Dependence of radiative efficiency and deep level defect incorporation on threading dislocation density for InGaN/GaN light emitting diodes

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The influence of threading dislocation (TD) density on electroluminescence and deep level defect incorporation in the multi-quantum well regions of InGaN/GaN light emitting diodes (LEDs) was investigated. LED efficiency increased with decreasing TD density. To elucidate the impact of TD density on deep level defect incorporation and resulting radiative efficiency, deep level optical spectroscopy and lighted capacitance voltage measurements were applied to the LEDs. Interestingly, the concentration of all observed deep levels decreased with TD density reduction, but their concentration also varied strongly with depth in the multi-quantum well region. These trends indicate that (1) TDs strongly influence point defect incorporation in InGaN/GaN LEDs and (2) TDs, possibly in conjunction with point defects, are detrimental to LED efficiency. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4759003]

Defects reduce InGaN/GaN light emitting diode (LED) external quantum efficiency (EQE). Threading dislocations (TDs) are a major defect source in InGaN/GaN LEDs because they are often grown heteroepitaxially on highly lattice-mismatched substrates. Previous work demonstrated increased internal quantum efficiency (IQE) and decreased non-radiative recombination coefficient (A) of $In_{0.13}Ga_{0.87}N$/GaN LEDs for decreasing TD density, indicating that TDs are detrimental to LED radiative efficiency.[1] Threading dislocations have also been suggested as a source for LED efficiency roll-off with increasing carrier density.[2] The deleterious role of TDs in InGaN/GaN LEDs is consistent with the observation that TD cores in GaN form deep levels that are non-radiative recombination centers,[3] though it has also been argued that TD cores within InGaN/GaN multi-quantum wells (MQWs) are innocuous when decorated by so-called "V-defects."[4] Therefore, attaining a better understanding of deep levels formed by TDs in the MQW region can help to elucidate their impact on InGaN/GaN LED performance.

We applied deep level optical spectroscopy (DLOS) and lighted capacitance-voltage[5] (LCV) measurements to $In_{0.13}Ga_{0.87}N$/GaN LEDs to quantitatively survey deep level defect incorporation in the MQW region as a function of TD density. DLOS and LCV were combined with electroluminescence (EL) characterization to understand how TD density mediates InGaN/GaN LED radiative efficiency through deep level defect formation. It was observed that LED radiative efficiency increased with reduced TD density, but varying TD density had no impact on the type of deep levels that incorporated in the MQW region. However, the deep-level defect density (N_t) decreased universally with reduced TD density, and the depth distribution of deep levels within the MQW region suggests a strong interaction between point defects and TDs during growth.

InGaN/GaN MQW LEDs were grown with similar structures and under similar growth conditions on GaN-on-sapphire templates by metal-organic vapor-phase epitaxy (MOVPE). The TD densities for the templates were controlled by varying nucleation-layer-growth and film-coalescence parameters,[6] yielding TD densities of 5.3 and $27 \times 10^8\,cm^{-2}$. TD density was evaluated using x-ray diffraction (XRD) rocking curve measurements of the Bragg peak widths of the (0004) and (10$-$11) reflections of GaN. Threading dislocation densities were extracted from the measured peak widths using analyses described in Ref. 7. For the edge- and mixed-type Burgers vectors expected to predominate for the TDs in these templates, previous work found that such XRD measurements of TD density agree with transmission electron microscopy within a standard error of $\sim$33%.[7]

LEDs had an n-type GaN layer grown at 1050 °C, followed by a MQW region consisting of five unintentionally doped (UID) 2.5-nm-thick $In_{0.13}Ga_{0.87}N$ QWs grown at 770 °C placed between 7.5-nm-thick Si-doped ($1 \times 10^{18}\,cm^{-3}$) GaN quantum-well barriers (QBs) grown at 850 °C, followed by a 30-nm-thick Mg-doped ($3 \times 10^{19}\,cm^{-3}$) $Al_{0.15}Ga_{0.85}N$ electron-block layer (EBL) and a 400-nm-thick p-type GaN contact layer. The 30× higher p-type EBL doping relative to n-type QW doping ensures an asymmetric depletion. Devices were patterned into 300 μm × 300 μm sized mesas using inductively coupled plasma etching. Ohmic n-type and p-type electrical contacts were formed by evaporating a Ti/Al/Ni/Au and a transparent NiO/Au metal stack, respectively.

Figure 1 compares the LED EQE versus TD density. Light-output power versus current measurements were performed on-wafer using a calibrated Si photodiode underneath the sapphire substrate of the LED wafer using pulsed current operation with 1 μs pulses and a 1% duty cycle. Peak LED efficiency increased by $\sim$50% with $\sim$5× reduced TD density, in accordance with the results of a previous study of LEDs with the same structure and a similar range of TD density.[6] Increased EQE with lower TD density for otherwise similar LEDs points to the deleterious impact of defects on MQW IQE or carrier injection efficiency, or possibly both. DLOS was used to assess how TDs meditate LED EQE through deep level formation.

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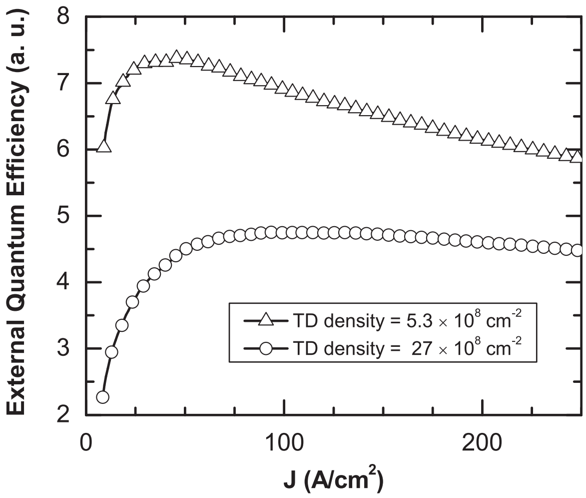

FIG. 1. External quantum efficiency of LEDs versus TD density exhibiting improved LED performance with reduced TD density.

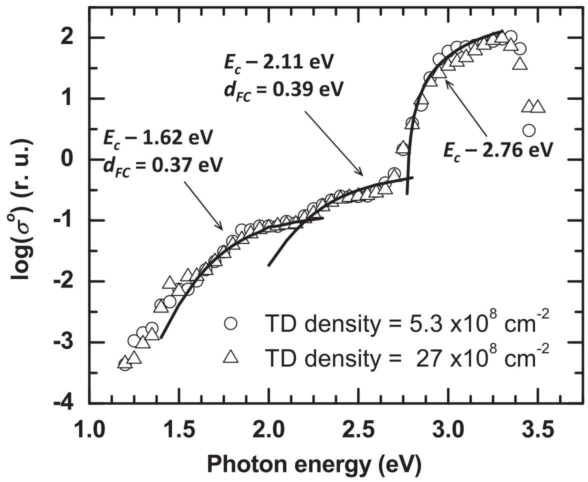

FIG. 2. DLOS spectra for LEDs with similar epitaxial structure and differing TD density. Their spectral similarity indicates that the same type of deep level defects incorporate in the MQW region despite the difference in TD density.

DLOS determines the energy level of a defect state from the photocapacitance response due to deep level photoemission upon exposure to sub-band gap, monochromatic illumination. DLOS measurements were conducted at room temperature at 1 MHz using broadband illumination from a 150 W Xe arc lamp dispersed through a monochromator using appropriate mode sorting filters to provide a photon energy range of 1.20–3.60 eV. The photon flux (ϕ) varied between 1 and $20 \times 10^{16}\,\mathrm{cm^{-2}s^{-1}}$. DLOS determines the spectral variation of the deep level optical cross-section (σ_o) from the time derivative of the photocapacitance transient at $t = 0$ s, the time when illumination begins, divided by ϕ. Fitting σ_o to an appropriate model[8] that accounts for lattice relaxation of the deep level defect determined the optical ionization energy (E_o) and the Franck-Condon energy (d_{FC}). DLOS measurements were conducted at reverse bias (V_r), followed by a 2 V filling pulse applied in the dark for 20 s to re-populate deep levels in the MQW region.

Figure 2 presents the DLOS spectra of the LEDs measured at $V_r = 1.7$ V. This value of V_r was chosen to ensure that the depletion region and the resultant DLOS sensitivity were strictly limited to the MQW region. Space-charge density (ρ) measured from capacitance-voltage (CV) in the dark, shown in Fig. 3(a), determined a depletion depth (x_d) of 57 nm at $V_r = 1.7$ V, indicating a space-charge region bounded by the edge of the heavily *p*-doped EBL on one end and the QB closest to the *n*-GaN bulk on the other. Thus, only defects located in the QWs or QBs appeared in the spectra of Fig. 2 because DLOS measures variations in space-charge caused by defect photoemission.[9] The symbols are experimental data, and the lines in the spectra are least-squares fits to the model of Pässler.[8] The E_o values are referenced to E_c because the associated ΔC values were positive.

The DLOS spectrum of the LED with TD density of $5.3 \times 10^8\,\mathrm{cm^{-2}}$ was discussed in detail in a previous study.[9] It was determined from V_r-dependent DLOS measurements that the E_c – 1.62 eV and the E_c – 2.76 eV deep level defects are associated with the $In_{0.13}Ga_{0.87}N$ QWs, in agreement with another DLOS study of $In_{0.2}Ga_{0.8}N$ thin films that exhibited qualitatively similar σ^o.[10] The E_c – 2.11 eV deep level defect was identified with the GaN QBs, also using V_r-dependent DLOS measurements. While resolving InGaN- versus GaN-related deep level defects in the MQW region helps to assess their physical origin, the detailed atomic structure of these deep levels are yet unclear and are the subject of future studies. The broad absorption spectra and concomitantly large d_{FC} values for the E_c – 1.62 eV and E_c – 2.11 eV deep states arise because the corresponding defect centers are strongly coupled to the lattice, enabling

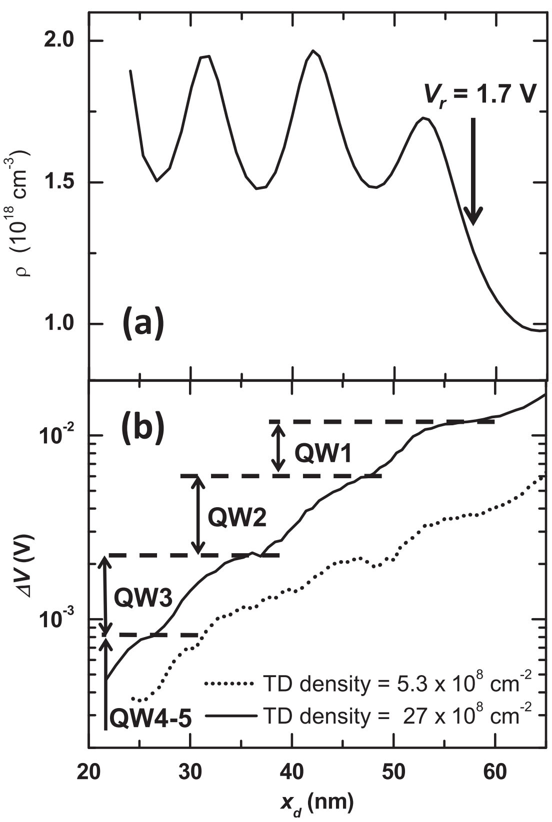

FIG. 3. (a) Space-charge density in the MQW region measured by CV in the dark. (b) Measured voltage shift versus apparent depletion depth for the QW-related E_c – 2.76 eV deep level. Lower ΔV for the LED with lower TD density indicates reduced N_t.

significant phonon-assisted photoabsorption for photon energies much less than E_o.[8] Conversely, the $E_c - 2.76$ eV level with negligible d_{FC} acts as a hydrogenic state with rather sharp optical absorption. Regarding the influence of TD on defect incorporation, the DLOS spectra are nearly identical for both TD density values, indicating that the same types of deep level defects incorporated in the MQW region for both LEDs despite the variation in TD density (note that the magnitudes of the DLOS spectra do not indicate N_t). The observation that TD density did not affect the type of deep levels in the MQW region suggests that changes in N_t with TD density are the key to understanding deleterious impact of TDs on LED EQE.

LCV was used to establish a quantitative link among TD density, N_t, and EQE. Similar to typical capacitance-voltage profiling, LCV measures the increase in space-charge density due to deep level photoemission under monochromatic, sub-bandgap illumination. LCV was performed at 50 kHz, and the phase angle for LCV measurements remained between $-88°$ and $-90°$ under illumination. This confirmed reliable LCV measurements of a nearly ideal capacitive element. The depth distribution of N_t for individual defect states within the MQW region was calculated using LCV by measuring the difference in V (ΔV) required to achieve the same x_d (and therefore C) with deep levels fully occupied or empty of electrons.[11] The additional voltage ΔV required to reach x_d when defects are emptied by illumination compared to when the defects are fully occupied is

$$\Delta V = \frac{q}{\Delta}\int_0^{x_d} x N_t(x)dx, \qquad (1)$$

where q is the Coulomb charge and ε is the semiconductor permittivity. Using this method, $N_t(x)$ was calculated for the deep levels using Eq. (1) by performing sequential LCV scans at selected deep-level-dependent photon energies. For example, [E_c – 1.62 eV] (brackets indicate the density of the deep level defect) was measured from ΔV under 2.10 eV illumination relative to the dark CV. Referring to Fig. 2, at 2.10 eV, only the $E_c - 1.62$ eV level is optically stimulated. Likewise, [E_c – 2.11 eV] was calculated from ΔV under 2.60 eV illumination relative to 2.10 eV illumination, and [E_c – 2.76 eV] was calculated from ΔV under 2.85 eV relative to 2.60 eV illumination.

Figure 3(b) plots ΔV against x_d for the QW-related E_c – 2.76 eV deep level for the two LEDs. The positions of the four QWs nearest to the n-side are evident in the ρ profile of Fig. 3(a), so contributions to ΔV from the $E_c - 2.76$ eV defect state in the individual QWs can be discerned and are marked by horizontal lines. It is seen from Fig. 3(b) that ΔV accumulated at each QW is smaller for the LED with the lower TD density, indicating that N_t decreases with decreasing TD density. Figure 4 shows the distribution of [E_c – 2.76 eV] in the MQW region for both LEDs calculated with Eq. (1). The same analysis was applied to ΔV data for the remaining deep levels in the MQW region, and the results are also shown in Fig. 4.

Having obtained N_t for the MQW-related defect states, the dependence of N_t on TD density can now be examined. Figure 4 demonstrates a strong TD density-dependence of N_t for every deep level. The deep level populations evidence ~2-3× decrease for a ~5× reduction of TD density. These findings of mitigated deep level incorporation and improved EL with reduced TD density agree with earlier studies of LEDs of identical structure and similar TD character and range in density.[1] Indeed, this previous investigation using power-dependent resonant PL found that A decreased by ~3×, in good agreement with the 2–3× decrease in N_t observed by LCV.[1]

Interestingly, the influence of TDs on LED performance appears to be convolved with point defect interaction. In addition to a strong dependence on TD density, N_t for both LEDs exhibited a strong depth-dependence skewed toward the n-side of the MQW region. Possible causes for depth-dependent N_t in the MQW region were examined in a previous study,[9] but the focus of this letter is the implications of this depth-dependence for deep level association with TDs. The simultaneous trends of depth-dependence and TD density-dependence of N_t for the observed defect levels is intriguing because these observations at first appear to be at odds with each other. Reduced N_t with reduced TD density for each deep level suggests that all of the corresponding defects are related to TDs. However, the strong depth dependence of N_t is not consistent with a uniformly spaced and vertically arranged series of defect sites expected along a TD core.

These considerations can be reconciled by realizing that there are a variety of mechanisms by which extended defects can influence point defect and impurity incorporation. Previous

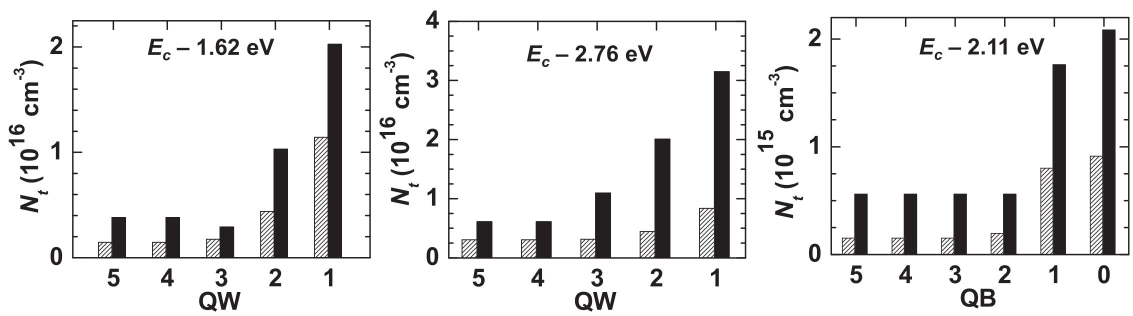

FIG. 4. Distribution of the deep level density in the MQW region for LEDs with similar epitaxial structure and differing TD density. QWs and QBs are enumerated starting from the n-side of the MQW region. The solid and cross-hatched bars correspond to the LED with TD density = 27×10^8 cm^{-2} and TD density = 5.3×10^8 cm^{-2}, respectively. The universal decrease of N_t with lower TD density suggests a TD-related source for the deep levels, while the strong depth dependence of N_t is indicative of a point defect source.

studies observed >30-fold reduction in the concentration of carbon impurities and a 3-fold reduction in oxygen contamination in *n*-GaN films when the TD density was reduced from $\sim 10^9\,\mathrm{cm}^{-2}$ to $\sim 4\times 10^6\,\mathrm{cm}^{-2}$.[12] Such dependence may arise because the formation energy of point defects and their impurity complexes can be reduced when incorporating near a TD core[13] and its surrounding strain field.[14] For example, Elsner *et al*. found that a gallium vacancy complexed with oxygen substituting on a N site (V_{Ga}-O_N) in the vicinity of an edge-type TD core produces energy levels similar to an isolated V_{Ga}-O_N but with reduced formation energy.[13] The influence of a TD on point defect incorporation can also extend well beyond the TD core due to altered morphology at the growth surface. Formation of V-defects at a threading edge dislocation core can occur at the onset of InGaN/GaN MQW growth and expose {10–11} facets,[4,15] and these new growth facets may incorporate point defects differently compared to the nominal (0001) growth surface. Given these considerations, N_t for what appears to be a distinct deep level in DLOS could actually have two separate contributions: a TD-related deep level with a depth-independent N_t that scales with TD density and an isolated point defect with a similar energy level that exhibits a depth-dependence of N_t that is insensitive to TD density. Thus, unambiguous attribution of deep levels to isolated point defects or TD cores is difficult in the context of these *c*-plane InGaN/GaN LEDs. Nonetheless, it is clear that the minimization of TD density is beneficial to LED performance.

In conclusion, the impact of TD density on InGaN/GaN LED defect incorporation in the MQW region and EQE was investigated. Reducing TD density improved LED efficiency but did not influence the type of deep levels incorporated in the MQW region. LCV analysis showed that N_t for every deep level defect decreased with lower TD density, suggesting that deep levels related to TDs cores reduce LED performance. However, N_t for these same deep levels were significantly higher on the *n*-side of the MQW region, suggestive of a point defect source. Interactions between point and extended defects during MQW growth could arise by various means such as the decoration of TD cores by point defects or the emergence of non-basal growth planes when a TD core opens during MQW growth. While it is not clear if the deep levels responsible for reduced EQE originate from TD cores, point defects, or interaction between extended and point defects, it is evident that the minimization of TD density is beneficial to LED performance.

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[1]Q. Dai, M. F. Schubert, M. H. Kim, J. K. Kim, E. F. Schubert, D. D. Koleske, M. H. Crawford, S. R. Lee, A. J. Fischer, G. Thaler, and M. A. Banas, Appl. Phys. Lett. **94**, 111109 (2009).
[2]B. Monemar and B. E. Sernelius, Appl. Phys. Lett. **91**, 181103 (2007).
[3]J. S. Speck and S. J. Rosner, Phys. B: Cond. Matt. **273–274**, 24 (1999).
[4]F. Hitzel and A. Hangleiter, Int. J. Mod. Phys. B **22**, 3261 (2008).
[5]A. Armstrong, A. R. Arehart, and S. A. Ringel, J. Appl. Phys. **97**, 083529 (2005).
[6]M. F. Schubert, S. Chhajed, J. K. Kim, E. F. Schubert, D. D. Koleske, M. H. Crawford, S. R. Lee, A. J. Fischer, G. Thaler, and M. A. Banas, Appl. Phys. Lett. **91**, 231114 (2007).
[7]S. R. Lee, A. M. West, A. A. Allerman, K. E. Waldrip, D. M. Follstaedt, P. P. Provencio, D. D. Koleske, and C. R. Abernathy, Appl. Phys. Lett. **86**, 241904 (2005).
[8]R. Pässler, J. Appl. Phys. **96**, 715 (2004).
[9]A. Armstrong, T. A. Henry, D. D. Koleske, M. H. Crawford, and S. R. Lee, Opt. Express **20**, A812 (2012).
[10]E. Gür, Z. Zhang, S. Krishnamoorthy, S. Rajan, and S. A. Ringel, Appl. Phys. Lett. **99**, 092109 (2011).
[11]S. D. Brotherton, Solid-State Electron. **19**, 341 (1976).
[12]S. Hashimoto, Y. Yoshizumi, T. Tanabe, and M. Kiyama, J. Cryst. Growth **298**, 871 (2007).
[13]J. Elsner, R. Jones, M. I. Heggie, P. K. Sitch, M. Haugk, Th. Frauenheim, S. Öberg, and P. R. Briddon, Phys. Rev. B **58**, 12571 (1998).
[14]R. Chang, Acta Metall. **10**, 951 (1962).
[15]N. Sharma, P. Thomas, D. Tricker, and C. Humphreys, Appl. Phys. Lett. **77**, 1274 (2000).