Spatially resolved luminescence spectroscopy of single GaN/(Al,Ga)N quantum disks

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The dependence of the optical transition energy E_t of GaN quantum disks (Qdisks) embedded within (Al,Ga)N nanocolumns grown on a (111) Si substrate on disk thickness, disk diameter, barrier thickness, and barrier composition has been investigated by both spatially resolved cathodoluminescence spectroscopy and applying a theoretical model. Results of E_t on the disk and barrier thickness, as well as barrier composition resemble those of corresponding quantum wells, whereas results on the disk diameter are essentially determined by the lateral strain distribution of the Qdisks.

1 Introduction The growth of nanocolumnar group III-nitride semiconductor crystals is considered as a promising approach to reduce the defect density and to relax the strain of GaN-based optoelectronic devices. Several groups succeeded in the fabrication of quantum wells (QW) embedded within GaN and (Al,Ga)N nanocolumns, which actually represent quantum disks (Qdisks). The optoelectronic properties of such mesoscopic structures can be significantly influenced by the large surface-to-volume ratio, which results, e. g., in an inhomogeneous lateral distribution of the strain of the disks and/or in a large impact of surface-related nonradiative recombination [5, 6]. For an optimisation of the quantum efficiency, the dependence of the strain and carrier distribution of Qdisks on the design of the structure has to be investigated carefully.

Recently, we reported on theoretical and experimental results regarding the optical properties of GaN Qdisks embedded within (Al,Ga)N nanocolumns It was found that luminescence spectra are broadened due to, e. g., inhomogeneous strain, and that new quantized states formed at the periphery and centre of the disks are especially important for the lateral carrier distribution within the disks. Here, we show the dependence of the optical transition energy of GaN Qdisks of single (Al,Ga)N nanocolumns on the disk thickness, column diameter, and AlN mole fraction of the barriers investigated by spatially resolved cathodoluminescence (CL) spectroscopy and compare the results with corresponding theoretical investigations.

2 Experimental Four nanocolumnar samples each containing five GaN/(Al,Ga)N Qdisks were grown by plasma-assisted molecular-beam epitaxy on a (111) Si substrate The columns consist of a 140-nm-thick GaN base, 280 nm (Al,Ga)N, five Qdisks and a 40-nm-thick (Al,Ga)N cap layer. For three of the samples, the AlN mole fraction (x) of the (Al,Ga)N barriers has been estimated to be about 0.2. These samples differ in the thickness of the disks, which amounts to 2, 3, and 4.5 nm. The fourth sample contains also 4.5-nm-thick disks, but exhibits a smaller value of x, which has been estimated to be 0.12.

A high spatial resolution of the optical investigations of several 10 nm has been achieved by CL spectroscopy in conjunction with a field emission gun scanning electron microscope (FEGSEM) equipped with a Gatan MonoCL3 system as well as with a liquid-He-cooling stage. All the CL measurements have been performed at 6 K using an electron beam energy and current of 3 keV and about 1 nA, respectively. Transmission electron microscopy (TEM) measurements were carried out by using a Jeol JEM 3010 microscope operating at 300 keV.

3 Results and discussion Figures 1(a) and 1(b) show FEGSEM micrographs of the samples with 3 and 4.5-nm-thick Qdisks, respectively (x = 0.2). The diameter of the columns (D) varies between 30 and 100 nm. Only few of them exhibit larger values. Despite of this variation of D, the height of the columns does not fluctuate by more than 10 to 20% suggesting that the thickness of the Qdisks (d_w) of a sample does not differ significantly among the various columns.

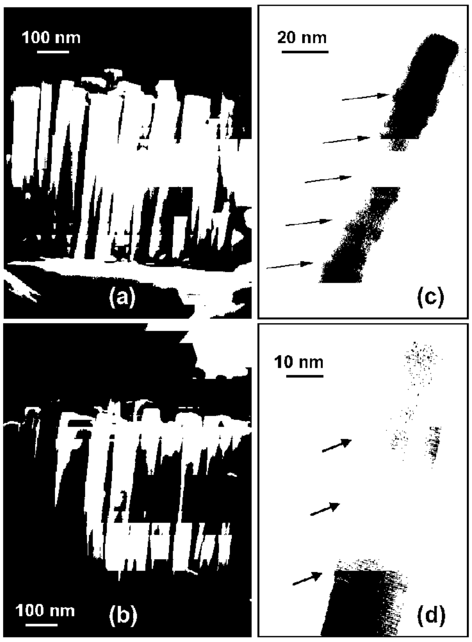

Figure 1(a) and (b) are FEGSEM images of GaN/(Al,Ga)N nanocolumns with x = 0.2 containing 3- and 4.5-nm-thick Qdisks. (c) and (d) represent TEM images of nanocolumns with x=0.12 and 4.5-nm-thick Qdisks.

The TEM images of Figs. 1(c) and 1(d) show that the heterostructure has been properly grown with a thickness of the five Qdisks (arrows) and of the barriers of 4.5 and 14 nm, respectively. Moreover, the protrusions marked by three arrows in Fig. 1(d) indicate a relaxation of the GaN disks at their periphery regions.

CL data of the samples are displayed in Fig. 2, where for Figs. 2(a), (b), and (c), the CL spectra differ in the values of d_w, D, and x, respectively. Each of the spectra consists of 2 broad bands. The low-energy (low-E) band of each of the CL spectra represents the Qdisk emission, whereas the broad or even split high-E band originates from the (Al,Ga)N column. The large spectral spread of the (Al,Ga)N spectra reflects an inhomogeneous AlN incorporation during growth, where a low AlN mole fraction is found close to the position of the Qdisks Therefore, it is suggested that x of the Qdisk barriers amounts to about 0.12 and 0.2 for the samples with low and high AlN mole fraction corresponding to the dashed and solid arrow in Fig. 2(c), respectively.

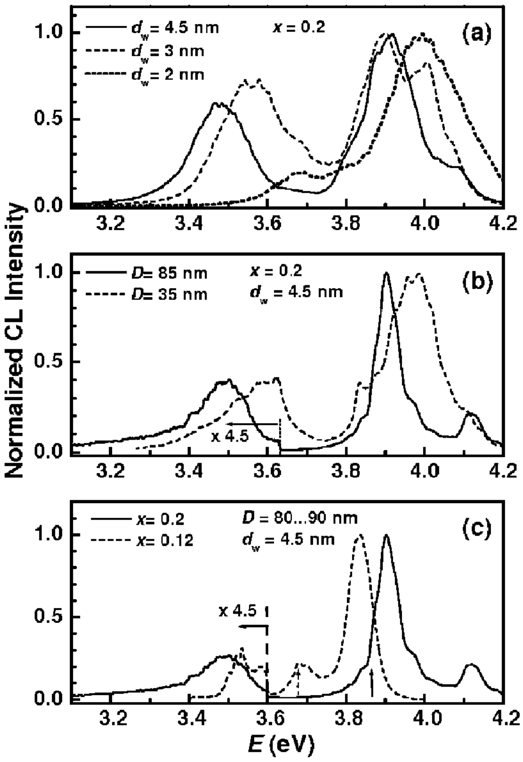

Figure 2 CL spectra of GaN/(Al,Ga)N nanocolumns containing Qdisks at 6 K. The spectra of (a), (b), and (c) differ in the disk thickness d_w, column diameter D, and AlN mole fraction x of the barriers, respectively. While in (a), a group of nanocolumns has been excited for each measurement, in (b) and (c), the CL spectra have been obtained from single nanocolumns.

In Fig. 2(a), each spectrum was obtained from a group of columns. We clearly observe a blue-shift of the disk spectra with decreasing d_w. Thus, even an averaging over many columns follows the expected trend indicating that indeed the disk thickness does not vary significantly among the columns of each of the samples. Consequently, we have been encouraged to acquire CL spectra of single columns differing in their diameter D of the samples with d_w = 4.5 nm and x = 0.2 assuming that both values do not vary significantly among the columns. As an example, two CL spectra are depicted in Fig. 2(b), where D amounts to 85 and 35 nm for the solid and dashed line, respectively. We clearly observe a blue-shift of the Qdisk spectrum for D = 35 nm compared with the one of D = 85 nm. This trend has been confirmed by the investigation of other single columns of this sample.

Finally in Fig. 2(c), we compare two CL spectra differing in the AlN mole fraction for single nanocolumns with comparable values of D and d_w. As expected, the CL spectrum of the Qdisks with barriers exhibiting higher AlN mole fraction is red-shifted relatively to the one of the disks with lower x. This red-shift is caused by an increased strain with increasing x, which leads to a higher piezoelectric field within the disk layers and therefore to lower transition energies (E_t).

On the one hand, the dependence of E_t on both the disk thickness and AlN mole fraction resembles the behavior found for the corresponding QW structures . On the other hand, the dependence of E_t on the diameter of the disks, the large broadening of the disk spectra, and a remarkable reduction of the quantum efficiency for Qdisks with $d_w < 3$ nm, which is demonstrated in Fig. 2(a), are typical features of our mesoscopic disk-like heterostructures

In the following, we would like to focus on the diameter dependence of E_t, for which both experiments and calculations have been performed. For the band structure and wave function calculations, we have used a strain-confinement model described Within the framework of this model, the lateral strain distribution of the disk and barrier layers have been numerically calculated, which in turn is used for the calculation of the in-plane carrier distribution of the Qdisks by means of a 2D Schrödinger solver.

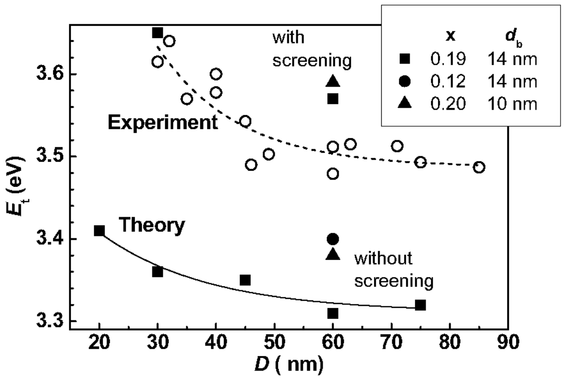

Figure 3 Optical transition energy as a function of the diameter of a GaN/(Al,Ga)N Qdisk. Full and open symbols represent numerical calculations and experimentally obtained data, respectively. The disk thickness amounts to 4.5 nm. Experiments have been performed at 6 K. The dashed and solid lines represent exponential fits to the data points.

Figure 3 shows the calculated transition energies of the Qdisks as a function of the disk diameter (full squares). The values of d_w and x were chosen to be 4.5 nm and 0.19, respectively. Moreover, for $D = 60$ nm, x and the thickness of the barriers (d_b) have been reduced to 0.12 and 10 nm (full circle and triangle), respectively, in order to check their influence on E_t. For comparison with the experiments, the CL peak energy as a function of D of the 4.5-nm-thick Qdisk ($x = 0.2$) have been added (open circles). Clearly, the model reflects qualitatively the experimentally found behavior of E_t of the Qdisks namely an increase with decreasing D and a smaller E_t for a higher value of x. As a further result of the model calculations, we have to expect a very efficient screening of the internal electric field of the Qdisks by the excited carriers due to their long lifetimes (not shown here). In Fig. 3, this is demonstrated by the calculated value of E_t for $x = 0.2$ and $d_b = 10$ nm (full triangle) and for $x = 0.19$ and $d_b = 14$ nm. The low and high values have been obtained without and with taking electric-field screening into account, where – corresponding to the experiments – a generation rate of excited carriers of 1.4×10^{12} electron-hole pairs per second has been assumed. Electric-field screening is not only responsible for the generally higher experimentally found E_t values, but can also explain their larger increase with decreasing D compared with the theoretical results, since field screening becomes more effective for disks with small diameters (see the full square for D=30 nm).

4 Conclusion The optical properties of GaN Qdisks embedded within (Al,Ga)N nanocolumns are essentially influenced by the lateral strain distribution of the disks and barriers. As an example, the dependence of the emission energy on the diameter of a 4.5-nm-thick Qdisk is rather determined by a partial relaxation of the strain and by an efficient screening of the strain-related electrical field than by lateral confinement effects. Further consequences of the distinct strain distribution of such mesoscopic structures are (i) a significant dependence of E_t on the barrier thickness, (ii) an unexpected spectral broadening, and (iii) a remarkable decrease of the quantum efficiency for thin disks.

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