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Coincident site lattice-matched InGaN on (111) spinel substrates

National Renewable Energy Laboratory, Golden, Colorado 80401, USA
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Coincident site lattice-matched wurtzite (0001) In$_{0.31}$Ga$_{0.69}$N, emitting in the important green wavelength region, is demonstrated by molecular beam epitaxy on a cubic (111) MgAl$_2$O$_4$ spinel substrate. The coincident site lattice matching condition involves a 30° rotation between the lattice of the InGaN epitaxial layer and the lattice of the spinel. This work describes an alternative approach towards realizing more compositionally homogeneous InGaN films with low dislocation density emitting in the “green gap” of low efficiency currently observed for semiconductor light emitting diodes (LEDs). This approach could lead to higher efficiency green LEDs presently of great interest for solid-state lighting applications. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.3702577]

InGaN is of considerable interest for solid-state lighting (SSL) applications, due to the fact that it is a direct band gap semiconductor that can span the entire visible spectrum. High efficiency SSL offers tremendous gains in energy efficiency over conventional lighting sources and consequently large environmental benefits. If SSL was adopted widely, it has been estimated that, by 2025, ~620 TW-h/yr of electricity could be saved in the U.S. alone.

Previously, InGaN light emitting diodes (LEDs), as well as other III-V LEDs, have poor efficiencies at wavelengths close to 550 nm, frequently referred to as the “green gap.” This region of low efficiency limits the wide-scale deployment of SSL technology. The ability to grow “lattice-matched” InGaN on (111) spinel substrates represents a significant advantage over growth on the more traditionally used sapphire and SiC substrates that do not allow similar coincident site lattice matching to be achieved.

We have grown a series of wz InGaN alloy layers on (111) spinel substrates by molecular beam epitaxy (MBE) to determine if the required epilayer alignment occurs, and characterized the materials by a number of techniques including transmission electron microscopy (TEM), scanning electron microscopy (SEM), and photoluminescence (PL). wz InGaN alloy layers were cleaned in a 2:1:10 NH$_4$OH:H$_2$O$_2$:H$_2$O solution for 1 min, and rinsed in de-ionized water prior to loading into a Riber Compact21 MBE system. The spinel substrates were subsequently heated to ~750 °C, allowed to cool under a Ga flux, then reheated to desorb the Ga. This Ga polish was repeated three times in order to clean the surface of outside contaminants. InGaN layers were then deposited at ~450 °C at a growth rate of ~0.5 µm/h with a V/III ratio just below unity. Cross-section and plan-view TEM samples were prepared in a FEI focused ion beam (FIB) work station, using the lift out technique of Overwijk et al., or by conventional mechanical polishing and Ar ion beam milling methods and examined in Philips CM30 and FEI Tecnai ST30 TEMs operated at 300 kV. High angle annular dark field (HAADF) imaging and energy dispersive x-ray spectroscopy (EDX) analysis were performed in a FEI Tecnai F20 UltraTwin scanning transmission electron microscope (STEM) operated at 200 kV. Electron

![Diagram illustrating coincident site lattice matching condition for the epilayer growth of a (0001) wurtzite InGaN alloy crystal on a (111) face centered cubic substrate surface. (a) No lattice rotation, (1-10) fcc//<11-20> wz; (b) after 30° lattice rotation, (1-10) fcc//<10-10> wz.](image-url)
backscatter diffraction (EBSD) experiments were performed using an EDAX TSL system integrated to a Tecnai Nova NanoSEM 630 SEM. PL was performed using the 442 nm line of a HeCd laser as the excitation source.

Figure 2(a) shows a calculated transmission electron diffraction (TED) pattern, using the CrystalKit software package, looking down the $h_{10}^{-10}$ pole of (0001) wz In$_{0.31}$Ga$_{0.69}$N grown coincident site lattice matched on (111) cubic MgAl$_2$O$_4$. An experimental TED pattern (Fig. 2(b)), taken using a selected area aperture positioned across the interface between InGaN and spinel, confirms that the required epitaxial alignment occurs in this region of this sample. Lattice images of the sample revealed an abrupt but stepped interface between the InGaN and the spinel (Fig. 3). This is most likely due to the poor surface quality of the spinel substrates used in this study.

To investigate if any un-rotated domains were also present, SEM EBSD studies were performed on plan-view samples. A plan-view EBSD orientation map and the corresponding pole figures are shown in Fig. 4. In this map, the InGaN is seen to be all (0001) oriented but the black lines indicate boundaries between two sets of $30^\circ$-rotated, micron-sized, orientation domains in the epitaxial InGaN alloy layer. The presence of these rotated orientation domains results in 12 maxima in intensity in the (1010) pole figure instead of the six maxima that would be present without the existence of rotated domains. One set of six minima is coincident site lattice matched with the epitaxial relationship shown in Fig. 1(b), whilst the other set is un-rotated with respect to the substrate lattice (Fig. 1(a)) and, hence, severely lattice-mismatched.

Plan-view TEM diffraction contrast images, not shown, revealed a high threading dislocation density, $>10^{10}$ cm$^{-2}$ on average, was still present in these layers and also that it was similar in both the rotated and un-rotated domains. This suggests that the lattice mismatch may not be the most important parameter that determines the threading dislocation density. Instead, it may be that the nucleation mode, 3-dimensional island as opposed to 2-dimensional layer-by-layer growth, may be a more dominant factor in determining the threading dislocation density. Cross-sectional TEM of a rotated InGaN domain, not shown, confirmed a high threading dislocation density of between $\sim8 \times 10^9$ cm$^{-2}$ and $\sim2 \times 10^{10}$ cm$^{-2}$. Cross-sectional HAADF STEM images, not shown, of this rotated domain revealed an absence of strong contrast features indicating no significant phase separation in the coincident site lattice matched InGaN grown on (111) spinel. This was confirmed by STEM EDX line profile compositional analysis performed in the InGaN layer parallel to the InGaN/spinel interface.

InGaN films showed room temperature PL emission (Fig. 5) centered at $\sim550$ nm, despite the presence of the un-rotated lattice-mismatched domains and a high threading dislocation density. This wavelength corresponds to the center of the “green gap” of low efficiency previously reported for nitride based LEDs. The full width half maximum (FWHM) of the room temperature PL peak was measured at $\sim0.28$ eV.
This compares favorably with a room temperature PL FWHM of ~0.38 eV reported previously by Naranjo et al.\textsuperscript{16} for a 300 nm thick $\text{In}_{0.27}\text{Ga}_{0.73}\text{N}$ layer grown by MBE on a GaN/sapphire template. Shimomoto et al.\textsuperscript{17} have more recently reported on the growth of 500 nm thick m-plane $\text{In}_{0.33}\text{Ga}_{0.67}\text{N}$ layers on nearly lattice-matched ZnO substrates. In this paper, the authors reported a room temperature PL peak FWHM of ~0.23 eV, only slightly less than our value, and concluded on the basis of x-ray diffraction measurements that the InGaN film did not show any serious phase separation despite the high In content. The FWHM of PL peaks can be affected by the compositional uniformity of the sample, for example, broadened by the occurrence of phase separation, but can also be determined by other factors such as the presence of impurities and a high density of defects. Nevertheless, the relatively narrow room temperature PL peak FWHM observed for our samples is suggestive that serious phase separation is suppressed by using the coincident site lattice matched (111) spinel substrate. Future work will be directed towards controlling the nucleation of the preferred coincident site lattice matched InGaN epitaxial material. We believe that optimized nucleation conditions will result in fully rotated domains, lower threading dislocation densities, and improved efficiencies of green InGaN LEDs.

The work presented here demonstrates the initial approach towards realizing InGaN films with improved properties emitting in the “green gap.” Our results show that coincident site lattice-matched InGaN can be grown on (111) spinel substrates, and further work is needed to apply this concept into working device structures. The ability to grow compositionally homogeneous InGaN with minimal dislocations and strain on widely available spinel substrates could lead to a significant increase in the efficiency of green LEDs and potentially have a significant impact on the future of SSL.

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\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig5.png}
\caption{Room temperature PL spectra obtained from the same InGaN sample as Figs. 2–4 showing luminescence centered at ~550 nm, i.e., in the center of the “green gap.”}
\end{figure}