were performed using a Nicolet Nexus-870 Fourier transform infrared (FTIR) spectrometer equipped with a liquid-nitrogen (LN_2) cooled HgCdTe detector. Samples were fabricated to the waveguide geometry with parallel 45° facets.

RESULTS AND DISCUSSION

Interband optical properties

Figure 1 shows the PL spectra of samples A, B and C measured at 77 K under the 325-nm laser excitation with the same power. The PL peak wavelengths of samples A, B and C are 591

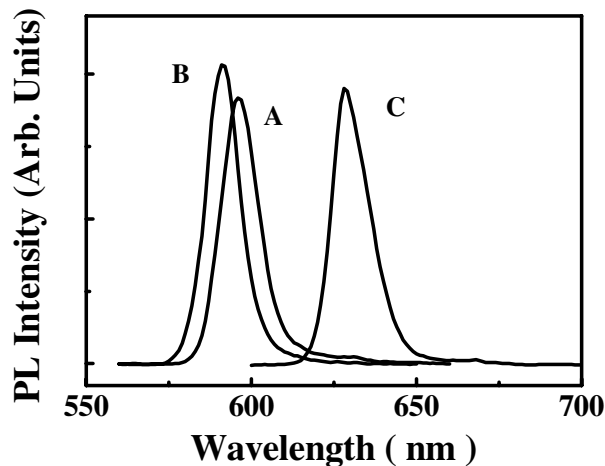


Figure 1 Photoluminescence spectra of doped samples B and C, and undoped sample A measured at 77 K

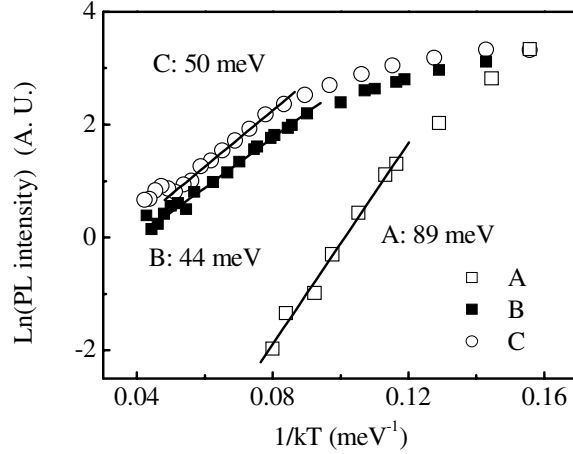


Figure 2 Integrated photoluminescence intensities as functions of samples A, B and C.

The corresponding activation energy of nonradiative centers is indicated for each sample.

nm, 595 nm and 628 nm, corresponding to emission energies of 2.098 eV, 2.084 eV and 1.974 eV, respectively. The small difference of the emission energy between sample A and B indicates that the dot size is almost the same for these two samples, which have the same deposition thickness of CdSe material. This means that heavy Cl doping has affect little to dot size of CdSe QDs. The little difference on PL intensity of samples A and B indicates that Cl doping will not decrease luminescence efficiency of the CdSe quantum dots. The PL emission energy of sample C is lower than that of the other samples due to its less quantum confinement for larger dot size.

To study nonradiative recombinations of the samples, PL spectra were recorded at different temperatures. Integrated intensities I_{int} of the corresponding PL peaks are plotted as functions of the inverse of temperature and are displayed as hollow squares for sample A, solid squares for sample B and hollow circles for sample C in Fig. 2, respectively. At higher temperatures, the linear fitting (solid lines in Fig. 2) of $\ln(I_{\text{int}}) \sim 1/kT$ gives activation energy of nonradiative recombination centers for each sample, which are 89 meV for sample A, 44 meV for sample B and 50 meV for sample C, respectively. Despite the different QD sizes of the doped samples B and C, they have almost the same activation energy. The undoped sample A has much higher activation energy than the doped samples. Strong carrier screening in heavily doped samples may reduce the barrier height of those nonradiative recombination centers and result in the decreasing of activation energy.

Time-resolved PL spectra of the samples were recorded at 77 K and are shown in Fig. 3. The

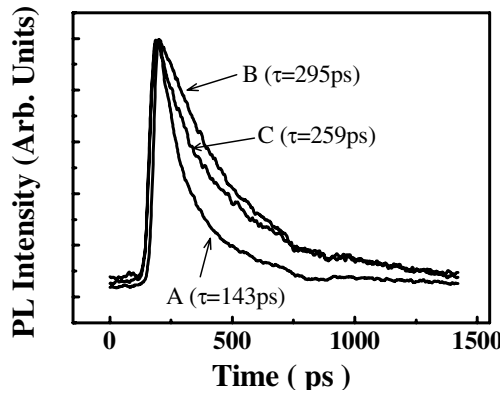


Figure 3 Time-resolved PL spectra of samples A, B and C recorded at 77 K.

t-PL traces show exponential decay on the first order. The decay times are determined to be A: 143 ps, B: 295 ps and C: 259 ps. The decay time for the undoped sample A is much shorter than that for the doped ones. This indicates more involvement of nonradiative recombinations in the undoped CdSe QDs and higher density of recombination centers as well [2]. However, before concluding that heavy doping in CdSe QDs may reduce the density of recombination centers, many growth factors, like fluctuation of growth conditions, should be excluded. The longer decay time of sample B comparing to sample C may be partially due to its relative larger electron density.

Intersubband optical properties

FTIR absorption spectra of the samples were measured at room temperature. The spectra were obtained by taking the ratio of *p*-polarized spectra over *s*-polarized spectra and are shown in Fig. 4. Absorption peaks are observed for the two doped samples B and C. For undoped sample A, no absorption feature was observed. The absorption of sample B is peaked at 0.49 eV (2.54 μm). Lorentzian line fitting of the absorbance spectrum yields a full width at half maximum (FWHM) of 46 meV. The narrow line width ($\Delta\lambda / \lambda_p \sim 9.4\%$) is an indication of relatively uniform distribution of dot size. For sample C, two absorption peaks were observed at 0.46 eV (2.69 μm) with a FWHM of 29 meV ($\Delta\lambda / \lambda_p \sim 6.3\%$) and at 0.35 eV (3.51 μm) with a FWHM of 20 meV ($\Delta\lambda / \lambda_p \sim 5.7\%$). We believe that the absorption peaks at 0.46 eV from sample B origins from electron transition from the ground state to the first excited state of the doped CdSe QDs. The origins of the absorption peaks at 0.49 eV and 0.35 eV from sample C are related to intersubband transitions between the confinement energy levels in the CdSe QDs. However, specific levels that are responsible for these transitions are not clear. Further experiments will be done to clarify this.

We have previously experimentally observed intersubband absorption of lattice-matched ZnCdSe/ZnCdMgSe multiple quantum wells [3]. Combining with the latest experimental results of CdSe quantum dots reported here, we conclude that II-Se material system can be a good choice as intersubband devices for infrared applications.

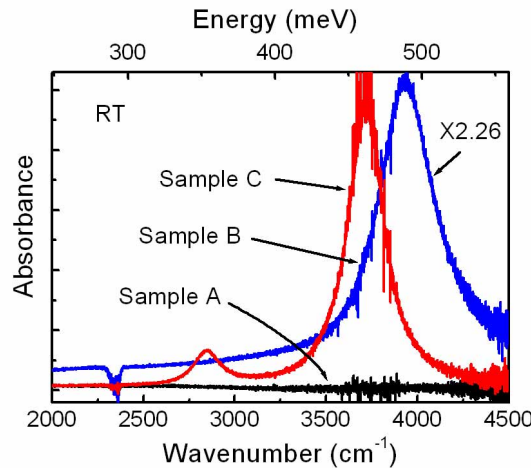


Figure 4 FTIR absorption spectra of samples A, B and C measured at room temperature

CONCLUSION

Interband and intersubband optical properties of heavily doped CdSe quantum dots have been experimentally studied. Heavy Cl doping in these QDs is found to not impair their interband optical properties. Absorption peaks induced by Intersubband transition have been successfully observed in mid-infrared wavelengths for the doped CdSe QDs, showing a bright future of doped CdSe QDs for intersubband device applications.

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