Applied Physics Letters

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Citation: Appl. Phys. Lett. 89, 092114 (2006); doi: 10.1063/1.2345224
View online: http://dx.doi.org/10.1063/1.2345224
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v89/i9
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Effects of grain size on the mosaic tilt and twist in InN films grown on GaN by metal-organic chemical vapor deposition

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(Received 19 May 2006; accepted 5 July 2006; published online 30 August 2006)

The structural property of InN films grown on Ga-face GaN layers by metal-organic chemical vapor deposition has been studied by high-resolution x-ray diffraction. The mosaic tilt and twist are found to be strongly dependent on the surface lateral grain size. The twist decreases with increasing grain size and finally approaches to a constant level. On the other hand, the mosaic tilt increases substantially when the grain size becomes large enough and exceeds the width of step terraces on the GaN surface, showing an important mechanism for the defect generation in the InN/GaN system with large out-of-plane lattice mismatch. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2345224]

During the past decade, InN has excited more research interest than before for its newly discovered small band gap value (~0.7 eV) (Refs. 1 and 2) and its excellent electrical properties predicted by theoretical calculations.[3,4] However, the growth of InN is more difficult than that of other group III nitride semiconductors due to its low growth temperature required and the lack of suitable lattice-matched substrate materials. Currently, the growth of InN is carried out mainly on as-grown epitaxial GaN layers. The crystalline quality of InN grown on GaN is much better than that directly grown on sapphire substrates.[5,6] Moreover, InN/GaN material system also facilitates the study of InN/GaN based heterostructure device applications.[7] On the other hand, however, the huge lattice mismatch (~10%) between InN and GaN poses a great difficulty to the InN researchers. For example, it has been found that three-dimensional (3D) InN island growth is preferred through the Stranski-Krastanov growth mode,[8,9] which often results in an undesired rough surface morphology. Furthermore, threading dislocations (TDs) can evolve from the coalescence of neighboring misoriented islands.[10] This affects the microstructure and crystalline quality of the InN epilayer.

A mosaic model is widely used to describe the epilayer microstructure, in which the in-plane and out-of-plane rotations of grains are quantified by the mosaic twist and tilt, respectively. The extent of tilt and twist can be directly related to the full width at half maximum (FWHM) of the (0002) symmetric and the (*hkil*) skew symmetric x-ray rocking curves with either h or k being nonzero.[11] A detailed characterization of the extent of tilt and twist in heteroepitaxial InN films can be conveniently implemented by virtue of the well-established high-resolution x-ray diffraction (HR-XRD) technology.

In this letter, we have made a systematic study of the structural property of InN epilayers grown on GaN by HR-XRD. It is found that the extent of mosaic tilt and twist is strongly dependent on the average grain size as they approach coalescence. Particularly, there is a remarkable increase in tilt as InN islands grow large enough to cross the step terraces on the GaN surface.

The growth of InN was conducted in a conventional horizontal metal-organic chemical vapor deposition (MOCVD) reactor operated at atmospheric pressure with N_2 as the carrier gas. The 4 μm-thick Ga-face GaN layers grown on *c*-plane sapphire substrate via a two-step procedure by MOCVD served as templates for the InN growth. Prior to the growth, the as-grown GaN templates were thermally cleaned at 700 °C in NH_3 atmosphere. Trimethyl indium and NH_3 were used as precursors for In and N, respectively. X-ray measurement was performed using a Rigaku SLX-1A x-ray diffractometer equipped with Si (220) analyzer. A NanoScope IIIa atomic force microscope (AFM) was employed for surface morphology observation. Besides, transmission electron microscopy (TEM) experiments were carried out in JEM 2010 electron microscope operating at 200 kV.

A series of InN films with a nominal thickness about 200–400 nm were deposited at a temperature ranging from 350 to 500 °C. As shown by AFM images in Fig. 1, the surface morphology is markedly characterized by the lateral

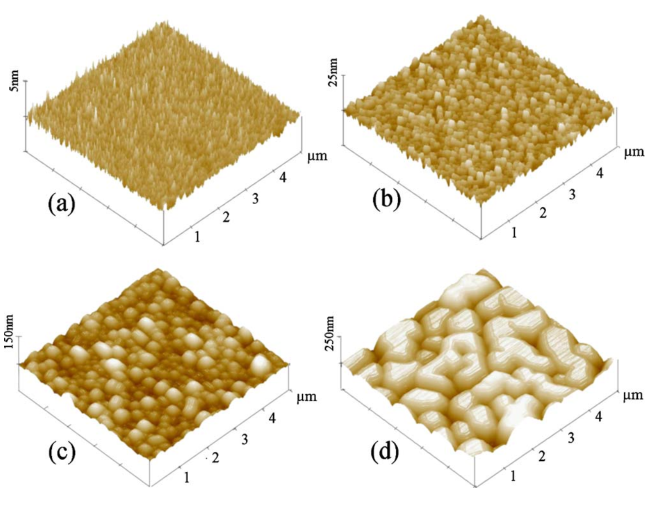

FIG. 1. 5×5 μm^2 AFM images of InN samples grown at 350 °C (a), 400 °C (b), 450 °C (c), and 500 °C (d), respectively.

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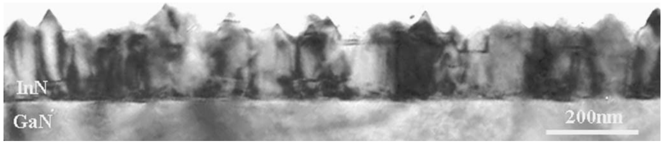

FIG. 2. Cross-sectional bright-field TEM image of InN film observed along the $[11\bar{2}0]$ direction with $g=(10\bar{1}0)$.

grains with various diameters dependent on the growth temperature. When the growth is carried out between 350 and 450 °C, the films take the form of small and close packed InN grains with diameters less than 250 nm because the mobility of the source atoms is too low to form larger islands.[12] At an elevated temperature (>475 °C), InN can grow into larger islands and the average surface lateral grain diameter varies from about 200 to 1000 nm by adjusting V/III ratio during the growth.

It is noted that the AFM measurement can only reveal the surface morphology of InN and the grain size might change significantly during epigrowth. In order to check the evolution of the grain size, a bright-field cross-section micrograph of InN film was measured with the operative $g=(10\bar{1}0)$, as shown in Fig. 2. It is found that InN film has nearly straight grain boundaries which arise near the InN/GaN interface and extend up to the top of the InN layer (a). It is also observed that these boundaries are connected with the "grooves between islands" emerging on the InN surface. It suggests that the average InN grain size starting from the coalescence can be quite well characterized by "lateral surface grain size" determined by AFM. In fact, such kind of microstructures has been reported as InN columnar structure when the growth was conducted below 500 °C.[9,13]

To evaluate the twist of InN films by the x-ray diffraction, we adopt the method presented in Ref. 14. The measured FWHMs of symmetric (0002) and skew $(10\bar{1}l)$ reflection rocking curves of InN films and the as-grown GaN template are plotted in Fig. 3(a) in the logarithmic scale as a function of the inclination angle (χ) between (0002) and $(10\bar{1}l)$ for the InN films with average grain diameters of 35, 120, 250, 265, 500, and 1000 nm, respectively. The dashed lines in the figure are fitting curves using the formula proposed in Ref. 15. From these fitting curves, the FWHM of the $(10\bar{1}0)$ rocking curves can be extrapolated to $\chi=90°$. The FWHMs of (0002) and $(10\bar{1}0)$ rocking curves reflect the mean tilt and twist, respectively. From these curves, it is evident that the FWHM monotonically increases with the inclination angle for all InN films, but the evolution strongly depends on the average grain diameter d_0. For InN films with small grain diameters ($d_0 \leqslant 250$ nm), the FWHM of (0002) rocking curve remains below 5 arc min which is comparable to that of the GaN template, whereas the FWHM increases dramatically with the plane inclination χ. This indicates a significant larger twist in these films. In comparison with the small grain films, a larger tilt but a less twist is found in the films with larger grains.

In order to clarify the correlation between mosaic structure and grain size, the FWHMs of (0002) and $(10\bar{1}0)$ rocking curves are plotted against the average grain diameter in Fig. 3(b). It is found that the tilt increases substantially as the grain size exceeds 250 nm while the twist decreases monotonically when the grain size increases and finally reaches to nearly a constant.

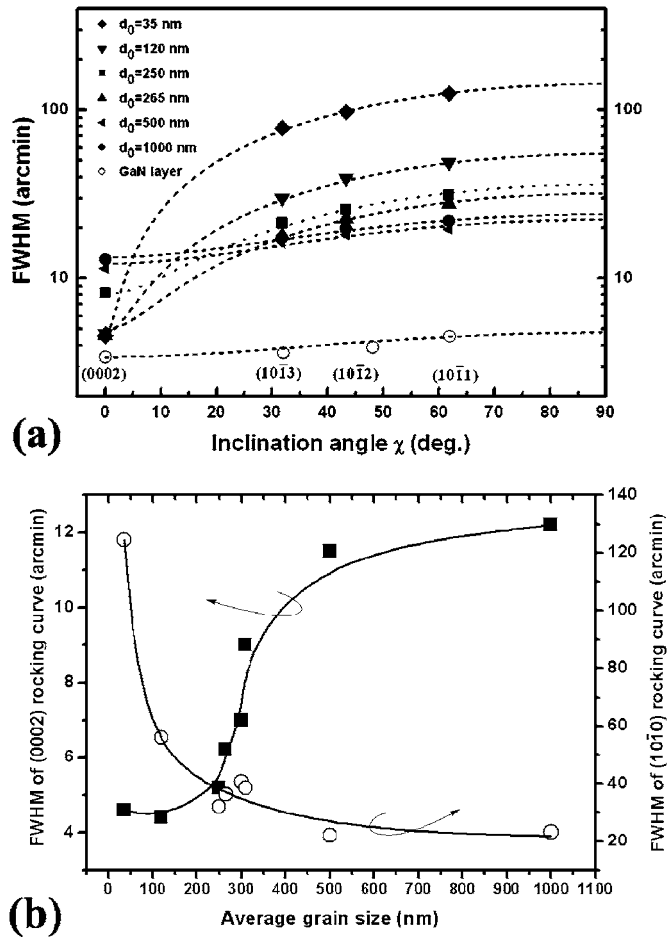

FIG. 3. (a) FWHM of x-ray rocking curves measured in a skew symmetric geometry as a function of the lattice plane inclination angle with respect to (0002). The dashed lines are fitting curves. (b) FWHM of (0002) rocking curves (filled squares) and the extrapolated FWHM of $(10\bar{1}0)$ rocking curves (filled circles) as a function of average grain size.

It is known that the density of TDs with edge and screw components in principle can correspond to the twist and tilt in the layers, respectively. The dependence of twist on the grain size, as shown in Fig. 3(b), is in agreement with the study on the mosaic structure of GaN grown on sapphire where the in-plane misorientation between the grains is compensated by edge TDs (***a*** type) located at grain boundaries.[16] In InN/GaN system there is a large in-plane lattice mismatch, which is mostly accommodated by a network of "geometrical misfit dislocations (MDs)" on the InN/GaN interface.[17] Since the growth mode in such a large lattice-mismatched system involves the independent formation of 3D InN nuclei, each nucleus should keep a special registry with the GaN layer by the periodical MDs network defined in $\{1\bar{1}00\}$ planes. The occurrence of ***a***-type TDs can be attributed to the discontinuity of the MD networks between the adjacent islands during coalescence. Thus, it is reasonable that the mosaic twist associated with the ***a***-type TD density is predominantly influenced by the average grain diameter as they approach the coalescence, i.e., decreases with increasing grain size.

To shed a light on the nature of the mosaic tilt in InN grown on GaN, two thin InN films with a nominal thickness of about 40 nm were grown at 400 and 500 °C, respectively.

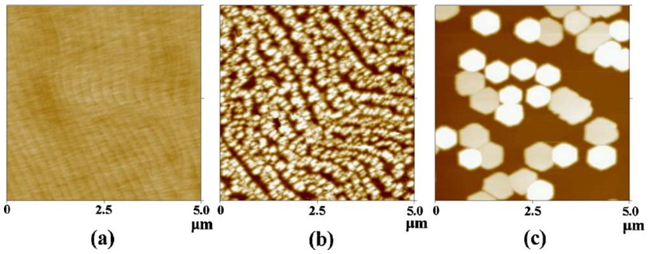

FIG. 4. 5×5 μm^2 AFM images of the surfaces of the as-grown GaN layer (a) and the thin InN films with a nominal thickness of about 40 nm grown at 400 °C (b), and 500 °C (c), respectively.

Their surface AFM images are shown in Figs. 4(b) and 4(b). The InN film grown at 400 °C exhibits a special morphology characteristic, i.e., small InN nuclei are aligned in lines on GaN surface [see Fig. 4(b)]. Comparing this surface morphology with that of the GaN layer shown in Fig. 4(a), we tentatively attribute such alignment of islands to the preferential nucleation of InN along the surface steps on the GaN surface due to the Ehrlich-Schwoebel barrier effect which is against the diffusion of adatoms towards the descending steps.[18,19] For the films grown at 500 °C, on the other hand, large and separated InN islands are observed with the grain size of about 750 nm which is close to that of thicker films grown under the same condition, as shown in Fig. 1(d). We believe that the existence of atomic steps on the GaN surface can give rise to the mosaic tilt in the InN films when the average grain size is large enough, for example, exceeds 250 nm, which is very close to the average width (200–250 nm) of step terraces on GaN surface determined by AFM. That is, when the grain size is small enough to be accommodated on one step terrace, no remarkable tilt will occur in the film since these InN grains are well-oriented along the *c* axis. While islands grow larger and exceed the width of the surface step terrace on the GaN surface, they will tilt with respect to the *c* axis.

There has been a general model accounting for crystallographic tilt in relation with the lattice elastic distortion exerted by the substrate surface steps (so-called Nagai's tilt).[20] However, this model is valid only for the systems with small mismatch.[21] In our case, the heterosystem involves a large out-of-plane lattice mismatch of about 10% between InN and GaN, therefore the role of MDs and the island growth mode becomes dominant in the formation of the tilt. Here, we propose that the tilt observed in our films is a result of the preferential occurrence of defects. The presence of steps on the substrate surface modifies the uniform biaxial stress field of the films near the steps. The change in stress distribution favors the generation of mixed dislocations with a Burgers vector of $\boldsymbol{c}+\boldsymbol{a}$ by gliding.[22] In fact, the dislocation generation and development mechanisms in the lattice-mismatched system depend upon the specific growth conditions. However, the mechanism presented in this letter should be one of the important mechanisms for the defect creation.

In conclusion, we have studied the structural property of InN films with various grain sizes grown on Ga-face GaN templates with a stepped surface. It is found that the twist decreases with increasing grain size and approaches to a constant level, while the mosaic tilt increases dramatically as the grain size is large enough and exceeds 250 nm. The critical grain size is dependent on the average width of step terraces on GaN surface and is attributed to the generation of mixed dislocations, showing an important mechanism for the defect generation in the InN/GaN system with large out-of-plane lattice mismatch.

The authors acknowledge the support from the National Natural Science Foundation of China (Grant Nos. 60506001 and 60576003).

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