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Growth of *A*-plane GaN on (0 1 0) $LiGaO_2$ by plasma-assisted MBE

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ABSTRACT

The (0 1 0) surface of $LiGaO_2$ is closely lattice matched to *A*-plane (1 1 $\bar{2}$ 0) GaN making it an interesting candidate as a substrate for heteroepitaxy of non-polar GaN. We demonstrate successful, first-time growth of *A*-plane GaN on (0 1 0) $LiGaO_2$ using plasma-assisted molecular beam epitaxy. Structural and morphological analysis is performed using X-ray and reflective high energy electron diffraction, scanning electron and atomic force microscopy. Very high phase purity of *A*-plane GaN is shown. Apart from defects of the epitaxial film originating from substrate scratches, the film is smooth and shows an rms roughness of 10 nm over an area of $8 \times 8\,\mu m^2$.

1. Introduction

Non-polar nitrides, which avoid the built-in polarization fields perpendicular to the surface, have recently been of great scientific interest and are therefore beneficial for optoelectronic applications [1]. Due to lack of readily available GaN substrates for homoepitaxy, much effort has been put into growing non-polar GaN heteroepitaxially on various substrates. While fabrication of *M*-plane GaN has been under extensive investigation on a number of different substrates, *A*-plane GaN has foremost only been grown on *R*-plane sapphire [2] due to the lack of matching substrates.

$LiGaO_2$ (LGO) presents a unique material system for growth of the three prominent GaN crystal orientations [0 0 0 1], [1 $\bar{1}$ 0 0] and [1 1 $\bar{2}$ 0], depending on the LGO surface orientation. Closely lattice matched relations between LGO and GaN are found for *M*-plane GaN on (1 0 0) LGO, *A*-plane GaN on (0 1 0) LGO and *C*-plane on (0 0 1) LGO [3,4]. Several groups have shown growth of *C*-plane GaN on (0 0 1) LGO [3,5–9]. We have recently demonstrated the successful growth of *M*-plane GaN on (1 0 0) LGO [10].

In this work we show for the first time the successful growth of *A*-plane GaN on (0 1 0) LGO by plasma-assisted molecular beam epitaxy (MBE).

Fig. 1 shows a ball and stick model of the (0 1 0) LGO surface including its unit cell as a dotted rectangle. The gray rectangles suggest possible nucleation sites for *A*-plane GaN growth. The lattice mismatch for the relationship [1 0 0] LGO‖[1 $\bar{1}$ 0 0] GaN and [0 0 1]LGO‖[0 0 0 1] GaN is 2.22% and 3.58%, respectively. Growth on the (0 1 0) plane of LGO, similarly to growth on the (1 0 0) LGO plane, benefits from the lack of a metal–non-metal polarity, which is an issue that has to be taken into account for growth on (0 0 1) LGO [6,9].

2. Experimental procedure

Crystal growth of the (0 1 0) LGO substrates was performed at the National Sun Yat-sen University in Taiwan. The LGO bulk crystal was then cut and polished by an external company. For growth of the LGO crystal Li_2CO_3 and Ga_2O_3 powders with at least 99.99% purity were mixed and introduced into a Czochralski pulling furnace. An iridium lid was placed on top of the Ir crucible to reduce the temperature gradient. The raw materials were melted at approximately 1650 °C. To prevent oxidation of the Ir crucible nitrogen gas was continuously supplied during growth. A (0 0 1) *c*-axis orientated seed was rotated at a rate of 10–20 rpm to control growth conditions. Given a high vapor pressure of the melt, a high crystal pulling rate of 2–4 mm/h was applied to prevent disconnection from the melt.

Before introduction to the MBE chamber, the LGO substrates were mounted onto a Si wafer using a thin In layer to provide a

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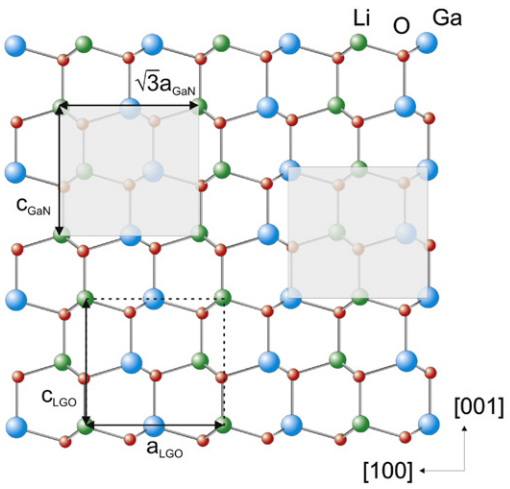

Fig. 1. Ball and stick model of the (0 1 0) LGO surface. The dotted rectangle signals the size of the LGO unit cell, while the rectangles with continuous lines indicate possible nucleation sites for (1 1 $\bar{2}$ 0) GaN.

homogeneous thermal coupling. The substrates, placed into molybdenum holders, were outgased for 60 min at 130 °C prior to transfer into the growth chamber. During growth activated nitrogen N_2^* was supplied by an Oxford HD25 RF plasma cell, operated at a steady gas flow of 0.3 sccm and 450 W forward power for all samples. Ion deflection plates were used to reduce the ion amount in the nitrogen flux. Growth procedures were monitored *in situ* using reflective high energy electron diffraction (RHEED).

To establish *A*-plane GaN growth the substrate temperature was optimized in the range between 650 and 800 °C and the Ga flux was varied from slightly Ga rich to Ga rich conditions. Several pre-growth treatments of the substrate in the growth chamber were studied. These were Ga desorption, nitridation, Ga desorption and nitridation, annealing and no treatment of the substrate.

Successful growth of *A*-plane GaN on LGO was achieved when growth conditions were optimized and temperature ramps were employed carefully. Directly before GaN deposition the substrate was annealed at 800 °C for 60 min in the growth chamber. Thereafter, growth of GaN commenced at slightly Ga rich

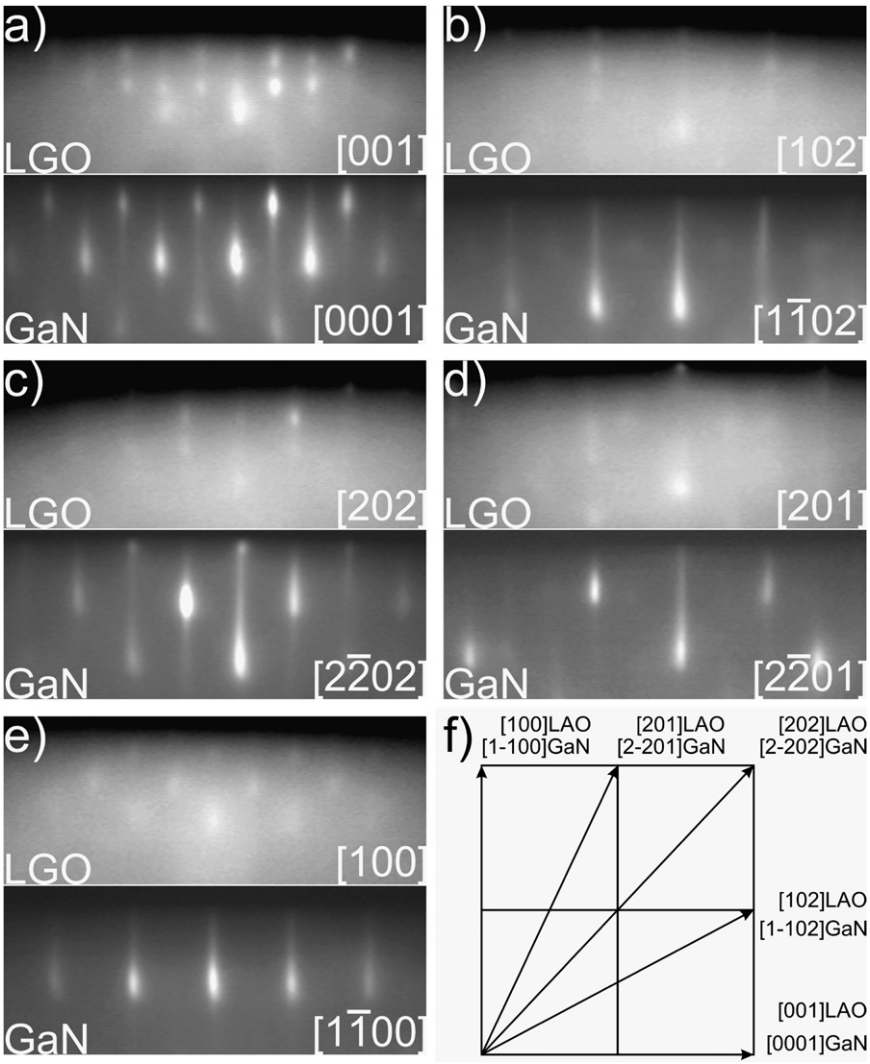

Fig. 2. RHEED images of five azimuths of (0 1 0) LGO and (1 1 $\bar{2}$ 0) GaN. Each pair of pictures (a–e) was taken at the same angles, the top one before and the bottom one after growth, respectively. The close lattice match between the substrate and the film is evident. Streaky patterns indicate a smooth and flat surface. (f) shows a schematic drawing of the [1 1 $\bar{2}$ 0] GaN surface indicating the directions of the observed azimuths, similar to how it was done for *M*-plane GaN by Waltereit et al. [11]. Calculation and measurement of angles and spacings allows the assignment of directions to the pictures.

conditions at a substrate temperature of 700 °C. The temperatures given correspond to the thermocouple readings. A growth rate of roughly 60 nm/h was used. Irradiation of the sample with the RHEED beam was reduced to a minimum to prevent possible damage to the sample, i.e. RHEED was only turned on after 30 min of GaN growth and in order to take RHEED pictures.

Shortly after taking the sample out of the growth chamber, the GaN film showed small cracks at the edges. A too high temperature in the area of contact between the substrate and the Mo holder probably is the reason for these features. However, approximately more than 98% of the film remained stable and did not lift-off or show cracks.

Characterization of the sample was done ex-situ by X-ray diffraction (XRD), scanning electron microscopy (SEM) and atomic force microscopy (AFM).

3. Results and conclusion

Samples from initial growth runs showed peeling off of the epitaxial film from the substrate. This is a common situation for GaN growth on LGO [10]. Following several different substrate pretreatment approaches to prevent this, we found that annealing of the substrate lead to the desirable result. While RHEED patterns of the untreated substrate were dim and hinted to a contaminated surface, substrate cleaning by Ga desorption, nitridation or both together showed no observable effect. When a high Ga flux was used, RHEED observations showed an increasingly polycrystalline type diffraction pattern as soon as the Ga shutter was opened.

A possible reason for the peeling off of the epi layer may have been the enhanced misfit of the anisotropic thermal expansion coefficients between LGO and GaN at high temperatures. Moreover, damage of the substrate's surface may have been caused by impinging highly energetic electrons from RHEED measurements. The polycrystalline RHEED patterns may result from bending of the partly peeling off film, thereby giving the impression of polycrystallinity.

In the following only the sample that was grown at optimized conditions will be discussed.

In Figs. 2(a–e) RHEED measurements of five azimuths are depicted. The top pictures show diffraction patterns of the substrate shortly before growth while the bottom pictures display electron diffraction patterns after 3 h growth of GaN for the same angles, respectively. The five azimuths of both, the LGO and GaN crystal, could reliably be identified in two ways thereby providing a valuable method of consistency check. First, the relative separation in angle was compared to the theoretical angle separation. The second method involved comparing the quotient of streak separation of the five azimuths to one another with the corresponding calculated values. The post growth pictures show streaky images of *A*-plane GaN hinting towards a flat and smooth film. The correspondence of streak separation of the LGO and GaN for the same angle displays the close lattice match between the substrate and its epitaxial layer. Fig. 2(f) shows how the directions of the observed RHEED patterns correspond to azimuths of both the (0 1 0) LGO and (1 1 $\bar{2}$ 0) GaN surface.

The thickness of the GaN epitaxial layer was measured in SEM to be 170 nm. In Fig. 3, part (a) and (b) show SEM pictures of the GaN sample surface using magnifications of 10 k and 1 k, respectively. Scratches in the GaN film, assumably caused by mechanical polishing of the substrate are clearly visible in both images. These surface irregularities are also observed in AFM images shown in Fig. 4. Picture (a) in Fig. 4 shows the height information obtained, while part (c) depicts the amplitude information, giving higher image sharpness. The depth profile of the horizontal line indicated in (a) is graphed in Fig. 4(b). While

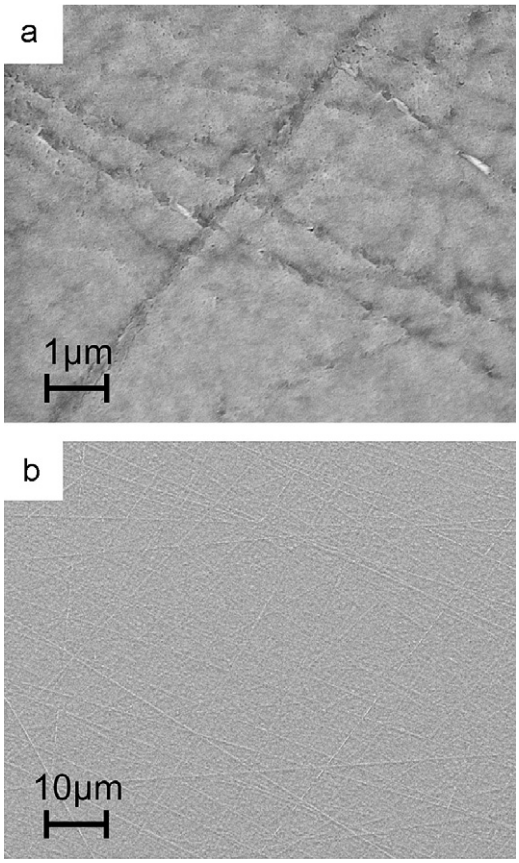

Fig. 3. (a) SEM picture of the GaN surface using a 10 k magnification. Deep scratches originating from the substrate are observable and intercept the continuity of the GaN film. (b) 1 k magnification of the GaN film. A large number of scratches in the film are apparent.

the peak to valley roughness can be as high as 75 nm in parts with 'deep' scratches, an rms value of 10 nm is obtained on an area of $8 \times 8\,\mu m^2$ when excluding the deepest scratches from analysis. Throughout the performance of microscopy no indication of *C*-plane or *M*-plane GaN, i.e. their typical crystal shape, could be observed in the film. It is expected that the surface roughness will improve when polishing procedures and growth conditions are optimized, thereby also increasing the GaN film quality.

The crystal orientation of the epitaxial GaN film was examined by X-ray diffraction. Fig. 5(a) shows an $\omega-2\theta$ scan in the range $\omega = 12-78^{\circ}$, performed without analyzer. All the apparent main peaks could be attributed to LGO planes. Two regions of the spectrum, that show additional information when zooming into them are displayed in Figs. 5(b) and (c). Here, reflections of *A*-plane GaN planes can be seen. Note that apart from diffraction peaks attributed to *A*-plane GaN and LGO no other peaks were observable. Therefore, a high degree of phase purity was found in the GaN film. The (1 1 $\bar{2}$ 0) GaN peak was convoluted with the (0 4 0) LGO reflection and does therefore not allow for an exact determination of the peak position of the GaN signal. The (2 2 $\bar{4}$ 0) GaN reflection had a broader shape and lower intensity giving a slightly larger error when determining its peak position. However, a strain state of the GaN epitaxial film could be estimated from the relative shift of the (2 2 $\bar{4}$ 0) GaN peak with respect to the (0 8 0) LGO peak. Disregarding twist or tilt inside the GaN crystal, the GaN film was relaxed to about 50% compared to the maximal possible transverse deformation caused by the lattice misfit between (0 1 0) LGO and (1 1 $\bar{2}$ 0) GaN.

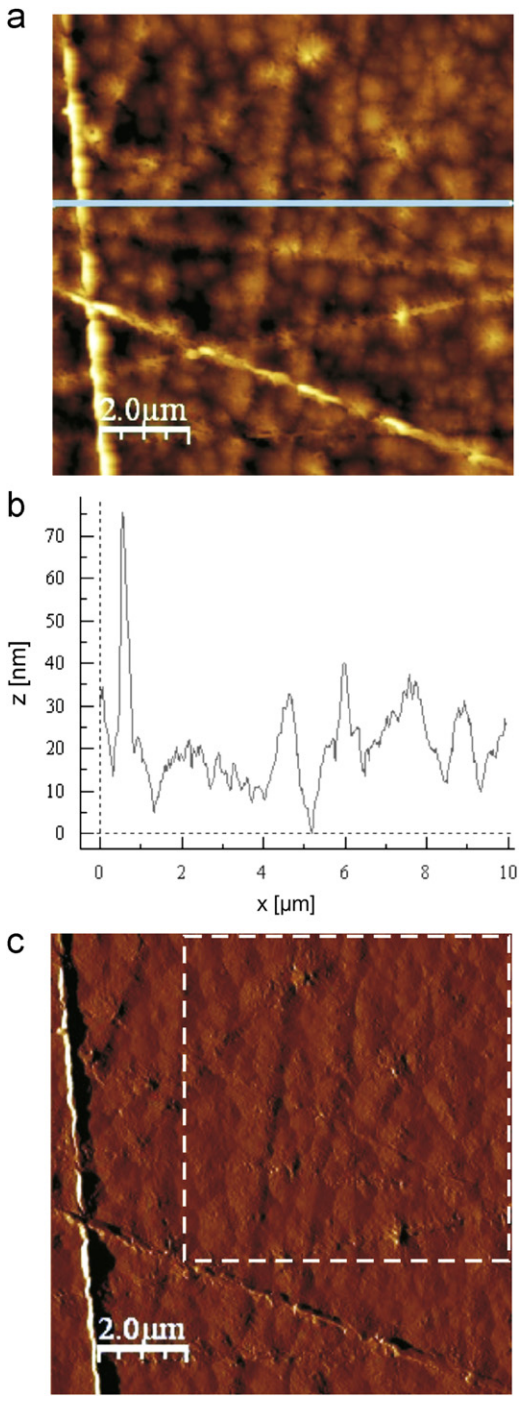

Fig. 4. (a) and (c) show $10 \times 10\,\mu m^2$ AFM images of the sample surface using height and amplitude information, respectively. The depth profile of the horizontal line displayed in (a) is shown in (b). The rms roughness of a $8 \times 8\,\mu m^2$ area as indicated by the white dashed square in part (c) is 10 nm.

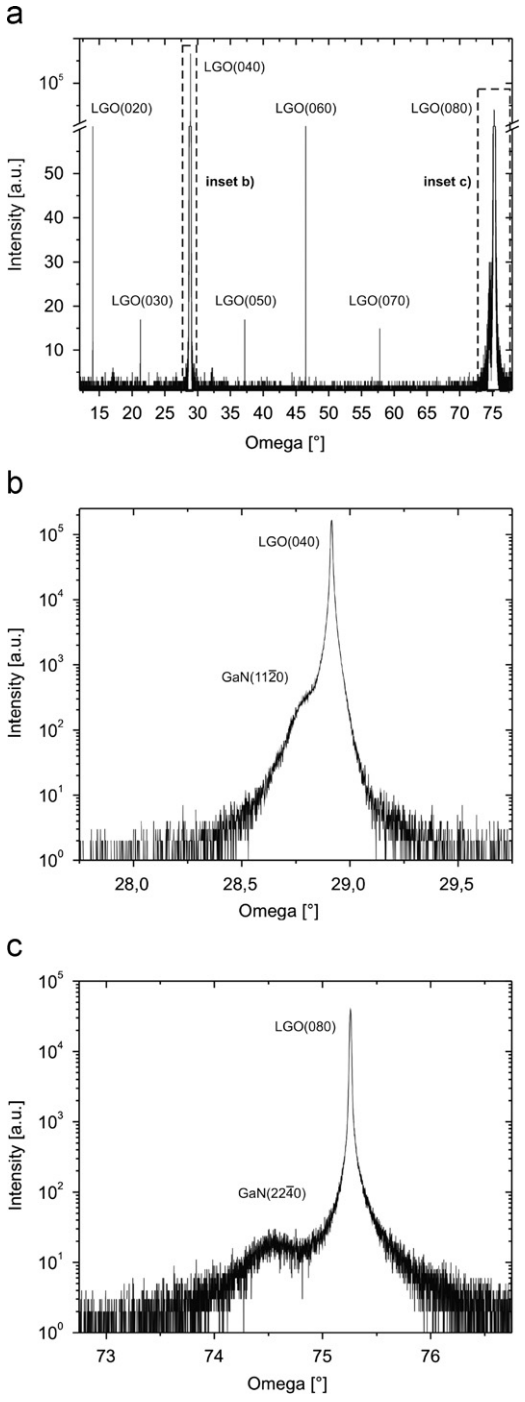

Fig. 5. (a) ω–2θ wide range X-ray scan without analyzer of a 170 nm thick GaN layer grown on (0 1 0) LGO. Diffraction peaks of several LGO as well as *A*-plane GaN planes can be seen. There is no indication of diffraction peaks of any other oriented crystal planes. (b) Zoom into the region $\omega = 27.75 - 29.75^{\circ}$ where the clearly visible right shoulder of the (0 4 0) LGO peak can be attributed to (1 1 $\bar{2}$ 0) GaN. (c) Zoom into $\omega = 72.75^{\circ} - 76.75^{\circ}$. Here, the diffraction peaks of *A*-plane GaN and LGO lie further apart allowing for a simple strain state analysis.

4. Summary

For the first time *A*-plane GaN has been epitaxially grown on (0 1 0) $LiGaO_2$. This has been achieved by plasma-assisted MBE. Streaky RHEED patterns after growth in addition to the SEM and AFM data show smooth *A*-plane GaN between the polishing related surface scratches. The XRD results indicate a very high degree of phase purity. For the 170 nm thick film a relaxation state close to 50% was found compared to the maximal possible strain that could have been induced due to the lattice mismatch between film and substrate. The results obtained prove the possible production of high quality *A*-plane GaN on (0 1 0) $LiGaO_2$.

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