

# RF-MBE growth of *a*-plane InN on *r*-plane sapphire with a GaN underlayer

G. Shikata<sup>a,\*</sup>, S. Hirano<sup>a</sup>, T. Inoue<sup>a</sup>, M. Orihara<sup>a,b</sup>, Y. Hijikata<sup>a</sup>, H. Yaguchi<sup>a,b</sup>, S. Yoshida<sup>a,b</sup>

<sup>a</sup>Department of Electrical and Electronic Systems Engineering, Faculty of Engineering, Saitama University, 255 Shimo-Okubo, Sakura-ku, Saitama-shi, Saitama 338-8570, Japan

<sup>b</sup>CREST, JST, 4-1-8 Hon-cho, Kawaguchi-shi, Saitama 332-0012, Japan

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## Abstract

*a*-Plane InN films were grown on *r*-plane sapphire substrates with GaN underlayers by RF-N<sub>2</sub> plasma molecular beam epitaxy (MBE). X-ray diffraction (XRD) measurements and reflection high-energy electron diffraction (RHEED) observation revealed that the InN films were grown with InN (1 1  $\bar{2}$  0) || GaN (1 1  $\bar{2}$  0) || sapphire (2  $\bar{2}$  0 4). We have carried out micro-Raman scattering measurements for *a*-plane InN film. Raman peaks were observed at 448, 490 and 598 cm<sup>-1</sup>, which can be identified as the A<sub>1</sub> transversal optical (TO), E<sub>2</sub> (high) and E<sub>1</sub> longitudinal optical (LO) mode phonon, respectively. We also carried out photoluminescence (PL) measurements for *a*-plane InN film. Strong PL was observed between 0.62 and 0.65 eV, which is the lowest ever reported for InN. It is suggested from the excitation intensity and temperature dependence of the PL spectra that the red shift of the PL peak is due to the Franz–Keldysh effect.

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## 1. Introduction

InN is a very attractive material for high-performance optoelectronic devices operating at optical-communication wavelength of 1.55 μm because of its narrow band gap of 0.6–0.7 eV [1]. InN has hexagonal wurtzite crystal structure, and is most commonly grown along the (0001) orientation. Spontaneous and piezoelectric polarization-induced electric fields along the polar *c*-axis, however, adversely affect the performance of optoelectronic devices. A useful approach for reducing the deleterious effects of the built-in field is to grow along the nonpolar directions. The growth of nonpolar GaN, such as *m*-plane and *a*-plane GaN, was intensively studied for improving the performance of light-emitting devices. On the other hand, there were a few reports concerning the growth of nonpolar InN, whose quality has still left room for improvement [2]. In this study, we report the growth of *a*-plane InN on *r*-plane

sapphire substrates by RF-molecular beam epitaxy (MBE) and its intriguing photoluminescence (PL) properties.

## 2. Experimental procedures

Nonpolar (1 1  $\bar{2}$  0) *a*-plane InN films were grown on (1  $\bar{1}$  0 2) *r*-plane sapphire substrates by MBE using an RF-N<sub>2</sub> plasma source. A series of samples were grown at different substrate temperatures between 700 and 1000 °C to investigate the temperature dependence of the crystal quality and surface morphology of the GaN films. As a result, we could obtain *a*-plane GaN films with a flat surface and good crystal quality when grown at 900 °C. Thus, before the growth of the *a*-plane InN layers, *a*-plane GaN underlayers with a thickness of 500 nm were grown on *r*-plane sapphire at 900 °C with a nitrogen flow rate of 1.6 sccm and an RF plasma power of 350 W. It is important to use a GaN underlayer, because InN films with a predominant zincblende (cubic) structure forms on *r*-plane sapphire, if the underlayer is not used [3]. The GaN layers were confirmed to be (1 1  $\bar{2}$  0) *a*-plane by *in situ* reflection

\*Corresponding author.

E-mail address: [shikata@opt.ees.saitama-u.ac.jp](mailto:shikata@opt.ees.saitama-u.ac.jp) (G. Shikata).

high-energy electron diffraction (RHEED) observation and X-ray diffraction (XRD) measurements after the growth. The surface of the *a*-plane GaN underlayers was smooth. Following the GaN layer growth, the substrate was cooled down to the InN growth temperature. The InN layers were grown for 1 h at 450–600 °C with a nitrogen flow rate of 1.6 sccm and an RF plasma power of 350 W. The surface structure of InN films during growth was observed using RHEED. The crystal structure and surface morphology were investigated by XRD and scanning electron microscopy (SEM), respectively. We have carried out PL measurements for *a*-plane InN films at between 15 and 300 K and micro-Raman scattering measurements for *a*-plane InN films at room temperature. The 532 nm frequency doubled line of a Nb:YVO<sub>4</sub> laser was used as an excitation source and an InSb photovoltaic device was used as a detector.

### 3. Results and discussion

The growth processes were monitored by RHEED. In the RHEED observation, a streak feature was observed for the GaN film grown at 900 °C, showing the surface was smooth. The SEM observation also confirmed that the surface of the GaN layers obtained in this study was flat. On the other hand, a spot pattern was observed for the InN film grown at 450 °C. A spot pattern was observed also grown at 500 and 550 °C. However, no RHEED pattern for InN was seen at 600 °C, which seems to be too high for the growth of InN films. From the SEM observation, the surface of the InN layers obtained in this study was not uniform. Large lattice mismatch between InN (11 $\bar{2}$ 0) and GaN (11 $\bar{2}$ 0) of 10% in the [0001] direction and 11% in the [1 $\bar{1}$ 00] direction probably cause the three-dimensional (3D) growth mode.

Fig. 1 shows the XRD  $\theta$ – $2\theta$  scan of InN grown at 450 °C on *r*-plane sapphire with a GaN underlayer. Only the XRD peak from wurtzite-type InN (11 $\bar{2}$ 0) is observed

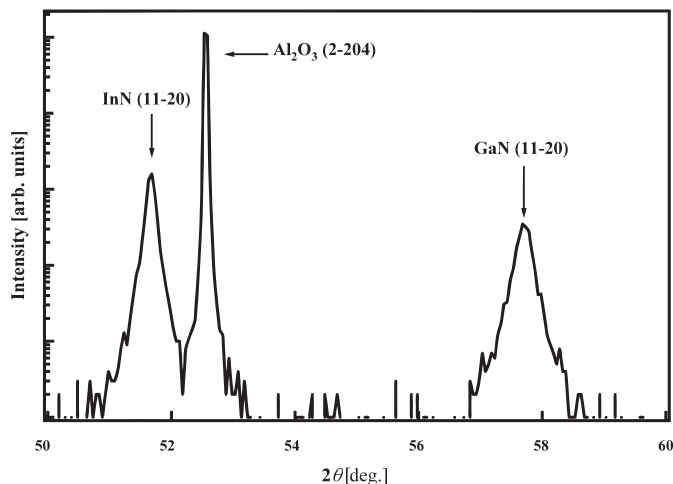


Fig. 1. XRD  $\theta$ – $2\theta$  scan of the InN film grown on *r*-plane sapphire with a GaN underlayer.

as well as that from sapphire (2 $\bar{2}$ 04). This shows that InN was grown with InN (11 $\bar{2}$ 0)∥GaN (11 $\bar{2}$ 0)∥sapphire (2 $\bar{2}$ 04). The full-width at half-maximum (FWHM) of the rocking curve of InN grown at 450 °C was 62 arcmin.

Fig. 2 shows a Raman spectrum obtained from the *a*-plane InN film grown at 450 °C. Three peaks were observed at 448, 490 and 598 cm<sup>−1</sup> in the spectrum, which are different from that observed for *c*-plane InN [4]. Since these Raman peaks correspond well with those previously reported [5,6], the peaks observed at 448, 490 and 598 cm<sup>−1</sup> can be identified as the A<sub>1</sub> transversal optical (TO), E<sub>2</sub> (high) and E<sub>1</sub> longitudinal optical (LO) phonon modes, respectively. It is found from the Raman selection rules for the wurtzite structure that *a*-plane InN films were successfully formed. It should be noted that the Raman peaks due to the polytypes, such as cubic and 4H, which were often reported for *a*-plane GaN and AlN [7,8], were not observed, indicating that the density of the basal-plane stacking faults (BSFs) is not so high.

From the 15 K PL spectrum of *a*-plane InN grown at 500 °C, a strong PL peak was clearly observed at 0.628 eV with FWHM of 64 meV. This peak energy has been the lowest for InN until now. For *a*-plane GaN, some groups have reported that the intensity of low-energy PL peaks correlated with the BSFs, which are parallel to the (0001) *c*-plane [8,9]. The nonpolar GaN, even in the layers grown by lateral epitaxial overgrowth technique, typically has a high density of BSFs, which often intersect the entire thickness of the layers. The BSF in GaN can be regarded as a very thin zincblende layer mixed in the wurtzite matrix or as a polytypic quantum well. Thus, one explanation for this red shift of the PL peak is that there exist BSFs in *a*-plane InN films grown in this study. Another possible explanation is the Franz–Keldysh effect. Although the feature of *a*-plane nitride films is no polarization field along the growth direction, the polarization field can exist along in-plane [0001] direction due to spontaneous and piezoelectric polarizations. Furthermore, since the InN films were grown in the 3D growth mode in this study, they tend to be polarized in the direction of [0001]. Thus, such a

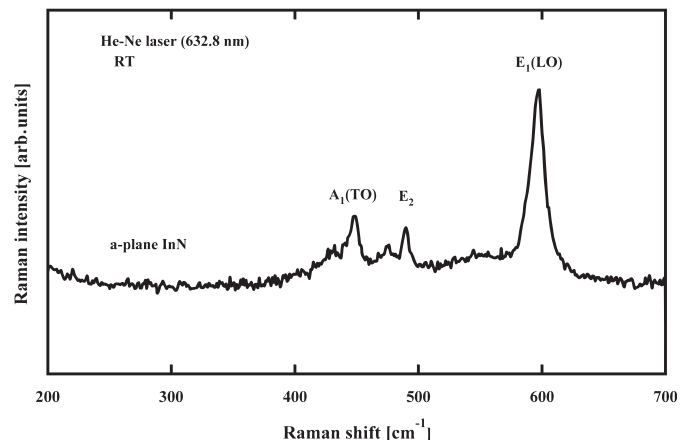


Fig. 2. Raman spectra obtained from *a*-plane InN.

polarization field causes a band bending and red shift of the PL emission.

In order to clarify the nature of emissions in *a*-plane InN, we carried out PL measurements at different excitation intensities and temperatures. Fig. 3 shows the excitation intensity dependence of PL spectra of *a*-plane InN at 15 K. The PL peak exhibited a large blue shift ( $\sim 15$  meV) with increasing excitation intensity over three orders of magnitude (see the inset in Fig. 3). Also for *a*-plane GaN with BSFs, PL spectra exhibited a similar blue shift with increasing excitation intensity over three orders of magnitude. However, the shift was considerably smaller ( $\sim 3$ – $4$  meV) than that observed in this study.

On the other hand, a characteristic parameter  $\Delta W$  of the Franz–Keldysh effect under field  $E$  [10] is expressed as

$$\Delta W = \left[ \frac{\hbar^2 e^2 E^2}{2m_r} \right]^{1/3}, \quad (1)$$

where  $e$  and  $m_r$  are the electron charge and the reduced mass for the electron and hole, and  $\Delta W$  can be used as a rough standard for the red shift of PL. For GaN, the internal electric field  $E$  is about 200 kV/cm [11]. If the magnitude of  $E$  of GaN is applicable to InN in similar way,  $\Delta W$  is estimated to be about 40 meV, which is in reasonably good agreement with the red shift of the PL peak. The blue shift with increasing excitation intensity can be also explained by the fact that the energy bands become flat due to the screening effect by photo-generated carriers.

Fig. 4 shows the temperature dependence of PL spectra of *a*-plane InN at  $4 \text{ W/cm}^2$ . With increasing temperature, as shown in this figure, the PL intensity at low energies became weaker than that at high energies. If the lower-energy PL emission is due to the BSFs, which can be regarded as polytypic quantum wells, this emission is expected to disappear with increasing temperature because

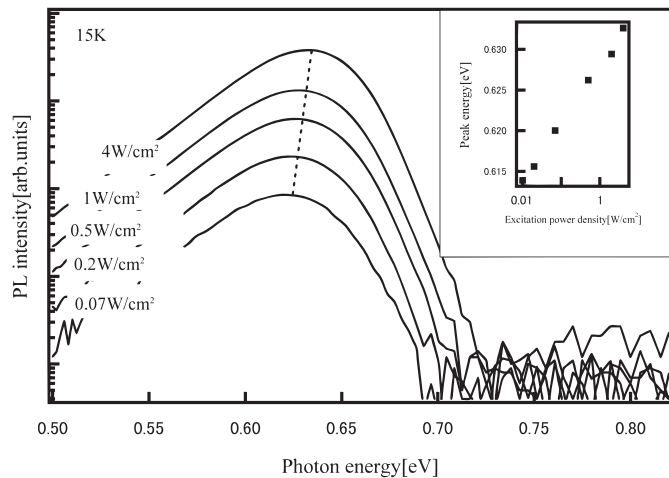


Fig. 3. PL spectra of *a*-plane InN at different excitation intensities at 15 K. With increasing excitation intensity, the PL spectra shifted to higher energies. The inset is excitation power dependence of the peak energy position.

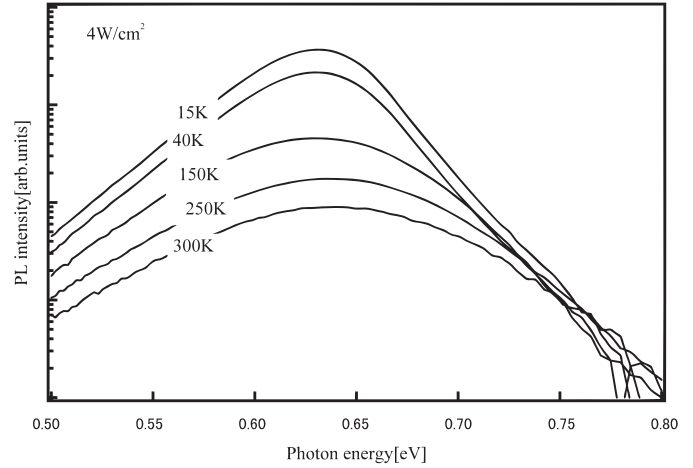


Fig. 4. PL spectra of the *a*-plane InN measured at different temperatures.

carriers escape from the BSFs with a low density. However, lower-energy emission was still observed at high temperatures, which indicates that it cannot be attributed to the BSFs. For the Franz–Keldysh effect, lower-energy transition has smaller transition probability, in other words, longer radiative lifetime than higher-energy transition because the lower-energy transition corresponds to the transition between electrons and holes which are more largely distant from each other. With increasing temperature, the higher-energy transition remains at high temperatures, but the lower-energy transition with a small transition probability disappears behind nonradiative recombination. Therefore, if the red shift of the PL peak is due to the Franz–Keldysh effect, the temperature dependence of the PL spectra can be also explained well. However, further investigation is required to clarify the reason for the lower-energy PL emission observed for *a*-plane InN.

#### 4. Conclusions

*a*-Plane InN films were grown on *r*-plane sapphire substrates with GaN underlayers by RF- $\text{N}_2$  plasma MBE. We found that InN was grown with InN  $(11\bar{2}0)\parallel\text{GaN}(11\bar{2}0)\parallel\text{sapphire}(2\bar{2}04)$  from XRD measurements and RHEED observation. We performed micro-Raman scattering measurements for *a*-plane InN film. Raman peaks were observed at 448, 490 and  $598 \text{ cm}^{-1}$ , which can be identified as the  $A_1(\text{TO})$ ,  $E_2(\text{high})$  and  $E_1(\text{LO})$  mode phonons, respectively, indicating that *a*-plane InN was successfully grown. We also performed PL measurements for *a*-plane InN film at different excitation intensities and temperatures. Strong photoluminescence was observed between 0.62 and 0.65 eV, which is the lowest ever reported for InN. The excitation intensity and temperature dependence of the PL spectra suggest that the red shift of the PL peak is probably due to the Franz–Keldysh effect.

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