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Growth-temperature dependence of Er-doped GaN luminescent thin films

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Visible photoluminescence (PL) and electroluminescence (EL) emission has been observed from Er-doped GaN thin films grown on (111) Si at various temperatures from 100 to 750 °C in a radio-frequency plasma molecular beam epitaxy system. PL and EL intensities of green emission at 537 nm from GaN:Er films exhibited strong dependence on the growth temperature, with a maximum at 600 °C. Scanning electron and atomic force microscopy showed smooth surfaces at 600 °C and rough surfaces at 100 and 750 °C. X-ray diffraction indicated that the GaN:Er film structure was oriented with the *c* axis perpendicular to the substrate for all growth temperatures. The crystalline quality initially improves with an increase in growth temperature, and saturates at ~500 °C. Considering both the luminescence and structural properties of the film, ~600 °C seems to be the optimal temperature for growth of Er-doped GaN luminescent films on Si substrates.
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Rare-earth (RE)-doped GaN has been shown[1] to be an extremely versatile optoelectronic material, having light emission throughout the visible spectrum as well as at important near-infrared wavelengths. The standard molecular beam epitaxy (MBE) growth process at 600–700 °C for *in situ* RE doping of GaN has resulted[2] in the successful fabrication of electroluminescent devices (ELDs) with red, green, and blue (RGB) emissions using Eu, Er, and Tm, respectively. We have also achieved[3] RGB in electroluminescence (EL) emission from GaN:RE films grown at nominally room temperature using same set of rare-earth elements. The ability to grow optically active RE-doped GaN films at reduced temperatures is very important in reducing the lattice strain in the material and also opens up the possibility for use of alternative, low-cost substrates. Effects in this direction have included deposition on glass substrates.[4,5] In this letter, we report on the effect of the growth temperature of Er-doped GaN films on optical properties and morphology. The temperature dependence of optical properties was investigated by photoluminescence (PL) and EL. Scanning electron microscopy (SEM), atomic force microscopy (AFM), and x-ray diffraction (XRD) were used to investigate the influence of growth temperature on the morphology.

GaN films were grown on *p*-type (111) Si substrates by MBE with a Ga elemental source and a nitrogen plasma source. Er doping was performed *in situ* during growth from a solid source. GaN:Er layers were typically grown for 1 h. For EL measurements a simple ring-shaped Schottky electrode was fabricated on top of the GaN:Er film using indium–tin–oxide (ITO) sputtering and a lift-off process. The ITO film has a more than 90% transmittance over the entire visible light range. The area of the electrode was 7.65×10^{-4} cm^2; details of its structure are reported elsewhere.[6]

Er-doped GaN films were grown at various temperatures from 100 to 750 °C. These temperatures were measured with a thermocouple mounted behind the sample holder. Since the Ga flux condition is critical[7] in the performance of luminescence of rare-earth-doped GaN, growth runs were carried out with various Ga fluxes at 100 and 600 °C. Since the EL emission in GaN:Er films is known to be a sensitive function of the Ga flux, the Ga cell temperature was first optimized independently at these two temperatures. It was determined that the optimal equivalent Ga cell temperature was 870 °C (producing a Ga flux of 2.0×10^{-7} Torr) for 100 °C growth and 900 °C (flux of 3.4×10^{-7} Torr) for 600 °C growth. Ga cell temperatures for other growth temperatures were determined by interpolation and extrapolation of these two growth temperatures. The Ga flux optimization at each growth temperature is necessary in order to be able to compare the highest EL emission levels obtainable at each growth temperature. The N_2 plasma source parameters were kept constant at a 1.2 sccm flow rate and 300 W plasma power. All samples were grown for 1 h. The growth rate varied with substrate temperature settings from ~0.3 to ~0.55 μm/h, due to thermal and Ga flux effects.

Figure 1 shows SEM and AFM images of GaN:Er films grown at 100, 600, and 750 °C. The 100 °C-grown film has a relatively rough surface and a grainy structure, Figs. 1(a) and 1(b). The 600 °C-grown film has a smooth surface with a compact structure and large grains, Figs. 1(c) and 1(d), while the 750 °C-grown film has a very rough surface, Figs. 1(e) and 1(f). The extreme roughness of the film grown at 750 °C is due to a combined effect of GaN desorption during growth (because of the high substrate temperature) and the use of a Ga flux well below a stoichiometric condition, (i.e., in a highly N-rich growth condition), which is known to increase the roughness. The occurrence of desorption was supported by the fact that the film grown at 750 °C was found to be thinner than that grown at 600 °C in spite of the use of a higher Ga flux. Since the Ga flux increased linearly with the growth temperature, it was probably not sufficient to compensate for the rapid increase in Ga loss (through either direct desorption or GaN decomposition) at this temperature.

The root mean square (rms) roughness of all the films was obtained by AFM. The results are plotted in Fig. 2. For

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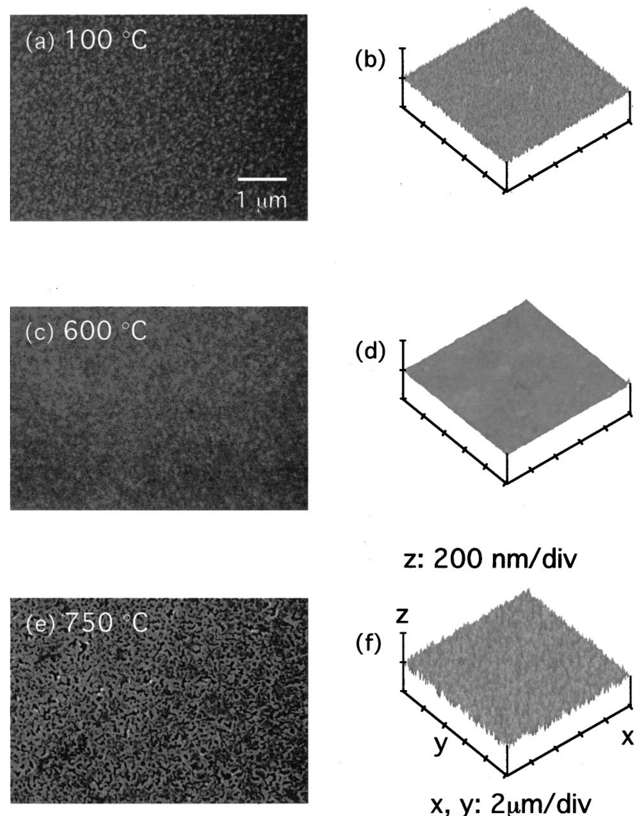

FIG. 1. Scanning electron and atomic force microscopy images of GaN:Er films grown at (a), (b) 100; (c), (d) 600; and (e), (f) 750 °C.

growth temperatures of 100 and 200 °C, the roughness was ~10 nm. In this growth temperature range films seemed to consist of nanocrystalline pillars or columns. As the growth temperature increased, the size of columns also seemed to increase. As a result, the roughness decreased to 2–5 nm for growth temperatures between 300 and 600 °C. Even though they exhibited similar surface roughness values, the film grown at 600 °C had larger grain sizes (as observed by SEM and AFM) than those grown at lower temperature. The ~10 times rougher surface obtained at 750 °C is thought to be mainly due to desorption of GaN as mentioned earlier.

XRD was used to investigate the crystalline quality of

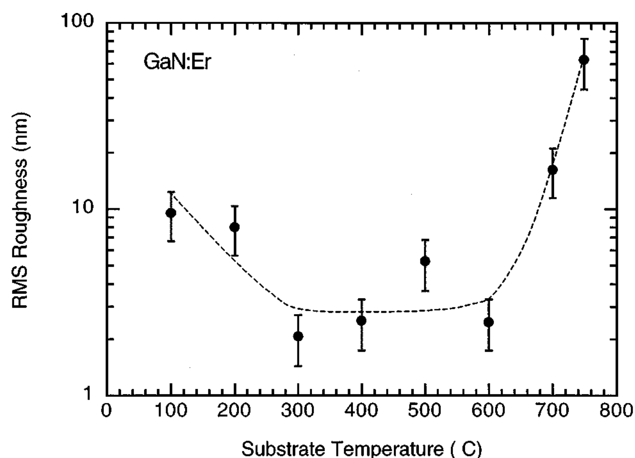

FIG. 2. AFM measurement of the rms roughness of GaN:Er films as a function of the growth temperature. Films grown at 300–600 °C show a smooth surface and their rms roughness is in the range of 2–5 nm.

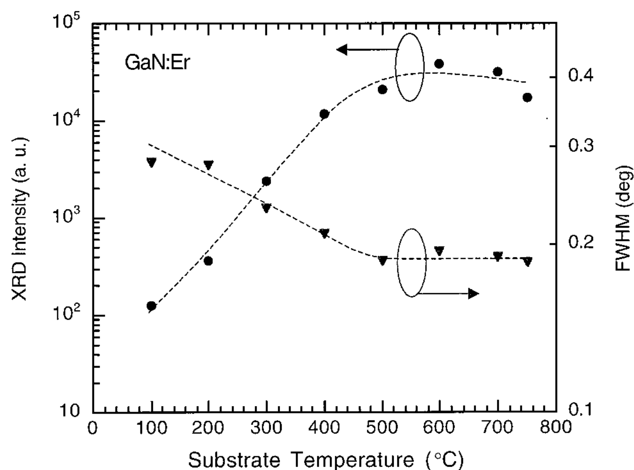

FIG. 3. Crystallinity of GaN:Er films grown at various temperatures: XRD intensity of the (0002) hexagonal GaN peak. The XRD signal first increases with the growth temperature and then saturates at around 500 °C. The FWHM of the XRD peak shows complementary behavior to the intensity and also saturates around 500 °C.

the films grown at various temperatures. The XRD spectra showed the preferential (0002) orientation of hexagonal GaN at $2\theta = 34.5°$ regardless of the growth temperature, indicating that c-axis preferred growth occurs even at temperatures as low as 100 °C. The (0002) peak intensity and full width at half maximum (FWHM) are plotted in Fig. 3. Hexagonal GaN usually consists of crystalline columns, with significant tilt and twist variations. XRD peak broadening is a measure[8] of the alignment disorder of the c axis among the columns due to their relative tilt and twist. A thin film acquires a more ordered structure as the growth temperature increases, and it becomes more ordered as its thickness increases (when grown at a high enough temperature). As shown in Fig. 3, at low temperature, especially at 100 °C, the rather weak broad peak combined with aforementioned morphology indicate that the film is weakly polycrystalline. As the growth temperature increases, the crystalline quality improves significantly up to 500 °C. Interestingly, films grown at 500 °C and above have almost the same crystalline quality, at least in terms of peak intensity and FWHM.

The PL intensity of samples grown at various temperatures was measured using above band gap excitation with a HeCd (325 nm) laser. The visible emission has two characteristic green peaks at 537 and 558 nm, which are caused by two Er^{3+} transitions: $^2H_{11/2} \rightarrow {}^4I_{15/2}$ and $^4S_{3/2} \rightarrow {}^4I_{15/2}$, respectively. As shown in Fig. 4, the PL intensity is a strong function of the growth temperature. The inset shows a typical PL spectrum from Er-doped GaN. No detectable PL emission was observed from films grown at 100 and 200 °C. Between 300 and 600 °C the PL intensity increases almost exponentially with the growth temperature. At higher growth temperatures, the intensity starts to decrease. The films grown at 700 and 750 °C were probably not completely optimized in terms of the Ga flux compared to the film grown at 600 °C. This conclusion is supported by the morphology results in Figs. 1 and 2.

EL was investigated utilizing ITO/GaN:Er Schottky devices. For a comparison of devices grown at different temperatures, we use the brightness normalized by current flow versus voltage, called BIV. Since the EL brightness is influ-

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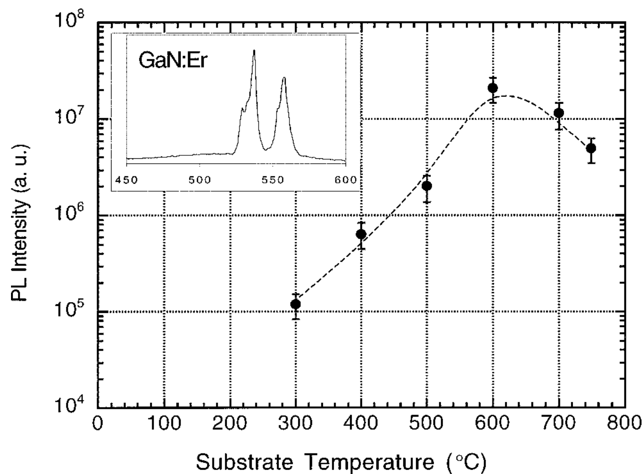

FIG. 4. PL intensity from GaN:Er films with respect to the growth temperature. The inset shows a typical PL spectrum from Er-doped GaN. The intensity with the growth temperature and reaches a maximum at 600 °C.

enced by many factors including those associated with EL device fabrication, the current-normalized brightness is a more appropriate parameter to evaluate rather than the raw brightness, i.e., brightness regardless of current flow through the device at a given applied voltage. All films showed EL regardless of the growth temperature, including those which did not exhibit PL (i.e., those grown at 100 and 200 °C). The maximum BIV for each sample is plotted in Fig. 5. Starting at a low growth temperature, BIV increases with the growth temperature, exhibits a maximum at 600 °C, and decreases at higher temperatures. This is the same trend as that obtained

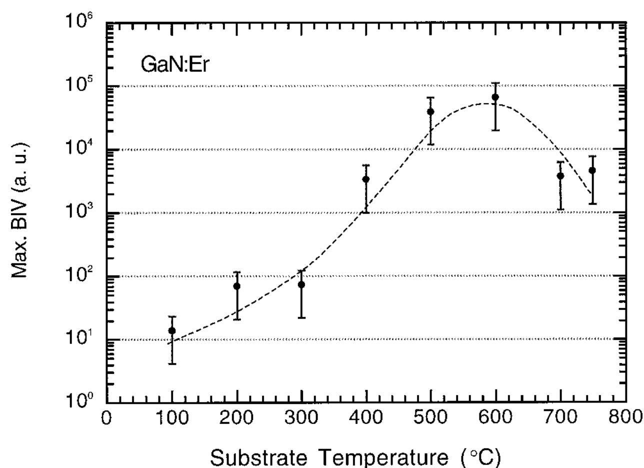

FIG. 5. Maximum current-normalized EL brightness (BIV) with respect to the growth temperature. BIV increases with the growth temperature and reaches a maximum at 600 °C like it does in PL.

from the PL data, thus reinforcing the conclusion that 600 °C is the optimal growth temperature for visible emission from GaN:Er. Note the large increase in BIV between the 300 and 400 °C growth temperatures. This may indicate the presence of a transition point at which crystalline GaN forms.

Electroluminescence in GaN:Er devices is obtained by carrier impact excitation. To accelerate injected electrons to a sufficient energy requires high electric fields. For example, carrier energy greater than 2.3 eV is required for green emission[9,10] from Er^{3+} ions. As voltage is applied to each device, reaching the electric fields needed requires relatively resistive samples. The devices fabricated from the 600 °C-grown film were the most resistive among these films and showed the strongest EL emission. As a result, they produced the highest maximum BIV. The decreased BIV from films grown at 700 and 750 °C is due mainly to the more conductive nature of those films, in spite of comparable raw EL intensity. We believe that the main cause for the increased conductivity of these films has to do with their rough morphology, which allows many pathways of electrical leakage. While a higher Ga flux was used in these cases, the films appeared to be N rich, with no Ga droplets.

In summary, we have grown Er-doped GaN films at various growth temperatures and investigated their optical and structural properties. Growth temperature of 600 °C seems to be optimal in terms of the PL and EL intensities as well as for structural properties such as surface roughness and crystalline quality. However, considering that the films grown at 700 and 750 °C were not optimized as fully as the films grown at 600 °C, it still may be possible to achieve optically better films at such high temperatures.

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[1] A. J. Steckl and J. M. Zavada, MRS Bull. **24**, 33 (1999).
[2] A. J. Steckl, J. Heikenfeld, M. Garter, R. Birkhahn, and D. S. Lee, Compound Semicond. **6**, 48 (2000).
[3] D. S. Lee and A. J. Steckl, Appl. Phys. Lett. **79**, 1962 (2001).
[4] Y. Sato, A. Kurosaki, and S. Sato, J. Cryst. Growth **189/190**, 42 (1998).
[5] V. I. Dimitrova, P. G. Van Patten, H. H. Richardson, and M. E. Kordesh, Appl. Phys. Lett. **77**, 478 (2000).
[6] D. S. Lee, J. Heikenfeld, R. Birkhahn, M. Garter, B. K. Lee, and A. J. Steckl, Appl. Phys. Lett. **76**, 1525 (2000).
[7] D. S. Lee and A. J. Steckl (unpublished data).
[8] S. D. Hersee, J. C. Ramer, and K. J. Malloy, MRS Bull. **22**, 45 (1997).
[9] E. Belloti, I. H. Oguzman, J. Kolnik, K. F. Brennan, R. Wang, and P. P. Ruden, Mater. Res. Soc. Symp. Proc. **468**, 457 (1997).
[10] A. J. Steckl, J. Heikenfeld, D. S. Lee, and M. Garter, Mater. Sci. Eng., B **81**, 97 (2001).

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