GaN thin films on *z*- and *x*-cut LiNbO₃ substrates by MOVPE

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We report epitaxial growth of GaN layers on *z*- and *x*-cut LiNbO₃ substrates using MOVPE. GaN layers with the thickness of 450 nm were characterized using X-ray diffraction. For both, *z*- and *x*-cut orientations of LiNbO₃ substrates, the GaN layers have *c*-axis orientation normal to the substrate plane and the in-plane lattice orientation of GaN layers coincides with the primary axes of LiNbO₃ substrates. Although GaN layers exhibit almost complete strain relaxation, the residual compressive strain determined with respect to a freestanding GaN is of the order of +0.37% and +0.2% for *z*- and *x*-cut substrates, respectively.

1 Introduction Epitaxial growth of GaN on LiNbO₃ substrates attracts attention due to potential applications of this heterostructure for monolithic integration of materials with complimentary optical and electronic properties [1, 2]. At the same time, studies of GaN deposition on LiNbO₃ are in a mainstream of the quest for alternative substrates for nitride epitaxy. LiNbO₃ has a rhombohedral *3m* crystal structure with lattice parameters $a = 0.5147$ nm and $c = 1.3862$ nm, while GaN has wurtzite structure with This corresponds to the in-plane lattice mismatch of 6.8% between LiNbO₃ and GaN ($a = 0.3189$ nm, $c = 0.5185$ nm) with a 30° rotation of the LiNbO₃ [3]. Although the lattice mismatch between GaN (00.1) and LiNbO₃ (00.1) is reduced compared to that in the GaN-on-sapphire (~16%), growth is usually performed using transitional and/or nucleation layers, such as AlN, to accommodate strain and to improve the quality of the grown GaN material [4-7].

Growth regimes for nitride heteroepitaxy on LiNbO₃ substrates require further optimization to prevent potential reduction of the oxide substrates and formation of unwanted LiNb₃O₈ layers at the heterointerface [2]. In this paper we report results of GaN heteroepitaxy on LiNbO₃ substrates with different orientation: *z*-cut and *x*-cut. Modification of the structural parameters of the grown GaN layers due to the different orientation of the substrate will be discussed.

2 Experimental Although the difference of in-plane lattice parameters in the heterosystem of GaN/LiNbO₃ is smaller compared to that for GaN/Al₂O₃, discrepancy in the thermal expansion coefficients between GaN and LiNbO₃ is large enough to generate delamination of the GaN layer. This occurs either during the growth or when cooling down from the growth temperature. As a result, the conventional growth conditions by MOVPE of GaN on Al₂O₃ substrates are not directly applicable for LiNbO₃ substrates. In order to obtain a good adhesion of GaN, novel growth conditions were explored in this work. A combination of ammonia (NH₃) and Dimethylhydrazine (DMHY) has been used as sources of atomic nitrogen. Because of its low temperature of decomposition, the DMHY allows to lower the GaN growth temperature down to

600 °C [8]. Pure Nitrogen was used as a carrier gas. The growth was performed in a T-shaped reactor [9]. Trimethylgallium (TMG) was used as a source material for gallium. The growth temperature and pressure were 730 °C and 450 Torr, respectively. Although precautions were taken, partial reduction of LiNbO₃ occurred during the growth as indicated by the colour change of the otherwise transparent substrates. The surface quality and the growth rate were monitored in real time during the growth using in-situ reflectometry.

Two GaN layers with the nominal thickness of 450 nm were grown simultaneously on optical grade LiNbO₃ substrates with *z*- and *x*-cut orientation polished on both sides. *Z*-cut substrate has *c*-axis (00.1) normal to the surface plane, while the *x*-cut substrate has *c*-axis in the plane. Typical misorientation of the substrate surface with respect to the crystallographic directions was about 0.6 degrees, as determined using X-ray diffraction (XRD).
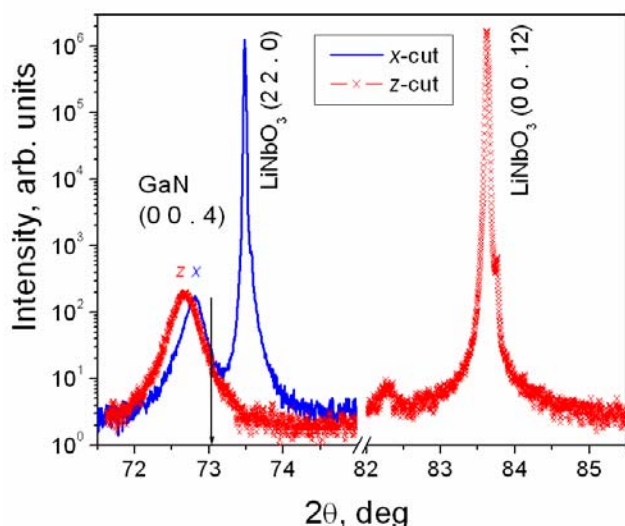

Figure 1 XRD 2θ-θ scans for GaN layers grown on *z*- and *x*-cut LiNbO₃ substrates. Scans show the (00.4) reflection peaks of GaN layers. Note that the GaN peaks are shifted with respect to the 2θ value for a free-standing GaN, which position is indicated with vertical arrow. This shift corresponds to the compressive strain of +0.37% and +0.2% for samples grown on *z*- and *x*-cut substrates, respectively. Peak at 2θ = 73.5 deg corresponds to (22.0) reflection of the *x*-cut LiNbO₃ substrate. Peak at 2θ = 83.6 deg corresponds to the (00.12) reflection of the *z*-cut LiNbO₃ substrate.

3 Results and discussion Structural characterization of GaN layers was carried out using Philips X'Pert diffractometer. At first, using pole figures for (11.4) reflection of GaN, we have confirmed that the primary orientation of both GaN films is with the *c*-axis [(00.1) direction] being perpendicular to the substrate. Figure 1 shows 2θ-θ scans for two samples measured in the vicinity of the (00.4) reflection of GaN. Experimental position of the (00.4) peaks for both GaN layers corresponds to a residual compressive strain of +0.37% and +0.2% for *z*- and *x*-cut substrates,
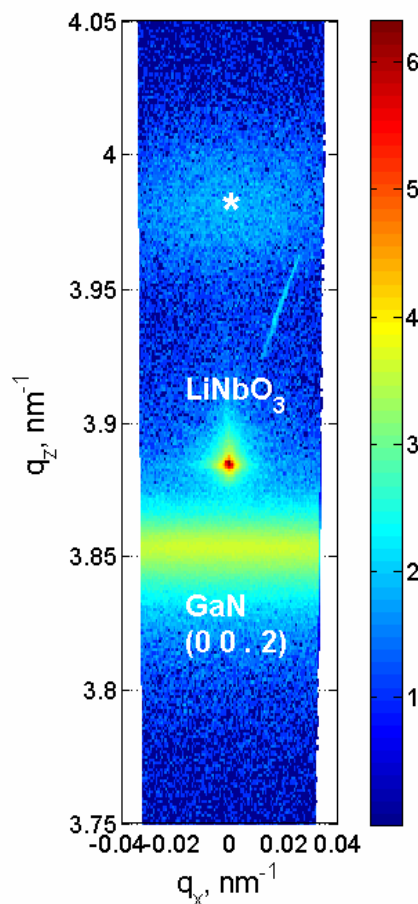
Figure 2 RSM for (00.2) reflection of GaN layer grown on *x*-cut LiNbO₃ substrate. Strong diffraction peak at $q_z = 3.88$ nm⁻¹ corresponds to the (11.0) reflection of the *x*-cut LiNbO₃ substrate. Weak peak at $q_z = 3.98$ nm⁻¹ marked with a star is from the growth-induced defects in the substrate.

respectively. Here strain is expressed in terms of the *d*-spacing mismatch in (00.1) direction and is calculated with respect to a free-standing GaN. No additional peaks that could be attributed to formation of transitional interface layers of LiNb₃O₈ were observed. The strong peak at 2θ = 73.5 deg corresponds to (22.0) reflection of *x*-cut LiNbO₃ substrate with *c*-axis being oriented in the plane of the sample and perpendicular to the diffraction plane. Note that the close proximity of the (00.4) peak for GaN and (22.0) peak for *x*-cut LiNbO₃ substrate indicate only that the *out-of-plane* lattice parameters of these two materials are commensurate, which does not necessarily mean that the *in-plain* lattice of both materials matches. Thus, the residual strain in GaN layers cannot be straightforwardly explained by the remains of the lattice mismatch between the layers and the substrates. The fact that both GaN layers grown on *z*- and *x*-cuts reveal the same crystallographic orientation indicates that the initial step of GaN formation on LiNbO₃ results in a strong structural change of the oxide substrate, such as a significant reduction of its oxygen content. The mechanisms that promote *c*-axis growth of

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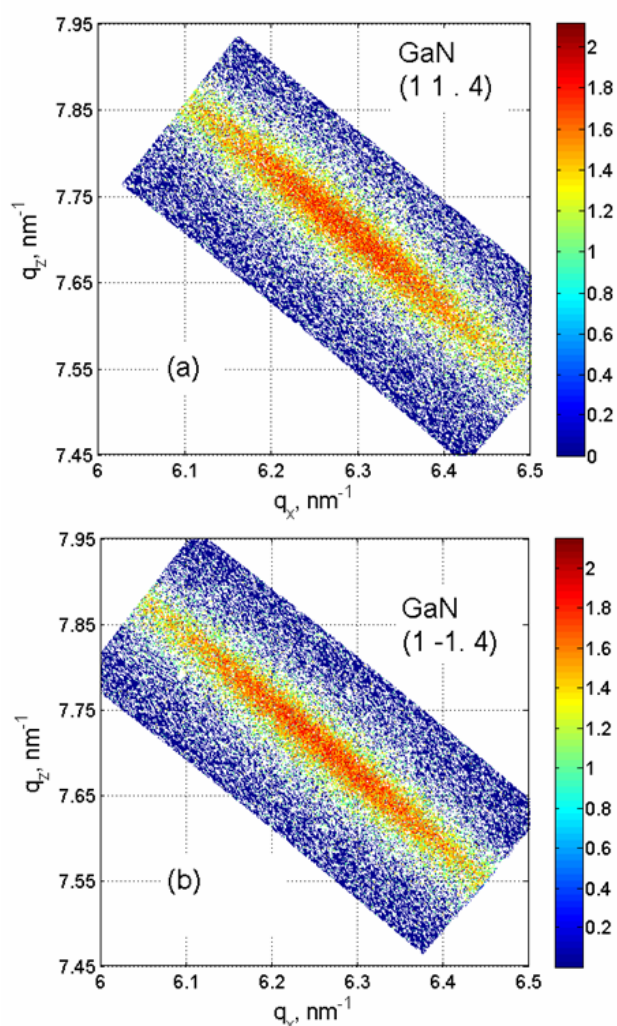
Figure 3 (a) RSM for asymmetric (11.4) reflections of GaN layer grown on *x*-cut LiNbO₃ substrate with the c-axis oriented perpendicular to the diffraction plane: Φ = 0 deg. (b) Same for (1-1.4) reflections with *c*-axis of the substrate being oriented at 60 deg to the diffraction plane: Φ = 60 deg.

GaN layers in both cases are not clear and require additional investigation.

Structural quality of the grown GaN layers can be assessed with the help of the reciprocal space map measured for (00.2) reflection [see Fig. 2]. The FWHM of the GaN layer peak, which is about 0.7 deg is significantly larger compared to that of the LiNbO₃ substrate. The weak peak, which appears in RSM above the LiNbO₃ (11.0) reflection, originates from the substrate and is probably related to the interface reconstruction due to the loss of oxygen during the growth. Note that the same peak was observed in LiNbO₃ substrate after simulation of the growth regime in MOVPE reactor without actual deposition of GaN.

To understand better the interplay between the crystallographic orientation of the substrate and GaN layers, we performed investigation of the reciprocal space maps for asymmetric (11.4) reflections that are more sensitive to the

in-plane lattice parameters. For a strong connection between in-plane lattice parameters of the layer and substrate, one could expect changes between the in-plane lattice parameter determined from (11.4) and (1-1.4) reflections of GaN layer grown on the *x*-cut substrate. Note that *c*-axis of LiNbO₃ is perpendicular to the diffraction plane for (11.4) reflection of GaN and correspondingly makes a 60 deg angle for the case of (1-1.4) reflection. Figure 3 compares RSM's for both asymmetric reflections for GaN layer. The inhomogeneous broadening of the corresponding peaks is significantly larger than any possible differences in the in-plane reciprocal lattice coordinates. This observation indicates again that the two lattices, *c*-axis oriented GaN and *x*- or *z*-cut LiNbO₃, are significantly decoupled due to the transitional interface layer of reduced oxide substrate.

4 Conclusion Relaxed GaN layers were grown on *x*- and *z*-cut LiNbO₃ substrates. Their structural properties and crystallographic orientations are similar to each other. The residual compressive strain determined with respect to a free-standing GaN is of the order of +0.37% and +0.2% for *z*- and *x*-cut substrates, respectively.

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