Enhanced blue and green emission in rare-earth-doped GaN electroluminescent devices by optical photopumping

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(Received 1 July 2002; accepted for publication 2 August 2002)

Electroluminescence (EL) from rare-earth-doped GaN (GaN:RE) EL devices (ELD) emission has been observed to be greatly enhanced by ultraviolet (UV) photopumping. With radiation from a HeCd laser (325 nm) both blue (from GaN:Tm) and green (from GaN:Er) EL brightness have been enhanced up to 2 orders of magnitude, depending on bias conditions. We explain the luminescence increase by the following mechanism: photoelectrons generated by above GaN band-gap excitation are accelerated by the electric field along with electrically injected electrons and both types of carriers contribute to EL emission through RE impact excitation. The EL intensity increases monotonically with increasing applied bias and with photopumping power. The photopumped-induced EL gain is most efficient at relatively low bias, reaching values of 50–100×. This increase in EL emission can be applied to flat panel displays with enhanced brightness, especially blue, and with improved color balance. Other applications include UV indicators and detectors, and infrared emitters.
[DOI: 10.1063/1.1509111]

Rare-earth-doped electroluminescent devices (ELDs) have been shown[1] to be a versatile approach for the fabrication of optical light sources with narrow linewidth emission wavelengths from the ultraviolet (UV) to the infrared (IR). Applications for rare-earth (RE) ELDs range from visible flat panel displays (FPD) to sources for IR optical communications. A variety of hosts have been investigated for RE incorporation.[2,3] For FPDs, obtaining bright blue emission, which carries the highest energy per photon (2.6–2.7 eV) of the three primary colors, has been a major challenge. A variety of RE hosts primarily based on II–VI semiconductors have previously been reported.[4–8] We have investigated the wide band-gap III–V semiconductor GaN as a RE host, producing green from GaN:Er,[9,10] red from GaN:Pr (Ref. 11) and GaN:Eu,[12] blue from GaN:Tm,[13] as well as mixed colors by RE codoping.[14]

GaN films were grown on *p*-type (111) Si substrates by molecular beam epitaxy (MBE) with a Ga elemental source and a nitrogen plasma source. Rare-earth doping was performed *in situ* during growth from solid elemental sources. GaN:RE layers were typically grown for 1 h with a growth rate of 0.5–1.0 μm/h. Ring-shaped Schottky diodes were fabricated for EL measurements using indium–tin–oxide (ITO) sputtering and a liftoff process. The ITO layer was typically 300–400 nm thick.

We have investigated the photopumping effect in GaN:Er and GaN:Tm ELDs, emitting green and blue light. For GaN:Er ELDs, the growth temperature was 550 °C with the Er cell temperature of 860 °C, while for GaN:Tm ELDs the growth temperature was 500 °C with the Tm cell at 580 °C. This results in Er and Tm concentration of 0.6 and 0.15 at. %. The GaN:RE growth was performed under slightly N-rich growth conditions: 1.5 sccm for nitrogen flow rate and 400 W for plasma power. Though it is well accepted that good crystalline GaN is usually grown under slightly Ga-rich growth conditions,[15] we have previously shown[16] that slightly N-rich growth conditions are favorable for GaN:RE EL emission. The resulting film thickness was ~1 μm for GaN:Er and ~0.6 μm for GaN:Tm. Visible emission from GaN:Er has two characteristic green peaks at 537 and 558 nm, which are caused by two $4f-4f$ Er^{3+} inner shell transitions: $^2H_{11/2}\rightarrow{}^4I_{15/2}$ and $^4S_{3/2}\rightarrow{}^4I_{15/2}$, respectively. Blue emission from GaN:Tm is due to the $^1G_4\rightarrow{}^3H_6$ transition. Near-infrared emission is also present at 802 nm ($^3F_4\rightarrow{}^3H_6$ transition) in GaN:Tm and 1.5 μm ($^4I_{13/2}\rightarrow{}^4I_{15/2}$) in GaN:Er.

Figure 1 shows the effect of photopumping (at 325 nm from a HeCd laser) on green EL emission from GaN:Er ELD and blue emission from a GaN:Tm ELD. Figures 1(a), 1(c), and 1(e) correspond to the ‘‘no-pumping’’ case, while Figs. 1(b), 1(d), and 1(f) correspond to the ‘‘photopumping’’ case. A very large enhancement in brightness (from barely visible to very bright) is evident for both green and blue ELDs. The dark notch in the emission pattern from the ring ELD in Figs. 1(a)–1(d) is due to the probe tip used to supply voltage. Both devices are dc biased in the threshold region for emission in the normal condition (no pumping). Brightness enhancement can reach a factor as high as ~100×, depending on bias voltage and laser power. Practical bias conditions for ELD operation are typically higher than those used here, but this reduced bias level provides a sharp visualization of the brightness enhancement. The diameter of the pump beam is somewhat larger than the ring diode outer diameter, insuring complete coverage of the device area. It is important to note that there is no emission from the open region in the center of the device even though it is irradiated with the same pump beam power density as the device itself. Furthermore, removing the electrical bias during photopumping greatly reduces the emitted light, to the point where it is barely (or not)

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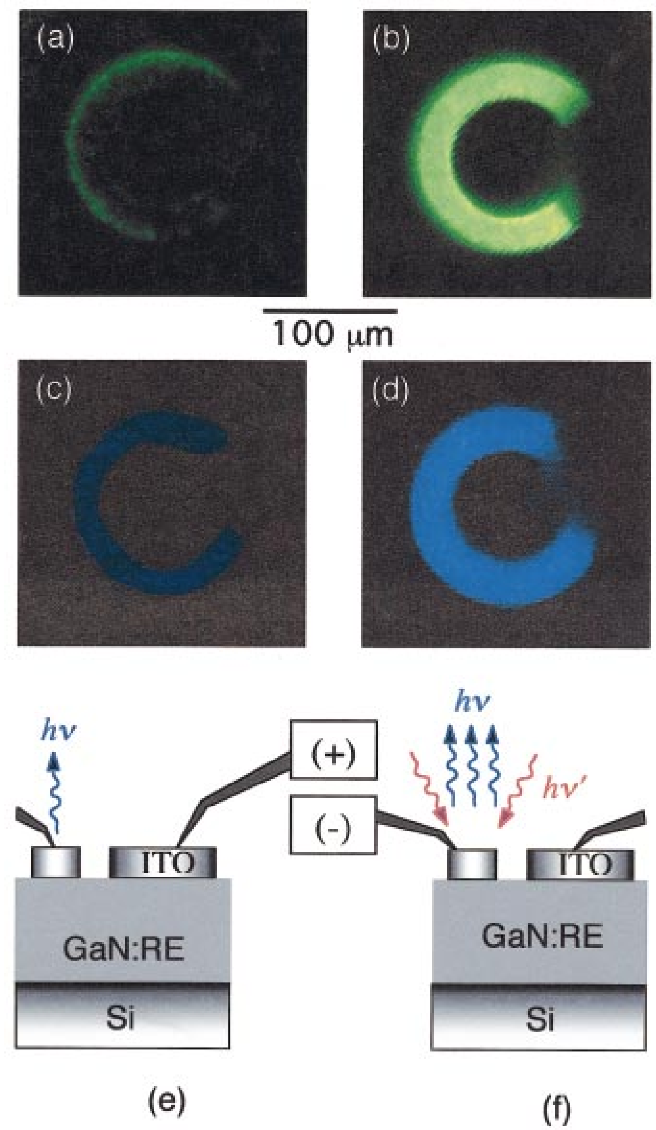

FIG. 1. (Color) EL emissions from GaN:RE ELDs with and without photopumping. Green emission from GaN:Er ELD: (a) with no pumping; (b) with photopumping. Blue emission from GaN:Tm ELD: (c) with no pumping; (d) with photopumping. HeCd laser (325 nm) was used for photopumping. Device structure and biasing are also shown: (e) with no pumping; (f) with photopumping. A major enhancement was observed in both green and blue brightness.

observable with the naked eye. Therefore, the effect observed here is clearly not the case of summing electroluminescence and photoluminescence mechanisms. Figures 1(e) and 1(f) contain schematics of the device structure and the probe tips used for biasing. The emitting ring electrode is 25 μm wide and is negatively biased. The outer diameter of the ring electrode is 150 μm and is surrounded by a large ground electrode.

Figure 2 contains EL spectra obtained from a GaN:Tm ELD biased at 45 V. In this specific example, photopumping increased the EL intensity $\sim 8\times$ at blue (478 nm) and $\sim 20\times$ at near IR (802 nm) wavelengths. In the same manner, photopumping GaN:Er ELDs increased both green (537/558 nm) and IR (1540 nm) emission (not shown here).

The operation of a photopumped GaN:RE ELDs can be explained with the aid of Fig. 3. Electroluminescence in GaN:RE devices is produced[14,17] by carrier impact excitation

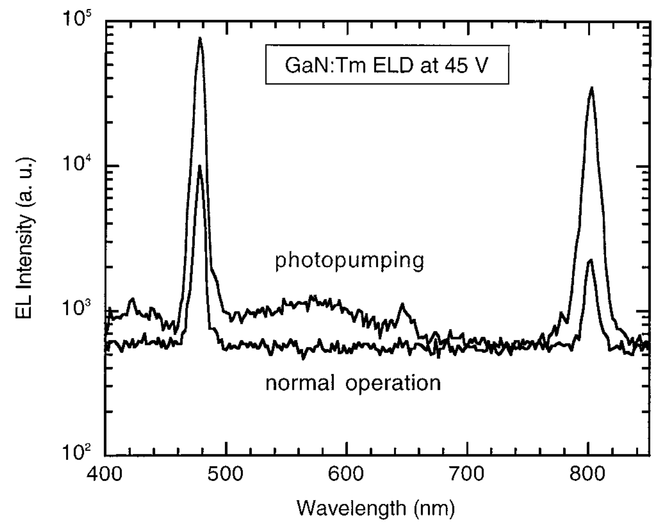

FIG. 2. EL spectra from a GaN:Tm ELD biased at 45 V. Photopumping increased emission by $\sim 8\times$ at blue (478 nm) and $\sim 20\times$ near IR (802 nm) wavelengths.

of RE^{3+} ions. An electron injected from the ITO Schottky electrode into the GaN:RE gains energy by being accelerated by the electric field and loses energy as it undergoes collisions with the GaN lattice and/or with RE^{3+} ions. When the ''hot'' electron reaches a sufficient energy for exciting $4f$ transitions in RE^{3+} ions, each collision may result in photoemission upon return of the excited ion to the ground state. For example, carrier energy of at least 2.3 eV is required for green emission from RE^{3+} ions. Photopumping by above-band-gap excitation generates additional charge carriers, which can take part in RE^{3+} impact excitation and result in increased EL emission. Interestingly, reversing the bias polarity such that holes are injected from the ITO electrode also results in photopumping enhancement, but much less pronounced than in the case of electron injection.

Figure 4 shows the GaN:Er ELD emission intensity at 537 nm versus applied bias for various photopumping power levels. The photopumping power was varied from 2.8 to 23 W/cm^2 (3.5–29 mW) using intensity filters. The ''no pumping'' condition is shown for comparison. Under relatively low bias conditions (within the dotted rectangle from 50 V to just below 80 V) no emission was measured under normal operation, but emission became visible and measur-

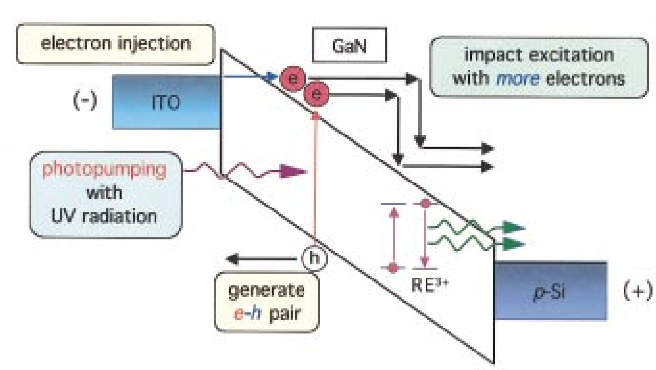

FIG. 3. (Color) Operation of GaN:RE ELDs under electrical bias and with above band-gap photopumping. Conventional EL emission is due to RE^{3+} impact excitation by electrically injected electrons accelerated by the applied bias. Additional hot electrons generated by photopumping produce EL enhancement.

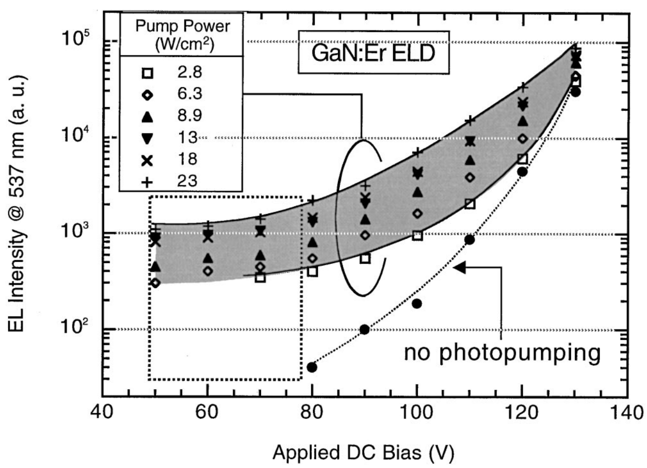

FIG. 4. EL intensity vs dc bias with various pumping powers obtained from a GaN:Er ELD. EL intensity increases monotonically with dc bias and laser power. Note that EL enhancement is proportionally larger at lower bias and seems to saturate at higher bias.

able by UV photopumping. The EL intensity increases monotonically with pump power at a fixed bias. The EL emission also increases monotonically with bias for all values of laser power. Note that EL enhancement is proportionally larger at lower bias and seems to saturate at higher bias (e.g., ~130 V). While the number of injected electrons is a strong function of applied bias (either thermionic or tunneling), the number of photogenerated electrons is thought to be constant regardless of bias. Therefore, the photopumping effect is more obvious at relatively low bias where fewer electrically injected electrons are present.

Figure 5 shows a plot of EL intensity gain (i.e., photopumping to no pumping emission ratio) versus pump power from a GaN:Er ELD. The EL gain increases monotonically with pumping power at all values of bias. The gain is more

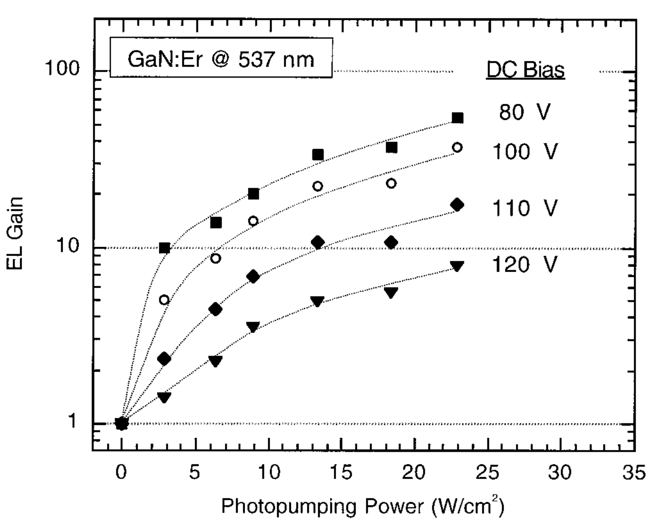

FIG. 5. EL intensity gain vs photopumping power obtained from a GaN:Er ELD at different bias voltages. A gain of ~60× was obtained at 80 V bias with 23 W/cm^2 pump power.

pronounced at lower bias levels, reaching a maximum value of ~60× at 80 V bias and 23 W/cm^2 pump power. Even at the higher bias levels significant gain is achieved. For example, at 120 V a gain of ~8 is obtained for 23 W/cm^2 photopump power.

Comparing the EL intensity as a function of current flow for photopumping and no pumping operation (not shown here), it is seen that photopumping enhanced the brightness 5–10× at the same total current flow. Therefore, the luminous efficiency can be said to be improved 5–10× by photopumping. A quantitative study of photopumping luminous efficiency in GaN:RE ELDs is currently underway.

In summary, a major brightness enhancement has been observed in EL emission from GaN:RE ELDs by UV photopumping: both visible (blue at 478 nm from GaN:Tm and green at 537/558 nm from GaN:Er) and near-IR (at 802 nm from GaN:Tm and at 1540 nm from GaN:Er). We believe that photoelectrons generated by above-band-gap excitation are accelerated along with electrically injected electrons resulting in stronger EL emission through enhanced impact excitation of rare earth ions. This increase in visible EL can be very beneficial for EL flat panel displays by increasing the brightness. This is particularly important for blue emission, which has been the most challenging color in terms of brightness. Photopumping is also important for increasing the output of GaN:RE IR emitters and for UV indicators and detectors.

This work was supported by ARO Grant No. DAAD19-99-1-0348. Equipment support was provided by an ARO URI Grant and the Ohio Materials Network. The authors thank J. Heikenfeld for assistance and are also pleased to acknowledge the support of J. Zavada.

[1] R. Mueller-Mach and G. O. Mueller, editor, Semicond. and Semimet. **65** (2000).
[2] M. Leskela, W. M. Li, and M. Ritala, Chap. 3 in Ref. 1.
[3] Y. A. Ono, *Electroluminescent Displays* (World Scientific, Singapore, 1995).
[4] G. Härkönen, M. Leppänen, E. Soininen, R. Törnqvist, and J. Viljanen, J. Alloys Compd. **225**, 552 (1995).
[5] C. N. King, J. Vac. Sci. Technol. A **14**, 1729 (1996).
[6] R. H. Mauch, Appl. Surf. Sci. **92**, 589 (1996).
[7] W. A. Barrow, R. C. Coovert, E. Dickey, C. N. King, C. Laakso, S.-S. Sun, R. T. Tuenge, R. Wentross, and J. Kane, Dig. 1993 SID Int. Displ. Symp., 761 (1993).
[8] S.-S. Sun, Displays **19**, 145 (1999).
[9] A. J. Steckl and R. Birkhahn, Appl. Phys. Lett. **73**, 1700 (1998).
[10] A. J. Steckl, M. J. Garter, R. Birkhahn, and J. Scofield, Appl. Phys. Lett. **73**, 2450 (1998).
[11] R. Birkhahn, M. J. Garter, and A. J. Steckl, Appl. Phys. Lett. **74**, 2161 (1999).
[12] J. Heikenfeld, M. Garter, D. S. Lee, R. Birkhahn, and A. J. Steckl, Appl. Phys. Lett. **75**, 1189 (1999).
[13] A. J. Steckl, M. Garter, D. S. Lee, J. Heikenfeld, and R. Birkhahn, Appl. Phys. Lett. **75**, 2184 (1999).
[14] A. J. Steckl, J. Heikenfeld, D. S. Lee, and M. Garter, Mater. Sci. Eng., B **81**, 97 (2001).
[15] E. Calleja, M. A. Sanchez-Garcia, F. J. Sanchez, F. Calle, F. B. Naranjo, E. Munoz, U. Jahn, and K. Ploog, Phys. Rev. B **62**, 16826 (2000).
[16] D. S. Lee and A. J. Steckl, Appl. Phys. Lett. **80**, 728 (2002).
[17] P. D. Rack and P. H. Holloway, Mater. Sci. Eng., R. **21**, 171 (1998).