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Influence of off-cut angle of (0001) 4H-SiC on the crystal quality of InN grown by RF-MBE

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Abstract

The effect of the off-cut angle of a 4H-SiC (0001) substrate on the growth of InN thick layer by RF-plasma assisted molecular beam epitaxy (RF-MBE) has been investigated. The off-cut angle used in this study was inclined from 0° (just surface) toward the [11-20] direction of 4° and 8°. Crystalline quality and surface morphology were remarkably sensitive to the value of off-angle. Higher off-cut angles result in a reduction of the full-widths at half-maximum of HRXRD (0002) ω -scans, compared to that of the layer on the (0001)-just surface. In addition, the full-widths at half-maximum of μ -Raman scattering spectra at 490 cm⁻¹, which is attributed to E₂ (high) phonon mode, was decreased with increase in off-cut angle. Furthermore, In-droplets, which are commonly observed on the (0001) InN grown surface under In rich-growth condition, were found to be suppressed owing to an improvement of a nucleation on the off-cut angle surface. In our case, the use of 8°-off substrate increased film density and growth rate, while a surface roughness was reduced. These results clearly demonstrate that the larger off-cut angles improve the crystalline quality of InN film with reducing the In-droplets due to a higher step surface density on the off-cut angle surface.

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1. Introduction

Recently, Indium nitride (InN) has received considerable attention because of its narrow band gap [1] and small electron effective mass [2]. Optoelectronic device operating in the wavelength region from UV

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to infrared, including a multi-junction solar cells, can be made by using alloys InGaN, InAlN and InGaAlN, since InN has a direct gap of 0.6 - 0.7 eV [1]. Commonly, sapphires are widely available and widely used as a substrate for the growth of InN film [3, 4] as well as other III-nitrides [5, 8]. However, silicon carbide (SiC) is also a promising candidate substrate because of its high electrical conduction and high thermal conductivity, which is useful for device application. The lattice mismatch and thermal mismatch between InN and SiC substrates are lower than those of InN and sapphire substrates. These make it suitable for the growth of InN film.

III-nitride semiconductors grown on commonly used (0001) substrates, which is induced to a high density dislocation, due to the large lattice mismatch and thermal mismatch between InN and SiC substrates. To increase the efficiency of these optoelectronics devices, it is necessary to improve the surfaces morphology and crystalline quality of the film. The use of off cut-angle substrates has been popular strategies to improve surface morphology *c*-plane AlN [6] and *c*-plane GaN [7] and crystalline quality *a*-plane [8]. This is due to a higher step surface density of the off-cut angle surface, which improves a nucleation. Thus, to improve *c*-plane InN crystalline quality, a 4H-SiC substrates with off-cut angles 4° and 8° were used in this work. The effect of the off-cut angle of a 4H-SiC (0001) substrate on the surface morphology and the crystalline quality of InN films grown by RF-MBE is addressed.

2. Experimental

InN films used in this work were grown under identical growth condition by RF-plasma assisted molecular beam epitaxy (RF-MBE, at Saitama University, Japan) on 4H-SiC (0001) substrates with 0° (just surface), 4° and 8° off-angles. Figure 1 shows a schematic side view of the off-cut substrate used in this study. The off-cut substrate surface was inclined by 4° and 8° from a just surface toward the [11-20] direction. Before the growth of InN film, a thermal cleaning was performed at 1100°C for 30 min. We used low-temperature InN as nucleation layers. The nucleation layer growth temperature and thickness were 350°C and 5 nm, respectively. After the growth of InN nucleation layers, 500 nm-thick InN film were grown at 500°C for 90 min. Indium flux and N₂ flow rate were maintained at 5.0×10⁻⁵ Pa and 1.6 sccm, respectively. An RF-plasma power was in the range of 370-400W.

The surface and cross-section of all films were characterized by scanning electron microscopy (SEM) and atomic force microscopy (AFM). The crystalline qualities of InN films were analysed by high resolution X-ray diffraction (HRXRD) and μ -Raman spectroscopy. The 514.5 nm line of an Ar⁺ laser was used as an excitation light source. The excitation laser beam was focused by a microscope lens system yielding a spot size ~2 μ m in diameter.

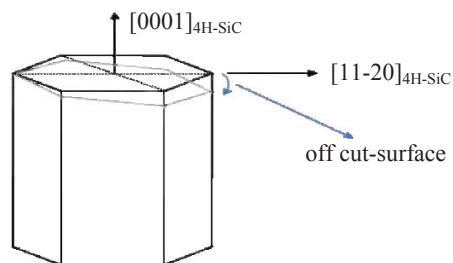


Fig. 1. Schematic side view of misorientation substrate used in this work

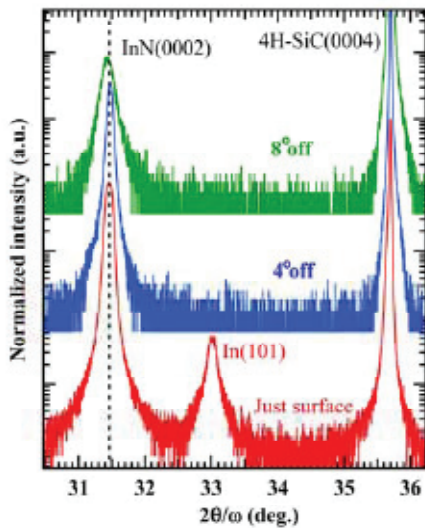


Fig. 2. HRXRD (0002) $2\theta/\omega$ scan profiles of InN films grown on 4H-SiC (0001) substrates with just surface, 4° and 8° off-angles

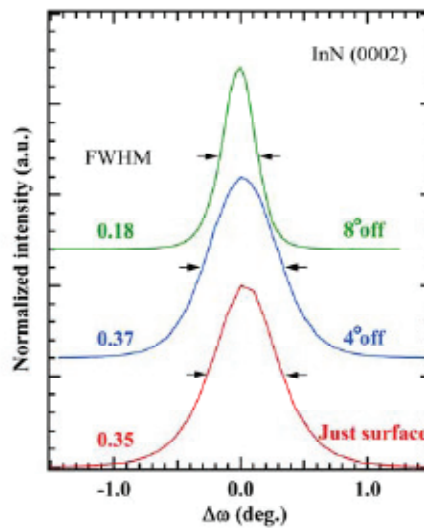


Fig. 3. HRXRD (0002) ω -scans of InN grown on 4H-SiC (0001) substrates with just surface, 4° and 8° off-angles

3. Results and discussion

Figure 2 illustrates HRXRD (0002) $2\theta/\omega$ scan profiles of the InN films grown on the (0001)-just and off-cut 4H-SiC substrates. The reflected peaks were observed at 31.46° and 35.70° , which are corresponding to a diffraction from the InN (0002) and 4H-SiC (0004) planes, respectively. This result indicates the epitaxial relationship between InN and 4H-SiC as $[0001]_{\text{InN}}/[0001]_{4\text{H-SiC}}$. The reflected peak observed at 33° corresponds to the (101) diffraction from the metallic In droplets, occurring for the InN film grown under In-rich growth condition [9]. The In (101) reflected peak was disappeared for the InN films grown on the off-cut 4H-SiC (0001) substrates. HRXRD (0002) ω -scans and their Full width at half maximum (FWHM) of the InN (0002) reflected peaks are shown in Fig. 3. The narrowest FWHM's value was 0.18° for the film on the 8° off-cut angle, indicating that the higher off-cut angles improve the crystalline quality of InN grown film with a reduction of a formation of the In-droplets.

μ -Raman scattering spectra of InN films on (0001)-just and off-cut 4H-SiC substrates are shown in Fig. 4. Raman scattering spectroscopy is used as a tool to identify a distribution of hexagonal and cubic phases in the InN grown films. It is known that cubic phase InN exhibits two Raman-active vibrational modes, namely LO (longitudinal optical) and TO (transverse optical) modes, which are usually observed at zzz and xxx cm^{-1} . On the other hand, for hexagonal InN, the LO and TO modes are represented as the A_1 (LO) and E_2 (high, TO) modes, respectively. They are the strongest ones among the six Raman-active vibrational modes for backscattering along the c -axis. As illustrated in Fig. 4, the intense Raman features have been clearly observed at 490 and 586 cm^{-1} , which are assigned to be InN A_1 (LO) and E_2 (high) modes, respectively. A weak Raman features attributed the A_1 (TO) mode of InN film were observed at 431 cm^{-1} for the (0001)-just and 8° off-cut substrates. On the other hand, the A_1 (TO) mode of InN film on 4° off-cut substrate was observed higher wave number, indicating a contribution of strain in the film. The E_2 (high), A_1 (TO) and A_1 (LO) modes are found to govern the Raman spectra, which are demonstrated that all the InN films have a single-hexagonal-phase. Two phonon modes, E_2 (high) and A_1 (LO), arising from backscattering along the hexagonal c -axis suggest that the grown surface of InN films

exhibit (0001) within the measurement’s sensitivity. On other hand, the weak Raman feature of A_1 (TO) are due to backscattering geometry along other axes except the hexagonal c -axis (are forbidden for backscattering along the hexagonal c -axis). This confirms that the InN films grew along the hexagonal c -axis. These values were consistent with those reports of InN film by Kaczmarczyk *et al.* [10].

FWHM of μ -Raman scattering spectra at 490 cm^{-1} , which dominant E_2 (high) phonon modes are summarized in Table1. FWHM value of E_2 (high) phonon modes as narrow as 3.83 cm^{-1} are obtained for the InN film grown on 8° off-cut angle. This result indicates that FWHM of E_2 (high) phonon modes was decreased with increase in off-angles.

The cross-section SEM images between InN film and the 4H-SiC (0001) substrate are shown in Fig. 5. It is clearly seen that no voids is formed at the InN/SiC interface. However, a formation of the In-droplets was observed for the InN film on the (0001)-just surface, which agrees well with HRXRD results (Fig. 5 (a)). On other hand, In-droplets are not present on the InN surface for the substrates with off-cut angles, as shown in Fig. 5 (b) and (c). This indicates that the larger off-cut angles improve the crystalline quality of InN film with reducing the In-droplets due to a higher step surface density on the off-cut angle surface. It is due to an improvement of a nucleation on the off-cut angle surface.

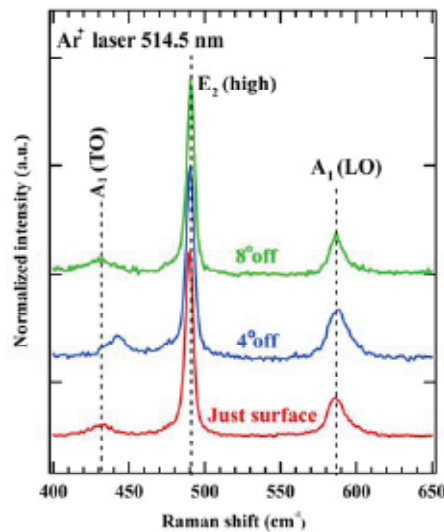


Fig.4. μ -Raman spectra of InN films grown on 4H-SiC (0001) substrates with just surface, 4° and 8° off-angles

Table 1. List of values of FWHM observed for the E_2 (high) phonon mode of InN on 4H-SiC substrates with (0001)-just and 4° and 8° off-cut angles

Substrate types	FWHM of E_2 (high) peak [cm^{-1}]
(0001)-just surface	4.04
4° off-cut angle	4.16
8° off-cut angle	3.83



Fig. 5. Cross-sectional SEM images of InN on 4H-SiC (0001): (a) 0° (just surface), (b) 4° and (c) 8° off-angles

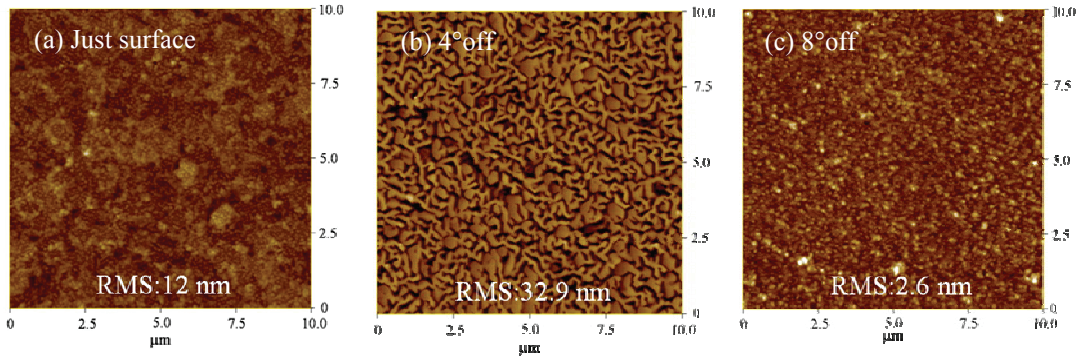


Fig. 6. AFM images showing surface morphologies of InN films on 4H-SiC (0001): (a) 0° (just surface), (b) 4° and (c) 8° off cut-angles

Figure 6 shows AFM images of InN films on 4H-SiC (0001): (a) 0° (just surface), (b) 4° and (c) 8° off cut-angles. Root mean square (RMS) roughness measured by AFM on an area of 10x10 μm is also indicated in Fig. 6. The surface RMS roughness was examined to be 12.0, 32.9 nm and 2.6 nm for the InN films on (0001)-just surface, 4° and 8° off-cut angles, respectively. This also confirms that the use of 8°-off substrate increased film density and growth rate (Figs 5), while the surface roughness RMS is remarkably improved. Growth rate of InN film on the 8° off-cut surface is higher than that on the (0001)-just surface. This implies that higher off-cut angle results in a decreasing terrace width of steps, which produced a higher step density. It is supposed that this reduces a chance of adatom deposition on the terrace and promotes the film density. It is verified by the smooth surface of the InN grown film on the 8° off-cut 4H-SiC substrate. Thus, an increase in growth rate using 4H-SiC substrate with high off-cut angle is expected.

4. Conclusions

We have reported the effect of the off-cut angle of a 4H-SiC (0001) substrate on the growth of InN thick layer by RF-MBE. Results show that the crystalline quality and surface morphology are very sensitive to the value of off-angle. Higher off-cut angles result in a reduction of FWHM of ω -scans of InN (0002), compared with the InN film on the (0001)-just surface. In addition, FWHM of μ -Raman scattering spectra at 490 cm^{-1} , which is attributed to E_2 (high) phonon mode, was decreased with increase in off-angles. Furthermore, In-droplets were found to be suppressed for the films on off-cut angle surface, owing to an improvement of a nucleation on the off-angle surface. InN film on the 8° off-cut substrate exhibited a higher film density and growth rate and a fairly flat surface. These results clearly demonstrate that the larger off-angles improve a nucleation on of InN on the grown surface due to a higher step density, results in a reduction of the In-droplets.

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References

- [1] Davydov V. Yu, Klochikhin A. A, Seisyan R. P, Emtsev V. V, Ivanov S. V, Bechstedt F, Furthmuller J, Harima H, Mudryi A.V, Aderhold J, Semchinova O, Graul J. Absorption and Emission of Hexagonal InN Evidence of Narrow Fundamental Band Gap *Phys. Stat. Sol.* 2002; B **229**: R1-R3.
- [2] Bhuiyan A. G, Hasimoto A, Yamamoto A. Indium nitride (InN): A review on growth, characterization, and properties. *J. Appl. Phys.* 2003; **94**: 2777-2808.
- [3] Xiao H, Wang X, Wang J, Zhang N, Liu H, Zeng Y, Li J, Wang Z. Growth and characterization of InN on sapphire substrate by RF-MBE. *J. Cryst. Growth* 2005; **276**: 401–406.
- [4] Dimakis E, Domagala J.Z, Delimitis A, Komninou Ph, Adikimenakis A, Iliopoulos E, Georgakilas A. Structural properties of 10 μm thick InN grown on sapphire (0001). *Superlattices and Microstructures* 2006; **40**: 246–252.
- [5] Yang Y.G, Ma H. L, Xue C. S, Zhuang H. Z, Hao X. T, Ma J. Investigation of preparation and characterization of GaN films on sapphire (0001) substrates. *Applied Surface Science* 2002; **202**: 295–300.
- [6] Nagamatsu K, Okada N, Kato N, Sumii T, Bandoh A, Iwaya M, Kamiyama S, Amano H, Akasaki I. Effect of *c*-plane sapphire misorientation on the growth of AlN by high-temperature MOVPE. *Phys. Stat. sol.* 2008C; **5**: 3048–3050.
- [7] Lu D, Florescu D. I, Lee D. S, Merai V, Ramer J. C, Parekh A, Armour E. A. Sapphire substrate misorientation effects on GaN nucleation layer properties. *J. Cryst. Growth* 2004; **272**: 353–359.
- [8] Shibata T, Asai K, Nakamura Y, Tanaka M, Kaigawa K, Shibata J, Sakai H. AlN epitaxial growth on off-angle *R*-plane sapphire substrates by MOCVD. *J. Cryst. Growth* 2001; **229** 63–68.
- [9] Kuntharin K, Sanorpim S, Nakamura T, Katayama R, Onabe K. Structural Investigation of Cubic-phase InN on GaAs (001) Grown by MBE Under In- and N-rich Growth Conditions, *Advanced Materials Research* 2008; **31**: 215-217.
- [10] Kaczmarczyk G, Kaschner A, Reich S, Hoffmann A, Thomsen C. Lattice dynamics of hexagonal and cubic InN: Raman-scattering experiments and calculations. *Appl. Phys. Lett.* 2000; **76**: 2122-2124.