Effect of island coalescence on structural and electrical properties of InN thin films

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Abstract

In this work, coalescence aspects of InN epitaxy are addressed. The coalescence phenomena have been studied in thin InN epilayers by means of electron microscopy and X-ray diffraction. Coalescence time and the corresponding diffusion coefficients at elevated temperatures were estimated for InN deposition. The substrate temperature was found to impact drastically the coalescence of the epilayer, and consequently, the electrical and transport properties of hexagonal InN material. Additionally, a simple growth model was suggested to explain the formation of domain boundaries and $(0\,0\,0\,1)$ stacking faults formed during the coalescence. In particular, it is shown that two adjacent and tilted, hexagonal-shaped InN domains may form a non-coherent boundary along a $\{1\,\bar{1}\,0\,0\}$ plane. We also suggest that the interaction between tilted domains induces formation of basal dislocations. This interaction has two consequences: a localized lateral growth of the most epitaxially oriented domain (forming a basal $(0\,0\,0\,1)$ stacking fault) followed by the formation of a surface step, and consequently the termination of a threading dislocation by its dissociation and propagation under the formed $(0\,0\,0\,1)$ stacking fault.

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1. Introduction

The superior electronic properties make InN a highly potential material for the fabrication of high-frequency heterojunction field effect transistors [1]. Another important discovery about 2H-InN is its very strong surface charge accumulation on the order of $10^{13}\,\mathrm{cm}^{-2}$ [2] revealing the promising applications of InN epilayers as chemical and biological sensors [3]. Recently reported results show also that THz emission from InN layers under excitation of ultrafast optical pulses exceeds those observed from p-InAs [4].

Recently, great progress in fabricating high-quality InN epilayers has been achieved [5]. Extensive research of

material and device properties has been accomplished by a number of studies concerning defects, impurities and associated growth mechanisms [6–10]. However, a lack of lattice-matched substrate remains a main challenge hindering the progress on the practical side. Moreover, there are still substantial problems concerning a deep understanding of the fundamental mechanisms of the InN heteroepitaxy.

Epitaxial 2H-InN layers, which can be used for device fabrication are usually highly faulted single crystals and typically adopt a mosaic structure with subgrain boundaries delineated by threading dislocations (TDs) [9–12]. The origin of this structure is related to surface atomic processes during growth. In order to improve growths in a systematic way it is essential to understand the underlying kinetic processes such as adsorption, desorption, and surface diffusion. In particular, adatom diffusion on the growing surfaces is considered to be a key parameter

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controlling the coalescence and consequently the material quality and the surface morphology [13].

It was recently shown [14] that heteroepitaxy of InN on highly mismatched substrates can be divided into four stages, each of them having a critical impact on the crystal quality of the epilayer. In the framework of the suggested growth model the following stages of the strain relief have been proposed: plastic relaxation of strain by the introduction of geometric misfit dislocations, elastic strain relief during the island growth, formation of TDs induced by the coalescence of the islands, and relaxation of the elastic strain by introduction of secondary misfit dislocations. Additionally, edge type TDs and dislocations of mixed character have been found to be dominating defects in 2H-InN layers. It was demonstrated that TDs are active suppliers of the electrons and an exponential decay of their density with the thickness implies the corresponding decay in the carrier density. Room temperature mobilities in excess of $1500 \, cm^2 \, V^{-1} \, s^{-1}$ were obtained for $\sim 800 \, nm$ thick InN layers with the dislocation densities of $\sim 3 \times 10^9 \, cm^{-2}$ [14]. The proposed model emphasizes the determining role of the coalescence process in the formation of a dislocation network in heteroepitaxially grown 2H-InN.

Nucleation and subsequent coalescence are interesting and important problems, which control a number of physical and mechanical properties of thin films [15,16]. However, most of the studies were devoted to computer simulations of the grain growth in the isotropic limit (i.e., when both energy and mobility of grain boundaries are isotropic), with rare attempts on anisotropy calculations [17]. In the case of the heteroepitaxy of thin 2H-InN films, both energy and mobility are strongly anisotropic, e.g., their values depend on the misorientation between two neighbouring crystals and the spatial orientation of their boundaries. In addition, non-equilibrium growth conditions and the presence of liquid phase at the grain boundaries, usual for a group-III-rich growth, may also result in a strong anisotropy of both energy and mobility. Due to their complexity, either analytical or computational studies of the coalescence in 2H-InN have been never addressed.

Recently, we have proposed elsewhere [14,26] that the coalescence process is a dominating factor controlling key crystal properties as polytypism, dislocation density and surface morphology of 2H-InN. However, the island coalescence process was not discussed in detail. The present work is primarily focused on coalescence of 2H-InN islands and its impact on epilayer properties. Coalescence times and the corresponding diffusion coefficients at elevated temperatures were estimated (in the case of a 10-nm-thick InN epilayer). Additionally, a simple growth model is proposed to explain the formation of domain boundaries and stacking faults during the coalescence stage.

2. Experimental procedure

The samples were grown in a Balzer's plasma-induced molecular beam epitaxy (PIMBE) system described else-where [14]. The substrate temperatures were calibrated by an infra-red pyrometer. The growth process was monitored by digitized patterns of RHEED. Molecular fluxes were monitored *in situ* by a quadrupole mass-spectrometer and calibrated using RHEED oscillations. The samples consisted of epitaxial $(0\,0\,0\,1)$ AlN/Al_2O_3 and GaN/Al_2O_3 templates overgrown *in situ* by a 2H-InN epilayer at $T_{sub} \sim 380\,°C$. In these conditions, the growth rate is proportional to the impinging group-III flux thus getting sticking coefficients approximately equal to unity [18]. The InN epilayers were grown under stoichiometric (1:1) conditions to prevent surface metal accumulation. The thickness of the InN layer has been varied from 0.35 to $2.2 \, \mu m$. More details on template preparation procedure and growth conditions can be found elsewhere [14,19].

Structural analysis was performed by high-resolution X-ray diffraction (XRD) using a Bruker D8 diffractometer. Rocking curves at the symmetric $(0\,0\,0\,2)$ and reciprocal space maps at the symmetric $(0\,0\,0\,2)$ and the asymmetric (20–25) InN reflections were taken to evaluate the crystal quality and the residual strain, respectively. The lowest FWHM of $0.270°$ was found at $\sim 1 \, \mu m$ indicating the continuous improvement of the InN layers up to this thickness and a change in the relaxation mechanism thereafter. Thick InN layers of $\sim 1 \, \mu m$ have only a small residual stress $<0.1\%$. Transmission electron microscopy (TEM) was performed in electron microscopes operating at 200 keV (TECNAI 20S-TWIN (FEI) and JEM-2011 from JEOL). Mechanical thinning and ion milling were used to prepare specimens for cross-section inspection. Electron diffraction patterns and micrographs obtained in bright-field (BF) and dark-field (DF) modes by conventional two-beam (2B) conditions and high-resolution TEM (HRTEM) were obtained and analysed.

3. Results and discussion

The growth mode of an epilayer is determined by both bulk thermodynamics and surface kinetics. However, at non-equilibrium conditions characteristic for InN PIMBE, surface kinetics plays often a dominant role, especially during the early stages of nucleation and growth. In general the shape and size of nucleated 2H-InN nano-crystals depends on the crystal structure, temperature and composition (e.g. stoichiometry) [20]. In many cases, these islands have a hexagonal form with small degrees of truncation (see Fig. 1). This shape occurs since it leads to surface energy minimization for nuclei formed at equilibrium, and because of kinetics where the shape is determined by the rate at which different crystal faces grow [21]. However, the combination of factors such as temperature, kinetics, impurities, and surface energy effects could lead to unusual nanoparticle size-shape distributions, which influences the particle coalescence.

Fig. 1(d) shows an initial stage of nucleation and coalescence of 2H-InN domains during the deposition on GaN $(0\,0\,0\,1)$. An estimation of the coalescence time (τ_c) for