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Optical and Structural Properties of Quantum Dots in Wide-Bandgap Semiconductors

M. STRASSBURG, A. HOFFMANN, I. L. KRESTNIKOV[1]), and N. N. LEDENTSOV[1])

Institut für Festkörperphysik, Technische Universität Berlin, Germany
Tel.: +49-30-31422054, Fax: +49-30-31422064, e-mail: marburg@physik.tu-berlin.de

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The optical and structural properties of ultrathin insertions in wide-bandgap semiconductors are studied. Structural investigations confirm that in nitride-based structures indium fluctuations lead to the formation of nano-islands. The zero-dimensional character in nitride- and II–VI-based quantum dot structures is demonstrated by its typical behaviour (splitting of the polarisation, optical gain mechanisms resulting from localised states, surface lasing without high-quality Bragg reflectors, etc.). Practical device applications of the structures based on ultrathin insertions for non-traditional devices are discussed.

Introduction For the fabrication wide-bandgap quantum dots (QDs) several selforganized growth approaches may be applied: islands can be formed by heteroepitaxial submonolayer deposition, by spinodal decomposition of a multi-component alloy, via Stranski-Krastanow (SK) or Volmer-Weber (VW) island growth mode, by growth on faceted or step-bunched surfaces, etc. [1–5]. Especially in ternary compounds based on InGaN and CdZnSe interdiffusion and segregation influence strongly the formation of nano-islands. Consequently, similar approaches are applied for the growth of QD structures. Appropriate growth conditions, e.g. ultrathin (submonolayer and a few monolayer) insertions, lead to dense arrays of flat two-dimensional nano-islands with a lateral size comparable or much smaller than the exciton Bohr radii. In spite of the relatively small size of the islands, the QD characteristics are proven by direct observation of luminescence lines from single QDs up to elevated observation temperatures, by excitonic gain and by lateral squeezing of excitons revealed in magneto-optical studies [6]. The aim of the present paper is to show that group III nitrides became attractive candidates for the application of ultrathin insertions. Such insertions have already applied to the group III nitrides [7] and enabled gain in the green spectral range in InGaN/GaN structures [8].

Experimental For the investigation of II–VI quantum dot structures samples were grown by molecular beam epitaxy (MBE). Further details of the growth are given in Ref. [9]. The nitride-based structures were grown in a horizontal flow MOVPE growth machine. To form nucleation layer, low-temperature AlGaN deposition was applied. GaN and AlGaN layers were grown at 1050 °C with H_2 carrier gas and at the total pressure of 200 mbar. For the growth of the InGaN-based active region the temperature was reduced. The total pressure was 600 mbar and argon was used as carrier gas [10].

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[1]) Also at A. F. Ioffe Institute, St. Petersburg, Russia

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. Optical reflection (OR) spectra were recorded at normal incidence using a tungsten lamp dispersed by a double monochromator. For the high excitation density measurements pulsed excimer and nitrogen lasers were used. Luminescence was detected by a photomultiplier attached to a 0.85 m double monochromator.

Results and Discussion For the CdSe/ZnSe system the formation of nano-islands by the deposition of ultrathin insertions is conformed by MOVPE and MBE grown samples. Their zero-dimensional character is proven by structural and optical investigations [6]. E.g., the polarisation of the PL in edge geometry enables a clear distinction between the quantum wells (QW) and QD cases. According to Kane's selection rule, the heavy-hole exciton luminescence in zinc-blende QWs grown on a high-symmetry (100) surface must be completely TE polarised as it was experimentally confirmed in numerous studies. In contrast, for spherical QDs no polarisation of the QD emission in edge geometry can be expected. As the CdSe nano-islands keep essentially a two-dimensional shape, the quantization in growth direction prevails, and the heavy-hole-like QD exciton luminescence in edge geometry is still TE-polarized. However, in contrast to the QW case, a significant contribution of the TM-polarised emission has been observed (Fig. 1). This underlines the role of the lateral exciton confinement. The most remarkable observation has been done however for the edge emission of vertically coupled QDs. This emission was found to be predominantly TM polarized. This indicates that the heavy-hole-like exciton wavefunction was more extended in the growth direction and, most probably, had a cylindrical shape. A similar polarisation of edge emission has been observed in the case of vertically coupled InGaAs–GaAs Stranski-Krastanow QDs [11].

While a lot of publications are available about II–VI wide-bandgap semiconductor quantum dots, in the case of the InGaN/GaN system the formation of quantum dots by ultrathin insertions is not so widely discussed. Fig. 2 shows the high-resolution (HR) TEM image of a InGaN/GaN multilayer structure, and the digital analyses of lattice images (DALI [13]). With respect to the investigations of similar II–VI QD structures, evaluation procedures have to be applied to reveal the nano-islands formed by ultrathin insertions in

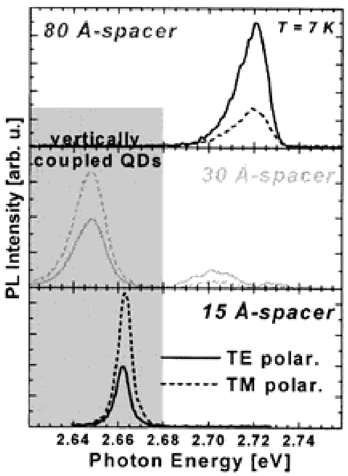

Fig. 1. Polarisation-dependent PL spectra of CdSe/ZnSe multilayer structures as a function of the thickness of the ZnSe spacer between adjacent CdSe layers. The polarisation splitting indicates the zero-dimensional character of the emission centres

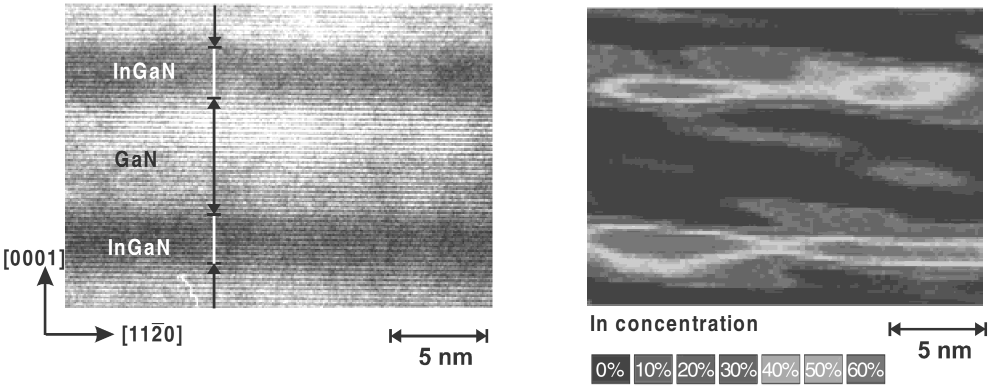

Fig. 2. High-resolution transmission electron microscopy image (left) and DALI evaluation (right) to reveal the indium distribution in an InGaN/GaN multi-quantum-well structure. The indium concentration is scaled from 0% to 60% (stepwidth: 10%) [12]

the InGaN/GaN system, too. These investigations support that the indium distribution is not homogeneous in the deposited layer and undergoes remarkable lateral variation. Areas with an indium content of up to 60% and a lateral size of 3–10 nm are revealed. A dense array (10^{18} cm^{-3}) of such nano-islands implicates a large potential for vertical cavity surface emitting laser (VCSEL) due to their enormous optical gain up to several 10^5 cm^{-1}.

Optical transmission, PL and stimulated emission recorded perpendicular to the surface at 16 K are shown in Fig. 3. The absorption in the active region (InGaN/GaN superlattice) leads to a decrease of the transmitted light at ~3.0 eV, which is suppressed for light energies above the GaN bandgap. In comparison to the transmission spectrum the maximum of the PL band is shifted to lower energies. The shape of the emission band with extended tails on the low and the high energy side and the pronounced red-shift suggest the presence of indium-rich areas with a significant size and composition dispersion. Both were revealed in HRTEM investigations and DALI evaluation (compare Fig. 2). At higher excitation densities, the maximum of the PL band shifts to higher energies and a narrowing was observed. As it is shown in the inset of Fig. 3, the

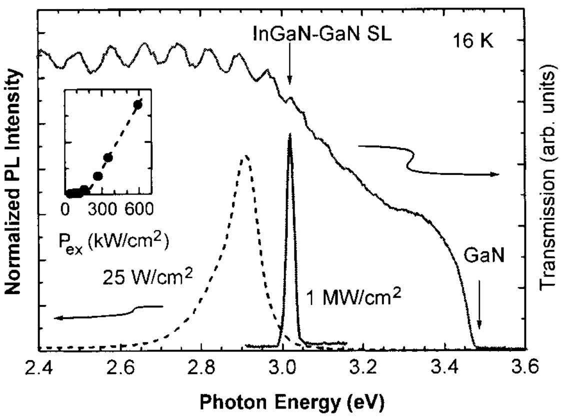

Fig. 3. Transmission and photoluminescence (PL) spectra of an InGaN/GaN multilayer structure. The absorption edges of GaN and the InGaN/GaN superlattice are marked by arrows. In the inset the integrated PL intensity as a function of the excitation density is depicted [10]

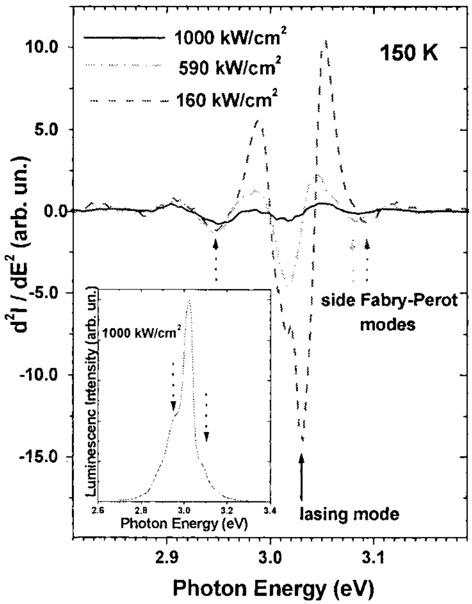

Fig. 4. Reflection spectra of an InGaN/GaN multilayer structure recorded at various excitation densities. The inset shows the photoluminescence spectrum at highest excitation density. Further cavity modes are marked by arrows

intensity of the emission band grows superlinearly. This behaviour hints to the observation of simulated emission in surface geometry, even when no Bragg reflectors are applied. We note, that the maximum of the PL band at high excitation densities remains in the vicinity of the onset of the absorption induced by the InGaN nano-islands.

The pronounced modulation of the light even in InGaN/GaN structures without Bragg reflectors is supported by optical reflection investigation at excitation densities when the gain overcomes the losses (Fig. 4). Deduced from the reflectivity coefficients of the GaN/AlGaN (2.4%) and the GaN/air (17%) interfaces and the thickness of the active region, the threshold gain, which is necessary to overcome the external losses was estimated to be 2×10^5 cm^{-1}. The behaviour of the second derivative normalised to the excitation density confirms that only one cavity mode exhibits superlinear growth. Side Fabry-Perot modes were detected even at highest excitation densities. The influence of the cavity modes on the PL emission are shown in the inset of Fig. 4. Although a superlinear growth of the main mode at 3.05 eV was observed, additionally emission bands appear fitting to other cavity modes according to the side Fabry-Perot modes. The observed chirp fits to the giant absorption–gain changes at threshold ($\sim 10^5$ cm^{-1}). Despite the remarkably low finesse of the cavity, superlinear growth and narrowing of the emission band indicate the appearance of surface lasing supported by a cavity formed by the GaN/AlGaN and GaN/air interfaces. For the fabrication of VCSEL with low threshold densities, the external optical losses have to be reduced. A promising way to achieve this is the enhancement of the reflectivity at the interfaces. Therefore, distributed Bragg reflectors (DBR) consisting of strain-compensated GaN/AlGaN multilayer sheets are applied.

The introduction of a bottom AlGaN/GaN Bragg reflector, with maximum reflectivity exceeding 90%, leads to a significant reduction of the external optical losses. Consequently, room temperature surface lasing [14] was achieved in structures with stacked InGaN QD insertions. For the investigated structure, which is schematically drawn in the left inset of Fig. 5, no top DBR was applied. Nevertheless, as already mentioned, the high material gain in stacked InGaN insertions enabled surface lasing even in case of very low finesse cavities (17% reflectivity on top due to the GaN/air interface, only). In Fig. 5 the room temperature PL spectra of the VCSEL structure recorded at different excitation densities are depicted. The observed surface lasing is confirmed by

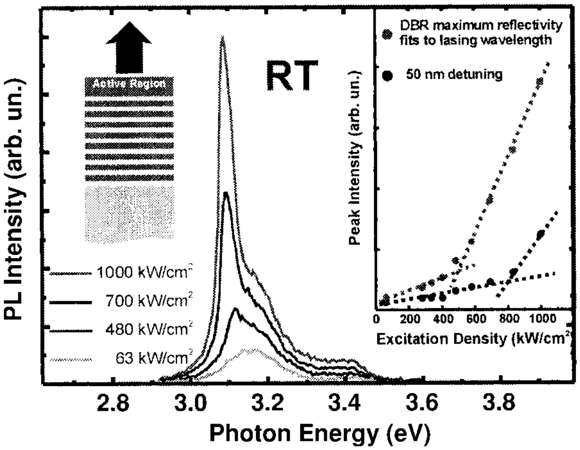

Fig. 5. Photoluminescence (PL) of an InGaN/GaN VCSEL structure recorded at various excitation densities at room temperature. The inset shows the peak intensity of the emission band as a function of the excitation density

the strong increase in the slope efficiency (see right inset of Fig. 5) and a narrowing of the PL emission band. From the farfield pattern of the emission of this structure a significant narrowing in comparison to the spontaneous emission of the same cavity was detected giving a further evidence of the lasing emission. The room-temperature threshold excitation density was 400 kW/cm^2. According to HRTEM investigations and the appearance of the lasing emission on the low energy tail of the PL band the lasing is attributed to recombination processes via localised states caused by In-rich nano-domains formed in the InGaN insertions (compare Fig. 2). The right inset of Fig. 5 shows peak intensity of the emission band as a function of the excitation density. The great impact of the DBR on the cavity quality is underlined by the observed behaviour. A detuning of the DBR out of the lasing wavelength leads to a decrease in the slope efficiency by a factor of two and to an enhancement of the threshold excitation density.

Conclusions The deposition of ultrathin insertions of ZnCdSe and InGaN leads to the formation of nano-islands. Structural investigations demonstrated that their diameter is in the range of the exciton Bohr-radii resulting in a three-dimensional confinement of excitons. It was shown, that the formation of cavity modes and surface lasing are enabled by the ultrahigh gain in QD structures. The reduction of the threshold excitation density and an increase of the slope efficiency was achieved by the introduction of bottom DBRs using GaN/AlGaN multilayer sheets and by a tuning of the emission wavelength to the cavity mode of the laser. The high potential of InGaN nano-islands for the realisation of VCSELs emitting in the blue and green were demonstrated.

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