RF-MBE growth of cubic InN films on MgO (001) substrates

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Cubic InN films have been grown on MgO substrates with cubic GaN underlayers by RF-N_2 plasma MBE. By changing the growth conditions, we clarified the growth temperature and In flux dependence of the quality of cubic InN films. It was found the surface of cubic InN films grown at relatively higher temperatures was smooth and that the hexagonal phase content in the InN films decreased with increasing In flux. Based on the findings, we have successfully obtained a c-InN film with high phase-purity and a smooth surface. We have carried out micro Raman scattering measurements for the high-quality cubic InN film. Raman peaks were clearly observed at 596 cm^{-1} and 467 cm^{-1}, which are attributed to longitudinal optical and transverse optical phonon modes of cubic InN, respectively.

1 Introduction

It is well known that III nitride semiconductors have usually hexagonal wurtzite structure unlike other III-V compound semiconductors which crystallize in the cubic zincblende structure. However, cubic III nitride semiconductors can be grown when (001) surfaces of cubic crystals, such as GaAs and 3C-SiC, are used as substrates. Cubic III nitride semiconductors have several advantages for electronic and optelectronic devices applications compared with hexagonal ones, for example, higher carrier mobility due to less lattice scattering, better doping properties and superior optical properties [1]. In addition, when zincblende semiconductors are grown on (001) surfaces, there exists no piezoelectric field, which often arises in strained wurtzite semiconductors and leads to lowering of radiative recombination probability due to the spatial separation of electrons and holes.

Although a number of papers have reported the growth and characterization of cubic GaN (c-GaN), AlN and their alloy [2–9], the growth of cubic InN (c-InN) films has been less reported [10–12]. Furthermore, little has been reported on the crystal quality and physical properties of grown c-InN films [13, 14]. In the present study, we report on RF-N_2 plasma molecular beam epitaxy (MBE) growth of c-InN films on MgO substrates with c-GaN underlayers. By changing the growth conditions, we clarified the growth temperature and In flux dependence of the quality of c-InN films. We have also performed micro Raman scattering measurements for high phase-purity c-InN obtained under the optimized growth conditions.

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2 Experimental procedures

Cubic InN films were grown on MgO (001) substrates by MBE using an RF-N_2 plasma source. Prior to the growth of c-InN epilayers, c-GaN underlayers with a thickness of 400 nm were grown on MgO at 700 °C. The hexagonal phase content in the GaN underlayers was 1-2%, and the surface of the c-GaN underlayers was very smooth. After the growth of c-GaN underlayers, c-InN films were grown for 1 hour at 300-550 °C with various In fluxes. The surface structure of c-InN films during growth was observed using reflection high energy electron diffraction (RHEED). The crystal structure and surface morphology were investigated by X-ray diffraction (XRD) and scanning electron microscopy (SEM), respectively. The hexagonal phase content in the GaN and InN layers was estimated from the relative XRD intensity [12, 15]. We have carried out micro Raman scattering measurements for c-InN films at room temperature. The 632.8 nm line of a He-Ne laser was used as the light source. An intensified charge-coupled device camera was used as the detector.

3 Results and discussion

Figures 1 (a) and (b) show RHEED patterns of c-InN films grown at 500 °C and 300 °C, respectively. As shown in these figures, a streak feature was observed for the c-InN film grown at 500 °C, showing the surface was very smooth. A streak feature was observed also at 550 °C. On the other hand, a spot-like pattern was seen at 300 °C. In addition, diffraction spots from hexagonal phase were observed at 300 °C. From the SEM observation, the surface of cubic InN films grown at 500 °C and 550 °C was very smooth although that grown at 300 °C was rough. These indicate the surface morphology is strongly affected by the substrate temperature which controls the migration length of adatoms and that the growth at relatively high temperatures is needed for obtaining a smooth surface.

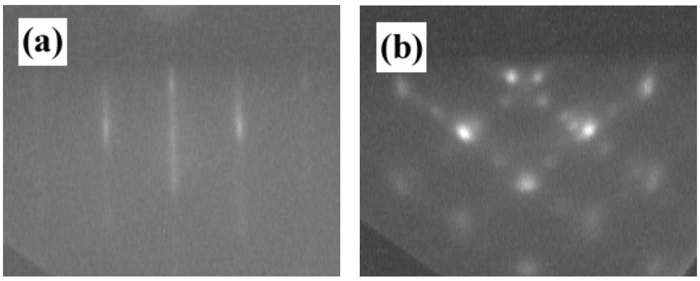

Fig. 1 RHEED patterns of c-InN films at (a) 500 °C and (b) 300 °C.

Figure 2 shows the XRD θ-2θ scan profile for a c-InN film grown at 480 °C on MgO substrate with a c-GaN underlayer. The peaks observed at 35.8°, 39.8° and 42.9°correspond to c-InN (002), c-GaN (002) and MgO (002), respectively. Lattice constant of c-InN is calculated to be 0.502 nm from the Bragg angles. The value for c-InN is somewhat larger than those previously reported [11, 13]. This discrepancy can be explained by in-plane compressive strain in the nitride layer due to the larger thermal expansion coefficient of MgO. Figure 3 shows the XRD rocking curves of c-InN (002) films grown at various temperatures. With increasing temperature, the rocking curve of c-InN (002) becomes narrower. The full-widths at half maxima of the rocking curve of c-InN grown at 500 °C and 550 °C were 46 arcmin. Consequently, growth at relatively high temperature is found to improve not only the flatness of films but also the crystalline quality of c-InN.

Figure 4 shows the hexagonal phase content in the InN films as a function of normalized In flux, where the substrate temperature, N_2 flow rate, and RF power were 480 °C, 1.6 sccm, and 400 W, respectively. The hexagonal phase content in the InN films is found to drastically decrease with increasing In flux. Considering that a larger In flux corresponds to a lower V/III ratio, a similar trend has been reported for the growth of c-GaN [4, 5]. Compared with the growth of hexagonal nitride semiconductors, it is likely that group III atom-rich conditions are commonly necessary for the growth of cubic nitride semiconductors.

Thus, the quality of c-InN films is strongly dependent on the substrate temperature and V/III ratio. It is found that both a low V/III ratio and a relatively high temperature are essential to the growth of c-InN films with high phase-purity, high crystalline quality and a smooth surface. Based on the findings, we have successfully obtained a c-InN film with cubic phase-purity is 95% and a smooth surface under the conditions that the substrate temperature and normalized In flux was 500 °C and 2.5, respectively.

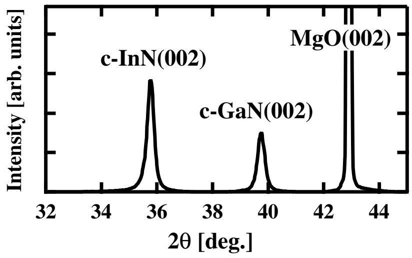

Fig. 2 XRD curve from a c-InN film grown on MgO substrate with a c-GaN underlayer.

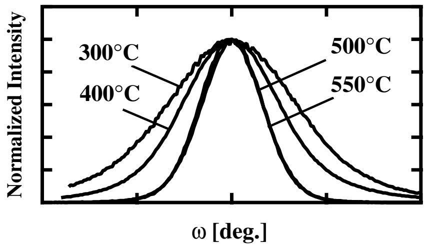

Fig. 3 XRD rocking curves of c-InN (002) films grown at various temperatures.

Table 1 Phonon frequencies of InN and GaN.

Phonon mode	Present work	InN	GaN
E_2 (low)		88 [b], 87 [c]	144 [e]
A_1 (TO)		440 [b], 447 [c]	535 [d]
*cubic (TO)	467	457 [a], 472 [b]	552 [d]
E_1 (TO)		476 [c]	558 [d]
E_2 (high)	489	490 [b], 488 [c]	569 [d]
A_1 (LO)	591	590 [b], 586 [c]	734 [d]
cubic (LO)	596	588 [a], 586 [b]	738 [d]
E_1 (LO)		593 [c]	740 [d]

[a] Ref. [13], [b] Ref. [14], [c] Ref. [17], [d] Ref. [7], [e] Ref. [18].

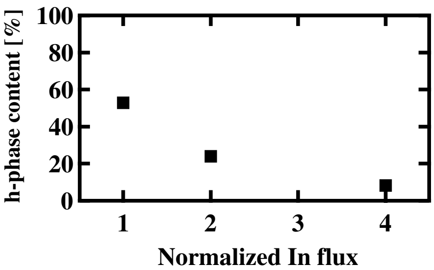

Fig. 4 Hexagonal phase content in the InN films as a function of normalized In flux.

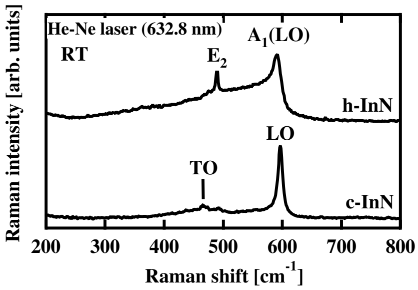

Fig. 5 Raman spectra obtained from c-InN and h-InN.

A Raman spectrum obtained from the c-InN film with a cubic phase-purity of 95% is shown in Fig. 5. For comparison, the Raman spectrum of hexagonal InN (h-InN) grown on 3C-SiC substrate [16] is also shown in this figure. In the spectrum of h-InN, A_1 longitudinal optical (LO)-mode and E_2 (high)-phonon modes were clearly observed at 489 cm^{-1} and 591 cm^{-1}, respectively. On the other hand, in the spectrum of the c-InN, two peaks were observed at 596 cm^{-1}, 467 cm^{-1}, which are different from those observed for h-InN. Judging from the high phase-purity of our sample, the peak observed at 596 cm^{-1} can be identified as the cubic LO phonon mode. This peak is much more clearly observed than those in previous reports [13, 14], and the full width at half-maximum of this peak is 9 cm^{-1}. This indicates that the crystalline quality of c-InN obtained in this study is considerably high. Table 1 lists the observed phonon frequencies of InN and the phonon frequencies of GaN for the purpose of reference. The value of cubic LO phonon mode frequency in the present work is larger than those in previous works. This is partly because the c-InN films used in this study are compressively strained as already mentioned. If the magnitude relation of phonon frequencies of GaN listed in Table 1 is applicable to InN in a similar way, however, it is reasonable that the frequency of the cubic LO phonon mode is larger than that of hexagonal A_1 (LO) phonon mode.

4 Conclusions

Cubic InN films were grown on MgO substrates with c-GaN underlayers by RF-N_2 plasma MBE. The quality of c-InN films was largely affected by both the growth temperature and V/III ratio. With increasing growth temperature, the surface morphology and crystalline quality of c-InN were improved. In addition, with decreasing V/III ratio, the hexagonal phase content of the InN films decreased. Based on this dependence, we have successfully obtained a c-InN film with high phase-purity and a smooth surface. We performed micro Raman scattering measurements for the high-quality cubic InN film. Raman peaks were clearly observed at 596 cm^{-1} and 467 cm^{-1}, which can be identified as cubic LO and TO mode phonon, respectively.

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