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Dissimilarity between cleaved edge and surface regions of GaN (0001) epitaxial layers studied by spatially-resolved photoluminescence and reflectivity

T. V. Shubina[†], M. G. Tkachman[†], A. A. Toropov[†], A. I. Karlik[†], S. V. Ivanov[†],
P. S. Kop'ev[†], T. Paskova[‡] and B. Monemar[‡]

[†] Ioffe Physico-Technical Institute, St Petersburg, Russia

[‡] Department of Physics and Measurement Technology, Linköping University,
S-581 83 Linköping, Sweden

Optical properties of GaN, the perspective material for modern optoelectronics, have been intensively studied during last three decades. Generally, three exciton transitions A ($\Gamma_7 \times \Gamma_9$), B ($\Gamma_7 \times \Gamma_7$), C ($\Gamma_7 \times \Gamma_7$) observed in GaN with C_{6v} symmetry originate from crystal-field and spin-orbit splitting. Exchange interaction removes the degeneracy of the fourfold degenerate exciton series and splits the A ($n = 1$) level into an allowed Γ_5 and a forbidden Γ_6 component. Both ground B and C levels split into three levels: Γ_5 , Γ_1 and Γ_2 . The allowed optical transitions involve the Γ_1 state in an $\mathbf{E} \parallel \mathbf{c}$ polarization and the Γ_2 state in $\mathbf{E} \perp \mathbf{c}$ [1].

Most of the GaN optical studies have been performed from the surface of epitaxial layers in the so-called α -polarization, when the wave vector \mathbf{k} is parallel to the crystal axis \mathbf{c} and the electric field vector \mathbf{E} is normal to it. Very few papers concern investigation from a cleaved edge (facet) of the epitaxial layers, although such a study performed by Dingle *et al.* for $\pi - (\mathbf{k} \perp \mathbf{c}, \mathbf{E} \parallel \mathbf{c})$ and $\sigma - (\mathbf{k} \parallel \mathbf{c}, \mathbf{E} \parallel \mathbf{c})$ polarizations permitted one to check selection rules and determine experimentally the principal parameters of GaN valence bands [2]. During the edge study some dissimilarity between α - and σ -polarized reflectance spectra was observed, which is not explained in the classical model and was not discussed in detail. Moreover, to the best of our knowledge, the edge reflectance measurements have not been repeated since that time due to obvious difficulties in preparation of high quality cleaved facets.

In this paper we present results of comparative studies of the edge and the surface optical properties of GaN epilayers done, using micro-photoluminescence (μ -PL) and reflectance (R) spectroscopies. The results undoubtedly demonstrate inequality in exciton transitions taking place in the internal and near surface regions of the epilayers, which is presumably controlled by anisotropic strain and defect density.

The study was performed using a typical $\sim 25 \mu\text{m}$ -thick sample grown by HVPE as described previously [3]. Detailed structural characterization by x-ray diffractometry (XRD) and transmission electron microscopy demonstrates a good quality of the sample, especially in the top near surface regions where the dislocation density is less than 10^8 cm^{-2} . At room temperature the layer experiences a biaxial compressive strain ($\sim 0.2 \text{ Gpa}$).

Measurements of μ -PL were carried out in a He continuous flow cryostat at 4 K under cw excitation by a 266 nm laser line of a solid-state diode-pumped frequency-doubled Nd:Vanadate cw laser followed by a MDB-266 frequency doubler unit. The beam impinging normally onto a surface or a cleaved edge facet of the sample was focused using a reflective objective creating an excitation spot of $\sim 1.5 \mu\text{m}$ in diameter. The same objective collects

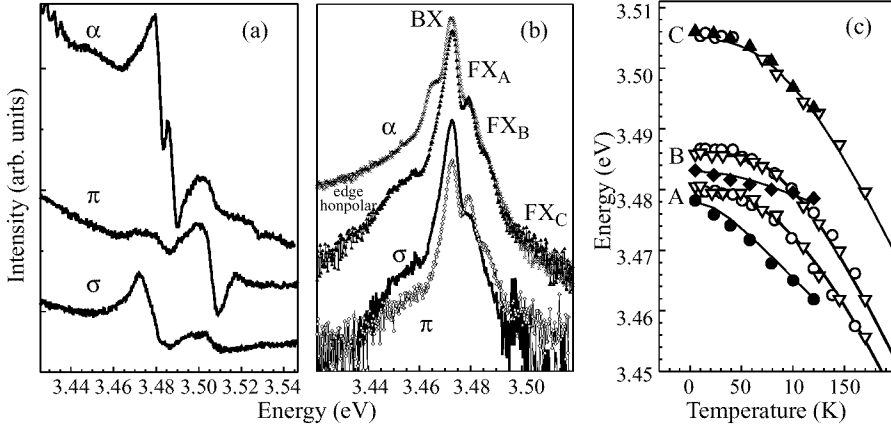


Fig. 1. Reflectance (a) and μ -PL (b) spectra measured at 5 K in a GaN epitaxial layer in different polarizations: α ($\mathbf{k} \parallel \mathbf{c}$, $\mathbf{E} \perp \mathbf{c}$), σ ($\mathbf{k} \perp \mathbf{c}$, $\mathbf{E} \perp \mathbf{c}$) and π ($\mathbf{k} \perp \mathbf{c}$, $\mathbf{E} \parallel \mathbf{c}$). Non-polarized facet μ -PL spectrum is shown by triangles. (c) Temperature dependencies of exciton energies obtained from polarized reflectance (σ solid circles, π solid diamonds (B) and triangles (C), and α open circles) and μ -PL (open triangles).

the PL signal and the sample image which is monitored by a charge coupled detector (CCD). The PL is relayed to the slits of an asymmetrical Czerny-Turner type monochromator and then to the nitrogen cooled CCD. The spectral resolution of the system is estimated as ~ 0.6 meV. The R measurements were performed in the same set-up, using a tungsten lamp as an excitation source. At the same focusing conditions the R spatial resolution is estimated as ~ 10 μm . A linear polarizer (followed by a depolarizer) was mounted before monochromator slits to analyze the PL and R polarization.

The comparison of R and μ -PL data presented in Fig. 1(a) and (b) permits us to assign the most intense peak to a bound A exciton which appears to be generally σ -polarized. The ordering and peak energy of free A, B and C excitonic transitions (marked as FX_A , FX_B and FX_C , respectively) are consistent with published data for weakly strained GaN on sapphire [4]. The μ -PL spectra are characterized by the distinctiveness of the excitonic features, whose width is about 3 meV at low power. The μ -PL measurements across the layer edge permit us to attribute a distinguished shoulder in the α -polarized μ -PL spectrum as reemission from relaxed bottom-interface regions, whereas temperature- and power-dependent studies show that an enhanced π -polarized component in the vicinity of a free A exciton peak FX_A is related to joint contribution of bound excitons of the B band and scattered states of the A bands.

The reflectance spectra (Fig. 1(a)) satisfy generally the selection rules and the expected intensity relationship of the exciton transitions [2]. At the α - and σ -polarizations the $\text{FX}_A(\Gamma_5)$ and $\text{FX}_B(\Gamma_5)$ exciton resonances are well pronounced, while the $\text{FX}_C(\Gamma_5)$ one is rather weak. Contrary, in the π -polarization, the $\text{FX}_C(\Gamma_1)$ resonance dominates with a well-distinguished $\text{FX}_B(\Gamma_1)$ feature and negligible FX_A . However, the α - and σ -polarized R spectra are appreciably different - the α -polarized spectrum is shifted to higher energies as compared to the σ -polarized one. Exciton resonances in the facet spectra are wider, likely, due to the edge region imperfection.

The difference between the α - and σ -polarized R becomes more pronounced with the temperature variation. Figure 1(c) presents exciton energies which are deduced from the R

spectra using a three oscillator model of the dielectric constant [5]. The exciton resonances, belonging to the same band, shift differently in the facet and surface spectra, although the energy gap between Γ_5 and Γ_1 states is expected to be less than 2 meV [6]. Only the C-band exciton transitions, shifted in energy due to spin-orbit splitting, coincide at all temperatures.

We have performed fitting of the facet R and μ -PL temperature dependencies of the exciton energies using an analytical four-parameter model proposed by R. Pässler [7]

$$E(T) = E(0) - \frac{\alpha_p \Theta_p}{2} \times \left[\frac{\rho}{2} \left(\sqrt[4]{1 + \frac{\pi^2}{6} \left(\frac{4T}{\Theta_p} \right)^2 + \left(\frac{4T}{\Theta_p} \right)^4} - 1 \right) + (1 - \rho) \left(\coth \left(\frac{\Theta_p}{2T} \right) - 1 \right) \right]$$

where $E(0)$ is a zero-temperature transition energy; α_p is a high-temperature slope of the dependence; the parameter $0 \leq \rho \leq 1$ determines relative weights of long-wavelength acoustical phonons (ρ) and a combination of optical and short-wavelength acoustical phonons ($1 - \rho$); Θ_p is defined via the Debye temperature Θ_D as $\Theta_p \cong (2/3)\Theta_D/(1 - 1/2\rho)$.

The fit of the μ -PL data has been performed using $E = 3.4810, 3.4858, 3.5053$ eV; $\Theta_p = 420, 400, 420$ K; $\rho = 0.4, 0.3, 0.42$ for $FX_A(\sigma), FX_B(\pi), FX_C(\pi)$, respectively, with the same $\alpha = 0.42$. The parameters are close to those found for perfect homoepitaxial GaN films [8]. It appears that the α -polarized R data can be successfully described by the parameters obtained from the edge μ -PL fitting. The coincidence of the μ -PL and the α -polarized R dependencies (Fig. 1(c)) means that the main part of the polarized μ -PL is provided by internal regions and is not a characteristic of the outer surface of the edge.

The edge reflectance data are fitted with $E = 3.4780, 3.4830, 3.5053$ eV; $\Theta_p = 100, 200, 420$ K; $\rho = 0.9, 0.85, 0.4$ for $FX_A(\sigma), FX_B(\pi), FX_C(\pi)$, respectively. The parameters are hardly meaningful in the model. It is worth to note that the energy gaps between the exciton energies taken as a function of the A exciton line, assumed frequently as a measure of stress in GaN [6], vary differently in the edge and internal regions. Reflectance edge data demonstrate the anti-correlated variation between bands, while similar energy gaps found from μ -PL and surface R are almost constant. Thus, the facet R dependencies seem to reflect a strong anisotropic strain near the facet surface [9].

The data on the band separations in the edge and internal regions are used to estimate spin-orbit Δ_{SO} and crystal-field Δ_{CR} parameters in the Hopfield's quasicubic model [10]. The obtained values at 5 K are: $\Delta_{CR} = 9.5$ meV and $\Delta_{SO} = 22$ meV – for edge regions; $\Delta_{CR} = 10.4$ meV and $\Delta_{SO} = 20.3$ meV – for internal regions. The set is generally consistent with that previously reported for surface measurements [6]. Particularly, the edge parameters are close to those found in facet studies [2], but with opposite assignment of Δ_{CR} and Δ_{SO} , which is possible because the Hopfield's model is symmetrical with respect to these parameters.

In conclusion, polarized photoluminescence and reflectance spectroscopy with high spatial resolution are employed to show the different optical properties of cleaved edges with respect to internal regions of thick GaN epitaxial layers. The values of spin-orbit and crystal-field parameters are determined separately for these areas. The obtained data indicate that an absorption edge of the material within the cleaved edge regions is below the energy of emission from internal regions (see Fig. 1(c)), which can be disadvantageous for the operation of edge-emitting devices, such as LED and lasers.

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