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In induced reconstructions of Si(1 1 1) as superlattice matched epitaxial templates for InN growth

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ABSTRACT

Indium induced surface reconstructions of Si(1 1 1)-7 × 7 are used as templates to grow high quality InN. We grow InN on Si(1 1 1)-7 × 7, Si(1 1 1)-4 × 1-In and Si(1 1 1)-1 × 1-In reconstructed surfaces and study the quality of the films formed using complementary characterization tools. InN grown on Si(1 1 1)-1 × 1-In reconstruction shows superior film quality with lowest band-edge emission having a narrow full width at half maximum, intense and narrow 0 0 0 2 X-ray diffraction, low surface roughness and carrier concentration an order lower than other samples. We attribute the high quality of the film formed at 300 °C to the integral matching of InN and super lattice dimensions, we also study the reasons for the band gap variation of InN in the literature. Present study demonstrates the proposed Superlattice Matched Epitaxy can be a general approach to grow good quality InN at much lower growth temperature on compatible In induced reconstructions of the Si surface.

1. Introduction

Group III-nitrides form an important class of semi-conducting materials which have technologically materialized and still promises several interesting applications [1]. InN is an important member of this family which has received great attention of the researcher owing to its different properties like lowest effective mass and highest peak and saturation velocities [2]. However, it is least studied compared to other nitrides and the available literature on this potent material shows ambiguities concerning its basic material parameters which is due to the difficulties in synthesizing good quality material [3]. One of the reasons, as for other nitrides, is the non-availability of lattice matched substrate for growth. However, the major reasons for the difficulty in synthesizing high quality InN are its low dissociation temperature and high nitrogen vapor pressure [4]. At high temperatures necessary for epitaxial growth, InN tends to dissociate, resulting in poor crystal quality film [4]. Thus, there is a necessity to identify conditions and methods to grow good quality epitaxial at relatively low temperatures.

Researchers have tried several growth schemes using different techniques to improve the crystalline quality of the InN film epitaxially on sapphire substrate. Among the different schemes used, nitridation of sapphire substrate to form a thin AlN intermediate layer, growth on GaN epilayer, use of low temperature InN buffer layer and alternating low and high temperature layers, are found to be promising [4–6]. Efforts for the incorporation of nitrides into the Si industry have been nominal at best, especially in the case of InN. As in the case of growth on sapphire, substrate nitridation of Si surface and growth of AlN and InN epilayers are found useful for the growth of InN films on Si substrate [7–9]. However, these techniques still rely on the use of high substrate temperature for the InN films. Thus, in search of a novel growth scheme to grow good quality InN on Si substrate at relatively low growth temperatures than usually used, we invoke the possibility of using metal induced Si reconstructions as growth templates [10].

Semiconducting surfaces are endowed with several fascinating ordered reconstructions which exhibit rich structural and electronic properties. Several research groups, including ours have previously shown the formation of several In induced submonolayer superstructural phases on Si(1 1 1) surfaces, viz. Si(1 1 1)-1 × 1-In, Si(1 1 1)-$\sqrt{31} \times \sqrt{31}R \pm 19°$-In, Si(1 1 1)-$\sqrt{3} \times \sqrt{3}R30°$-In, Si(1 1 1)-4 × 1-In [11,12]. Based upon several experiments in UHV and using *in situ* Low Energy Electron Diffraction (LEED) and Auger Electron Spectroscopy (AES) measurements, we have previously

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reported a 2D phase diagram [13] for this system that shows several kinetic pathways for the formation of these reconstructed structural phases that have different 2D lattice parameters. The present study is a demonstration of In induced stable interfacial superstructural phases on Si(1 1 1), as templates for InN growth. We employ here Plasma Assisted Molecular Beam Epitaxy as the preferred technique for InN growth, since the growth temperature chosen can be independent of the N source. The experiments involve firstly, the formation of the In metal induced superstructural phases in the submonolayer regime, followed by growth of epitaxial InN films and characterized by complementary tools. We then correlate the 2D unit cells of the superstructure and InN overlayer in terms of Superlattice Matching Epitaxy (SME). We identify the superlattice unit cell that enables the growth of lattice matched template to grow good quality InN at a reduced growth temperature. The consequence of the difficulty in synthesizing good quality InN films is the ambiguity in determining its different material properties [14]. Literature shows a wide spectrum of band gap values (0.6–2 eV) for InN and several reasons to account for its variability, which has as much confused as helped to shed light on this issue [14]. Our systematic investigation using several complementary characterization probes enables us to suggested reasons for the band gap variation and help towards assigning a proper band gap value for InN.

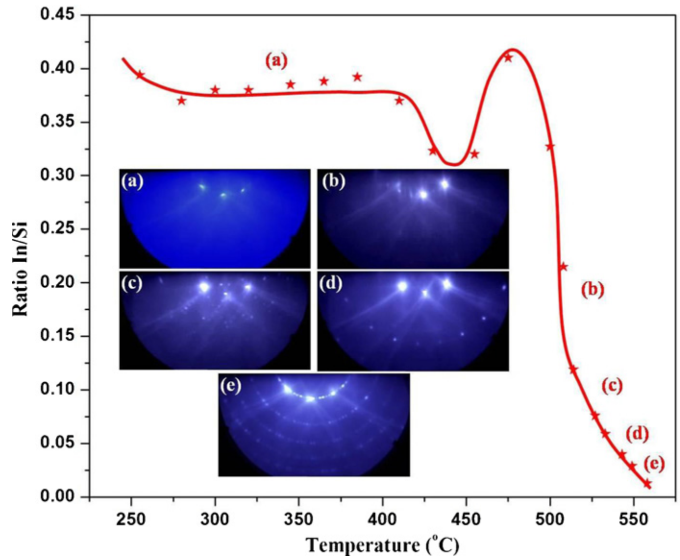

Fig. 1. Desorption curve and reconstructions. Auger residual thermal desorption curve for In/Si(1 1 1)-7 $\times$ 7 system showing the appearance of different superstructural phases and their respective RHEED patterns are shown. (a) 1 $\times$ 1, (b) 4 $\times$ 1, (c) $\sqrt{31} \times \sqrt{31}$, (d) $\sqrt{3} \times \sqrt{3}$ and (e) 7 $\times$ 7.

2. Experimental

We use the phase diagram for Si(1 1 1)-7 $\times$ 7-In system as guidance to obtain these superstructural phases in our PA-MBE system (SVTA, USA) which is equipped with several precise flux determination tools such as *in situ* Reflection High Energy Electron Diffraction (RHEED, from STAIB instruments), Atomic Absorption Spectroscopy, Quartz Crystal Thickness Monitor (QCTM) and Reflectometry. We adsorb about 2ML of In at RT on Si(1 1 1)–7 $\times$ 7 surface and anneal the surface at different temperatures while monitoring the evolution of the superstructures using RHEED. The InN growth temperature is chosen as low as 300 °C, since all the studied surface phases are stable at this temperature. Nitrogen flux is set to be 4.5 sccm and In flux used is 4.4×10^{13} cm^{-2} s^{-1}, which results in the InN growth rate of 0.15 μm per hour as determined by surface profiler and cross section FESEM. FESEM is also used to study the surface morphology, and the film crystallinity is determined by XRD measurements. Optical emission from the sample is carried out using room temperature Photoluminescence (PL) both in the visible and IR regions and charge carrier concentration is estimated by Hall Effect measurements using Van der Pauw method. Stoichiometry measurements are done by X-ray Photoelectron Spectroscopy using Mg K_α source. InN samples grown on phases, 7 $\times$ 7, 4 $\times$ 1 and 1 $\times$ 1 are denoted as sample A, B, and C, respectively, in the following discussions.

3. Results and discussion

The kinetic pathway is represented by the desorption curve which plots the In/Si Auger ratio versus annealing temperature (which we have reported in Ref. [12]) shown in Fig. 1. The Auger ratio of In MNN (404 eV) peak to Si LVV (92 eV) peak is plotted versus annealing temperature showing the residual thermal desorption process. The figure shows the progressive appearance of different ordered phases viz., (a) 1 $\times$ 1, (b) 4 $\times$ 1, (c) $\sqrt{31} \times \sqrt{31}$, (d) $\sqrt{3} \times \sqrt{3}$ and (e) 7 $\times$ 7 as In desorbs from the surface from hierarchical sites, which are represented in Fig. 1 as observed by RHEED in our MBE system.

Surface morphology of the grown InN films obtained by FESEM is shown in Fig. 2(a)–(c) for samples A, B, and C, along with their respective RHEED pattern as insets. Fig. 2(a) shows large InN crystallites of random shapes and the corresponding RHEED pattern depicts broken ring pattern, characteristic of mis-oriented or polycrystalline surfaces. Fig. 2(b) shows the FESEM image for InN grown on 4 $\times$ 1 phase, which has closely packed hexagonal crystallites with average grain size of ≈100 nm having pyramidal tops. The inset of Fig. 2(b) shows corresponding broken ring RHEED pattern where the elongated spots (arcs) have a high spot to background intensity contrast, suggesting a mis-aligned arrangement of the crystallites. The InN grown on 1 $\times$ 1 surface shows a morphology in Fig. 2(c) which consists of relatively larger hexagonal grains with flat tops. Inset to Fig. 2(c) show a markedly RHEED pattern, which is spotty and streaky, superimposed on a faint ring background. This manifests better ordered InN crystallites formed preferentially on the 1 $\times$ 1 phase as compared to films grown on other superstructures. The Root Mean Square (RMS) surface roughness of the different samples grown is measured by AFM, over a 5 $\times$ 5 μm^2 area. Sample A shows RMS roughness of ≈35 nm whereas samples B and C have a value of ≈15 nm indicating that films formed on 1 $\times$ 1 and 4 $\times$ 1 surfaces, have smoother surface morphology.

Fig. 3(a)–(c) consist of the θ–2θ XRD scans for samples A, B, and C, respectively, confirming the wurtzite structure of InN films are formed. Fig. 3(a) is the XRD pattern for InN grown on clean Si 7 $\times$ 7 which clearly shows crystalline InN (0 0 0 2) and InN (0 0 0 4) peaks at 31.33° and 65.37°, respectively. Fig. 3(a) also shows the presence of weak InN (1 0 $\underline{1}$ 1) and (1 0 $\underline{1}$ 3) reflections at angles 33.16° and 57.04°. Fig. 3(b) and (c) are the XRD pattern for InN grown on the 4 $\times$ 1 and 1 $\times$ 1 superstructural phases which have only crystalline peaks related to InN (0 0 0 2) and InN (0 0 0 4). Scans of the (0 0 0 2) wurtzite InN reflections on the different samples (after subtracting the contribution from the $K_{\alpha 2}$ X-ray component) are shown as inset in Fig. 3 for samples, A through C. FWHM of the (0 0 0 2) reflections for samples A shows a value of ≈1000 arcsec, whereas samples B and C have a significant lower value of ≈645 arcsec. Thus, XRD results show better crystalline quality for samples B and C.

Observation of PL in InN has been rare, especially in high carrier concentration samples, due to the poor material quality of the films [14]. The first observation of PL in InN was only possible in 2002 which revised the band gap of InN to be ≈0.7 eV rather than the