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Suppression of domain formation in GaN layers grown on Ge(111)

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ABSTRACT

Heteroepitaxial growth of GaN on Ge(111) by molecular beam epitaxy has previously been reported to be feasible. However, structural characterization revealed that these GaN layers consisted of misoriented domains. In this work, it is shown that the formation of domains can be suppressed by increasing the substrate temperature, decreasing the nitrogen flux or increasing the surface step density by using off-oriented substrates. Hereby, the step flow growth is enhanced with respect to 2D nucleation. The suppression of domains significantly improves the crystal quality of the GaN layer. For 47 nm of GaN a (0002) and $(10\bar{1}2)$ ω FWHM value of, respectively, 408 and 935 arcsec is obtained.

1. Introduction

III-Nitrides show interesting physical properties allowing many electronic and optoelectronic devices. Nitride materials are predominantly grown by heteroepitaxy on foreign substrates. Metal organic vapor phase epitaxy and molecular beam epitaxy (MBE) are the most important growth techniques. Sapphire, SiC and Si are most commonly used for heteroepitaxial growth of nitrides. These substrates have huge lattice mismatches with respect to GaN of +16%, +3.5% and −17%, respectively [1]. An advantage of Ge as substrate for III-nitride structures lies in the significantly smaller thermal mismatch of −5.5% compared to−34%, +25% and +54% for sapphire, SiC and Si, respectively [1]. The theoretical in-plane lattice mismatch between GaN(0001) and Ge(111) is, however, substantial at −20% and one would suspect that here also a special buffer layer is required. Recently, we reported that the quality of GaN grown on Ge using plasma-assisted MBE (PAMBE) was surprisingly good [2]. The GaN epilayer grown directly on Ge(111) coalesced rapidly to give a smooth surface. Good crystal quality was revealed by X-ray diffraction (XRD) measurements with 371 arcsec full-width at half-maximum (FWHM) for the (0002)-rocking (ω) curve of a 38 nm GaN epilayer. These results suggest that direct growth of high-quality GaN on Ge is possible, which could lead to applications in various devices such as heterojunction bipolar transistors. The formation of a few monolayers crystalline Ge_3N_4 just before the start of GaN growth, has been used to explain how the lattice mismatch between GaN and Ge(111) of −20% is overcome [3]. This Ge_3N_4 has most likely the hexagonal β-phase [3], which has a lattice mismatch with Ge(111) of only 0.5% [4]. However, the in-plane lattice constant of GaN and β-Ge_3N_4 differs substantially with 3.189 Å for GaN [1] while 8.038 Å for Ge_3N_4 [4]. Nevertheless, the smooth growth and high crystal quality indicate that the GaN lattice fits well on this intermediate Ge_3N_4 layer. We propose that there exists a mesh ratio approximately equal to $\frac{5}{2}$ between hexagonal GaN and a β-Ge_3N_4 surface. The lattice mismatch that follows from such a mesh relation is only −0.8%. Although high crystal quality was observed with XRD (0002)-rocking curve

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measurements, additional structural characterization revealed the GaN layers consisted of misoriented domains [5]. The domains are GaN grains rotated relative to each other around the GaN-(0 0 0 1) zone axis by about 4° clockwise and counterclockwise ($\pm 4°$) with respect to the Ge substrate, which lead to substantial broadening of asymmetric XRD rocking curves. Suppression of this domain formation should lead to further improvement of the GaN structural and electrical quality. The GaN domains extend from the interface with Ge to the surface. Therefore, the conditions at the start of growth could be important in their formation. For this reason, we have investigated the influence of growth parameters on the formation of GaN grains and the crystal quality. The growth parameters discussed are Ga flux, substrate temperature, N flux, surface step density and presence of Ge surfactants.

2. Experimental procedure

Ge(1 1 1) substrates are chemically cleaned to remove metallic contamination, particles and native oxide from the surface, just before loading into the MBE system. Subsequently, the samples are degassed at 450 °C in vacuum with a background pressure of 1×10^{-9} Torr. The cleanliness of the surface is confirmed by reflection high-energy electron diffraction (RHEED), which shows a reconstructed surface. Substrate temperatures are measured by thermocouple. Three different substrate temperatures are studied: 750, 800 and 850 °C. Higher substrate temperatures lead to more Ga desorption from the surface. We have investigated Ga beam fluxes from 0.7 to 8.0×10^{-7} Torr. It has previously been shown that exposure of the clean Ge(1 1 1) surface to nitrogen plasma above 600 °C leads to the formation of a thin crystalline layer of Ge_3N_4 in a self-limiting process [4]. In the first series of experiments, a N_2 flow of 1.2 sccm and a radio frequency power of 250 W have been used. These settings correspond to a beam equivalent pressure of around 4.5×10^{-7} Torr N. After the formation of Ge_3N_4, by 1 min nitrogen plasma exposure, the Ga shutter is opened and GaN growth is started. Layers with a thickness between 36 and 72 nm have been grown. The thickness of the GaN layer is determined ex situ by spectroscopic ellipsometry. The presence of domains rotated in the plane of growth could be observed in the RHEED pattern. In the absence of domains, one GaN RHEED pattern is visible. In the presence of domains, two GaN RHEED patterns are visible at about $\pm 4°$ rotation in the plane of growth with respect to the RHEED pattern of the Ge substrate. The existence of domains could also be revealed by XRD. The crystal quality as reflected by the FWHM of both the symmetric (0 0 0 2) and the asymmetric $(1 0 \bar{1} 2)$ reflection was determined by XRD.

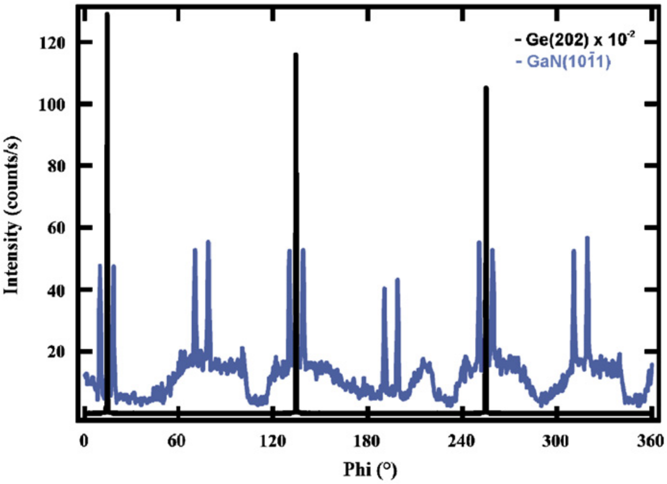

Fig. 1. XRD phi scans of skew-symmetric reflections of GaN $(1 0 \bar{1} 1)$ and Ge (2 0 2). The GaN and Ge scans are overlaid. For each Ge diffraction peak (repeated every 120°), 2 GaN peaks are visible (repeated every 60°), indicating the presence of GaN domains with two different orientations.

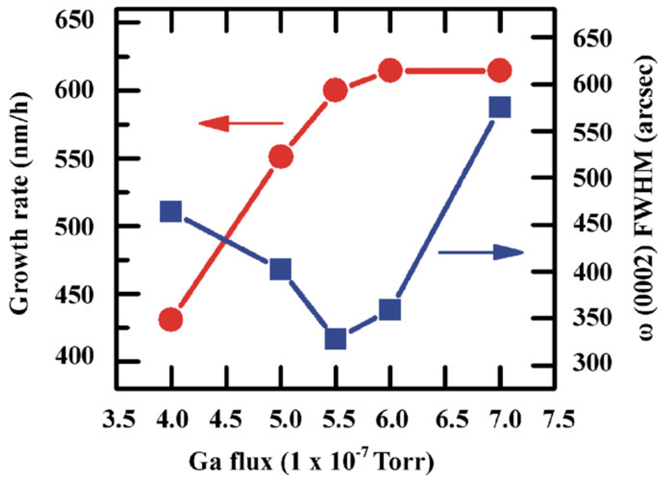

Fig. 2. GaN growth rate and XRD ω (0 0 0 2) FWHM for different Ga fluxes. Growths were performed for 5 min at a substrate temperature of 800 °C. The XRD ω (0 0 0 2) FWHM is minimum at 5.5×10^{-7} Torr Ga flux, indicating the best crystal quality is obtained when the Ga and N flux ratio is close to unity. GaN domains are observed for all Ga fluxes.

3. Results and discussion

In the first series of experiments, with a substrate temperature of 800 °C, GaN layers were grown under N- and Ga-rich conditions by keeping the N_2 flow constant and changing the Ga flux from 4.0 to 7.0×10^{-7} Torr. For all Ga fluxes used, domains could be observed by RHEED. This was subsequently confirmed by XRD phi scans of skew-symmetric reflections, a typical result is shown in Fig. 1. For every 60° two GaN peaks are visible, indicating the presence of GaN grains with two orientations. The GaN domains are rotated in the plane of growth about $\pm 4°$ with respect to the Ge lattice [5]. This suggests that at these growth conditions a large number of small islands are nucleated on the Ge_3N_4 surface. Apparently, these islands are not all aligned with respect to each other. A slight twist of $\pm 4°$ with respect to the germanium lattice exists. In Fig. 2, it can be seen that the symmetric (0 0 0 2) ω XRD FWHM exhibits a minimum of 328 arcsec at 5.5×10^{-7} Torr Ga flux. At this point of optimal material quality, the growth rate is at the transition of being limited by the N flux or by the Ga flux. This result is similar to that reported for homoepitaxial growth of GaN, where the optimum Ga flux is reported to be slightly higher than the N flux [6]. Although the symmetric scan of these layers grown at 800 °C shows good values, one can expect that the presence of domains has mainly influence on the FWHM of the ω XRD FWHM of the $(1 0 \bar{1} 2)$ planes, rather than on the FWHM of the (0 0 0 2) reflection. This is indeed the case: although the FWHM of the symmetric (0 0 0 2) ω XRD scan is limited, the FWHM of the asymmetric $(1 0 \bar{1} 2)$ ω XRD scan is larger than 4000 arcsec for all these samples grown at 800 °C. Suppression of domain formation will be revealed in a reduction of notably the FWHM of the $(1 0 \bar{1} 2)$ reflection.

When the substrate temperature was increased to 850 °C, no domains were observed by RHEED for all Ga fluxes used. A typical XRD phi scans of skew-symmetric reflections is shown in Fig. 3, where for each Ge diffraction peak (repeated every 120°), only one