Characteristic emission from quantum dot-like intersection nodes of dislocations in GaN

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Abstract. Freshly introduced a-screw dislocations in gallium nitride are an effective source of ultraviolet radiation, characterized by intense emission of narrow luminescence doublet lines in the spectral range of 3.1-3.2 eV. Furthermore, an additional narrow spectral line with an energy of 3.3 eV has been found at the points of intersection of such dislocations, where extended dislocation nodes were formed. In this communication, we report on the spectral properties of the characteristic luminescence of such nodes, which were obtained for the (0001) gallium nitride samples with dislocations introduced by nanoindentation. The spectral position of the dislocation-related luminescence doublet experiences a redshift with increasing distance from the indentation site. It follows the spectral shift of the excitonic near-bandgap emission, associated with stress relaxation. The luminescence of the intersection points exhibits a similar tendency. At certain local positions, its doublet fine structure is observed, which has a spectral linewidth of the order of or even less than that of the exciton. In this case, the spectral splitting between components of the doublet varies irregularly depending on the position of the exciton (i.e., on the mechanical stress). We see a clear indication of quantum dot-like emission. The fine structure of the luminescence of the intersection points can be easily explained by the energy dependence of emission on their size, as well as on their density, in particular, by the formation of paired nodes, which were previously observed in experiments in a transmission electron microscope.

1. Introduction

Grown-in dislocations in gallium nitride (GaN) are one of the major concerns in commercial applications, which are believed to limit lifespan and efficiency of GaN-based devices [1]. On the other hand, recent studies of recombination properties of a-screw dislocations freshly introduced by a local plastic deformation in wurzite-type GaN have shown an intense luminescence in ultraviolet region. Albrecht et al. [2] observed in semi-insulating GaN:Fe samples a single, spectrally sharp emission line accompanied by bright, straight-line contrast in cathodoluminescence images, and studied the core structure of dislocations with transmission electron microscopy. They interpret the emission as a radiative recombination of an exciton bound at the perfect core of an a-screw dislocation due to strain-induced bending of the valence and conduction bands. The energy of the dislocation-bound exciton emission is 3.346 eV at T = 6 K and the peak vanishes above 100 K.
In contrast, in low-ohmic n-GaN, dislocation-related luminescence (DRL) due to a-screw dislocations persists at room temperature and exhibits a spectral doublet structure at 3.15-3.18 eV at T = 70 K [3]. Dislocations dissociate into partials with the formation of an I2 stacking fault between them. The presence of a stacking fault in the dislocation core is assumed to be the reason for the red shift of DRL in n-GaN with respect to the semi-insulating one, since stacking faults are thought to form quantum wells in wurtzite-type GaN [4]. Two spectral components of DRL have different temperature behavior [3] and the direction of light emission polarization [5]. DRL is attributed to optical transitions in the cores of Ga and N partials.

Besides, luminous spot-like contrasts are found in the cathodoluminescence images at the intersections of crossing a-screw dislocations [6]. They exhibit an emission peak at ~3.3 eV that is called intersection-related luminescence (IRL). Its spectral position is close to the emission peak of I2 stacking faults, and transmission electron microscopy revealed the formation of the extended triangular nodes containing stacking faults of various sizes in the range of tens of nanometers [7,8]. Accordingly, IRL is interpreted as the luminescence from quantum-dot-like features, although a detailed understanding of the IRL mechanism is still lacking due to the absence of information on its detailed properties.

One of the important properties of any spectral line is its behavior under external mechanical stress with respect to the band gap. Such a dependence can be obtained from measurements of the local spectral cathodoluminescence at positions differently distant from the place of local deformation (indentation or scratching). Previous experiments revealed that the DRL spectral position closely follows the free exciton position, which thus confirms its excitonic character [3].

Here we report for the first time about such a dependence for IRL that is obtained on a dislocation network introduced by specially designed local plastic deformation, which creates an array of dislocation nodes in a wide range of distances from the indenter. We found that in addition to the red shift of the IRL band with distance due to stress relaxation, a clear spectral fine structure is observed. The presence of dislocation nodes of various shapes and sizes is assumed to be the origin of the fine structure.

2. Materials and methods
Thick (~10 μm) low-ohmic, specially undoped wurtzite-type n-GaN layers grown by halide vapor phase epitaxy on a sapphire substrate were used for this study. Their basal plane (0001) was indented with a Nano Indenter G200 using a Berkovich tip. The facets of the indenter were oriented perpendicular to the dislocation propagation directions <1̅2̅10>, introducing dislocation arms with three intersecting families. Cathodoluminescence studies of the structures were carried out using a JEOL JSM 6400 scanning electron microscope with a custom cathodoluminescence system [9]. All experiments were conducted at an accelerating voltage of 5 kV and at a temperature of 6 K.

3. Results and Discussion
Figure 1(a), (b) depicts a secondary electron (SE) image together with a simultaneously recorded panchromatic cathodoluminescence (CL) map at T = 6K near the indentation prick highlighted by a triangle. The dislocation rosette (figure 1 (b)) consists of three arms of networks of a-screw dislocations [3] propagating in <1̅2̅10> directions from nanoindentation borders for approximately 10 μm. They have dark linear contrast in the panchromatic CL map, which corresponds to reduced near-band-edge (NBE) emission. Other black dots are most likely due to threading dislocations.

The spatially averaged spectrum (figure 1(c)) shows dominant NBE emission with a sharp emission peak at 356 nm, which coincides well with the donor-bound exciton in strain-free GaN [10]. It has a pronounced LO phonon replica at ~364.5 nm.

The wide band between 384 nm and 397 nm (3.13 eV – 3.23 eV) is due to the luminescence of straight dislocation segments (DRL), as it can be seen from their bright linear contrast in monochromatic CL maps in figure 2(a)-(c). A less pronounced luminescence band between 370 nm and 380 nm (3.27 eV – 3.36 eV) arises from bright spots (figure 3(a)) and is therefore referred as IRL.
Figure 1. (a), (b) – secondary electron (SE) image and panchromatic cathodoluminescence (CL) map of the indentation prick (shown by a triangle); (c) – the spatially averaged spectrum (NBE is the near-band-edge, IRL is the intersection-related luminescence, and DRL is the dislocation-related luminescence) at T = 6 K.

Figure 2(a)-(c) represents the cathodoluminescence maps taken at three different wavelengths (shown in the legend) within the spectral region which is characteristic for the DRL band in figure 1(c). The DRL peak shifts towards longer wavelengths with distance from the indentation margin, which explains the broadening of the DRL band in the spatially integrated spectrum (figure 1(c)). To quantify this red shift, a linescan along one of the dislocation rosette arms was performed through its full length and width. Figure 2(d) represents the series of local CL spectra acquired from the indentation border to the end of the dislocation branch. The DRL center gradually changes its spectral position starting from 387 nm (3.21 eV) near the indentation prick to 392 nm (3.17 eV) at the end of the linescan. In addition, the NBE band spectrum position moves parallel to DRL, which indicates their similar dependence on the mechanical stress decreasing with distance from the indentation site. This result is in agreement with those previously obtained for dislocations introduced by scratching [3].

The IRL intensity position discretely varies with distance (figure 2(d)), and its central spectral position migrates from 372.5 nm (3.33 eV) to 374.5 nm (3.32 eV), which is also quite similar to the behavior of the NBE band.

Figure 2. (a)-(c) – cathodoluminescence (CL) maps of the same area as in figure 1(a) and 1(b) at wavelengths specified at the top; (d) - CL linescan of one of the dislocation branches: evolution of emission from the indentation prick to the top.
One of the dislocation branches is investigated in detail using CL (figure 3(a)) at higher magnification, and local spectra are depicted at three characteristic spots (which are marked in figure 3(a) as (b), (c), (d)). All intense IRL spots in the area between points “b” and “c” in the image have single sharp emission lines with spectral positions varying between 371 nm and 372 nm (figure 3(b)). At the edge of the dislocation branch, the IRL spots have distinct spectral features: emission with a poorly resolved doublet structure (figure 3(d)), which is the main peak around 374 - 375 nm with a shoulder at 372.5 - 373.5 nm. The doublet structure is best pronounced at the spot “c” where the spectrum shown in figure 3(c) was acquired. The spectral width of the both doublet components at 372 - 375 nm and 374 - 375 nm is comparable or even less than that of the NBE peak.

Figure 3. (a) – CL intensity image at wavelengths of the IRL: 370 – 377 nm for the dislocation branch near the prick in figures 1(a) and (b) where “b”, “c”, “d” indicate the positions where the local spectra in figures 3(b), (c) and (d) are recorded respectively; (e) – the difference between the NBE and IRL emission energies, plotted versus the NBE energy; the rectangles show the points with both spectral components; the numbers indicate the distance to the indentation margin; IRL-1 and IRL-2 represent higher and lower energy peaks, respectively.

The latter value found in the SEM CL spot mode contradicts the data on the line width, obtained in the CL linescan (figure 2(d)), where it is noticeably wider than NBE. In addition, the energy separation of the IRL peaks from NBE (figure 3(e)) lacks any pattern, and geometrical dimensions of the IRL spots (figure 3(a)) also have volatile variation over the sample, being significantly larger than the diffusion length of the minority carriers (about 200 nm). These facts cannot be explained only by a decrease in stress with distance from the indentation site. A more plausible explanation is that IRL spectral position is determined by the shape and dimensions of its source. The thickness of the staking fault $I_2$ in the extended dislocation nodes is about one nanometer, while the lateral sizes are tens of nanometers, which satisfies the conditions of quantum confinement of electron-hole pairs and, therefore, IRL can be considered as the luminescence influenced by quantum size effects. Due to the small size of the extended
dislocation nodes, some of them can fall into the region of generation of electron-hole pairs and each of them has its own narrow characteristic spectral line depending on its size.

4. Conclusion
Scanning electron microscopy-cathodoluminescence studies conducted on a dislocation network created by nanoindentation allowed us to examine the optical properties of both dislocations and their intersection points over a sufficient range of external mechanical stress. The CL spectrum of straight a-screw dislocations (DRL) in the range from 384 nm to 397 nm (3.13 eV – 3.23 eV) arises from the steady red-shift due to a decrease in the strain with distance from the indentation site and follows the near-band edge emission. This result is in complete agreement with those obtained earlier for dislocations created by scratching [3].

For the first time, the spectral properties of a-screw dislocation intersections (IRL) were examined in detail. The IRL band as a whole exhibits a spectral shift between 370 nm and 380 nm (3.27 eV - 3.36 eV) similar to the DRL and NBE bands. In addition, the spectral fine structure of IRL is found in many intense spots with a characteristic spectral fine structure (doublet of narrow spectral lines with the variable component separation up to 25 meV). Based on the previous results of structural studies of dislocation nodes [6–8], the fine spectral structure of IRL is interpreted as due to their different geometrical properties influenced by the quantum size effect.

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