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Growth of InN on Ge substrate by molecular beam epitaxy

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Abstract

InN epitaxial growth on a (111)-oriented, Ga-doped germanium substrate using molecular beam epitaxy is described. X-ray diffraction and transmission electron microscopy investigations have shown that the InN epitaxial layer consists of a wurtzite structure, which has the epitaxial relationship of $(0001)_{\text{InN}} \parallel (111)_{\text{Ge}}$. Transmission electron microscopy shows an intermediate layer at the interface between the InN/Ge substrate. Consistent with recent reports implying a narrow bandgap of InN [Phys. Stat Sol. B 229 (2002) R1, Appl. Phys. Lett. 80 (2002) 3967], a strong photoluminescence with peak energy of 0.69 eV at 15 K was observed for this InN epilayer, in contrast to the peak energy of 0.71 eV for Ga-doped Ge under the same measurement conditions.

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1. Introduction

GaN and related III–V materials have attracted enormous attention due to their use in short-

wavelength optical devices, as well as high-power and high-temperature electronic devices [1–3]. Recently, however, InN has also become a material of interest due to the significant finding of the bandgap of InN to be approximately 0.65–0.8 eV [4,5]. The use of InGaN ternary alloys extends the range of optical emission of nitride-based devices from ultraviolet to near-infrared

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regions [6]. Moreover, InN-based devices are promising due to outstanding material properties such as small effective mass, large electrical mobility, and high peak and saturation velocities [6]. In the absence of a lattice-matched choice for a substrate, an alternative criterion for the substrate selection might be based on the energy band suitability for vertical conduction devices. The first criterion for such a selection is a substrate with a comparable bandgap. Germanium has such a bandgap, being within 40 meV of the bandgap of InN [7]. The second criterion is for a suitable and small band offset. Currently, these values are unknown, as the bandgap of InN is in theoretical dispute [4,5]. Future devices such as multi-junction tandem solar cells or heterojunction bipolar transistors and photodetectors could benefit from the use of an optimally selected vertically conducting substrate.

Until now, InN has been less studied compared with other (AlGa)N-related materials due to its low dissociation temperature at high equilibrium vapor pressure of nitrogen, which makes it less suitable to be grown by vapor deposition [8]. Therefore, RF-plasma molecular beam epitaxy (MBE) is an essential technology for obtaining high-quality InN films due to the facilitation of low growth temperatures compared with metalorganic chemical vapor deposition (MOCVD) [8].

InN epitaxial layers have been extensively grown on sapphire substrates. The InN film has a large lattice mismatch of $\sim 25\%$, which creates an extremely high density of structural defects [6,9]. As an alternative, Si (111) substrates, which have a smaller lattice mismatch of 8%, have been used for InN growth [6]. However, previous attempts at using Si substrates were unsuccessful due to the formation of SiN_x layers on the substrate face due to unintentional nitridation [10].

Herein, the growth of InN on p-type (Ga-doped) Ge (111) by RF plasma-assisted MBE is reported. The Ge (111) substrate has a smaller lattice mismatch with InN, of approximately 11.3%. Although the expected dislocation density is still quite large, the use of Ge for vertical conduction devices may be a practical alternative.

2. Experimental procedure

A (111)-oriented epi-ready Ga-doped Ge substrate was mounted on an EPI uniblock using custom spring plates. Upon loading into the introduction chamber, the Ge substrate was outgassed at 320 °C for 1 h. The substrate was then loaded into the MBE growth chamber where a constant substrate temperature of 475 °C was maintained with an In flux of 1.4×10^{-7} Torr beam equivalent pressure (BEP). An EPI-unibulb nitrogen source operated at 350 W and a flow rate of 0.34 sccm was used. The growth was monitored in situ by reflection high-energy electron diffraction (RHEED). The growth rate was 0.2 $\mu\text{m/h}$, with a final film thickness of 0.4 μm .

3. Results and discussions

The optical properties of InN were investigated using photoluminescence (PL) measurements at 15 K with an excitation of ~ 3 mW provided by a near-infrared laser diode operating at 980 nm. As a control, the PL of the Ge substrate was also taken with the same laser at $10 \times$ the excitation. A lock-in detection scheme was used with a computer-controlled monochromator (Spex 270) for spectral analysis of the back-scattered signal from the sample. To minimize distortion of the spectrum, an InGaAs photodetector with extended response to 2.2 μm was used to detect the output signal from the monochromator.

The structural properties of InN were measured with a Philips XPert MRD diffraction (XRD) system. The microstructure of the InN film was characterized by transmission electron microscopy (TEM). Defects such as threading dislocations, grain boundaries and stacking faults were found in the film.

During the growth of InN epitaxial layers, the surface was monitored in situ by RHEED (not shown here). The growth temperature was maintained at 475 °C and the In flux was varied from 1.0×10^{-7} Torr BEP ($T_{\text{In}} = 776$ °C) to 1.4×10^{-7} Torr BEP ($T_{\text{In}} = 790$ °C) in an attempt to obtain smooth RHEED patterns. The RHEED patterns show both streaks and spots for

$T_{\text{In}} = 776^\circ\text{C}$. However, at a higher flux of In ($T_{\text{In}} = 790^\circ\text{C}$), the RHEED pattern maintains a similar mixed streaky and spotty pattern but became dim. According to commonly accepted RHEED pattern interpretations for GaN [11], this dim pattern likely indicates excess indium on the surface of the growing film, while the spotty pattern indicates significant three-dimensional morphology.

The crystallographic structure was studied by double-crystal XRD. Fig. 1 shows $\omega - 2\theta$ XRD scans for the $0.4\ \mu\text{m}$ thick InN grown on Ge substrates. Both (0002) InN and (111) Ge diffraction peaks are clearly observed. The (111) and (0002) peak positions were $\theta = 13.581^\circ$ and $\theta = 15.768^\circ$ for Ge and InN, respectively. This result indicates the epitaxial relationship $[0001]_{\text{InN}} \parallel [111]_{\text{Ge}}$. The best (0002) $\omega - 2\theta$ full width at half maximum (FWHM) values were ~ 144 arcsec. Further characterization of the film was done with XRD pole-figure measurements (not shown here). The pole-figures show peaks with six-fold rotational symmetry, which confirm the presence of hexagonal InN. Wurtzite epitaxial InN has previously been demonstrated on (111)-oriented Si substrates [6]. The hexagonal InN was also confirmed by TEM (see below). The best (0002) rocking curve measurements showed

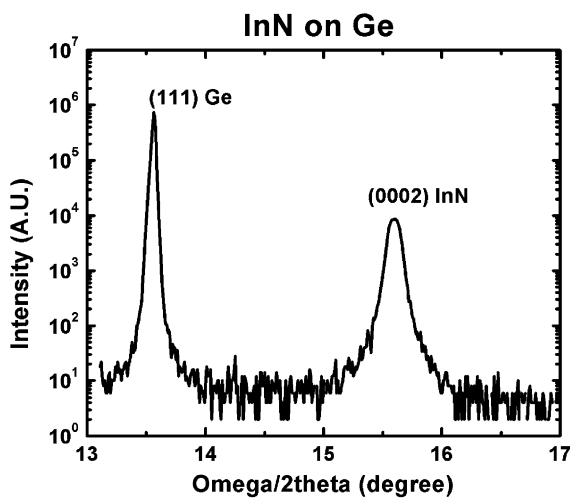


Fig. 1. Omega/2theta of In/Ge showing a (111) Ge peak at 13.581° and an InN peak at 15.768° .

~ 2597 arcsec FWHM, indicating significant tilt and mosaic grain structure.

The microstructure of InN films grown on Ge substrates was characterized by TEM. The diffraction pattern in Fig. 2 was taken at the InN/Ge interface along the $[110]_{\text{Ge}}$ zone axis. The pattern with the bright dots connected by solid lines is due to Ge and the weak spots connected by dotted lines are from InN. The orientation relationship between InN and the Ge substrate shown in Fig. 2 can be described as follows: $[110]_{\text{Ge}} \parallel [11\bar{2}0]_{\text{InN}}$ and $(111)_{\text{Ge}} \parallel (0001)_{\text{InN}}$. This epitaxial relationship between the InN and the Ge substrate was similar to that of InN on Si substrates [6]. From TEM diffraction, the calculated lattice mismatch between InN (0002) and Ge (111) is about 11%. This value is significantly smaller than the lattice mismatch between InN and sapphire ($\sim 25\%$) [6,9].

TEM images were used to analyze structural qualities such as stacking faults and threading, screw, and edge dislocations. Analysis by diffraction contrast microscopy shows that the $0.4\ \mu\text{m}$ InN film contains a high density of threading

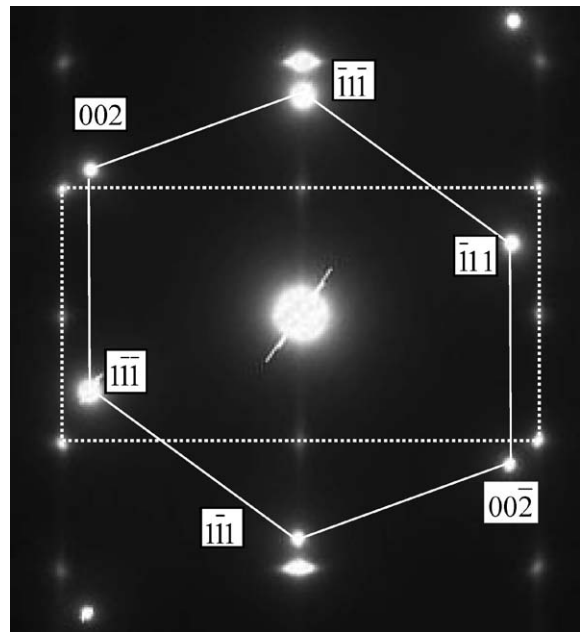


Fig. 2. Selective electron diffraction pattern taken at the interfacial region of InN/Ge along $[110]_{\text{Ge}}$ zone-axis, which is parallel to $[11\bar{2}0]_{\text{InN}}$.

dislocations as well as grain boundaries, as shown in Fig. 3. The dislocations were predominantly screw type with densities in the order of 10^{10} cm^{-2} . The high dislocation density was generated to accommodate misfit strain in the basal planes between InN and the Ge substrate. At the early stage of growth, the film consists of nanoscale columnar crystallites. These crystallites subsequently coalesce into larger crystallites. Fig. 3(a) shows a high threading dislocation density near the interface, which reduces with increasing thickness, similar to that common for GaN epitaxial layers [12].

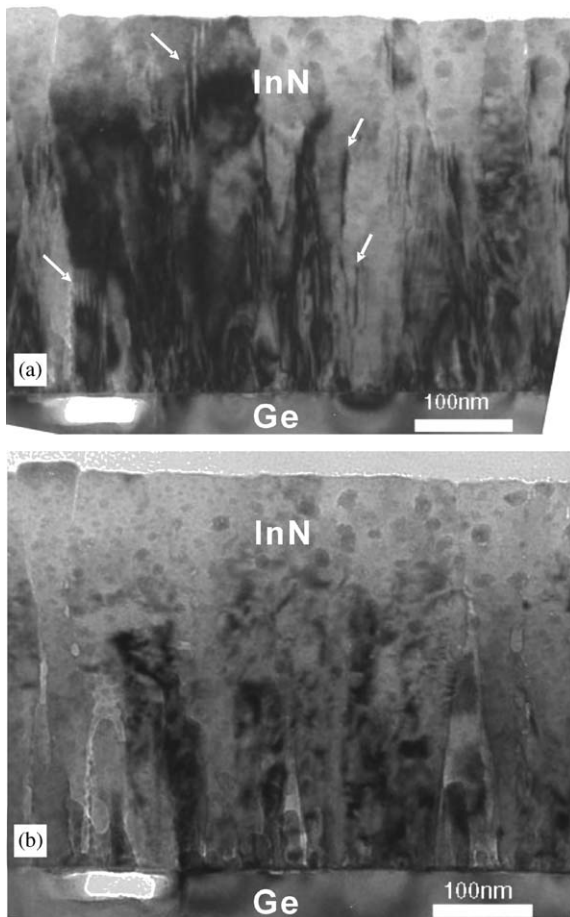


Fig. 3. Bright field diffraction contrast TEM images taken under different conditions to view threading dislocations of (a) screw type, $g = 0002$, and (b) edge type, $g = 1\bar{1}00$.

The cross-sectional TEM images show an intermediate layer between InN and the Ge substrate. This boundary region can be interpreted as an amorphous interlayer of unknown origin or perhaps a low symmetry monoclinic phase possibly resulting from the nitridation of Ge to form $\beta\text{-Ge}_3\text{N}_4$ [13,14]. Alternatively, indium–germanium compounds have a well-documented eutectic below the growth temperature used in these experiments. The intermediate layer may thus be a result of the formation of In–Ge–N alloys. Further evidence of the formation of a eutectic is given by the occasional observation of etch pits in the Ge substrate shown in Figs. 3(a) and (b). These pits suggest that a reaction is occurring between the Ge and a species present during the growth to form a compound with a lower melting temperature.

The 15 K PL spectrum of a $0.4 \mu\text{m}$ InN epitaxial layer grown on a Ge substrate, and that of the Ge control sample are shown in Fig. 4. Strong luminescence was observed around 0.69 eV for the InN epitaxial layer grown on Ge. This value is close to recent reports of a bandgap near 0.7 eV [4,5]. Also note the characteristic low-energy tail of the PL peak, which most likely is due to defects in InN or possibly to a graded bandgap due to the possible presence of In–Ge–N alloys. In contrast,

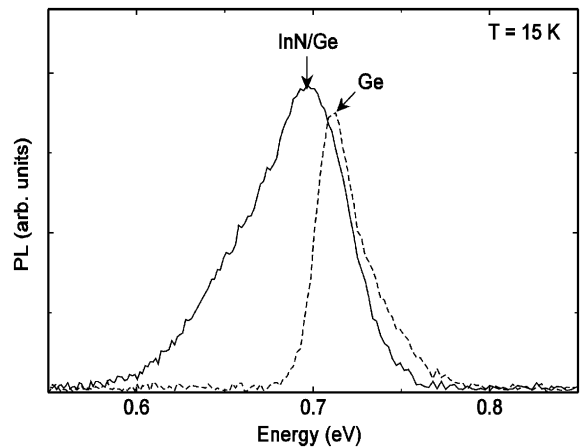


Fig. 4. PL of the InN/Ge sample (solid curve) and the Ge control sample similar to the substrate (dashed curve) taken at 15 K. The PL of the Ge was obtained by excitation $10 \times$ that of the InN/Ge sample. The peak of the InN/Ge is near 0.69 eV , while the peak for the Ge sample is approximately at 0.71 eV .

the Ge standard sample requires $10 \times$ the excitation to show a comparable luminescence at 0.72 eV, which corresponds to the Ge bandgap at this temperature range [7]. Collectively, the difference of PL peak energy, PL line shape and PL intensity between the two materials at 15 K allows differentiation of InN from the Ge substrate.

4. Conclusion

An investigation of InN grown on Ge substrates via plasma-assisted MBE was carried out to study the material characteristics of InN. This effort found that wurtzite InN has been grown with the polar axis parallel to the (111)-oriented Ge substrate, and has a lattice mismatch of 11.3% between InN and the Ge substrate. TEM shows that the dominant structural defects are threading dislocations, stacking faults, and a disordered intermediate layer existing at the interface between the InN layer and the Ge substrate. The threading dislocation density decreased with increasing thickness of InN epitaxial layers. PL at 15 K shows that the InN film grown on Ge is a potential material for infrared optical devices. With the recently reported bandgap of InN in the proximity of that of Ge, InN/Ge is a promising candidate for vertical conduction devices.

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