

## High-surface-quality nanocrystalline InN layers deposited on GaN templates by RF sputtering

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We report a detailed study of the effect of deposition parameters on optical, structural, and morphological properties of InN films grown by reactive radio-frequency (RF) sputtering on GaN-onsapphire templates in a pure nitrogen atmosphere. Deposition parameters under study are substrate temperature, RF power, and sputtering pressure. Wurtzite crystallographic structure with *c*-axis preferred growth orientation is confirmed by X-ray diffraction measurements. For the optimized deposition conditions, namely at a substrate temperature of 450 °C and RF power of 30 W, InN films present a root-mean-square surface roughness as low as ~0.4 nm, comparable to the underlying substrate. The apparent optical bandgap is estimated at 720 nm (1.7 eV) in all cases. However, the InN absorption band tail is strongly influenced by the sputtering pressure due to a change in the species of the plasma.

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**1** Introduction Nanocrystalline InN is an interesting material for application in the development of optoelectronic devices like high-efficiency low-cost solar cells, optical coatings, and opto-chemical sensors [1-3]. However, InN has been less studied than other III-nitride semiconductor materials such as GaN and AlN due to its low dissociation temperature ( $\sim$ 500 °C under vacuum) [4]. In order to reduce the decomposition rate, InN should be deposited at low substrate temperatures. From this point of view, reactive radio-frequency (RF) sputtering holds an advantage compared to other well-known growth methods used for InN growth, like molecular-beam epitaxy (MBE), metalorganic vapor phase epitaxy (MOVPE), pulsed laser deposition, and high-pressure chemical vapor deposition (a selection out of many studies can be found in Refs. [5–8]). Among them, reactive sputtering is considered as a promising technique to synthesize nanocrystalline InN films for relatively low-cost technology [9].

Previous work demonstrated the feasibility of obtaining InN layers on glass and sapphire substrates by using reactive sputtering [10–13], and InN deposition on Si(111) substrates has been recently investigated [11, 14, 15]. However, to the best of our knowledge, a systematic study of InN growth on GaN-on-sapphire templates by RF sputtering has not been reported so far. This study is indeed interesting in view of the application of nanocrystalline InN as a low-resistivity coating/contact layer.

In this work, we report the successful growth of InN layers on GaN-on-sapphire templates by reactive RF sputtering. We discuss the effect of various deposition parameters, like substrate temperature, RF power, and sputtering pressure, on the crystal structure, surface morphology, and optical properties of the InN films.

**2 Experimental** The InN layers investigated in this work were deposited in a reactive RF sputtering system equipped with a 2 in. confocal magnetron cathode (AJA International, ATC ORION-3-HV). Substrates were commercial 3.5-µm thick GaN layers grown on sapphire by MOVPE (GaN templates). Pure nitrogen (6N) was used as



reactive gas, whereas the target was a pure indium disk (4N5). The distance between the substrate and the indium target was fixed at 10.5 cm. The base pressure for the deposition was of the order of  $10^{-5}$  mTorr. The nitrogen flux into the deposition chamber was fixed at 14 sccm. The temperature of the substrate was measured using a K-type thermocouple placed 2 mm below the sample holder. To guarantee a uniform temperature, the sample was kept 30 min at the growth temperature before starting the deposition. Reproducibility tests point to a temperature error bar of  $\pm 10$  °C. Prior to the growth, the substrates were chemically cleaned, loaded in the sputtering system and outgassed in the deposition chamber at 600 °C for 30 min.

**3 Results and discussion** A typical X-ray diffraction (XRD) pattern of an InN film deposited on GaN-on-sapphire template is shown in Fig. 1. Only reflections associated to (0001)-oriented InN are observed, together with reflections related to GaN-on-sapphire template. We can hence conclude that all samples under study present a wurtzite crystallographic structure with *c*-axis preferred growth orientation and no parasitic crystallographic orientations within the resolution limit of the XRD system were observed.

From XRD measurements ( $\theta$ -2 $\theta$  scans) of InN on GaN templates, we deduce a *c*-lattice parameter of 5.74 Å for all InN samples under study, indicating a compressive strain of -0.85%. This value is similar to that obtained by Shinoda and Mutsukura [12] for InN samples deposited by RF sputtering on sapphire and glass substrates, but it is slightly higher than the 5.70 Å reported for fully relaxed MBE samples [16].

The influence of growth conditions on structural, morphological, and optical qualities of InN films deposited on GaN-on-sapphire templates has been studied. In particular, we have analyzed the effect of parameters such as substrate temperature, RF power, and sputtering pressure.

**3.1 Influence of substrate temperature** The mobility of the adsorbed species is controlled by the substrate temperature, which should hence have an influence



Figure 1 XRD pattern of an InN film deposited at 400  $^\circ\text{C}, 30$  W and 3.5 mTorr.



Figure 2 Dependence of rms surface roughness on substrate temperature for InN layers deposited at 40 W and 3.5 mTorr.

on growth rate and grain size in polycrystalline materials. Therefore, a set of samples varying the substrate temperature from 400 to 550 °C was deposited to evaluate its effect on the layer properties. For all samples, RF power and sputtering pressure were fixed at 40 W and 3.5 mTorr, respectively. The layer thickness is  $\sim$ 150 nm.

Figure 2 shows the evolution of root-mean-square (rms) roughness, obtained from  $2 \times 2 \,\mu m^2$  atomic force microscopy (AFM) images measuring in the tapping mode, with the substrate temperature. The best result is obtained for InN films deposited at 450 °C, which presents an rms roughness of ~3.5 nm for a layer thickness ~180 nm. The InN surface deterioration at higher temperatures (see Fig. 2) is attributed to the increase of the InN desorption rate. On the other hand, for temperatures below 450 °C the InN surface degrades due to the decrease of the mobility of the adatoms.

Figure 3 shows the  $2 \times 2 \mu m^2$  AFM images of InN films deposited at 450 and 550 °C, illustrating a strong increase of the grain size with the substrate temperature. To our knowledge, this is the first study of morphological quality of InN films deposited by RF sputtering on GaN-on-sapphire templates. For comparison, Guo et al. [13] reported recently an rms surface roughness of 2.6 nm for InN deposited by RF sputtering on Si(111) at low substrate temperature (T = 100 °C).

Structural properties have been further analyzed by XRD measurements. The  $\omega$ -scan full width at half-maximum (FWHM) of the (0002) InN reflection peak remains around



Figure 3 (online color at: www.pss-a.com) AFM micrograph of  $2 \times 2 \ \mu m^2$  area of InN layers deposited at (a) 450 °C and (b) 550 °C.

1.6°, which is a factor of 3 narrower than measurements of InN deposited on Si(111) under the same deposition conditions [15]. Finally,  $\theta$ -2 $\theta$  XRD scans allow us to estimate the grain size, *D*, from the Scherrer formula

$$D = \frac{0.9\lambda}{\Delta 2\theta} \cos\theta,\tag{1}$$

where  $\lambda$ ,  $\theta$ , and  $\Delta 2\theta$  are the X-ray wavelength, Bragg diffraction angle, and FWHM of the InN  $2\theta$  reflection, respectively. A grain size of ~33 nm is obtained independently of the deposition conditions, which is consistent with AFM observations and comparable to results reported by Yamaguchi et al. [17] for InN deposited on GaN by MOVPE.

**3.2 Effect of RF power** The increase of the RF power leads to an increase of the deposition rate due to the higher density of ionized atoms in the sputtering atmosphere. Nevertheless, the energy of the active elements increases with the RF power, thus increasing the damage that these species can cause in the growing surface. To study the effect of this parameter, we deposited a set of InN samples in which the RF power was varied between 20 and 45 W, whereas the substrate temperature and sputtering pressure were fixed at 450 °C and 3.5 mTorr, respectively.

Morphological properties of the material improve when decreasing the RF power (see Table 1). The best result is obtained for InN films deposited at low RF power (30 W), which display an rms roughness of the same order as the underlying substrate. Figure 4 shows the  $2 \times 2 \,\mu\text{m}^2$  AFM images of InN films deposited at 30 and 45 W, illustrating a strong increase of the grain height with the RF power. The morphological deterioration for higher RF powers is attributed to surface damage induced by the high energy of the impinging In atoms [18, 19].

X-ray diffraction characterization confirms that the  $\omega$ scan FWHM of the (0002) reflection peak stays below 2.1°, independently of the RF power used in this study; being a factor of 2 narrower than InN samples deposited on Si(111) at the same conditions [15]. This enhancement in terms of crystal quality is attributed to a better substrate/epilayer interface, which could be deteriorated in the case of Si(111) by the formation of amorphous Si<sub>x</sub>N<sub>y</sub> [9]. Finally, for all samples under study a grain size of ~29 nm is obtained.

**Table 1** Morphological characterization of InN samples deposited at different RF powers. Substrate temperature was fixed at 450 °C and sputtering pressure at 3.5 mTorr. The rms roughness of the GaN-on-sapphire template is  $\sim$ 0.3 nm.

RF power (W)	rms roughness (nm)	thickness (nm)
20	1	90
30	0.4	115
40	3.5	180
45	10.5	125



**Figure 4** (online color at: www.pss-a.com) AFM micrograph of  $2 \times 2 \mu m^2$  area of InN layers deposited at different RF powers: (a) 30 W and (b) 45 W.

**3.3 Influence of N<sub>2</sub> sputtering pressure** The variation of the nitrogen pressure in the sputtering process leads to a change in the partial composition of the different N species within the N<sub>2</sub> plasma, which influences the morphological and optical properties of the material [20]. In order to evaluate the effect of this parameter on the layer properties, a set of InN films with sputtering pressure varying from 3.5 to 14 mTorr were deposited. For this set of samples, the substrate temperature and RF power were fixed at 450 °C and 30 W, respectively.

Figure 5 shows the evolution of the growth rate as well as the rms surface roughness estimated on  $2 \times 2 \mu m^2$  scanning areas as a function of the sputtering pressure. The layer thickness of the samples is 130, 114, and 79 nm, for sputtering pressures of 3.5, 7.0, and 14 mTorr, respectively. The increase of N<sub>2</sub> pressure causes a strong deterioration of the InN surface. This effect is attributed to a change of the nitrogen species in the plasma, which has been confirmed by *in situ* monitorization of the spectral lines of the plasma during deposition. An increase of the reactive nitrogen (N<sup>\*</sup>)/ N<sub>2</sub> ratio and a decrease of the deposition rate were observed when increasing the sputtering pressure. Both effects could be related to the reaction of N<sup>\*</sup> with other impurities present in the sputtering chamber and their incorporation into the growing layer.



**Figure 5** Evolution of rms surface roughness and growth rate with the sputtering pressure for InN samples deposited at  $450 \,^{\circ}$ C and  $30 \,$ W.



The lowest rms surface roughness ( $\sim 0.4 \text{ nm}$ ) is obtained for InN samples deposited at low sputtering pressures (3.5 mTorr). For higher N<sub>2</sub> pressures also a decrease of the deposition rate is observed. Nevertheless, the deterioration of the surface is not attributed to the decrease of the growth rate with the pressure, as it is supported by the low rms values obtained in samples deposited at RF power of 20 W, with a growth rate of 60 nm/h (see Table 1).

**3.4 Characterization of optimized layers** The sheet resistance ( $R_s$ ) of InN layers was determined using the four-point probe method; and resistivity values ( $\rho_s$ ) were obtained from  $\rho_s = R_s t$ , where *t* is the film thickness. Optimized InN samples present a resistivity of  $\sim 6.2 \times 10^{-4} \Omega$  cm (sheet resistance  $\sim 48 \Omega/sq$ ), which is consistent with typical resistivity values of polycrystalline InN deposited by RF sputtering ( $\sim 10^{-3} \Omega$  cm [9]).

As an estimation of the concentration of electrically active impurities, the free carrier concentration of the layers was obtained by Hall-effect measurements, being in the range of  $10^{20}$  cm<sup>-3</sup>. As Butcher and Tansley [22] discuss, if all the oxygen was acting as a donor, the oxygen concentration would be ~6%. However, in InN material the solubility of oxygen donor is limited to 0.3% [22]. Thus, the origin of the measured carrier concentration should be attributed not only to the existence of oxygen but also to other impurity species incorporated mainly at the grain boundaries of the InN layers.

Regarding the optical properties of the InN films, roomtemperature optical transmittance was measured under normal incidence with an optical spectrum analyzer (OSA) in the visible/near-infrared spectral range. Figure 6 shows optical transmission spectra of InN samples deposited at 3.5, 7.0, and 14 mTorr, where the GaN-on-sapphire template limits the maximum transmittance to ~80%. Experimental transmission results were compared with theoretical calculations using the transfer matrix method [21] in order to obtain values of bandgap wavelength  $\lambda_{g}$ , ordinary refractive



**Figure 6** Optical transmission measurements of InN samples deposited at sputtering pressures of 3.5, 7.0, and 14 mTorr on GaN-on-sapphire templates. Substrate temperature and RF power were fixed at  $450 \text{ }^{\circ}\text{C}$  and 30 W, respectively.

index  $n_0(\lambda)$ , and absorption coefficient  $\alpha_0(\lambda)$ . First-order Sellmeier dispersion formula was considered for refractiveindex calculations in the transparent region. Besides, a sigmoidal approximation was used for the absorptioncoefficient estimation [21]. The estimated apparent optical bandgap energy is  $\sim$ 720 nm (1.72 eV) for samples deposited at 3.5 mTorr. This bandgap value is similar to the apparent optical bandgap obtained by other authors in InN samples deposited by reactive sputtering [22], and it is much larger than the well-established values of 0.7-1.0 eV reported for high-quality single-crystalline InN grown by MBE or MOVPE [9]. This large apparent optical bandgap is attributed to the polycrystalline nature of the InN material and due to the high free electron concentration of the layers  $(\sim 10^{20} \text{ cm}^{-3})$ , as observed by other authors [23]. At the same time, from these simulations an absorption coefficient of  $\alpha_0 = 3 \times 10^4 \,\mathrm{cm}^{-1}$  and a refractive index of  $n_0 = 2.4$  were obtained at  $\lambda = 700 \text{ nm}$  for InN samples deposited at 3.5 mTorr. These values are in good agreement with measurements previously reported [24]. For all calculations, the GaN-on-sapphire template refractive index was assumed to be 2.29.

For samples deposited at sputtering pressures above 3.5 mTorr, a blue-shift of the apparent optical bandgap is observed, as shown in the optical transmission measurements of Fig. 6. This blue-shift of the bandgap energy when increasing the working pressure, (and thus when changing the partial composition of N<sub>2</sub> species in the plasma [25]), could be related to an increase of the density of impurities and defects incorporated into the layer during the sputtering process [26]. This phenomenon could be responsible of the change in the absorption band edge, as can be observed in the low-wavelength region of Fig. 6.

Regarding the oxygen contamination of the InN layers, no peak related to the formation of the InON<sub>x</sub> alloy has been observed near the (0002) InN reflection peak in the  $\theta$ -2 $\theta$  scan obtained by XRD measurements (see Fig. 1). We can hence conclude that there is not enough oxygen in the sputtered samples to explain the increase of the apparent optical bandgap energy expected only due to the InON<sub>x</sub> alloy formation [22, 27].

**4 Conclusions** In this work, we have evaluated the influence of deposition conditions, like substrate temperature, RF power, and nitrogen pressure, on the structural, morphological, and optical properties of InN films deposited on GaN-on-sapphire templates by reactive RF sputtering. For all deposition conditions, wurtzite InN films with (0001) preferred growth orientation have been deposited, as has been confirmed by XRD measurements. Regarding the morphological characterization of the samples, best results have been obtained for substrate temperatures of 450 °C and low RF powers (20–30 W). The sputtering pressure seems to strongly influence the InN absorption edge due to a change of the nitrogen species in the plasma. For the optimized deposition conditions, high surface quality InN films have been achieved, with rms roughness as low as  $\sim$ 0.4 nm,

comparable to the surface roughness of the underlying substrate. The optimized InN layers present resistivity values of  ${\sim}6.2 \times 10^{-4}\,\Omega$  cm. The room-temperature optical bandgap has been estimated through transmission measurements  ${\sim}720$  nm (1.7 eV) for InN samples deposited at 3.5 mTorr of N<sub>2</sub> pressure. These results indicate that reactive RF sputtering is a promising deposition technique for obtaining low-cost high-surface-quality nanocrystalline InN films on GaN.

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