Journal of Physics: Conference Series

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To cite this article: M G Mynbaeva *et al* 2020 *J. Phys.: Conf. Ser.* **1697** 012064

Emission properties of textured gallium nitride with high density of stacking faults

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Abstract. In this study we related structural properties of GaN grown on ceramic substrate and studied with transmission electron microscopy with the results of photo- and cathodoluminescence investigations. We found that stacking faults in the basal plane were responsible for both strong room temperature visible emission and exciton-related ultraviolet luminescence at cryogenic temperature.

1. Introduction

Most recently, light emitting nanostructures formed by 'optically active' crystal lattice defects in semiconductors have attracted much attention as prospective components for future quantum technologies [1]. In particular, it has been shown that defects in GaN can act as bright photon emitters that operate at room temperature (RT) in a wide spectral range from ultraviolet (UV) to infrared (IR). These defects are basal-plane stacking faults (BSFs) that represent cubic inclusions within hexagonal GaN, and/or intrinsic defects or impurities surrounding an extended defect, such as a SF or a dislocation. Room-temperature 'optically active' emitters were discovered in standard doped and un-doped thin GaN films grown on various kinds of commercial substrates [2]. In this work, we report on defect-related bright RT emission in a free-standing textured GaN bulk material.

2. Experimental

Large-area 3 mm-thick GaN slabs were produced by deposition on ceramic substrates in the reactor designed for Hydride Vapour-Phase Epitaxy (HVPE) [3]. We use the term "bulk material" because there was no epitaxial interaction between the substrate and the grown material due to the presence of a gallium-based liquid phase on the surface of the substrate at the nucleation stage of GaN. For the study of optical properties, several specimens were cut from the slabs in two geometries: a number of 1.1×1.1 cm^2 plates with the thickness 100 to 200 μm were sliced along the growth direction, and a cleaved edge of 1.5 mm-thick sample was also studied. Correlation of the optical properties with specifics of the defect structure of the material was established with transmission electron microscopy (TEM) and RT- and low-temperature photoluminescence (PL) studies. For the TEM studies, Philips EM–420 (accelerating voltage 80 kV, resolution 5 Å) and Jeol JEM–2100F (accelerating voltage 200 kV, resolution 2 Å) microscopes were used. Specimens for these studies were prepared using standard procedures of mechanical thinning and subsequent etching with Ar^+ ions with energy 3 to 4 keV. For the PL studies, Horiba Jobin-Yvon T64000 spectrometer was used.

3. Results and discussion

Earlier, we have shown that the slab material was an *n*-type semiconductor with thermal and mechanical properties close to those of single-crystal GaN produced by conventional bulk growth techniques [3]. X-ray diffractometry and TEM studies showed that the material represented a highly ordered (0001) wurtzite GaN texture formed by crystallographic blocks with lateral dimensions gradually increasing from units to hundreds of micrometers towards the slab surface [4]. According to the results of TEM, the predominant types of defects in the blocks were multiple intrinsic BSFs of I_1 type; prismatic defects also have been observed, but with a significantly lower density. With the thickness of the material increased up to 1000 μm, the density of SFs in individual blocks reached 10^6 cm^{-2} [5]. Along with this, it was found that when using characterization methods employing the action of ion and electron beams, the material showed an unusual bright glow at RT. The observed light emission propagated in the bulk of the material to a considerable (hundreds of micrometers) distance from the point of excitation. The characterization methods included Auger electron microscopy (AES), Secondary-Ion Mass Spectroscopy (SIMS) and Micro-Cathodoluminescence (MCL) imaging. AES and SIMS measurements were performed with the use of a low (9.5 keV) energy Ar^+ ion beam. The ion beam scanning of an area 1000 × 1500 μm^2 was used for material sputtering during the depth profiling performed with focused electron beam current of 0.5 μA. MCL imaging technique was used to visualize emission regions (if any) with luminescence in a wide range of wavelengths in visible spectrum. The panchromatic images were obtained at RT using wide (diam. 100 μm) electron beam with energy 10 keV and beam current 60 nA. PL images of the surface of the samples were obtained with laser excitation and a digital camera was used to acquire an image of the emission. Some examples of RT glowing of textured GaN samples under impact of beams different in nature are shown in figure 1.

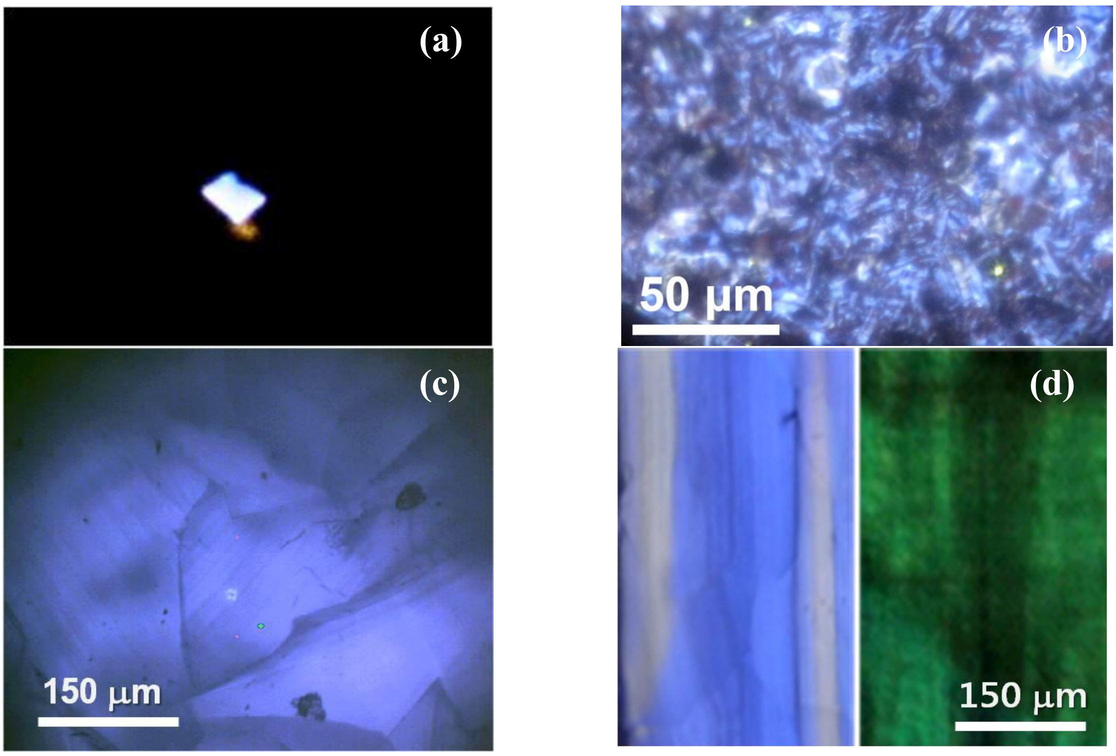

Figure 1. RT glowing of textured GaN under impact of energetic beams: glow of 1 cm^2 sample under scanning Ar^+ beam with exposure over 1000 × 1500 μm^2 surface area (a); plane-view panchromatic CL image of the low portion of the GaN slab (b); plane-view PL image of the surface of 1.5 mm-thick sample (c); panchromatic CL images: of a cleaved edge of 1.5 mm-thick textured GaN slab (left image) and of 600 μm-thick GaN layer grown by HVPE on a sapphire substrate (right image) (d).

Figure 1(a) is a photograph of the luminous specimen with 1 cm^2 in size placed in a vacuum chamber of the AES machine taken during its exposure to a scanning Ar^+ beam. Figures 1(b) and 1(d) (left image) show RT MCL images of the plane view of the sample that was cut from the low portion of the GaN slab, and of the cleaved edge of 1.5 mm-thick sample, respectively. Bright luminescence from the blocks with various dimensions is clear visible. For comparison, right image in figure 1(d) shows a CL image obtained under similar conditions from a transverse cleavage of a 600 μm-thick HVPE layer grown in the conventional way on a sapphire substrate [6]. In the latter case, the dominating defects were threading dislocations with density ~10^7-10^8 cm^{-2}, which is typical of this material. The difference in emitting ability of these two types of materials grown with the use of different approaches to HVPE is clear. A PL image of the surface of the 1.5 mm-thick textured GaN sample is shown in figure 1(c). The emission was excited by a nitrogen UV laser with the excitation wavelength λ_{ex}=325 nm. One can see a uniform emission from neighbouring blocks of the GaN texture with the size of the order of hundreds of micrometers. In order to clarify the nature of the unusual emission properties of the textured GaN, CL and PL spectra were recorded with excitation at λ_{ex}=325 nm. The main results obtained are presented in figure 2.

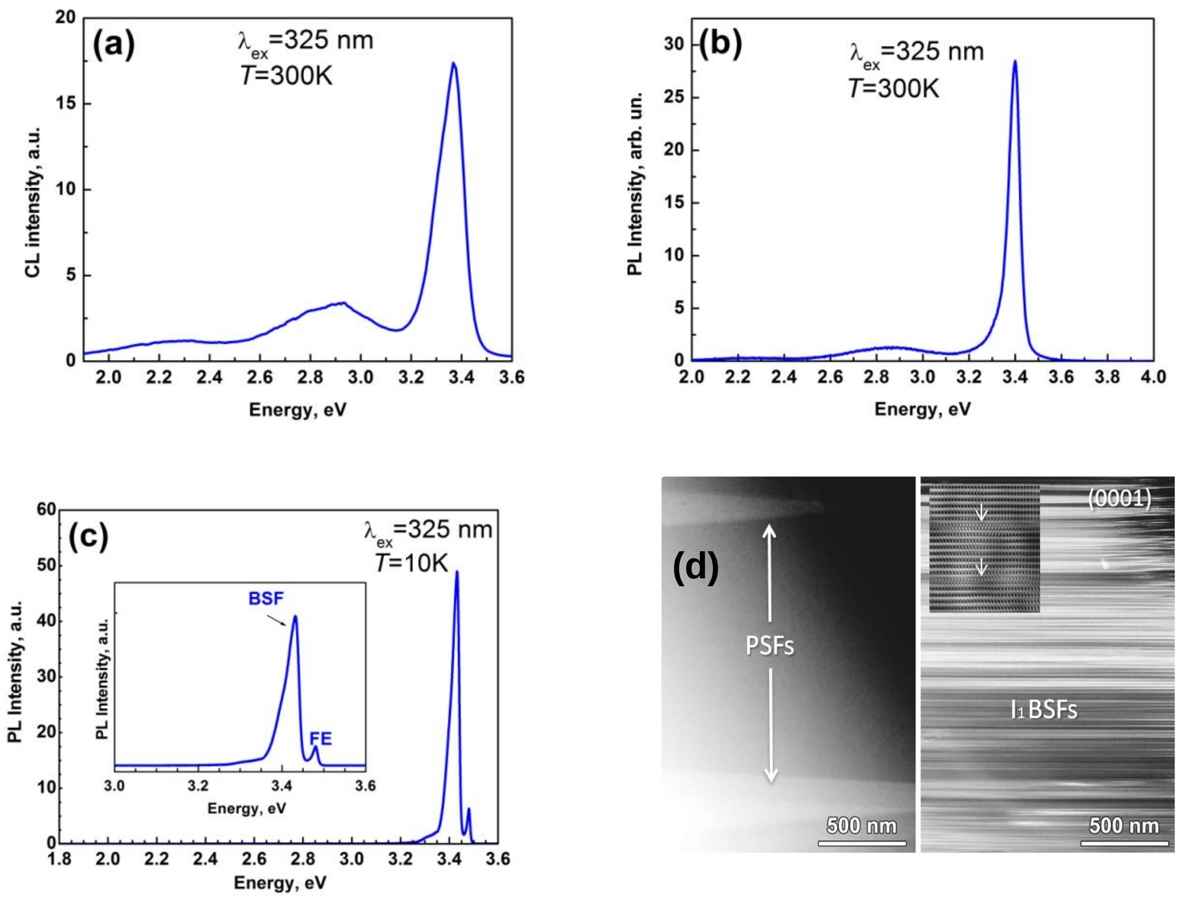

Figure 2. Luminescence of the studied material: RT CL (a) and PL (b) spectra, and a low-temperature PL spectrum dominated by the emission of excitons bound at BSFs with the inset showing the details of ‘edge’ PL (c). Image (d) shows TEM images of cross-sections of 1.5 mm-thick thick sample with prismatic stacking faults (PSFs, left image) and BSFs (right image), the inset shows a high-resolution TEM image of BSFs.

RT CL and PL spectra, as expected, were dominated by strong near-band-edge emission with the peaks at ~3.39 and ~3.36 eV and full widths at half-maxima (FWHM) of 0.102 and 0.052 eV, respectively. The shapes of both wide CL and PL peaks were indicative of the fact that most probably they represented a superposition of a number of luminescence lines, which made it hard to identify them. Low-temperature measurements shed the light on the origin of the observed luminescence (figure 2(c)). Low-temperature PL spectrum contained a band with the peak at 3.47 eV with FWHM 11 meV. Variable-temperature PL studies showed that this peak followed the temperature dependence of the bandgap of GaN. This allowed for attributing this PL band to the recombination of a free exciton (FE). The dominating band in the low-temperature spectrum had FWHM of 21 meV with the peak at 3.42 eV. At that, the intensity of this peak directly depended on the density of SFs in the samples, as determined with TEM. In samples with the highest density of I_1 BSFs (figure 2(d)) the intensity of this peak was ten times greater than that of the FE peak.

In accordance with the already existing results on bright RT luminescence in semiconductor materials that was associated with structural defects, the emission properties of textured GaN crystals can be described in terms of the quantum-well model proposed by Rebane et al [6]. The model assumes that due to the two-dimensional nature of BSFs they represent a kind of natural quantum wells with atomically flat interfaces. The local deviation from the 2H stacking of the wurtzite matrix induces a bound state in the gap of the host crystal, resulting in the localization of excitons at internal cubic-wurtzite interfaces. Highly efficient luminescence originates in radiative recombination of excitons with particular energies.

Concluding this topic, it remains to discuss the phenomenon of radiation-induced luminescence of GaN textured samples under argon ion treatment. At this stage of understanding of the optical properties of GaN we can speculate that irradiation with argon ions helps to reduce the number of trapped carriers at BSFs, thus encouraging the carriers to participate in the radiative recombination.

4. Conclusion

In this study we related structural properties of textured GaN grown on ceramic substrate, as studied with transmission electron microscopy, with the results of photo- and cathodoluminescence studies. We found that basal stacking faults were responsible for the observed strong blue emission at the room temperature and narrow exciton-related UV luminescence band at cryogenic temperature. This means that our GaN slabs is a promising material for the boosting technologies of non-classical solid-state photon emitters self-assembled within the host crystalline matrix. Luminescence under Ar^+ ion irradiation suggests that our material can be used in sensors for the visualization of particle beams in dosimetry applications.

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