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Resonant Gain in ZnSe Structures with Stacked CdSe Islands Grown in Stranski-Krastanov Mode

M. STRASSBURG¹ (a), M. DWORZAK (a), A. HOFFMANN (a), R. HEITZ (a),
U.W. POHL (a), D. BIMBERG (a), D. LITVINOV (b), A. ROSENAUER (b),
D. GERTHSEN (b), I. KUDRYASHOV² (c), K. LISCHKA (c), and D. SCHIKORA (c)

(a) *Institut für Festkörperphysik, Technische Universität Berlin, D-10623 Berlin, Germany*

(b) *Laboratorium für Elektronenmikroskopie der Universität Karlsruhe,
D-76128 Karlsruhe, Germany*

(c) *Universität Paderborn, Fachbereich 6 Physik, Warburger Str. 100, D-33098 Paderborn,
Germany*

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The optical gain of CdSe/ZnSe structures containing stacks of islands grown in the Stranski-Krastanov mode was studied. The islands have a lateral size of ≈ 16 nm and a small height-to-width ratio. Cd contents of $\approx 70\%$ were estimated from transmission electron microscopy investigations. The radiative recombination of excitons localised in the islands leads to a broad emission band around 2.4 eV. At low temperatures a resonant gain of up to 300 cm^{-1} generated by localised transitions was found.

Introduction Wide bandgap II–VI quantum dot structures based on self-organised island growth were achieved by several growth modes [1] resulting either in the formation of small 2D-like islands [2] or in islands formed in a three-dimensional (3D) growth process [3 to 5]. Zero-dimensional excitons localised in such islands enable resonant gain. Therefore, efficient excitonic waveguiding [2] can be applied to reduce the threshold density of lasers devices. For structures containing small islands, labelled type A, zero-phonon gain (see, e.g., [6, 7]) and optical pumped lasing at room temperature [8 to 10] were achieved. Nevertheless, the limited thermal stability of carrier localisation in type A islands prevents room temperature (RT) operation. Recent investigations on CdSe/ZnSe structures [11] have demonstrated that at room temperature excitons are evaporated from type A islands whereas thermal stability was found for larger islands, grown in the Stranski-Krastanov (SK) mode (labelled type B). In order to take advantage of the better thermal stability in type B islands, we studied the resonant gain in structures containing such islands.

Experimental The investigated structures were grown by molecular beam epitaxy (MBE) and consist of a single layer or a 10-fold stack of nominally 2.8 ML thick CdSe coverage deposited at 340 °C. The CdSe layers in the stacks were separated by

¹) Corresponding author: Tel.: +49-30-31422054; Fax: +49-30-31422064;
e-mail: marburg@physik.tu-berlin.de

²) On leave from: Ioffe-Institute, Polytekhnicheskaya 26, St. Petersburg, 194021, Russia.

12 nm ZnSe spacers. All structures were cladded by 45 nm thick ZnSe buffer and cap layers. To study the conditions for the formation of type A islands which have a Cd content below 40% and those of type B islands which are described in detail in previous publications [5, 11], samples were grown both on exactly (001)-oriented GaAs (exactly oriented) substrates and on substrates which were tilted by an angle of 6° towards the $\langle 100 \rangle$ direction (tilted). The growth on tilted surfaces is believed to enable the step-flow growth mode, and thus to suppress the formation of type A islands. The formation of 3D islands was confirmed by in-situ monitoring of the RHEED pattern. More details of the growth are given in Ref. [12].

The samples were studied by transmission electron microscopy (TEM) in plan-view and cross-section geometry using a Philips CM 200 FEG/ST electron microscope with an electron energy of 200 keV and a Scherzer resolution of 0.24 nm. For the optical investigations the samples were mounted in a He-flow cryostat providing temperatures between 4 and 300 K. The photoluminescence (PL) was excited by the 325 nm line of a cw He–Cd laser. Gain studies were performed in edge geometry using the variable-stripe-length method [13]. These measurements used a pulsed dye laser with ≈ 20 ns pulse duration pumped by an excimer laser at an excitation energy of 2.85 eV. Luminescence was detected by a photomultiplier attached to a 0.85 m double monochromator. Gain spectra were recorded at 8 K.

Results and Discussion A typical cross-section TEM image of the sample with stacked SK islands (type B) grown on tilted substrate is depicted in Fig. 1. A relatively large number of type B islands is revealed by regions with a bright contrast inside the islands. This contrast indicates a high Cd concentration which was estimated to be $\approx 70\%$ [12]. Type B islands having an average lateral size of 16 nm are laterally surrounded by an ≈ 3 nm thick wetting layer (WL) with a significantly lower Cd concentration. The height of the islands exceeds that of the WL only slightly. In some places, the island arrangement is laterally shifted in adjacent layers. This is observed particularly in samples grown on tilted substrates. We attribute this island arrangement to a preferential nucleation of islands at growth surface steps induced by stacking faults (SFs), the density

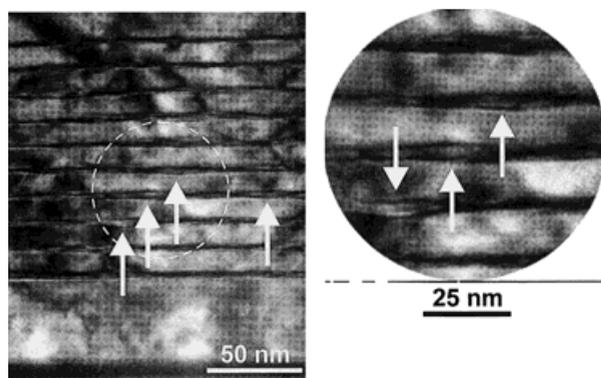


Fig. 1. TEM cross-section dark-field image of a sample with a 10-fold stack of CdSe islands, grown on a tilted substrate, taken with $\mathbf{g} = (002)$. The type B islands are indicated by the bright contrast inside the ZnCdSe layer and some of them are marked by arrows. The inset shows some islands on a larger scale

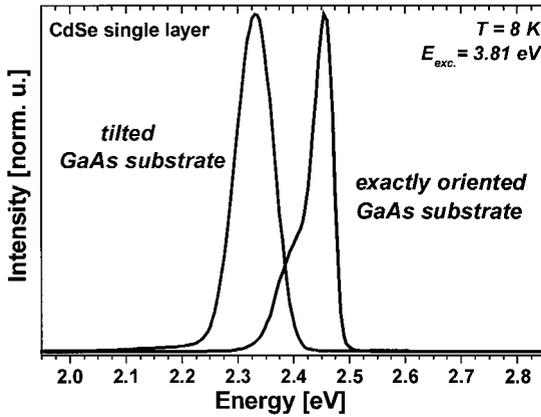
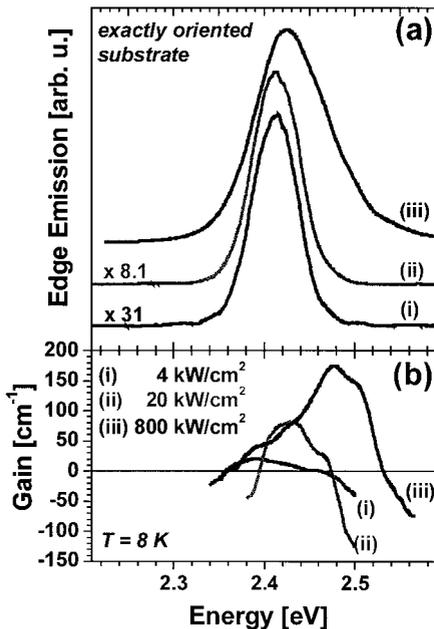


Fig. 2. Comparison of the photoluminescence of CdSe Stranski-Krastanov-island singlelayer embedded in a ZnSe matrix, grown on tilted and exactly oriented substrate, respectively

and the type B islands investigated here, can clearly be distinguished in plan-view TEM images. A more detailed investigation of the structural properties is given in Ref. [12].

The PL of CdSe SK-island single-layer structures, grown either on exactly oriented or tilted GaAs substrates is shown in Fig. 2. Two significant effects are observed: the emission band of the structure grown on the tilted substrate is significantly shifted to lower energies and a pronounced shoulder (a second emission band) is found on the low energy tail of the PL band of the sample grown on exactly oriented substrate. In previous investigations [11] we showed that for samples grown on exactly oriented substrates, the photoluminescence is governed by type A islands at low temperatures, and that the low energy shoulder can be attributed to the radiative recombination of excitons localised in type B islands becoming dominant at RT. In the emission band of the sample grown on tilted substrates no additional shoulder occurs, and a redshift of the PL maximum was detected. We attribute this observation to the suppression of type A islands achieved by a step flow growth on tilted substrates.

The excitation density dependence of the edge emission and the optical gain are shown in Figs. 3a and b, respectively, for a sample with 10 sheets of 2.8 ML CdSe coverage, grown on an exactly oriented substrate. The



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Fig. 3. Excitation density dependence of the edge emission and the gain of a sample with a 10-fold stack of islands on exactly oriented GaAs substrate. The edge emission spectra are vertically shifted for clarity

edge emission of this sample shows with increasing excitation density no significant change in shape and maximum position of the emission band. This behaviour is known from recombination of 3D confined excitons. Here, the excitons are localised in type A and B islands providing the 3D confinement which is necessary for the radiative recombination of excitons at high excitation densities. At very high excitation densities, the high energy tail of the edge emission becomes broader and a shift of the emission maximum to higher energies occurs. According to the applied excitation densities we attribute the observed behaviour to an increasing population of higher states (smaller type A islands and the WL). This explanation is supported by the results of gain investigations. Whereas at the threshold the gain maximum is observed on the low energy tail of the edge emission, the gain maximum shifts with increasing excitation density into the resonance of the edge emission. Further increase of the excitation density leads to a saturation of the resonant gain at $\approx 40 \text{ cm}^{-1}$ and a strong blueshift of the gain maximum. We note that this energy shift is far more pronounced than that observed for the edge emission. This can be attributed to the limited number of islands and the population of higher states.

The behaviour of the edge emission and the gain for the sample with stacked type B islands, grown on the tilted substrate, is similar to that of the sample grown on the exactly oriented substrate. In Fig. 4 the edge emission and the gain at an excitation density above the transparency density are depicted. Gain in resonance to the ground-state transition of the edge emission are detected. A further increase of the excitation density leads to a saturation of the resonant gain. In comparison to the sample on the exactly oriented substrate, we observed here higher gain values. This could be a hint to a higher island density or higher oscillator strength for the localised excitons in the sample grown on the tilted substrate. Additionally, a broadening of the high energy tail of the edge emission was detected with increasing excitation densities. Simultaneously, an additional feature appears in the gain spectrum on the high energy tail. This gain band exhibits a continuous rise of the maximum gain value up to 300 cm^{-1} at 470 kW/cm^2 . The inset shows the excitation density dependent energy of the edge emission and the gain maxima. At low excitation densities the gain rises on the low energy tail, in the range of localised biexcitons, which were also found to be the dominant gain mechanism in struc-

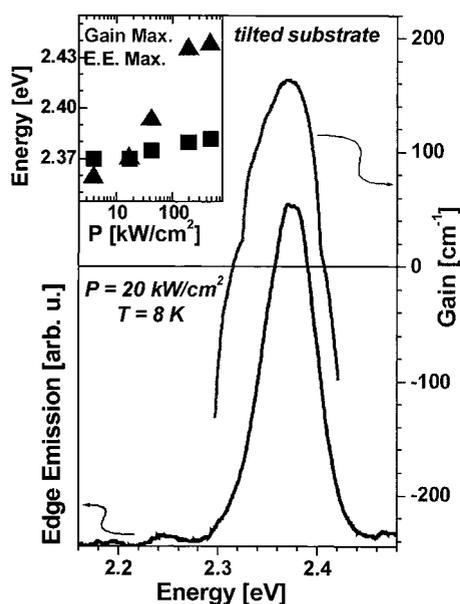


Fig. 4. Edge emission and gain of a sample with a 10-fold stack of islands grown in Stranski-Krastanov mode on a tilted substrate. Inset: Excitation density dependence of gain and stimulated emission; triangles and squares denote the maximum positions of gain and stimulated emission, respectively

tures containing type A islands only [15]. The pronounced blue-shift of the gain maximum at highest excitation densities is induced by the saturation of the resonant gain and the appearance of a second gain process on the high energy tail of the edge emission. This behaviour is assigned to state filling and population of higher excited states. For a clear identification of the observed gain mechanisms further investigations are necessary.

Conclusions Structural and optical properties of samples containing single- and multi-layer sheets of CdSe islands grown in the SK mode were investigated. A pronounced influence of the substrate tilt on the formation of small (type A) and larger (type B) islands was determined. Using tilted substrates a redshifted emission band and the suppression of type A islands were observed. For the first time, optical gain up to a value of 300 cm^{-1} was achieved in structures with a 10-fold stack of Stranski-Krastanov (type B) islands.

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