

Luminescence Spectra Of Superbright Blue and Green InGaN/AlGaN/GaN Light-Emitting Diodes

K. G. Zolina, V. E. Kudryashov, A. N. Turkin, A. E. Yunovich
Moscow State Lomonosov University

Shuji Nakamura
Nichia Chemical Industries

This article was received on May 25, 1996 and accepted on September 20, 1996.

Abstract

Electroluminescence spectra of superbright blue and green LEDs based on epitaxial $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{Al}_y\text{Ga}_{1-y}\text{N}/\text{GaN}$ heterostructures with thin quantum well active layers [1] were studied at currents $J = 0.01\text{-}20$ mA. Spectral maxima of blue and green LEDs are $\omega_{\text{max}} = 2.58\text{-}2.75$ eV and $\omega_{\text{max}} = 2.38\text{-}2.45$ eV, dependent on the active layer In content. The low energy tails of the spectra are exponential with the parameter $E_0 = 42\text{-}50$ meV almost independent of the temperature. The high energy tails of the spectra are exponential with a temperature dependent parameter $E_1 = 20\text{-}40$ meV. Both parameters (E_0, E_1) are current independent at $J > 0.5$ mA. The spectral band can be described by taking into account quantum size effects, impurities and electron-phonon interactions in active layers. A structure in the spectra was detected which can be described by the influence of light interference in the GaN layer on the sapphire substrate. Light intensity was a linear function of the drive current over the interval $J = 1\text{-}20$ mA, and was slightly temperature dependent. In the blue LEDs, the efficiency fall off at low currents ($J < 0.7$ mA) had a $I \sim J^{4-5}$ dependence at room temperature. The green LEDs showed no such dependence. The influence of tunnel effects on the efficiency at low currents is discussed. Tunnel radiation spectra with maxima moving with the voltage were detected at low currents in III-N structures.

1. Introduction

The development of electroluminescent heterostructures based on GaN and III-N ternary compounds has been quite successful over the past three years ([2] and references therein). The best short wavelength (violet, blue, bluish-green) light-emitting diodes (LEDs) were MOVPE-grown InGaN/AlGaN/GaN multilayers with a thin (25-30 Å) $\text{In}_{1-x}\text{Ga}_x\text{N}$ active layer. External quantum efficiencies in the 4-9 % range have been achieved [1][2]. The electroluminescence (EL) spectra of InGaN/AlGaN/GaN heterostructures were studied in [1], and in the earlier publications of the same group [3][4][5]. In this paper, we report the spectra of the multilayered structures InGaN/AlGaN/GaN with a thin active InGaN layer at currents $J=0.01\text{-}20\text{mA}$ at temperatures $T=200\text{-}300\text{K}$.

A model of the radiative recombination in two dimensional (2D) structures which takes into account potential fluctuations [6] was previously applied to describe photoluminescence spectra of GaAs/AlGaAs multiple quantum wells [7], and is used here to analyze the spectra of GaN 2D LED structures. Optical interference can occur in multilayered structures and we have detected the influence of interference on the spectra of blue-green LEDs. In addition, the spectra of tunnel radiation were detected for the first time in GaN-based structures.

2. Experimental

We have studied LEDs made from structures described in [2]. A GaN buffer layer (≈ 300 Å) was grown on the sapphire substrate followed by an n-GaN:Si (≈ 5 μm) base and electron emitter layer (barrier for holes). The thin active layer is $\text{In}_{1-x}\text{Ga}_x\text{N}$ with $x = 0.2\text{-}0.49$ and thickness of this layer were varied to move the

layer $\text{In}_x\text{Ga}_{1-x}\text{N}$ was $\approx 20\text{-}30$ Å. The In content ($x = 0.2\text{-}0.45$) and thickness of this layer were varied to move the spectra from violet-blue to green. A p-type $\text{Al}_{0.1}\text{Ga}_{0.9}\text{N:Mg}$ (≈ 100 nm) layer was grown onto the active layer as a hole emitter (barrier for electrons) and the structure was completed by a top-GaN:Mg ($\approx 0.5\mu\text{m}$) cap. The ohmic contact metallizations were Ni-Au (p-type) and Ti-Al (n-type). The device had a mesa structure with an active area $S = 350 \times 350 \mu\text{m}^2$.

EL spectra were recorded on a KSVU-12 spectrometer connected to an x486 PC.

3. Experimental results

3.1. Spectra at room temperature

The luminescence spectra of some LEDs at the room temperature (RT) and $J=10$ mA are given in Figure 1. The violet-blue and blue LEDs have spectral maxima $\omega_{\text{max}} = 2.58\text{-}2.75$ eV, and the green LEDs in the $\omega_{\text{max}} = 2.38\text{-}2.45$ eV range dependent on the active layer In content ($x=0.20\text{-}0.25$ for the blue and $0.42\text{-}0.44$ for the green LEDs). The LEDs span the entire visible short wavelength spectrum [1][2]. The spectra of a blue diode at various currents are shown in Figure 2. The exponential decay on both sides of the peak are unchanged over the range of currents plotted ($J = 0.3\text{-}20$ mA). The long wavelength side of the peak is described by an exponent:

$$I(\omega) \sim \exp(\omega/E_0). \quad (1)$$

The parameter E_0 varied in different diodes between $E_0 = 42\text{-}50$ meV $> kT$, and was independent of temperature. The short wavelength side of the peak is described to first approximation by the formula:

$$I(\omega) \sim \exp(-\omega/E_1); \quad (2)$$

with a temperature dependent parameter equal to $E_1 = 31\text{-}34$ meV at RT.

3.2. Temperature dependence of spectra

The temperature dependence of spectra at $T = 200\text{-}300$ K for one of blue diodes is shown in Figure 3. The temperature, measured by a thermocouple attached to the plastic cap of the LED, was current dependent above 1mA as a result of Joule heating. Therefore measurements were taken at $J = 1$ mA, where such heating was negligible. The data are bunched roughly in two groups in this temperature interval. The high energy side of the spectra are changing according to (2), the parameter E_1 being approximately proportional to T .

3.3. Interference structure

A distinct structure on the spectra could be detected when measurements were done at a spectral resolution better than 1meV. This structure was better resolved when a Gaussian or hyperbolic fitting function is subtracted from experimental curves, or if the derivatives of spectra were analysed (see Figure 4). The structure is nearly periodical. This can be understood if the light interference is taken into account. The period of the spectral structure is of the order of several nm. If the interference is localized in a layer having thickness t and a refractive index n , then

$$2t n (1 + (\lambda/n)(dn/d\lambda)) = \lambda((1 + \lambda)/\lambda) \quad (3)$$

The n-GaN layer of the structure is $t = 5.0 \pm 0.5 \mu\text{m}$. The values of the refractive index n and its dispersion $(1 + (\lambda/n)(dn/d\lambda))$ calculated from (3) for different blue LEDs were 2.48, 2.73, and 3.03. These values correspond to the known refractive index of GaN $n = 2.5$ to within the limits of the above analysis.

3.4. Dependence of intensity versus current

The integrated radiation intensity of the spectrum was proportional to the intensity at the peak I_{max} and the full width at half-maximum $D(\omega)_{1/2}$. The $D(\omega)_{1/2}$ values for the blue ($D(\omega)_{1/2} = 0.12 - 0.13$ eV) and green ($D(\omega)_{1/2} = 0.15\text{-}0.16$ eV) LEDs were current independent. We therefore were able to determine the current dependence of integrated intensity by merely measuring I_{max} .

The current dependence $I_{\max}(J)$ was different for blue and green LEDs (see Figure 5 and Figure 6). The linear dependence $I_{\max}(J) \sim J$ is seen in the current range $1 \text{ mA} < J < 10 \text{ mA}$. For $J > 10 \text{ mA}$, Joule heating overrides the dependence. The intensity drops dramatically at currents below 1 mA in the blue diodes, following a $I_{\max}(J) \sim J^{4-5}$ dependence at RT (see also Figure 2). At lower temperature, the transition between the two regimes occurs at lower current levels ($J < 0.1 \text{ mA}$). The green diodes had no such temperature dependence (see Figure 6a).

The difference in efficiency corresponds to the different J-V characteristics of blue and green diodes (see Figure 5 and Figure 6). The low temperature J(V) of the blue diodes was exponential:

$$J(V) \sim \exp(-eV/E_J); \quad (4)$$

with $E_J = 130\text{-}140 \text{ meV}$ independent of temperature. This can be caused by a tunnel "excess" current which is common for thin p-n-junctions with high electric fields in the space charge region. At higher currents the curve flattens and can be described by a temperature dependent exponent. In this regime, series resistance R_s is limiting the current:

$$V = U + JR_s; \quad (5)$$

where U is the p-n-junction potential. The green diodes had higher series resistance and the J(V) curve had no tunnel component.

3.5. Tunnel radiative recombination

Tunneling effects in blue LEDs were detected also in the low current EL spectra (see Figure 7). A wide spectral band is seen on the low energy side of the spectra with its maximum moving nearly equal to the voltage $\omega_{\max} \approx V \approx U$. Analogous spectra were studied in highly doped GaAs, InP and GaSb narrow p-n junctions [8] [9]. It was shown that optical transitions between the tails in the density of states at the band gap edges are caused both by the high electric field at the junction and the fluctuating fields of charge impurities. Now we can conclude that such a situation occurs in GaN based structures characterized by narrow space charge regions and a thin 2D active layer.

4. Discussion

4.1. Energy diagram

Let us analyze the energy diagram of a multilayered structure at forward bias (see Figure 8). There are three main current components in the space charge regions: tunneling, injection and recombination, and only injection and recombination currents in the 2D active layer. The tunneling component can dominate at low currents in highly doped junctions with thin space charge regions. The larger part of the space charge corresponds to the $\text{p-Al}_{0.1}\text{Ga}_{0.9}\text{N:Mg}$ ($\sim 100 \text{ nm}$) layer. The space charge width is lower for blue diodes as determined by preliminary capacitance measurements which give values of 25 nm for the blue and 40 nm for green LEDs. We conclude that the difference between the two LED groups is differing concentrations of charge impurities in the p-regions. Tunneling dominates at low currents in blue LEDs. The main part of this component is non-radiative recombination, but another part is the tunnel radiative recombination. Injection and recombination in space charge p-region dominate in green LEDs at low currents. Effective radiative recombination is achieved when both carrier types are injected to the active QW layer. At high currents some of the voltage will drop across the adjacent layers, and some recombination will occur there. This produces the maximum of the quantum efficiency at a given current density. It means that the charge distribution in InGaN/AlGaIn/GaN heterostructures is very important for optimizing the efficiency.

4.2. A model approximation of the main spectral band

The effective energy gap of a doped QW can be estimated by taking into account the dependence of $E_g(x,T)$ for InGaN, shifts of QW levels for electrons and holes (ΔE_{1c} , ΔE_{1v}), shifts due to deformations ΔE_p , and shifts due to impurity band tails $\Delta E_{A,D}$:

$$E_g^{\text{eff}} = E_g(x,T) + \Delta E_{1c} + \Delta E_{1v} + \Delta E_p - \Delta E_{A,D}. \quad (6)$$

An estimation of $E_g(x,T)$ was made by taking data from a review [10]. The parameters $\Delta E_{1c} + \Delta E_{1v}$ were approximated

using standard formulae for levels in rectangular quantum wells [11] and approximate effective masses in InGaN. A model of joint 2D-density of states suggested in [6] and used in [7] to analyze spectra of GaAs/AlGaAs MQWs can be applied to fit the spectral band by the equations:

$$I(h\omega) \sim N^{2D}(h\omega, E_g^{\text{eff}}) f_c(h\omega, kT, F_n) (1 - f_v(h\omega, kT, F_p)); \quad (7)$$

$$N^{2D}(h\omega, E_g^{\text{eff}}) = (1 + \exp(-(h\omega - E_g^{\text{eff}})/E_0))^{-1}; \quad (8)$$

with $f_c(h\omega, kT, F_n)$ and $(1 - f_v(h\omega, kT, F_p))$ representing the Fermi functions for states near the effective band edges. One of the fits is shown in Figure 3. The fit is quite successful and the number of parameters is not so high. The parameter E_0 depends on the roughness of interfaces, strains, alloy inhomogenities and Coulombic impurity fields.

The exponential decay on the high energy side is described by the exponents

$$((m_c^*(h\omega - E_g^{\text{eff}})/(m_c^* + m_v^*)kT) \text{ and } (m_v^*(h\omega - E_g^{\text{eff}})/(m_c^* + m_v^*)kT).$$

We would like to discuss the parameters of this fit in the future.

5. Conclusions

The luminescent spectra of InGaN/InGaN/GaN heterostructures have maxima and exponential decays in the blue and green spectral regions which can be described by taking into account the impurity band tails in the 2D active layer. A model approximation for the spectra is suggested. A periodical structure was detected in the spectra which can be described by interference effects in the GaN base layer.

The dependence of intensity versus current differs for blue and green LEDs. The falloff of the blue LED efficiency at low currents is caused by a non-radiative tunnel component of the current. The tunnel radiative recombination spectra with the maxima moving with the voltage have been detected for the first time in GaN based heterostructures.

Acknowledgments

The authors thank A. N. Kovalev and F. I. Manyachin for measurements of electrical properties of the LEDs and A. E. Kovalev and S. S. Shumilov for help in computer problems.

References

- [1] S. Nakamura, M. Senoh, N. Iwasa, S. Nagahama, T. Yamada, T. Mukai, *Jpn. J. Appl. Phys.* **34**, L1332-L1335 (1995).
- [2] Materials Research Society Symposium Proceedings 395, (1995)
- [3] S. Nakamura, M. Senoh, N. Iwasa, S. Nagahama, *Jpn. J. Appl. Phys.* **34**, L797-L799 (1995).
- [4] S. Nakamura, T. Mukai, M. Senoh, *J. Appl. Phys.* **76**, 8189 (1994).
- [5] Shuji Nakamura, Takashi Mukai, Masayuki Senoh, *Appl. Phys. Lett.* **64**, 1687-1689 (1994).
- [6] R. Cingolani, W. Stolz, K. Ploog, *Phys. Rev. B* **40**, 2950 (1989).
- [7] B. Vardanyan, A. E. Yunovich, *Fiz. Tech. Polupr.* **29**, 1976 (1995).
- [8] A. E. Yunovich, A. B. Ormont, *Zh. Exp. Tech. Fiz.* **51**, 1292 (1966).
- [9] V. M. Stuchechnikov, A. E. Yunovich, *Fiz. Tech. Polupr.* **3**, 1293 (1969).

[10] H. Morkoc, S. Strite, G. B. Gao, M. E. Lin, B. Sverdlov, M. Burns, *J. Appl. Phys.* **76**, 1363-1398 (1994).

[11] M. Herman, "Semiconductor Superlattices", Acad. Verlag, Berlin, 1984

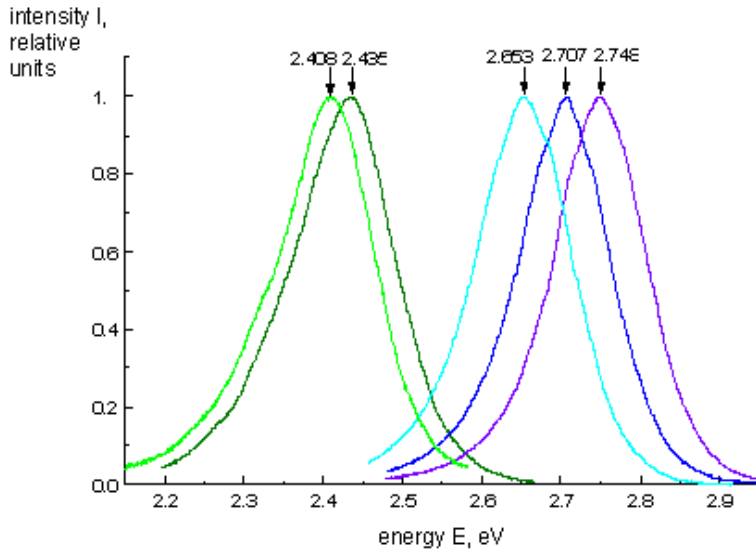


Figure 1. Room temperature spectra of blue and green InGaN/AlGaIn/GaN LEDs, $J=10$ mA (Arrows show E_{max}).

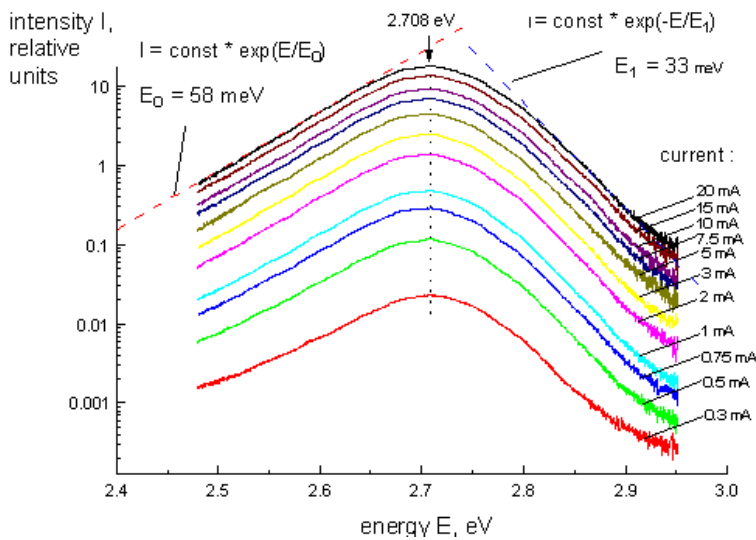


Figure 2. Room temperature spectra of blue InGaN/AlGaIn/GaN LED #3 over the current range $J = 0.3-20$ mA.

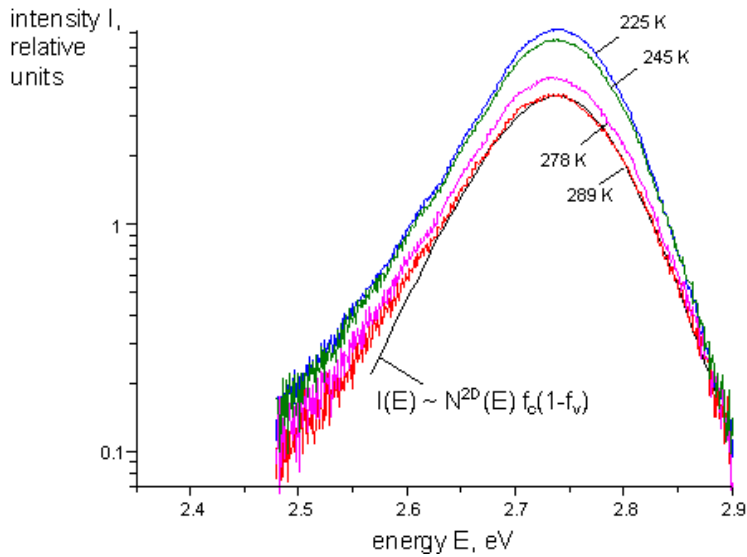


Figure 3. Temperature dependent spectra of blue LED #2 at 1 mA.

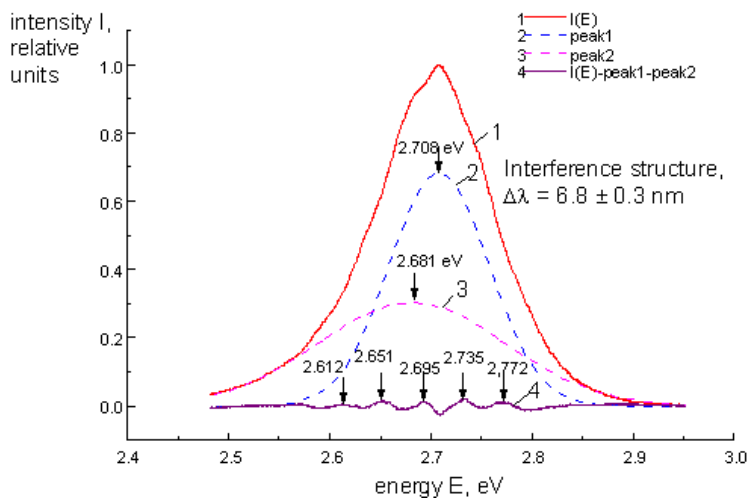


Figure 4. Two Gaussian approximation of a blue LED #3 spectrum at RT, $J = 10$ mA. The lower curve is the difference between the experimental data and the sum of the two Gaussians.

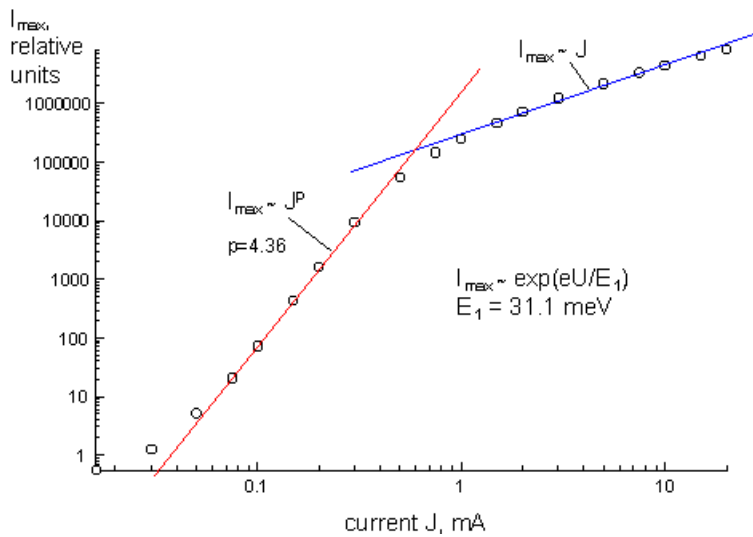


Figure 5a. Current dependence of I_{\max} at RT for blue LED #5.

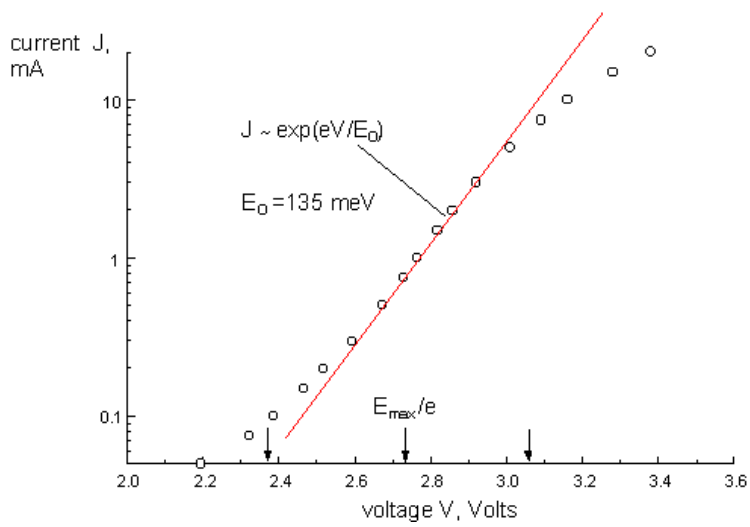


Figure 5b. RT J-V curve of blue LED #5.

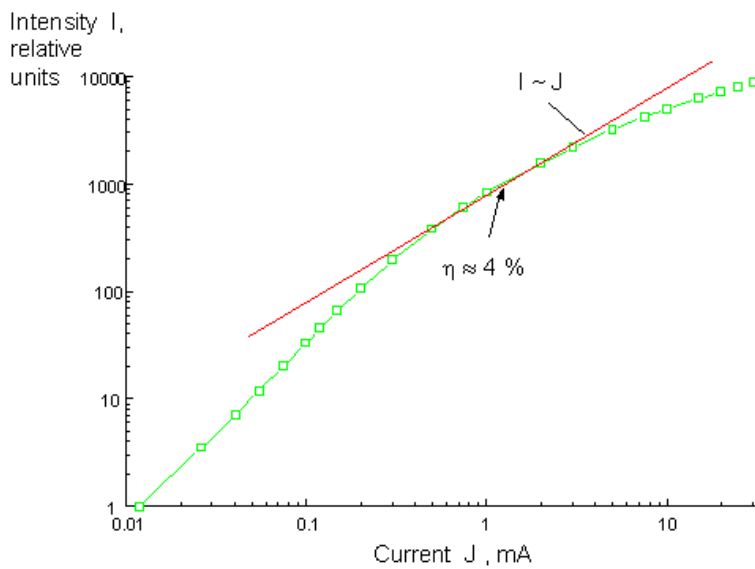


Figure 6a. Current dependence of I_{max} at RT for green LED #3.

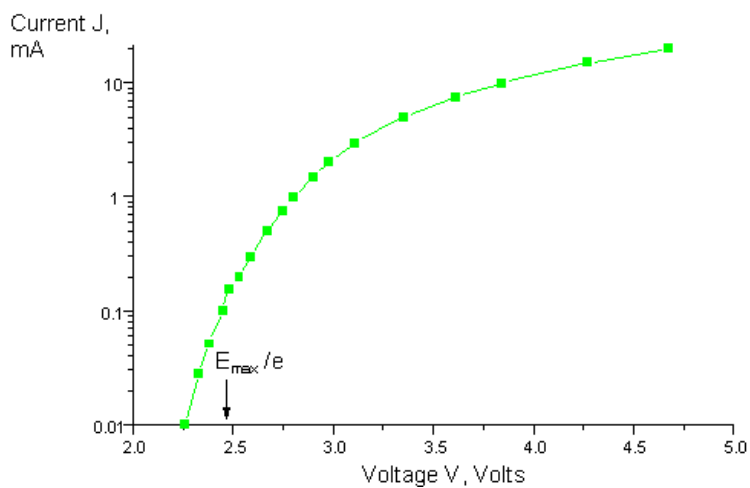


Figure 6b. RT J-V curve of green LED #3.

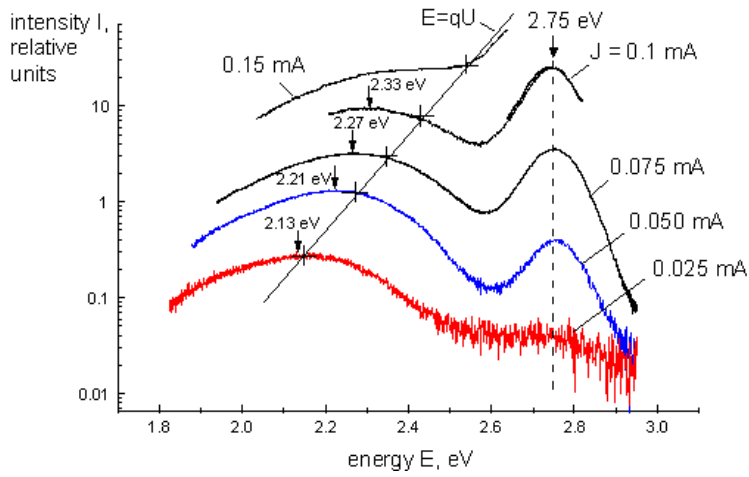


Figure 7. Low current RT spectra of blue LED #2 showing tunnel radiative recombination.

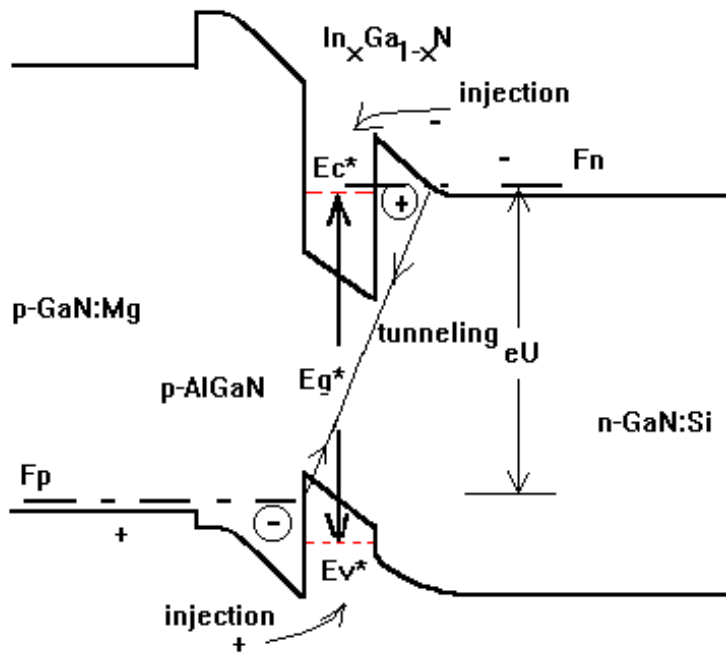


Figure 8. Energy diagram of a InGaN/AlGaIn/GaN heterostructure under forward bias.

© 1996-1998 The Materials Research Society