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Yellow and green luminescence in a freestanding GaN template

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We have studied a broad photoluminescence band in high-mobility freestanding 200-μm-thick GaN template prepared by hydride vapor-phase epitaxy. Variable-excitation intensity and energy experiments showed two defect-related bands: a yellow luminescence (YL) band at about 2.15 eV and a green luminescence (GL) band at about 2.43 eV. In contrast to epitaxial GaN samples prepared by both vapor-phase and molecular-beam epitaxy, the YL in the sample studied is weak and can be easily saturated. However, the GL is dominant. We attribute the GL to isolated defects involving gallium vacancies and the YL to the same defect, but bound to dislocations, or possibly to structural surface defects. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1371961]

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-shallow-acceptor transitions were also used to explain the results of magnetic-resonance experiments.[5] The issue of whether the YL is related to a point defect or to a distribution of states in the gap is still an open question. Shalish *et al.*[6] invoked a broad distribution of acceptor-like surface states to account for the YL band. A redshift of the YL band with decreasing energy of the below-gap excitation may also indicate that the broadening of the YL band is due to emission from several closely spaced traps.[7] On the other hand, the temperature dependence of the bandwidth and the photoluminescence (PL) excitation spectrum has been quantitatively explained by using a configuration-coordinate model that attributes this YL band to a point defect with strong electron–phonon interaction.[1,8] On the structural side, the YL band has been repeatedly attributed to gallium vacancies (V_{Ga}).[1–4] Theoretical calculations predict a low formation energy and deep levels for the isolated V_{Ga} and its complexes with shallow donors such as Si and O ($V_{Ga}Si_{Ga}$ and $V_{Ga}O_N$).[3,4] It has also been demonstrated that the formation of these defects is much more favorable at the threading-edge dislocations.[9] In this letter, we report on our study of the YL band in a high-purity freestanding template grown by hydride vapor-phase epitaxy (HVPE). What is unique is that the position of this broad band shifted from 2.15 eV (yellow emission) to

2.43 eV (green emission) depending on experimental conditions. We account for our results on the basis that two point defects are present, one in the form of an isolated defect involving V_{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.[10,11]

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

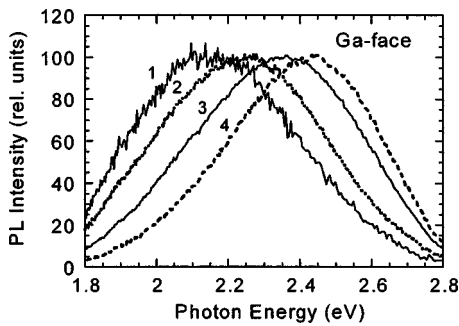[a]Author to whom correspondence should be addressed; electronic mail: hmorkoc@vcu.edu

FIG. 1. Defect-related PL spectrum from the Ga face of the GaN template for an excitation energy of 3.81 eV at different excitation intensities [3 $\times 10^{-4}$ W/cm^2 (1); 3×10^{-3} W/cm^2 (2); 3×10^{-2} W/cm^2 (3); 3 W/cm^2 (4)]. The PL intensity is normalized to the value at the maximum.

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.[12] 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 2.5×10^{15} cm^{-3}, representing the best figures to date in GaN.[11] Moreover, the possible concentration of deep donors is much less than 1×10^{16} cm^{-3}.[13] 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.[14] 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.[15] 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 va-
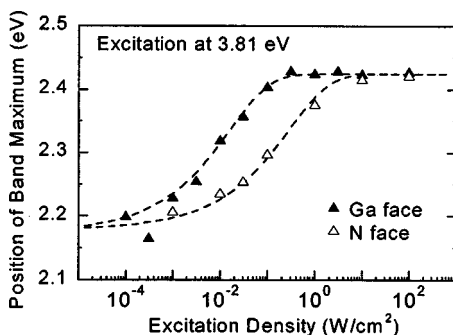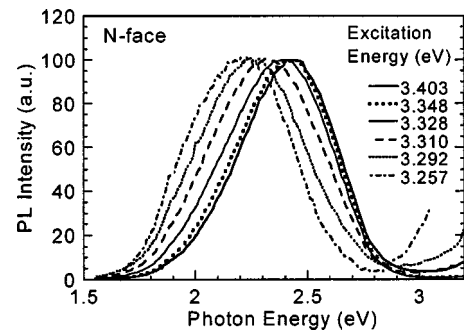

FIG. 3. PL spectrum in the defect range taken for below-band excitation at different photon energies. The excitation density is about 0.2 W/cm^2.

lidity 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, $\hbar\omega_{exc}$, 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 $\hbar\omega_{exc} < 3.26$ eV, where the absorption coefficient is of the order of or less than 10 cm^{-1}, the position of the band maximum saturates at about 2.2 eV and the band FWHM is about 590 meV. For $\hbar\omega_{exc} > 3.35$ eV, where the absorption coefficient is more than $\sim 10^3$ cm^{-1}, the position of the band maximum saturates at about 2.43 eV and the band FWHM is about 500 meV. For $\hbar\omega_{exc} < 3.26$ eV, only a small fraction of the excitation light is absorbed in the sample, while for $\hbar\omega_{exc} > 3.35$ 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-

FIG. 2. Position of the band maximum as a function of the excitation intensity for two faces of the sample. Dashed lines are guides to the eye.
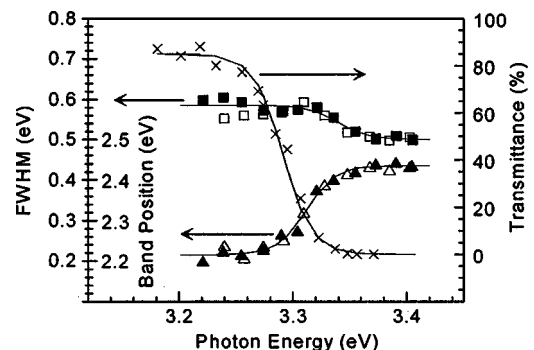

FIG. 4. Position of the band maximum, band FWHM, and transmittance of the 200-μm-thick sample as a function of the incident light energy. The excitation density is about 0.2 W/cm^2. Full squares and triangles are for the Ga face, open squares and triangles are for the N face. Solid lines are guides to the eye.
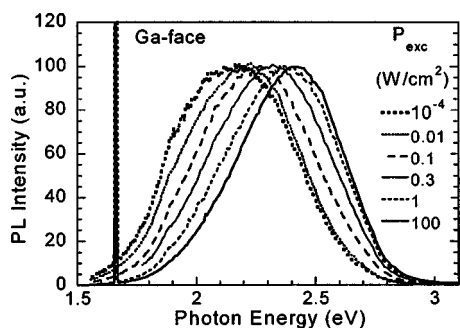
FIG. 5. PL spectrum excited with 3.32 eV photons at different excitation densities (P_{exc}). The sharp peak at 1.66 eV is the second order of the excitation light coming from the grating.

related defect is much larger than that for GL. It is possible that in the excitation energy range of $\hbar\omega_{exc} = 3.3-3.43$ 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 due to its larger concentration and/or larger nonradiative capture cross section. In the intermediate region ($\hbar\omega_{exc} = 3.25-3.35$ eV), both mechanisms, resonant and nonradiative excitations, compete.

Figure 5 shows a transformation of YL to GL in the transition region ($\hbar\omega_{exc} = 3.32$ eV), which is attributed to a saturation of the YL-related defect with increasing excitation intensity. The excitation power required for this saturation decreased as the excitation photon energy was increased. This is expected since for lower photon energies the transparency is higher and fewer defects resonantly capture photons. The converse is true for a higher photon energy excitation. Our cumulative observations are consistent with the argument that the YL defect is dominant near the surface, which is also in agreement with the results of Shalish *et al.*[6] Further, the concentration of the YL defect on the N face is some ten times larger than that on the Ga face since the excitation intensity for saturation on this face is some ten times smaller (Fig. 2). However, this is not to say that the YL-related defect is not present at all throughout the entire sample.

Unlike the YL band, the GL band showed almost no saturation for below-the-gap excitation in the intensity range used, which implies that the concentration of this deep acceptor is relatively high. We believe that this band is related to an isolated native defect (isolated V_{Ga} or isolated complex involving V_{Ga}), whereas the YL is related to the same point defect bound to some structural defect.

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_{Ga} or V_{Ga}–donor complex) and to the same defect bound to structural imperfection, respectively.

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