

Room-temperature epitaxial growth of high-quality *m*-plane InGaN films on ZnO substrates

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The authors have grown high-quality *m*-plane In_{0.36}Ga_{0.64}N (1 $\bar{1}$ 00) films on ZnO (1 $\bar{1}$ 00) substrates at room temperature (RT) by pulsed laser deposition (PLD) and have investigated their structural properties. *m*-plane InGaN films grown on ZnO substrates at RT possess atomically flat surfaces with stepped and terraced structures, indicating that the film growth proceeds in a two-dimensional mode. X-ray diffraction measurements have revealed that the *m*-plane InGaN

films grow without phase separation reactions at RT. The full-width at half-maximum values of the 1 $\bar{1}$ 00 X-ray rocking curves of films with X-ray incident azimuths perpendicular to the *c*- and *a*-axis are 88 arcsec and 78 arcsec, respectively. Reciprocal space-mapping has revealed that a 50 nm thick *m*-plane In_{0.36}Ga_{0.64}N film grows coherently on the ZnO substrate, which can probably explain the low defect density that is observed in the film.

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In_xGa_{1-x}N films have attracted much attention as materials for light-emitting diodes (LEDs) and laser diodes (LDs) that could possibly cover a wide wavelength region from the ultraviolet to the infrared, since they have direct band gaps ranging from 0.65 eV to 3.4 eV [1–3]. However, highly-efficient light emitting devices are only fabricated in the blue region at present, and the efficiencies of devices that operate in the green region and at longer wavelengths are still quite low, because it is difficult to grow high-quality InGaN with In compositions greater than 30%. This difficulty stems partially from the fact that phase separation of InGaN can occur quite easily due to the low miscibility between InN and GaN. To suppress this tendency for phase separation, the development of a growth technique that operates under highly non-equilibrium conditions such as extremely low growth temperatures is highly sought after. Another problem that affects long-wavelength InGaN

light-emitting devices is the ‘quantum confinement Stark effect’, which causes separation between holes and electrons in the quantum well. It is known that this undesirable effect can be eliminated by growing the InGaN on non-polar planes such as the (1 $\bar{1}$ 00) *m*-plane [4] or the (1120) *a*-plane [5]. Until now, many research groups have worked on the epitaxial growth of nonpolar nitrides on substrates such as LiAlO₂(100) or Al₂O₃(1 $\bar{1}$ 02). However, the epitaxial films grown on these substrates suffer from the formation of high densities of crystalline defects such as threading dislocations (TDs) and stacking faults (SFs) since the crystalline symmetry of these substrates is significantly different from that of group III-nitride materials. It is quite natural to believe that the use of nonpolar ZnO substrates would be a straightforward strategy for solving this problem because ZnO has the same crystalline structure as group III nitrides (wurtzite) and it has a relatively

small lattice mismatch with most group III nitrides. In particular, $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ has the same a -axis lattice parameter as ZnO, and therefore InGaN films grown on ZnO should theoretically have low misfit-dislocation densities. However, it has proved difficult to obtain high-quality group III nitrides on ZnO by conventional growth techniques such as metal organic chemical vapor deposition or molecular beam epitaxy, since ZnO readily reacts with nitrides at temperatures above 500 °C [6].

We have recently found that the above-mentioned problems (the phase separation of InGaN and the interfacial reactions between group III nitrides and ZnO) can be solved by the use of pulsed laser deposition (PLD), which allows us to dramatically reduce the growth temperature that is required for group III nitrides [7–10]. In fact, we have grown c -plane group III nitrides by using the layer-by-layer mode on ZnO (000 $\bar{1}$) by PLD even at room temperature (RT) [11–13] and we found that the heterointerfaces between the nitride films and the substrates were atomically abrupt. The growth of high-quality nitrides (even at RT) by the use of PLD can be attributed to enhancement of the kinetic energies of the group III atoms, which assists in the surface migration of the film precursors. In addition, we can expect the suppression of phase separation in the InGaN by the use of the low-temperature PLD technique. In this letter, we report on the growth of homogeneous high-quality m -plane InGaN films on ZnO substrates at RT by the use of PLD.

The ZnO that was used in this study was prepared by the hydrothermal method and was cut into (1 100) wafers. In order to obtain atomically flat ZnO surfaces, each substrate was annealed in air at 1050 °C for 3.5 h. The wafers were placed in a box made of ceramic ZnO during the annealing process in order to maintain the surface stoichiometry. After annealing, the ZnO (1 $\bar{1}00$) substrates were loaded into an ultra-high-vacuum PLD growth chamber. A KrF excimer laser light ($\lambda = 248$ nm, $\tau = 20$ ns) with an energy density of 3.0 J/cm² and at a pulse repetition rate of 20 Hz was used to ablate a metal-eutectic $\text{In}_{0.245}\text{Ga}_{0.755}$ alloy (99.999% purity) target. During the growth of the InGaN layers, a nitrogen plasma was supplied through an rf generator operating at 320 W, with a pressure of 1×10^{-5} Torr. The typical thickness of the InGaN films was approximately 50 nm, which was confirmed by grazing-incidence X-ray reflection measurements. The structural properties of the InGaN films were characterized using reflection high-energy electron diffraction (RHEED), high-resolution X-ray diffraction (HRXRD), and atomic-force microscopy (AFM). The molar composition of indium in the InGaN films was determined from the lattice constants that were measured using reciprocal space mapping (RSM).

AFM observations revealed that the surfaces of the ZnO substrates annealed in the ZnO box were atomically flat, although a high density of scratches caused by the mechanical polishing was observed on the as-received ZnO substrates. The full width at half maximum (FWHM) val-

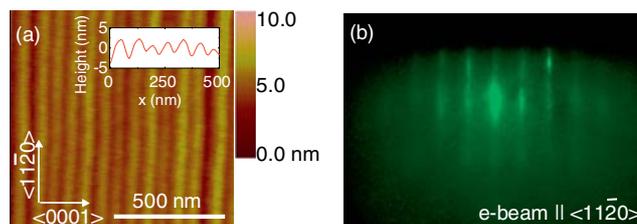


Figure 1 (online colour at: www.pss-rapid.com) (a) AFM image for an m -plane InGaN film grown on an atomically flat m -plane ZnO substrate at RT. The inset shows the cross-sectional profile. (b) RHEED pattern for an m -plane InGaN film with an electron beam incidence along the $\langle 11\bar{2}0 \rangle$ azimuth.

ues of $1\bar{1}00$ X-ray rocking curves for the m -plane ZnO substrate after annealing were 28 arcsec and 30 arcsec with the X-ray incidence perpendicular to a - and c -axes, respectively. As shown in Fig. 1(a), an InGaN film grown on an annealed m -plane ZnO substrate at RT also showed an atomically flat surface featuring stepped and terraced structures, which is analogous to that observed for the ZnO substrate. A RHEED pattern indicative of m -plane growth, as shown in Fig. 1(b), was clearly observed for InGaN from the initial stage of the film growth, and the pattern remained unchanged during the whole growth process, which indicates that the interfacial reactions that can occur between the nitride films and the ZnO substrates [11] were well suppressed and that high-quality InGaN had been grown in ‘two dimensional mode’ by the use of the RT growth technique. Figure 2(a) shows an XRD $2\theta/\omega$ scan for the out-of-plane $1\bar{1}00$ diffraction for a 50 nm thick InGaN film grown on a ZnO substrate at RT with an X-ray incident azimuth perpendicular to the c -axis. Two diffraction peaks were observed at 30.6° and 31.8°, which correspond to InGaN $1\bar{1}00$ and ZnO $1\bar{1}00$, respectively. In addition, grazing-incidence X-ray diffraction measurements revealed that $\langle 11\bar{2}0 \rangle$ direction of the InGaN film is parallel to that of the ZnO substrate. These results indicate that m -plane InGaN (1 $\bar{1}00$) grows epitaxially on m -plane ZnO

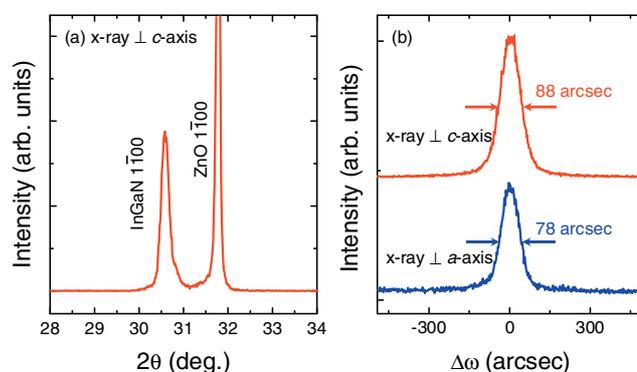


Figure 2 (online colour at: www.pss-rapid.com) (a) XRD $2\theta/\omega$ scan for $1\bar{1}00$ diffraction for a 50 nm thick m -plane InGaN film grown at RT with an X-ray incident azimuth perpendicular to the c -axis. (b) X-ray rocking curves for the InGaN $1\bar{1}00$ diffraction with an X-ray incident azimuth perpendicular to the a - and c -axis.

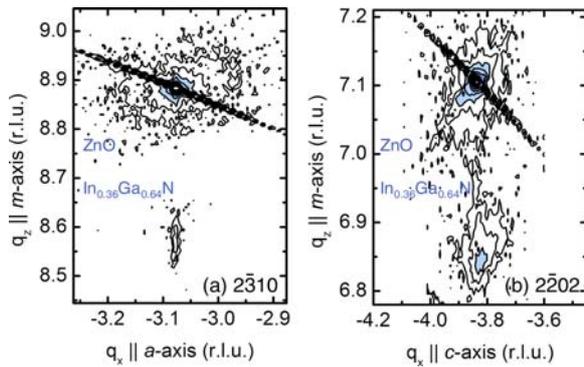


Figure 3 (online colour at: www.pss-rapid.com) Reciprocal space maps for (a) $2\bar{3}10$ and (b) $2\bar{2}02$ diffractions for a 50 nm thick *m*-plane InGaN layer. The transverse directions in (a) and (b) correspond to the *a*- and *c*-axes, respectively.

($1\bar{1}00$), even at RT. In addition, the appearance of the single diffraction peak for InGaN suggests that the phase separation of InGaN does not occur during RT growth by PLD. We then measured $1\bar{1}00$ X-ray rocking curves (XRCs) in order to characterize the mosaicity of the InGaN films. As shown in Fig. 2(b), the FWHM values of the XRC for the InGaN $1\bar{1}00$ diffraction were 78 arcsec and 88 arcsec with the incident X-ray azimuths perpendicular to the *a*- and *c*-axes, respectively. These low values indicate that *m*-plane InGaN layers with low TD densities grow on *m*-plane ZnO substrates at RT.

We then performed RSM in order to investigate the In composition and the residual strains in *m*-plane InGaN grown on ZnO. Figure 3(a) and (b) show the RSMs of the $2\bar{3}10$ and $2\bar{2}02$ diffractions for 50 nm thick *m*-plane InGaN, respectively. It was found that *m*-plane InGaN grows coherently in both the *a*- and *c*-axis directions due to the constraints arising from the *m*-plane ZnO substrate, which is probably responsible for the high crystalline quality of the film. It is known that the In composition *x* in an $\text{In}_x\text{Ga}_{1-x}\text{N}$ film can be determined by Hooke's law. In the case of *m*-plane epitaxy, the strain along the *m*-axis, ε_m , is expressed as follows:

$$\varepsilon_m = \frac{-C_{12}\varepsilon_a - C_{13}\varepsilon_c}{C_{11}}, \quad (1)$$

where C_{11} , C_{12} , and C_{13} are the elastic constants, and ε_a and ε_c are the strains along the *a*- and *c*-axis, respectively. If we assume that the strains and the elastic constants of $\text{In}_x\text{Ga}_{1-x}\text{N}$ follow Vegard's law, the formula can be expressed in an equation that contains *x* as the only variable. The value of *x*, calculated using the reported lattice constants and the elastic constants for InN [14] and GaN [15], was 0.36. We utilized various reported sets of the elastic constant [16, 17]. We found that the distribution of the *x*-value is less than 1%. The lattice parameters and the residual strain of InGaN and ZnO are listed in Table 1. This result indicates that In was preferably transferred from a PLD target into films. This phenomenon is probably ex-

Table 1 Lattice parameters (in nm) and residual strain of InGaN and ZnO.

	<i>a</i>	<i>c</i>	<i>m</i>
InGaN	0.325 (−1.8%)	0.522 (−3.2%)	0.292 (+1.7%)
ZnO	0.325	0.521	0.281

plained by the higher ablation efficiency of In. The coherent growth of *m*-plane $\text{In}_{0.36}\text{Ga}_{0.64}\text{N}$ (in spite of its large In fraction) can be attributed to the low growth temperature, which suppresses not only the phase separation reactions, but also the introduction of misfit dislocations. It has recently been found that the activation energy for the introduction of misfit dislocations into III nitrides during two dimensional growth is too large to be overcome at low substrate temperatures [18].

In summary, we have succeeded in the epitaxial growth of high-quality *m*-plane InGaN with In compositions of approximately 36% on ZnO substrates by reducing the growth temperature down to RT by using a PLD growth technique. The FWHM values of the $1\bar{1}00$ XRCs for *m*-plane $\text{In}_{0.36}\text{Ga}_{0.64}\text{N}$ grown on ZnO with the X-ray incident azimuths perpendicular to the *c*- and *a*-axis are 88 arcsec and 78 arcsec, respectively, which indicates that the densities of TDs in the *m*-plane $\text{In}_{0.36}\text{Ga}_{0.64}\text{N}$ grown on ZnO at RT are quite low. RSM has revealed that 50 nm thick *m*-plane $\text{In}_{0.36}\text{Ga}_{0.64}\text{N}$ films grow coherently on ZnO substrates, which is probably the reason for the high crystalline quality of the films.

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