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Exciton-related photoluminescence in homoepitaxial GaN of Ga and N polarities

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A photoluminescence (PL) study of GaN homoepitaxial layers grown by metal–organic chemical-vapor deposition demonstrates the high optical quality of N-face layers deposited on vicinal $(000\bar{1})$ GaN substrates. In contrast to broad PL emission in exact $(000\bar{1})$ layers, narrow-bound (0.9 meV) and free- (*A* and *B*) excitonic transitions are observed. By following the PL spectra as a function of temperature and excitation power, the main optical transitions in the Ga- and the misoriented N-face layers are found to be the same. Observed differences are related to the distinct creation of donor and acceptor states in the samples of different polarities. © *2000 American Institute of Physics.* [S0003-6951(00)02317-2]

The recent progress in GaN-related (opto)electronic devices is based on a constant improvement of structural and morphological quality of heteroepitaxially grown III-nitrides layers.[1] The lattice mismatch and different expansion coefficients of the epilayer and substrate cause, however, a high density of defects and a biaxial strain in the heteroepitaxial layer changing the basic optical properties of the material.[2–5] In contrast, homoepitaxial GaN layers correspond to almost stress-free material[6] with a much lower dislocation density $(10^4-10^5\,\mathrm{cm}^{-2})$.[7] Superior optical quality has been demonstrated for (0001) Ga-face layers, showing excitonic transitions narrower than 1 meV.[7,8] From a technological point of view, N-face GaN epilayers are also very attractive since, in contrast to Ga-face layers, they are easily processable by mechanochemical etching. However, epitaxial layers grown on a N-face GaN substrate generally reveal a rough surface morphology with hexagonal hillocks and a high free-carrier concentration.[9] Lack of good homoepitaxial N-face layers has also hampered the study of polarity-related effects in GaN. In order to improve the morphology of these layers it was recently proposed to grow N-polar films on a misoriented GaN substrate.[10] The miscut introduces additional steps on the substrate surface leading to lateral overgrowth of potential hillock nucleation centers.

In this letter, we present the results of a photoluminescence (PL) study of homoepitaxial GaN of different polarities (Ga and N). Samples with the crystallographic *c* axis perpendicular to the surface plane as well as a N-polar layer grown on a misoriented substrate with an off-angle of 4° toward the $\langle 11\bar{2}0\rangle$ direction were examined. By using a N-face misoriented substrate, a smooth epilayer morphology,[10] together with a significant suppression of donor concentration, was achieved. As a result, the optical quality of the N-face film was found to be drastically improved, enabling a direct comparative study of epilayers of Ga and N polarities.

Nominally undoped epitaxial GaN layers with a thickness of about 1.5–2 μm were grown by metal–organic chemical-vapor deposition using the conditions reported in Refs. 9 and 10. The bulk GaN single-crystal substrates were, typically, 100–200 μm thick with a surface area of 30–50 mm^2 and a free-electron concentration close to $5\times10^{19}\,\mathrm{cm}^{-3}$. The details of their structural properties and the growth technique have been described earlier.[11] Prior to the epitaxial growth, mechanochemical polishing (for the N face)[9] or mechanical polishing with subsequent reactive ion etching (for the Ga face)[12] were applied in order to obtain smooth substrate surfaces. Structural and morphological properties of the resulting epilayers were thoroughly studied in Refs. 9, 10, and 12. The determination of the unintentional doping concentration in the epitaxial layers was difficult due to the conducting substrate. By comparison with similar homoepitaxial GaN reported earlier[13,14] and considering the very narrow PL peaks shown below, we estimated the free-carrier concentration of the top 100 nm of the layer to be in the range of low $10^{17}\,\mathrm{cm}^{-3}$ for both the Ga-face and misoriented N-face samples.

PL measurements were performed at temperatures from 4 to 110 K. A HeCd laser (325 nm) was used for excitation with power densities up to $I_0=50\,\mathrm{W/cm}^2$, incident at approximately 30° to the normal of the sample surface. The PL emission was dispersed by a 0.6 m monochromator and detected by a cooled GaAs photomultiplier. The spectral resolution was 0.4 meV in the region from 3.2 to 3.55 eV, unless stated otherwise.

Low-temperature PL spectra of a (0001) Ga face, a $(000\bar{1})$ N face, and a misoriented N-face GaN epilayer are presented in Fig. 1. The epitaxial layers grown on the $(000\bar{1})$ N-face substrate result in a broad PL emission, indicating a high free-electron concentration as a consequence of either intrinsic donor generation or easy donor incorporation.[14] In order to monitor the influence of the substrate preparation on the epilayer properties, either mechanochemical polishing or mechanical polishing with subsequent reactive ion etching were used. In both cases, however, the exact $(000\bar{1})$ films reveal PL spectra of the same intensity and shape. In con-

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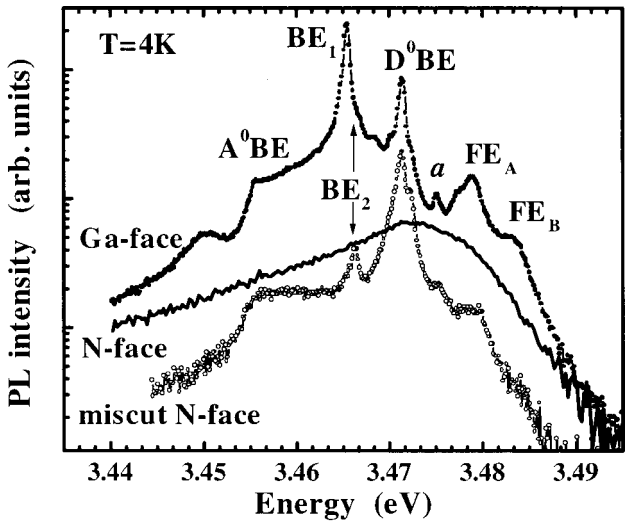

FIG. 1. Low-temperature PL spectra of Ga-face, N-face, and 4°-misoriented N-face GaN samples.

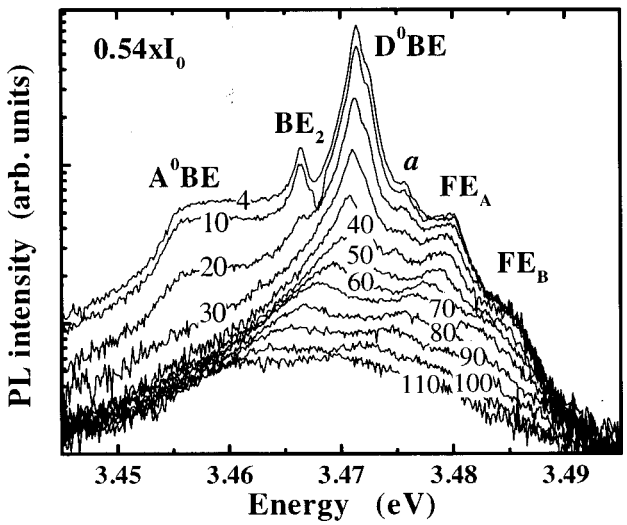

FIG. 3. PL spectra of the misoriented N-face GaN epilayer measured at temperatures ranging from 4 to 110 K. The numbers on the curves denote the corresponding temperature in K.

trast, the miscut N-face epilayer shows a PL spectrum very similar to that of the (0001) Ga-face sample featuring narrow-bound and free-excitonic (FE) lines. Two peaks, FE_A (3.4789 eV) and FE_B (3.4832 eV), are associated with *A* and *B* free excitons, which dominate the PL spectra at higher temperatures for both samples (Figs. 2 and 3). The assignment of these peaks is also supported by reflectance measurements performed on similar (Ga-face) samples.[8,15,16] The PL line at 3.4714 eV (Fig. 1) was attributed to excitons bound (BE) to a neutral donor (D^0BE) with an exciton localization energy of 7.5 meV. The full width at half maximum (FWHM) of the D^0BE peaks at 4 K goes down to 0.35 meV in the Ga-face layer, and to 0.9 meV in the misoriented N-face one. The D^0BE line in the misoriented N-face sample is unusually narrow for homoepitaxial GaN of N polarity. Although the narrow PL peaks of the misoriented N-face sample show a strongly reduced free-carrier concentration, the dominant character of the D^0BE peak at low temperatures still indicates a considerable amount of donors.

It is important to note that the energy positions of almost all main PL peaks observed in the miscut N-face layer are equal to those in the Ga-face layer. This also includes a peak labeled *a* (at 3.4752 eV), which can be associated with donor-bound excitons formed with participation of a deeper valence-band hole, and a peak originating from excitons bound to neutral acceptors (A^0BE at 3.4556 eV). As a consequence, we can conclude that the structural properties, i.e., lattice parameters of the Ga- and misoriented N-polar layers, are identical. The main difference between the high-quality Ga- and N-face layers is related to the spectral region around 3.466 eV. The origin of the emission lines BE_1 (3.4654 eV) and BE_2 (3.4664 eV) in this region seems to be ambiguous and has been previously identified either as excitons bound to a neutral acceptor[16,17] or as ionized-donor-bound excitons.[18,19] The BE_1 peak is more pronounced than BE_2 in the Ga-face sample but not present in the misoriented N-face one. In the Ga-face layer, the BE_2 peak can only be seen as a high-energy shoulder of the BE_1 line because its FWHM

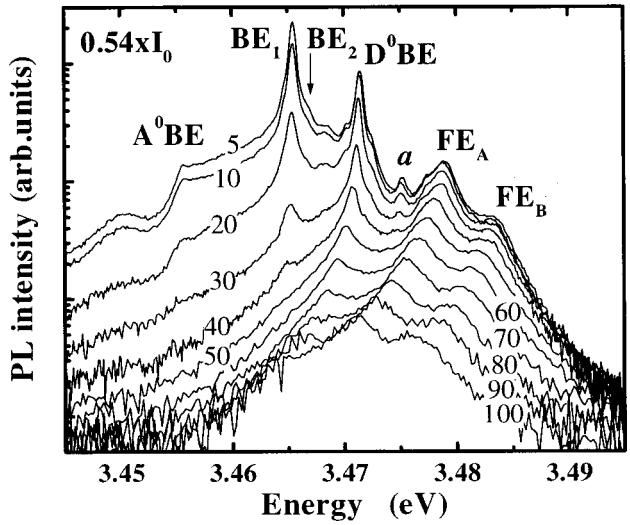

FIG. 2. PL spectra of the Ga-face GaN epilayer taken at temperatures ranging from 5 to 100 K. The numbers on the curves denote the corresponding temperature in K.

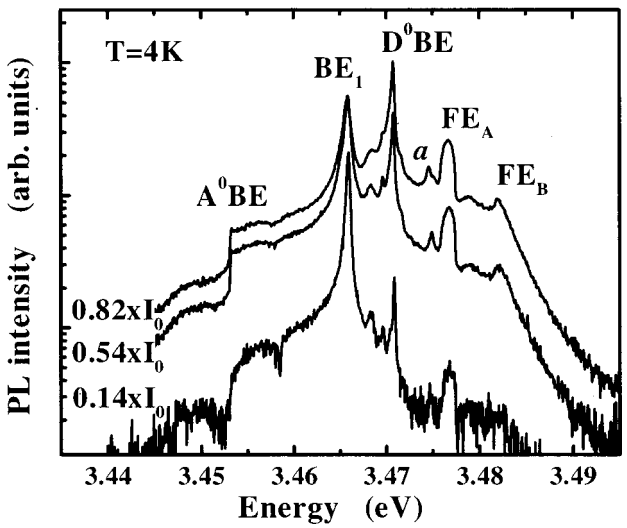

FIG. 4. Low-temperature (4 K) PL of the Ga-face GaN layer at different excitation intensities measured with a spectral resolution of 0.2 meV.

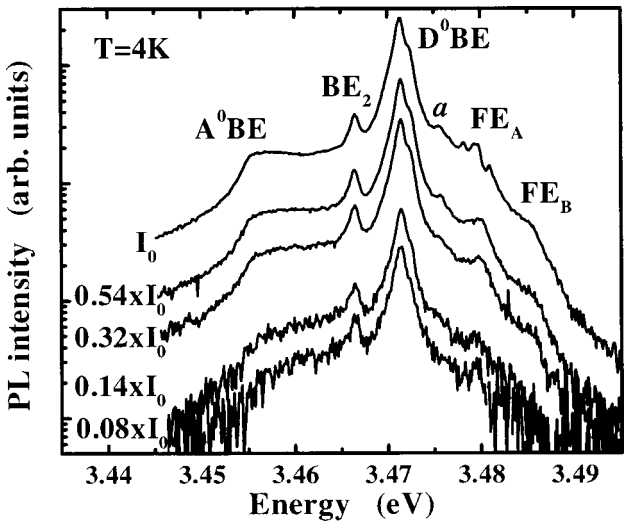

FIG. 5. Low-temperature (4 K) PL of the misoriented N-face GaN layer at different excitation intensities.

(~2 meV) is larger than the separation between the BE_1 and BE_2 transitions (~1 meV). The localization energies of these bound excitons are found to be 13.5 and 12.5 meV for BE_1 and BE_2, respectively. The thermal quenching of the lines happens, however, earlier than for the D^0BE, at 50 K for the BE_1 (Fig. 2) and at 30 K for the BE_2 (Fig. 3). This unusual behavior is probably related to exciton tunneling from BE_1/BE_2 to donor-bound or even free excitons that has been derived from the time decay of PL at temperatures above 30–40 K.[16] The excitation-dependent PL for the Ga-face layer in Fig. 4 shows saturation of the BE_1 peak intensity with increasing excitation power. On the other hand, the PL spectra for the misoriented N-face layer in Fig. 5 reveal no saturation of the BE_2 line even at higher excitations. Thus, from the temperature and excitation power-dependent PL it follows that the BE_1 and BE_2 peaks have different origins. In our opinion, the BE_1 line can be assigned to excitons bound to an acceptor as this transition is not seen in the N-face sample, but dominates the spectrum of the Ga-face epilayer, in which the incorporation of acceptors is expected to be easier.[14] The origin of the BE_2 peak can be another acceptor or donor present in both epilayers. In order to identify the specific acceptor corresponding to the BE_1 peak and to determine the origin of the BE_2 peak, further investigations are needed.

In conclusion, N-face homoepitaxial layers of high optical and morphological quality have been grown using a misoriented $(000\bar{1})$ GaN single-crystal substrate. In comparison with exact $(000\bar{1})$ films, a significantly lower donor concentration was achieved. The main PL transitions, such as FE_A, FE_B, D^0BE, and A^0BE, were observed at the same energies for both Ga- and misoriented N-face samples, suggesting that the lattice parameters of the Ga- and the misoriented N-polar layers are the same. Temperature and excitation-dependent PL of the Ga-face sample reveals, however, an optical transition (BE_1), which is not present in the misoriented N-face film. It can be associated with excitons bound to a neutral acceptor, incorporated only in the Ga-polar epilayer.[14]

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