Widely Tunable and Intense Mid-Infrared PL Emission from Epitaxial Pb(Sr)Te Quantum Dots in a CdTe Matrix

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We present the temperature dependence of the mid-infrared PL of epitaxially precipitated PbTe/CdTe QDs for different dot sizes. A strong shift of the emission to longer wavelengths with decreasing temperature is found and explained by a theoretical model of the ground state transition energy. Secondly, we demonstrate that $Pb_{0.93}Sr_{0.07}Te$ ternary QDs also give strong PL emission at room temperature, thus extending the size tunability of such epitaxially precipitated dots. In addition, we showed that size control is also obtained for a fixed layer thickness by variation of the growth temperature.

Introduction

Epitaxial semiconductor quantum dots (QDs) are of great importance for optoelectronic devices. Their conventional synthesis uses the Stranski-Krastanow growth mode for strained layer heteroepitaxy with efficient interband emission mostly found in the visible and near-infrared spectral regions. Recently, we have demonstrated a completely different approach for the synthesis of epitaxial QDs for the mid-infrared (MIR). It is based on phase separation between two lattice-type mismatched, immiscible materials, namely PbTe (rock salt lattice) and CdTe (zinc blende lattice). Upon thermal annealing, 2D PbTe layers grown by molecular beam epitaxy within CdTe barrier layers are transformed into PbTe QD nano-precipitates with highly symmetric almost spherical shapes and atomically sharp hetero-interfaces. These defect-free QDs show continuous-wave photo-luminescence (cw-PL) with high efficiency even at room temperature [1], [2]. Moreover, due to the very small strain in the dots (lattice mismatch only 0.29% at 300 K) and the narrow band gap of bulk-like PbTe of 320 meV (3.85 µm), the optical emission is in the MIR. We are also able to control the dot sizes and the vertical positions of the dots to a large extent [3]. The lead salts have widely been applied for fabrication of MIR lasers and detectors for gas spectroscopy due to their favorite band structure and low Auger recombination rates.

Sample Structure and Experimental Details

The samples were grown by MBE using solid source CdTe, PbTe, Cd, and Te effusion cells on (100)-oriented GaAs substrates. On a CdTe buffer layer, PbTe and PbSnTe layers with different thicknesses were deposited using a low growth temperature of 220 °C to ensure a two-dimensional growth without island formation. A 50 nm CdTe cap layer was grown on top to form a PbTe quantum well.

By in-situ and post-growth thermal annealing in inert N_2 atmosphere, the PbTe quantum well layers are transformed into isolated quantum dot nano-precipitates [1], [2] due to the immiscibility of PbTe and CdTe. The annealing conditions (duration and temperature) were optimized for each sample to achieve the highest cw-PL intensity at room temperature. As shown by TEM [1] – [3], the annealed dots have a centrosymmetric shape of a small-rhombo-cubo-octahedron which is the dot shape in thermal equilibrium (minimization of interface energies). From the TEM investigations, we also know that the dots are essentially defect free and exhibit atomically sharp hetero-interfaces.

The PL measurements were done with cw diode lasers with wavelengths between 980 nm and 1480 nm and output powers of up to 245 mW. Therefore, only the PbTe dots and not the CdTe barriers are excited. The pump laser was focused on the sample placed in a He flow cryostat under an angle of 45°, and the PL emission was recorded by lock-in technique with a liquid-nitrogen-cooled InSb infrared detector mounted on a grating spectrometer.

Results

In this work, at first, we present the temperature dependence of the cw PL of PbTe/CdTe QDs for different dot sizes. A strong shift of the emission to longer wavelengths with decreasing temperature is found for both dot sizes (see Fig. 1).

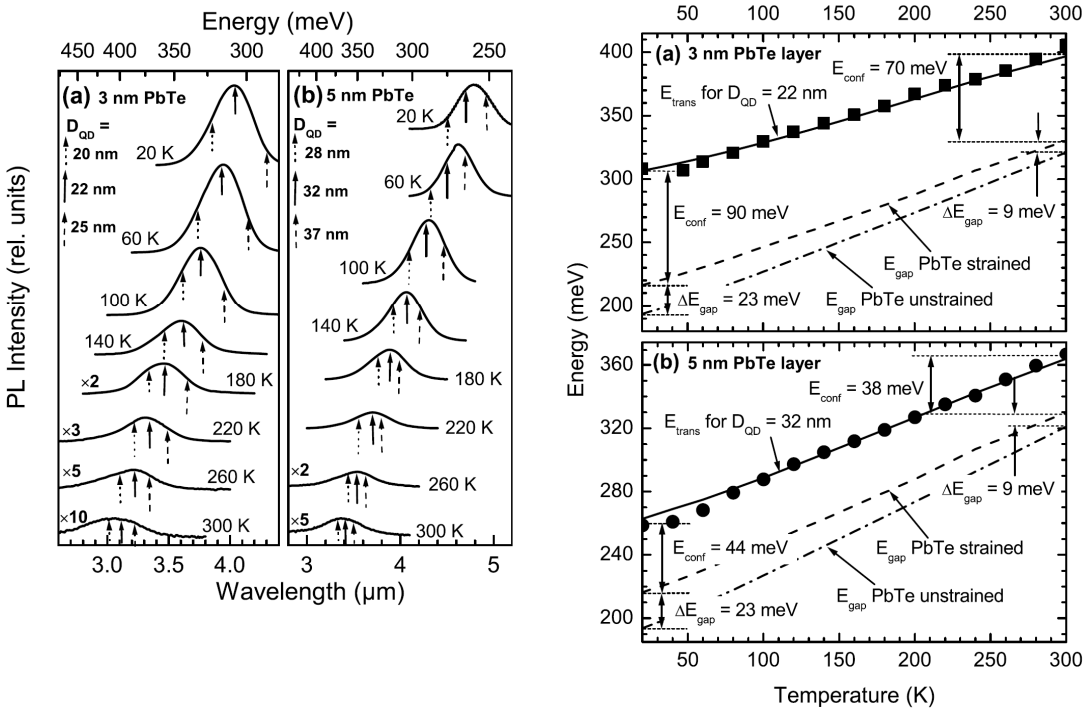


Fig. 1: (left:) Temperature dependent cw-PL spectra for PbTe/CdTe quantum dots formed by an initially (a) 3 nm and (b) 5 nm thick PbTe layer. The arrows depict the calculated ground state transition energy for different dot diameters D_{QD} as indicated.
(right:) Peak energy of the PL emission spectra as a function of temperature (symbols) for (a) 3 nm and (b) 5 nm PbTe. The full, dashed and dash-dotted lines depict the calculation of the ground state transition energy for (a) 22 nm and (b) 32 nm dot diameter, the band gap of isotropically strained PbTe, and the band gap of bulk-like PbTe, respectively.

This shift is not just explained by the strong temperature dependence of the band gap of PbTe, but also by the strain in the dot as well as by the high quantum confinement (ΔE_g(PbTe-CdTe) = 1.2 eV), both being temperature dependent via the lattice mismatch and the effective masses of PbTe. We also observed an increase of the emission intensity with rising temperature for temperatures below 55 K or 100 K, depending on dot size (Fig. 2 (left)). This is attributed to the presence of a dark ground state lying below the bright state as was also observed for other quantum dots. The influence of the excitation power on the emission spectra at various temperatures (Fig. 2 (right)) indicates a carrier redistribution between the dots, which could also explain the observation of Fig. 2 (left).

Furthermore, a calculation of the transition energy responsible for the PL emission is presented. We make use of the fact that the ground state transition in a spherical dot with finite barriers can exactly be calculated using the results for the first excited state in a quantum well with the same size. We also include the strain in the dots as well as in the matrix and the anisotropic effective masses of PbTe, both being temperature dependent, and by that we obtain a good agreement to the experimental data as seen in Fig. 1.

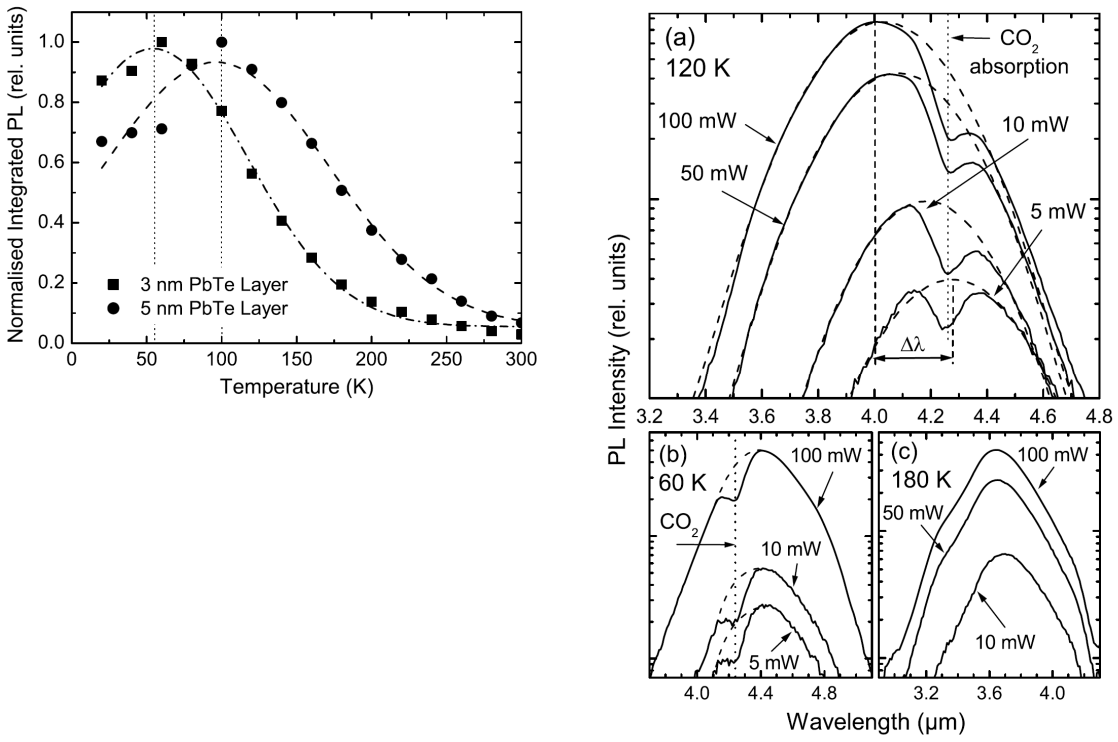
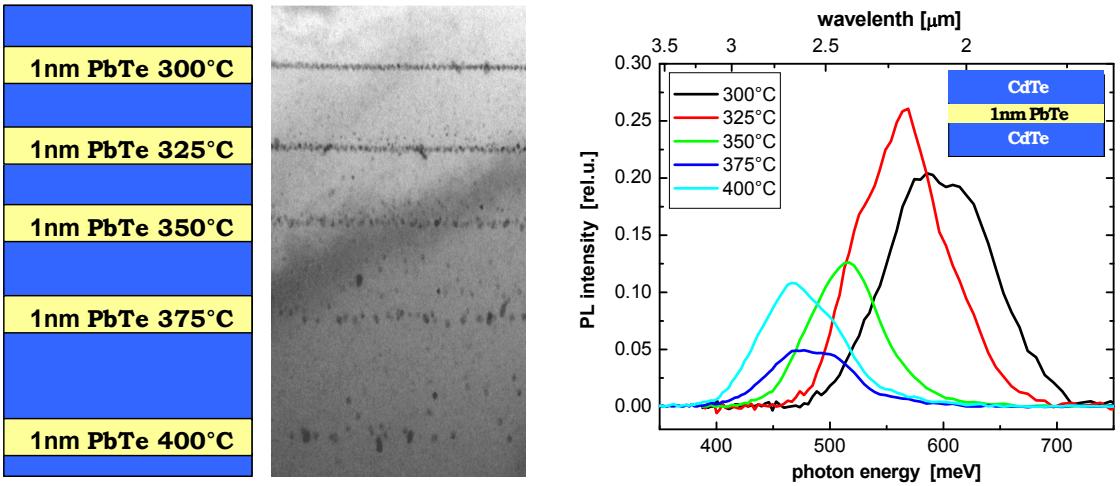


Fig. 2: (left:) Normalized integrated PL intensity vs. temperature for PbTe/CdTe dots with a 3 nm (squares) and a 5 nm thick PbTe layer (circles). The lines are guides to the eye.
(right:) Excitation power dependent PL spectra at different temperatures of (a) 120 K, (b) 60 K and (c) 180 K on a logarithmic scale. The dips at 4.25 μm indicated by the dotted line arise from residual CO_2 absorption in the optical measurement path. The dashed lines are Gaussian line fits to the data.

Secondly, we demonstrate for the first time $Pb_{0.93}Sr_{0.07}Te$ ternary QDs also giving strong PL emission at room temperature. Due to the increasing band gap with increasing Sr content, this leads to a larger tunability of the emission towards shorter wavelengths. Remarkably, for originally 1 nm thin PbSrTe layers, the PL signal is much

stronger as compared to a 1 nm PbTe reference dot sample, despite the shorter emission wavelength for the PbSrTe sample. We also observed for thin PbSrTe layers that the blue shift with regard to the bulk band gap is much lower as expected indicating that part of the Sr is incorporated in the CdTe matrix material.

In addition, we show that it is possible to fabricate high quality PbTe dots by simply growing 50 – 300 nm thick ternary $Cd_{1-x}Pb_xTe$ layers with Pb-contents of 1 % to 6 %. The strong PL emission at room temperature is peaked at about 2 – 2.3 µm indicating a rather small average dot size of 9 – 11 nm in diameter depending on the actual Pb content. Recently, we showed that the size of the PbTe dots can effectively be tuned by the original 2D PbTe layer thickness [3]. In addition to that, we demonstrate here that size control is also obtained for a fixed layer thickness by variation of the growth temperature (see Fig. 3). For a nominally 1 nm PbTe layer, dots with average diameter of 10 nm for 300 °C and of 15 nm for 400 °C growth temperature are found.



(a) (b)

Fig. 3: (a) TEM image of a sample with 1 nm PbTe layers in CdTe grown at different temperatures with a sketch of the sample structure. (b) Room temperature cw PL emission spectra of five individually grown 1 nm PbTe layers grown at different substrate temperatures demonstrating a longer wavelength emission for samples grown at higher temperatures. This indicates a larger dot size for higher growth temperatures as also seen in the TEM image.

Conclusion

We presented the temperature dependence of the cw PL of epitaxially precipitated PbTe/CdTe QDs for different dot sizes. A strong shift of the emission to longer wavelengths with decreasing temperature is found and explained by a theoretical model of the ground state transition energy. Secondly, we demonstrated that $Pb_{0.93}Sr_{0.07}Te$ ternary QDs also give strong PL emission at room temperature, thus extending the size tunability of such epitaxially precipitated dots. In addition, we showed that size control is also obtained for a fixed layer thickness by variation of the growth temperature.

Acknowledgements

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