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Yellow and green luminescence in a freestanding GaN template

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DEFECTS play an important role with regard to the optical and electrical properties of semiconductors with implications for the devices fabricated in them. Despite intense use of gallium nitride (GaN) in light-emitting and high-power/high-temperature electronic devices, the origin of many of the point defects in undoped (unintentionally doped) GaN is not yet clearly established. Therefore, the omnipresent defect-related band at about 2.2–2.3 eV, commonly referred as the yellow luminescence (YL) band, continues to receive extensive attention. There seems to be agreement that transitions from the conduction band or a shallow donor to a deep acceptor are responsible for this band. 1–4 Yet, deep-donor-to-deep levels for the isolated Ga, and the other being the same defect, but bound to a structural defect.

The freestanding GaN template studied was grown by HVPE on a c-plane sapphire substrate and separated from the substrate by laser lift-off. The GaN wafer was mechanically polished from both sides and the Ga face was dry etched. The final thickness of the template was about 200 μm. A defect delineating etch demonstrated very low density of dislocations in this sample: 5 × 10^5 and 1 × 10^7 cm^-2 on the Ga and N faces, respectively. 10 Photoluminescence was excited either with an above-gap He–Cd laser (3.81 eV) or a below-gap second harmonic of a Ti–sapphire ‘‘Tsunami’’ laser (in the range of 3.1–3.4 eV). The excitation intensity varied between 10^-4 and 100 W/cm^2. A closed-cycle cryostat was used for measurements from room temperature down to 10 K.

The low-temperature PL spectrum exhibited sharp peaks in the excitonic region with the highest peak at 3.471 eV (1 meV wide), attributed to exciton bound to neutral shallow donor. 11 The excitonic spectrum of the as-received N face was nearly featureless. However, after etching the N face in hot H_3PO_4 for a very short time, to remove the mechanical polish-induced damage, the spectrum approached that of the Ga face. We attributed the broadening of the spectrum from the N face before etching to the surface damage caused by mechanical polishing. 10,17

Interestingly, the shape and position of the defect-related broad band (1.5–2.8 eV) from Ga and N faces are different. The difference was obvious even to the naked eye in that the Ga-face emission was green (maximum at about 2.4 eV), whereas the N-face emission was yellow (maximum at about 2.2 eV) for an excitation density P_{exc} of about 0.1 W/cm^2. Note that the spectrum from the N face after etching became identical to that from the Ga face, including the shape and position of the defect-related band. At a first glance it would mean that the yellow luminescence is related to the structural

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defects near the surface, while the green luminescence (GL) is related to defects in the bulk of the material, on which we will elaborate below.

With increasing excitation intensity, we observed that the position of the defect-related PL band shifted from about 2.15 to 2.43 eV (Fig. 1). However, the excitation energy range where this shift occurred was different for the two faces of the sample (Fig. 2). Specifically, the transformation of the band position and shape took place in the range of $\sim 10^3 - 10^{-1}$ W/cm$^2$ for the Ga face and $\sim 10^{-2} - 1$ W/cm$^2$ for the N face. At lower- and higher-excitation densities, the position and shape of the band did not change (Fig. 2). A large shift of the PL band with excitation intensity is typical for deep-donor–acceptor pairs or in a semiconductor with large potential fluctuations. However, this rationale can be ruled out for our sample, which has a room-temperature electron mobility of 1200 cm$^2$ V$^{-1}$ s$^{-1}$ and an acceptor concentration of about $2.5 \times 10^{15}$ cm$^{-3}$, representing the best figures to date in GaN. Moreover, the possible concentration of deep donors is much less than $1 \times 10^{16}$ cm$^{-3}$. Such a low concentration of deep defects cannot account for the observed intense PL due to the extremely small overlap of the corresponding wave functions. Furthermore, our measurements of transient PL demonstrate that the room-temperature PL decay is exponential, which suggests that the transitions from the conduction band to a deep acceptor level are involved. Instead, we suggest that the shift with excitation intensity is due to a competition between two deep acceptors. At low-excitation intensities, the YL-related defect dominates the PL spectrum, and at high-excitation intensities the YL saturates, giving way to the GL band. To check the validity of this assumption, resonant (below-band-gap) PL excitation was undertaken at room temperature.

Figure 3 shows the defect PL spectrum for different excitation energies. With increasing excitation energy, the spectrum gradually blueshifts from about 2.2 eV (yellow emission) to 2.43 eV (green emission) on both N and Ga faces. In Fig. 4, we show the variation of the position of the band maximum and full width at half maximum (FWHM) as a function of the photon excitation energy, $\hbar \omega_{\text{exc}}$, along with the transmittance of the sample. The transmittance was calculated as the ratio of the transmitted and incident light intensities while taking the surface reflection into account. The variation of the shape and position of the band strongly correlates with the transmittance curve. For $\hbar \omega_{\text{exc}} < 3.26$ eV, where the absorption coefficient is of the order of or less than $10^2$ cm$^{-1}$, the position of the band maximum saturates at about 2.2 eV and the band FWHM is about 590 meV. For $\hbar \omega_{\text{exc}} > 3.35$ eV, where the absorption coefficient is more than $10^3$ cm$^{-1}$, the position of the band maximum saturates at about 2.43 eV and the band FWHM is about 500 meV. For $\hbar \omega_{\text{exc}} < 3.26$ eV, only a small fraction of the excitation light is absorbed in the sample, while for $\hbar \omega_{\text{exc}} > 3.35$ eV, the excitation light is absorbed in a thin (of the order or less than 10 μm) surface layer.

Based on the above observations, we suggest that the YL-related defect is of relatively high concentration in the first 1 μm from the surface, whereas the GL-related defect is distributed uniformly throughout the bulk GaN. As for the predominance of the YL at excitation photon energies below 3.3 eV, we suggest that the optical cross section for the YL-
related defect is much larger than that for GL. It is possible that in the excitation energy range of $\hbar \omega_{\text{exc}} = 3.3 - 3.43$ eV the excitation is not resonant but interband, phonon assisted. In this case, the nonradiative cross section of deep defects dictates the defect-related emission and GL may dominate the excitation. Our cumulative observations are consistent with the notion that two deep defects contribute to these bands. The defect causing the YL band appears mostly surface oriented, mainly on the N face, and can be easily saturated, increasing the excitation intensity. The defect causing the GL band is uniformly distributed in bulk GaN. We attribute the GL and YL to isolated native defects ($V_{\text{Ga}}$ or $V_{\text{Ga}}^{\text{donor complex}}$) and to the same defect bound to structural imperfection, respectively.

In conclusion, we have studied the yellow and green photoluminescence bands in a high-purity freestanding GaN template. The behavior of the PL spectrum at different excitation energies and intensities is consistent with the notion that two deep defects contribute to these bands. The defect causing the YL band appears mostly surface oriented, mainly on the N face, and can be easily saturated, increasing the excitation intensity. The defect causing the GL band is uniformly distributed in bulk GaN. We attribute the GL and YL to isolated native defects ($V_{\text{Ga}}$ or $V_{\text{Ga}}^{\text{donor complex}}$) and to the same defect bound to structural imperfection, respectively.

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13. The upper limit of the deep donor concentration has been estimated from accounting for the electron scattering on neutral impurities.