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Twinning in Low-Temperature MOCVD Grown GaN on (001) GaAs Substrate*

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Abstract: GaN buffer layers (thickness ~60nm) grown on GaAs(001) by low-temperature MOCVD are investigated by X-ray diffraction pole figure measurements using synchrotron radiation in order to understand the heteroepitaxial growth features of GaN on GaAs(001) substrates. In addition to the epitaxially aligned crystallites, their corresponding twins of the first and the second order are found in the X-ray diffraction pole figures. Moreover, {111} ϕ scans with χ at 55° reveal the abnormal distribution of Bragg diffractions. The extra intensity maxima in the pole figures shows that the process of twinning plays a dominating role during the growth process. It is suggested that the polarity of {111} facets emerged on (001) surface will affect the growth-twin nucleation at the initial stages of GaN growth on GaAs(001) substrates. It is proposed that twinning is prone to occurring on {111}B, N-terminated facets.

Key words: X-ray diffraction; metalorganic chemical vapor deposition; nitrides

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1 Introduction

Research activities in GaN-based materials and devices are flourishing. Up to date, most of the GaN-based devices have been fabricated from hexagonal phase nitrides. However, cubic GaN (c-GaN) can offer many advantages over its hexagonal counterpart, including easy cleavage, lower phonon scattering, and convenience for device structure designing and integrating when GaAs substrate is used.

One of the difficulties in growing high quality single crystal c-GaN is the strong tendency of twinning via stacking faults on {111} planes. Therefore, the preparation of c-GaN often suffers from occurrence of multiple orientation and mixture of phases if the nucleation and the growth conditions are not optimized. In order to grow high quality c-GaN layer, it is necessary to gain a better understanding of the nucleation and growth features of GaN on GaAs (001), especially the early growth stages of GaN buffer layer. In our previous paper[1], we reported on the structural characteristics of the GaN/GaAs(001) buffer layers with different thickness. We found the nominal {111} diffractions could be detected very easily but cubic

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GaN (002) diffraction was unexpectedly weak. For the sake of simplicity, we did not take the twinning into consideration in the calculation of phase content. However, it might play a very important role in the heteroepitaxy of GaN on (001) GaAs substrate.

It is well known that the twin formation is a very serious problem for the preparation of Ⅲ-Ⅴ compound semiconductors[2,3]. Twinning in the diamond cubic and zinc blende semiconductors can be represented by a 60° rotation about the normal to the {111} twinning planes and thus be easily observed. Despite the importance of the problem, reasonable explanation for the features of twinning in GaN grown on GaAs(001) has not yet been reported in literatures. It remains probably the most important problem in the epitaxial growth of GaN on GaAs(001) substrate, especially for the low-temperature growth of buffer layer which is used as a growth templet of the subsequently grown epitaxial layer.

Studies by cross-sectional transmission electron microscopy (XTEM) and high resolution electron microscopy (HREM)[4~6] of the initial stages of growth have been very useful in providing structural information about the heteroepitaxial growth processes. However, these techniques for the study of orientation relationships are limited due to small region detection. Moreover, sample preparation as well as the evaluation are quite laborious. On the other hand, X-ray diffraction (XRD) pole figure measurements are nondestructive and can offer phase identification and orientation relationship determination. Some research groups[7~10] have employed the XRD pole figure measurements to characterize the film quality regarding preferential orientations of the crystallites.

In this paper, we study further the structural characteristics of LT-MOCVD grown GaN buffer layers on (001) GaAs by using synchrotron X-ray pole figure and ϕ scan measurements. It provides insights into the underlying mechanism of GaN buffer layer growth on (001) GaAs substrate.

2 Experiment

GaN buffer layers were deposited on GaAs (001) by low pressure (10.132kPa) MOCVD at a low temperature of 530℃ which is the usual temperature used to grow the buffer layer before the high temperature epitaxial growth. Trimethylgallium (TMG) and ammonia in H_2 carrier gas were used as Ga and N precursors, respectively. The thickness of these buffer layers estimated from the in situ laser reflectivity measurements is about 60nm. These low-temperature MOCVD grown buffer layers were then characterized ex situ by X-ray diffraction pole figure measurements using synchrotron radiation. An intense synchrotron source was employed from B_4 beamline at the Beijing Synchrotron Radiation Facility (BSRF). The wavelength of the incident X-ray was set at 0.154nm by using a Si(111) monochromator.

3 Results and discussion

It has been reported that the cubic GaN (002) diffraction of the MOCVD grown GaN/GaAs(001) buffer layers is extremely weak[1] and even undetectable[11], whereas the {111} diffraction could be detected very easily. In this study, relatively thicker buffer layers (thickness ~60nm) are investigated further by using synchrotron radiation to get better understanding of the phenomena mentioned above. First of all, we perform {002} pole figure measurements on these GaN/GaAs(001) buffer layers. Figure 1 is a typical experimental XRD {002} pole figure of these buffer layers. It can be seen in Fig. 1, there are extra intensity maxima in addition to the central cubic GaN (002) diffraction and the anisotropy of the intensity distribution is also obvious. The weak central spot in the measured {002} pole figure, with 2~3 orders of magnitude weaker than that of the extra maxima, indicates that the epitaxially aligned component of the as-deposited GaN buffer layers with [001] orientation identical to

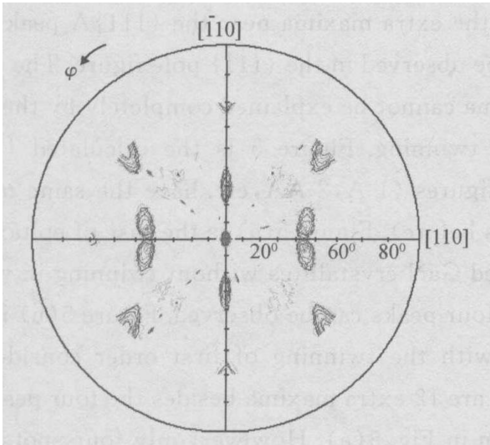

Fig. 1　{002} XRD pole figure of the GaN/ GaAs(001) buffer layers

that of the GaAs(001) substrate is very low. This is the same as the result obtained from the XRD ω-scan analysis of GaN/ GaAs (001) buffer layers[1,11]. We do not consider the cubic twinning in the calculation of phase content in our previous analysis[1] where we just point out that the rotation about the normal to the {111} plane would weaken the (002) diffraction. However, twinning might play a very important role at the initial stages of GaN heteroepitaxy on GaAs(001). The extra intensity spots appear in the {002} pole figure (Fig. 1) are most probably due to the diffractions from cubic twins. As before-mentioned, twinning in zinc-blende semiconductors can be represented by a 60° rotation about the normal to the {111} twinning plane. Generally, lattice plane (hkl) will change to ($h^t k^t l^t$) after a twinning on the (HKL) twinning plane, and the new indexes can be expressed as

$$h^t = - h + \frac{2H}{H^2 + K^2 + L^2}(Hh + Kk + Ll)$$

$$k^t = - k + \frac{2K}{H^2 + K^2 + L^2}(Hh + Kk + Ll)$$

$$l^t = - l + \frac{2L}{H^2 + K^2 + L^2}(Hh + Kk + Ll)$$

Thus, pole figures in the presence of twinning can be determined in principle. Figure 2 is the theoretically determined cubic GaN {002} pole figures. Figure 2(a) shows the case of twinning of first order. 1-A and 1-B represent the twinning of first order occurred on {111}A and {111}B planes of cubic GaN matrix, respectively. Obviously, the measured {002} pole figure is very different from Fig. 2(a). So, it would not be explained well if just the twinning of the first order is considered. Nevertheless, the measured {002} pole figure is very similar to the theoretically predicted one in which twinning of the second order is considered (Fig. 2(b)). In Fig. 2(b), 2-AA and 2-AB represent the twinning of second order occurred on {111}A and {111}B planes of 1-A GaN twins, respectively. Similarly, 2-

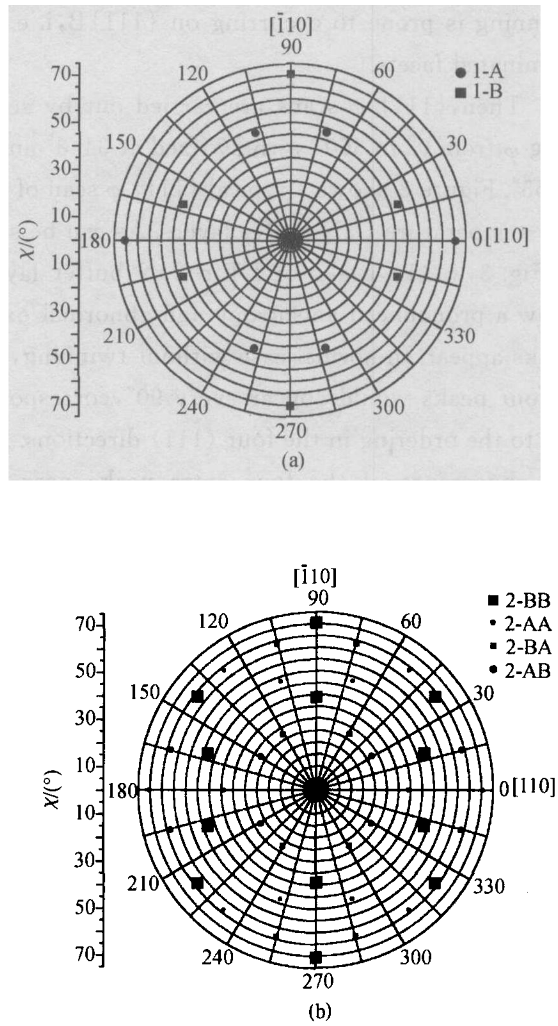

Fig. 2　Theoretically determined {002} pole figure considering twinning　(a) Twinning of the first order; (b) Twinning of the second order

BA and 2-BB are the twinning of the second order occurred on {111}A and {111}B planes of 1-B GaN twins, respectively. It should be noticed that figure 1 can be explained satisfactorily if {111}B planes are supposed to have a higher tendency to twinning, i.e. the diffraction intensity from 2-BB is much higher than the other twinning cases (Fig. 2 (b)). Dudley *et al*.[3] investigated the influence of polarity on twining in zinc-blende semiconductors and found the nucleation of twins occurs preferentially on {111}B faces. It seems the polarity of the {111} facets emerged on (001) surface also affects the growth-twin nucleation at the initial stages of GaN growth on GaAs (001) substrates and the twinning is prone to occurring on {111}B, i.e. N-terminated facets.

Then, {111} ϕ scans are carried out by scanning ϕ from 0° to 360° with 2θ fixed at 34.3° and χ at 55°. Figure 3 shows a typical {111} ϕ scan of the low-temperature GaN buffer layers. As can be seen in Fig. 3, {111} peaks of LT c-GaN buffer layers show a pronounced anisotropy and abnormal extra peaks appear. In normal case without twinning, only four peaks would appear every 90°, corresponding to the ordering in the four ⟨111⟩ directions. But the appearance of the four extra peaks near the {111}A peaks at 45° and 135° needs further investigation.

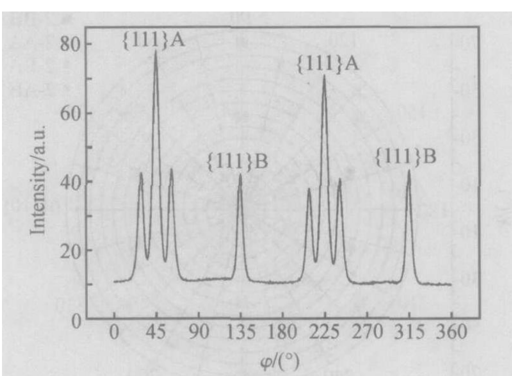

Fig. 3 {111} ϕ scan with χ at 55°

Therefore, we have measured {111} pole figures for the low-temperature GaN buffer layers by fixing 2θ at 34.3°, then scanning χ from 0° to 70° and ϕ from 0° to 360°. Figure 4 is a representative {111} XRD pole figure. The anisotropy in intensity distribution is also obvious. There are extra intensity maxima besides the normal four peaks at χ = 55° with ϕ every 90°. As revealed in the {111} ϕ scan, the extra maxima near the {111}A peaks can also be observed in the {111} pole figure. The extra maxima cannot be explained completely by the first order twinning. Figure 5 is the calculated {111} pole figures (1-A, 2-AA, etc. have the same meaning as before). Figure 5(a) is the case of epitaxially aligned GaN crystallites without twinning in which only four peaks can be observed. Figure 5(b) is the case with the twinning of first order considered, there are 12 extra maxima besides the four peaks as shown in Fig. 5(a). However, only four spots corresponding to twinning of first order at χ = 16° can be detected due to the tilt limiting in the χ circle. On the assumption of {111}B facets possess higher tendency to twinning, the {111} pole figure in the presence of twinning of the second order is shown in Fig. 5(c). Obviously, if we superpose these three pole figures in Fig. 5, we will obtain a {111} pole figure very similar to the measured one (Fig. 4). Therefore, we conclude that twinning of the first and the second order in GaN/GaAs(001) buffer layers is most likely responsible for the extra maxima in the measured {111} pole figure (Fig. 4).

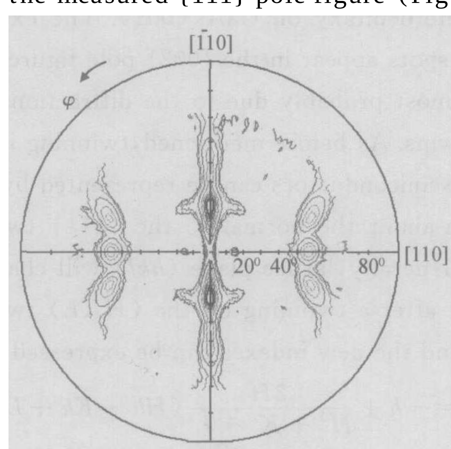

Fig. 4 Measured {111} XRD pole figure of the GaN/GaAs(001) buffer layers

4 Conclusion

GaN/GaAs(001) buffer layers (thickness ~60nm) grown by MOCVD have been investigated by X-ray diffraction pole figure measurements using synchrotron radiation to better understand the

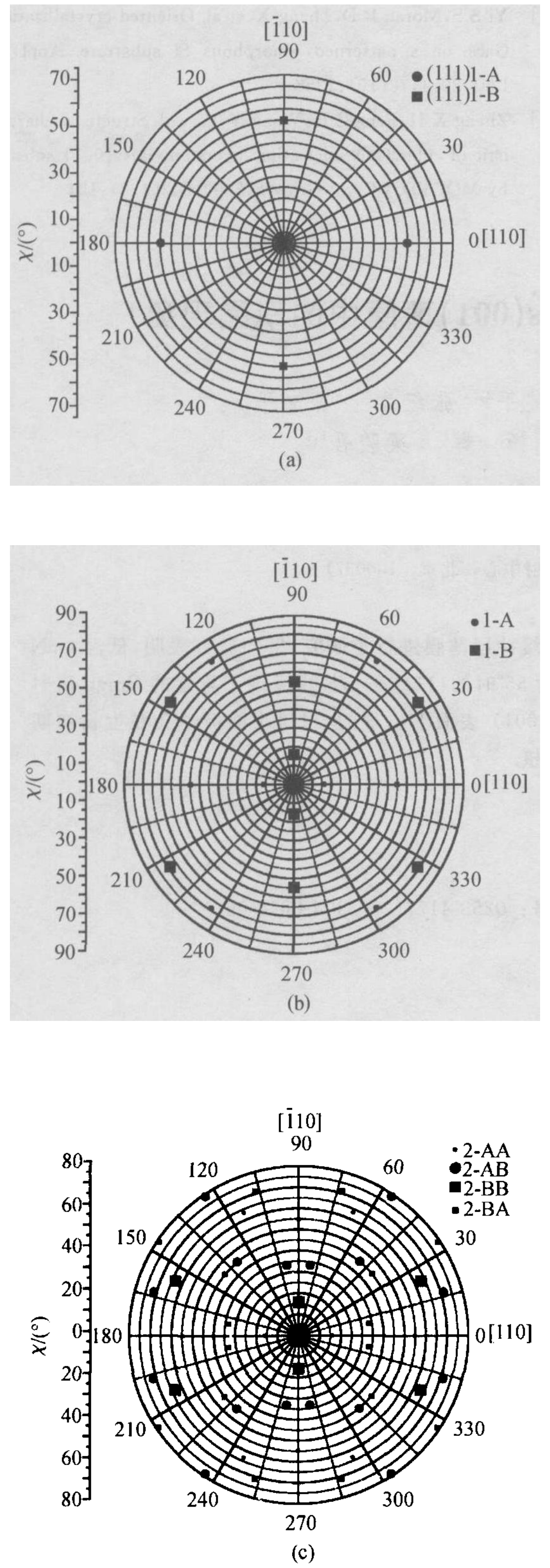

Fig. 5　Theoretically determined {111} pole figures
(a) Twinning is not included; (b) Twinning of the first order is included; (c) Twinning of the second order is included

growth features of heteroepitaxial GaN on (001) GaAs substrates. Besides the epitaxially aligned crystallites their corresponding twins of first and second order have been found in the X-ray diffraction pole figures. The observed twinning phenomena in nominal cubic GaN/GaAs buffer layers are probably responsible for the nearly undetectable cubic GaN (002) diffraction and the high content of hexagonal phase in this kind of buffer layers and the consequently grown epilayer. The extra intensity maxima in the pole figures shows that the process of twinning plays a dominating role during the growth process. It is suggested that the polarity of {111} facets emerged on (001) surface would affect the growth-twin nucleation at the initial stages of GaN growth on GaAs (001) substrates. Compared to {111}A facets, {111}B facets would show a higher tendency to twinning.

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低温 MOCVD 法沉积的 GaN/GaAs(001)薄膜中的孪晶现象*

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摘要: 采用同步辐射 XRD 极图法对低温 MOCVD 生长的 GaN 缓冲层薄膜进行了研究. 极图研究表明, 低温 GaN 薄膜中除有正常结晶外还存在一次孪晶和二次孪晶. 在 χ 固定为 55°时的{111} ϕ 扫描中发现了异常的 Bragg 衍射峰, 表明 GaN/GaAs(001)低温生长中孪晶现象非常明显. GaAs(001) 表面上出现的{111}小面极性会在生长初期影响孪晶成核, 实验结果表明孪晶更易在{111}B 面即 N 面上成核.

关键词: X 射线衍射; 金属有机物化学气相沉积; 氮化物
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