Cathodoluminescence spectroscopy and imaging of GaN/(Al,Ga)N nanocolumns containing quantum disks

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Using spatially resolved cathodoluminescence spectroscopy, the authors have measured the spectral and spatial distribution of the luminescence intensity of GaN/(Al,Ga)N nanocolumns containing quantum disks. The optical emission of the quantum disk and of the thick (Al,Ga)N layer in the columns were clearly identified. The disk spectra of single columns are as broad as 80 meV. A significant contribution to this broadening is probably due to the laterally inhomogeneous strain distribution within the disks. The optical emission of the thick (Al,Ga)N layer in the columns is spread out over a wide spectral range of several 100 meV, which is caused by an inhomogeneous incorporation of Al along the growth direction of the columns. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2724913]

For group-III nitrides, the growth of microcrystals provides an approach to fabricate defect-free GaN,[1] which is still a challenge for the epitaxy of corresponding layer systems. In this regard, the self-organized formation of GaN nanocolumns during plasma-assisted molecular-beam epitaxy (PAMBE) is a promising mechanism, which even allows for the fabrication of heterostructures such as GaN/(Al,Ga)N quantum disks (QDisk),[1–3] GaN/AlN Bragg reflectors,[4] or (In,Ga)N/GaN multiple QDisk light-emitting diodes[5] embedded within nanometer-scale columnar crystals. The electronic properties of such heterostructures are significantly influenced by a laterally inhomogeneous distribution of strain and carrier concentration as well as by band bending near the surfaces,[6,7] which in turn depend strongly on the geometrical parameters of the columnar structure. A macroscopic physical characterization of the electronic properties of nanocolumns containing heterostructures, as has been done so far by photoluminescence[1] or Raman scattering,[8] generally averages over variations among the columns within a sample. In order to determine the true intrinsic electronic properties of nanostructures, an optical analysis of a single column consisting of a heterolayer system is highly desirable.

In this letter, we report on cathodoluminescence (CL) spectroscopy and imaging of nanocolumnar heterostructures with high spatial resolution, which allows for the characterization of a few or even a single nanocolumn. Samples with GaN/(Al,Ga)N nanocolumns containing five QDisks were fabricated by PAMBE under N-rich conditions on a Si(111) substrate.[9] As an example, Fig. 1(a) shows a scanning-electron-microscope (SEM) image of the columnar structure of a sample containing 3-nm-thick GaN QDisks. The height of the columns amounts to about 0.55 μm. Their diameter varies between 30 and 80 nm. A magnified image of the region marked by the dashed rectangle in Fig. 1(a) is depicted in Fig. 1(b). Five bright stripes are clearly visible representing the 3-nm-thick GaN QDisks. A schematic diagram of the whole columnar heterostructure of the investigated samples is depicted in Fig. 1(c). The columns consist of a 140-nm-thick GaN base, 280 nm (Al,Ga)N, five QDisks separated by about 8-nm-thick (Al,Ga)N layers, and a 40-nm-thick (Al,Ga)N cap layer. The nominal AlN mole fraction of the (Al,Ga)N regions of the columns amounts to 28%.

The optical emission of the nanocolumns was investigated by spatially resolved CL spectroscopy in a SEM. The used microscope (Zeiss-Ultra 55) is equipped with a Gatan

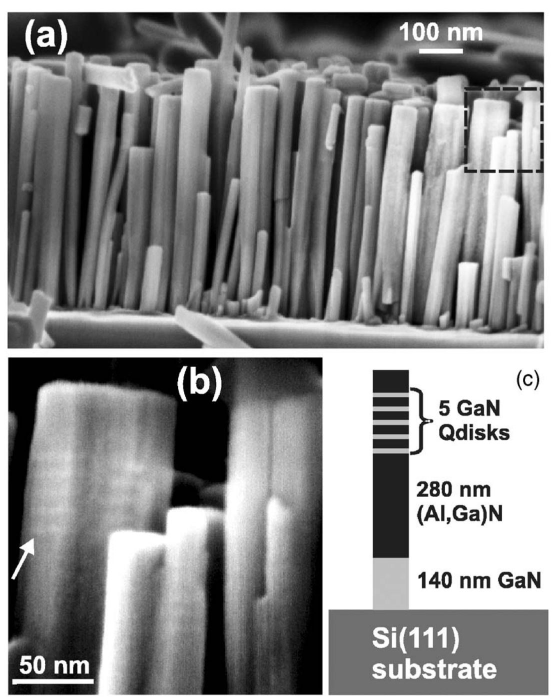

FIG. 1. (a) SEM image of GaN/(Al,Ga)N nanocolumns with 3-nm-thick GaN QDisks. (b) Magnified SEM image of the sample region marked by the dashed rectangle in (a). The arrow points to the 3-nm-thick QDisks. (c) Schematic diagram of the columnar heterostructure.

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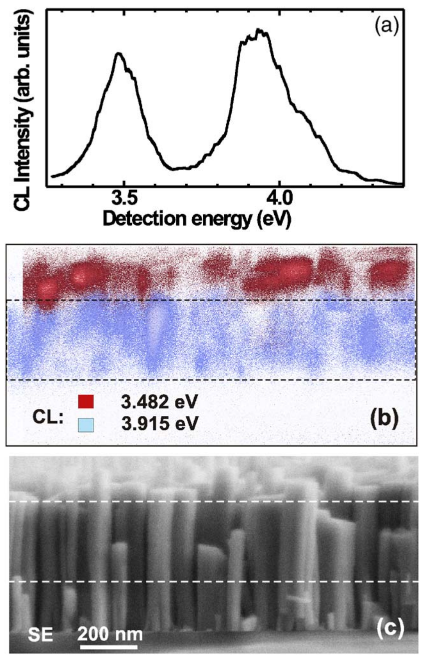

FIG. 2. (Color online) (a) CL spectrum of a large number of GaN/(Al,Ga)N nanocolumns containing 4.2-nm-thick QDisks. (b) Superimposed CL images differing in the detection energy. The dark and bright colored images correspond to CL detection energies of 3.482 and 3.915 eV, respectively. (c) SEM image of the GaN/(Al,Ga)N nanocolumns investigated by CL in (a) and (b). The spectrum and images have been obtained at 6 K.

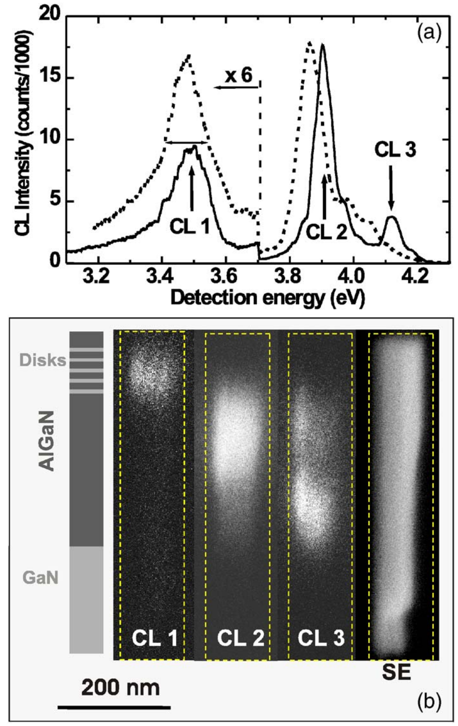

FIG. 3. (Color online) (a) CL spectra of two single GaN/(Al,Ga)N nanocolumns at 6 K. (b) CL images and SE image of the single nanocolumn represented by the solid line in (a) at 6 K. The detection energies used for CL imaging are marked by arrows in (a).

MonoCL3 system as well as with a He-cooling stage. Thus, optical investigations can be performed at temperatures down to 6 K with a spatial resolution on the order of a few tens of nanometers. The electron beam energy and current used in the CL experiments amounted to 3 keV and about 2 nA, respectively.

In the following, we discuss the optical properties of a sample, which contains 4.2-nm-thick QDisks. Figure 2(a) represents a CL spectrum obtained for a large number of nanocolumns of this sample. The spectrum consists of two broad bands centered at about 3.48 and 3.91 eV. In Fig. 2(b), the CL image obtained for a detection energy (E_d) of 3.482 eV (dark) is superimposed on the one obtained for E_d=3.915 eV (bright). The high-E_d CL image has been acquired simultaneously with the SE image of Fig. 2(c). The small horizontal shift between the CL images is due to a drift of the sample during the measurement. The comparison of the CL images with the SE image clearly confirms that the low-energy CL band originates from the top of the columns and can therefore be assigned to the emission of the QDisks. There is only a negligible contribution from the GaN base of the columns, which, in principle, is also expected to contribute to the optical emission in this spectral range. The high-energy CL band represents undoubtedly the thick (Al,Ga)N layer situated between the QDisks and GaN base of the columns.

In Fig. 2(a), both the spectrum of the QDisks and the one of the (Al,Ga)As are rather broad. Their full widths at half maximum (FWHM) amount to 155 and 205 meV, respectively. In order to separate contributions of the spectral broadening caused by averaging over a large number of columns from intrinsic broadening mechanisms, we have performed CL experiments on a single nanocolumn. The CL spectra of Fig. 3(a) originate from two single nanocolumns of the same sample, as shown in Fig. 2. The FWHM of the QDisk-related CL bands is smaller—it amounts to about 130 meV—compared with the one of Fig. 2(a), but is still much larger than the one of corresponding quantum well (QW) spectra, which amounts to about 20 meV.[12,13] The (Al,Ga)N-related part of the CL spectra of Fig. 3(a) is split into two lines spanning a large energy range. Figure 3(b) shows three CL images and a SE image of one of the columns. The E_d values chosen for the CL imaging are marked by arrows in Fig. 3(a). The picture CL 1 confirms again the QDisk-related origin of the low-energy part of the spectrum. The pictures CL 2 and CL 3 reveal clearly that the thick (Al,Ga)N layer of the column consists of various regions differing in the optical emission energy, indicating an inhomogeneous distribution of the AlN mole fraction along the column. Therefore, an averaging over many columns results in a broad (Al,Ga)N-related CL spectrum, as has been obtained in Fig. 2(a). The observed phase separation may be caused by strain arising from the lattice mismatch between the GaN base and the thick (Al,Ga)N layer of the column. Further investigations are necessary for a better understanding of this phenomenon.

Concerning the spectral broadening of the QDisk CL, rough interfaces can be ruled out, since high-resolution transmission-electron-microscope investigations (not shown) revealed smooth (Al,Ga)N/GaN/(Al,Ga)N interfaces. According to theoretical studies by Rivera *et al.*,[7] a considerable contribution to the broadening of the QDisk luminescence is expected to be an inherent property of mesoscopic structures such as the investigated QDisks due to an inhomogeneous lateral strain distribution induced by the large surface-to-volume ratio.

In order to further limit averaging effects, we restricted the excitation volume of the CL measurement to the QDisk

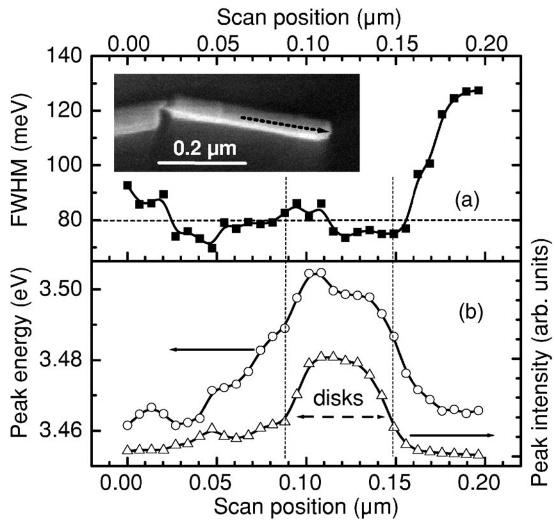

FIG. 4. (a) FWHM and (b) peak energy (circles) and peak intensity (triangles) of CL spectra measured along a line [dashed arrow in the inset of (a)] crossing the QDisk region of a single nanocolumn at 6 K. The dashed vertical lines mark the QDisk region. Inset of (a): SEM image of the GaN/(Al,Ga)N nanocolumn.

region by scanning the electron beam along the upper part of a single column of the same sample, as has been investigated in Figs. 2 and 3. 30 CL spectra have been measured along the black arrow marked in the inset of Fig. 4(a). Figures 4(a) and 4(b) show the FWHM and the spectral line position as well as the peak intensity of the corresponding QDisk spectra as a function of the scan position, respectively. The FWHM amounts to about 80 meV except for excitations far below and above the QDisk region, where its value increases. The latter is not yet fully understood, but could be due to an inhomogeneous electric-field screening for the case of an indirect excitation of the QDisks. An excitation-related electric-field screening is clearly indicated by the blueshift of the spectra [cf. circles in Fig. 4(b)], when the electron beam approaches the QDisk region, i.e., when the excitation-related carrier density of the QDisks increases. Note that the linewidth of the QDisk CL is mainly above a value of about 80 meV, which is by far larger as observed for corresponding QWs.[2,13] Besides the lateral strain distribution, other factors can additionally contribute to the broadening of the CL spectra such as band filling, inhomogeneous electric-field screening of the stack of QDisks due to the surface depletion layer as well as variations among the QDisks. Further investigations on single QDisks are necessary in order to distinguish between those contributions.

In conclusion, while the luminescence lines of single homoepitaxial GaN nanocolumns and GaN/(Al,Ga)N QWs are as narrow as 2–5 and 20 meV,[10–13] respectively, the FWHM of GaN/(Al,Ga)N QDisk spectra is as large as about 80 meV, which can partly be caused by inherent optical properties of such mesoscopic structures due to a laterally inhomogeneous distribution of strain. This interpretation is consistent with recently published theoretical results predicting unexpectedly low quantum efficiencies for thin GaN/(Al,Ga)N QDisks and generally broad optical spectra caused by a laterally separated confinement of electrons and holes, a lateral inhomogeneous carrier distribution, and a laterally as well as vertically inhomogeneous screening of the electric field within the QDisks.[4,7] Consequently, the lateral strain distribution can counteract the advantage of a defect-free growth of GaN-related nanocolumnar structures. Therefore, a careful choice of the structural parameters, particularly, of a proper ratio between the diameter of the columns on the one side and the thickness of the QDisk as well as barrier layers on the other side is important for the employment of such columnar heterostructures in optoelectronic device applications.

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