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Dislocation Reduction in GaN on Sapphire by Epitaxial Lateral Overgrowth*

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Abstract: High quality GaN is grown on GaN substrate with stripe pattern by metalorganic chemical vapor deposition by means of epitaxial lateral overgrowth. AFM, wet chemical etching, and TEM experiments show that with a two-step ELOG procedure, the propagation of defects under the mask is blocked, and the coherently grown GaN above the window also experiences a drastic reduction in defect density. In addition, a grain boundary is formed at the coalescence boundary of neighboring growth fronts. The extremely low density of threading dislocations within wing regions makes ELOG GaN a potential template for the fabrication of nitride-based lasers with improved performance.

Key words: metalorganic chemical vapor deposition; GaN; epitaxial lateral overgrowth; dislocation
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1 Introduction

Group Ⅲ nitrides have recently become the focus of numerous research groups worldwide because of their potential applications in optoelectronics. Due to a lack of large area native substrates, nitride semiconductor structures have been developed by heteroepitaxial growth on foreign substrates such as sapphire, 6H-SiC, and Si. However, GaN epilayers always suffer from high dislocation densities in the range of 10^9 to $10^{10}\,cm^{-2}$, which may result from the large difference in the lattice constants and the thermal expansion coefficients between GaN and the substrates[1]. In a two-step growth of GaN on sapphire, it has been demonstrated that an appropriate three-dimensional nucleation mode could decrease the dislocation density to the order of $10^8\,cm^{-2}$[2~4]. However, only when the dislocation density is low can better device performance be achieved, especially for laser diodes. For this reason, epitaxial lateral overgrowth (ELOG) was introduced[5~7], and this technique has resulted in significant improvement in the performance of nitride laser diodes[8].

More recently, the first pulsed operation of a nitride-based laser diode at room temperature in mainland China was realized in our group[9]. Instead of an ELOG GaN template, this nitride-based laser was grown on sapphire substrate. Therefore, the ELOG of high quality GaN is necessary to improve the performance of nitride-based laser diodes. We report here high quality the ELOG of GaN by metalorganic chemical vapor deposition (MOCVD). Atomic force microscopy (AFM), wet chemical etching, scanning electron microscopy (SEM), double crystal X-ray diffraction (DC-XRD), and transmission electron microscopy (TEM) were used to characterize the dislocation reduction in ELOG GaN.

2 Experiment

A GaN template was grown on (0001) Al_2O_3 substrate with a closed-space showerhead MOCVD reactor with H_2 carrier gas. Trimethylgallium (TMGa) and NH_3 were used as precursors. After the deposition of about 2μm of GaN film by a conventional two-step method, a 50nm thick Si_xN_y mask layer was deposited by plasma enhanced

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chemical vapor deposition. The mask was then patterned using standard photolithography to open stripe-like windows aligned along the ⟨10$\bar{1}$0⟩ GaN. The window stripes were 3μm in width and were spaced 20μm apart. Subsequently, the patterned substrate was reloaded into the MOCVD system. During the first step of overgrowth, a triangular cross section was developed with a low Ⅴ/Ⅲ ratio (~300) and low temperature (~1020 ℃), followed by a second step in which lateral overgrowth and coalescence were carried out with a high Ⅴ/Ⅲ ratio (~2700) and elevated temperature (~1080 ℃).

As-grown ELOG samples were characterized by SEM using a Cambridge S-360. The surface topography was measured with an XE 100 AFM operating in non-contact mode. Samples were etched in molten KOH for 10min to form etching pits on the surface and were then observed by SEM. The crystalline properties of the ELOG GaN and GaN template were analyzed by a Rigaku SLX-1A X-ray double crystal diffractometer with Cu Kα_1 radiation. TEM observations were carried out with a Philips CM200 operating at 200kV.

3 Results and discussion

Figure 1 shows plane-view and cross-sectional SEM micrographs of coalesced ELOG GaN. The sapphire substrate, the GaN template, the Si_xN_y mask layer, and the overgrown GaN can be readily distinguished in the cross-sectional image. Tapered voids can be seen at the merging fronts, which correspond to the vertical lines in the plane-view image. By examining perspective views, in which the surface and section of a sample can be observed simultaneously, the contrast lines in Fig.1 can be assigned to the coalescence boundary of neighboring merging fronts. The full-width at half maximum (FWHM) of (0002) reflection for the 2μm GaN template is about 360″, while the FWHM of (0002) reflection for the ELOG sample, which is about 9μm thick, is 180″ when the diffraction plane is parallel to the (11$\bar{2}$0) plane. Furthermore, when the diffraction plane is perpendicular to the (11$\bar{2}$0) plane, no peak splitting is visible in the rocking curve, which implies a relatively small tilt angle of the wing region with respect to the window region.

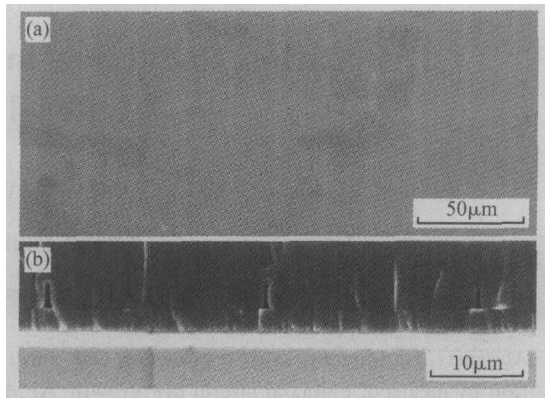

Fig.1 Plane view (a) and cross-sectional view (b) of SEM images for coalesced ELOG GaN samples One of the merging front related contrast lines is highlighted by a vertical arrow.

Wet chemical etching is a commonly used technique for surface defect investigation because of its low cost and simple equipment. In this paper, both the GaN template and ELOG sample were etched in molten potassium hydroxide (KOH) at 180 ℃ for about 10min. It should be noted that the KOH etchant in this study is pure KOH that has been heated to its molten state rather than an aqueous KOH solution. Figures 2(a) and (b) show the etch pits on the etched surface of the GaN and ELOG samples, respectively. The etch pits revealed by the KOH etch, mostly hexagonally shaped, are ascribed to threading dislocations (TD) with screw character or mixed character. The etch pit density of GaN template, as estimated from Fig.2(a), is about $8\times10^{6}cm^{-2}$. On the contrary, three different regions can be distinguished on the etched surface of the ELOG GaN (Fig.2(b)) according to the defect density-namely, the window region, wing region, and the merging front. In the window region, dislocation related etch pits can be observed with a density of about $7\times10^{5}cm^{-2}$, an order of magnitude less than that of GaN template. However, the wing region over the Si_xN_y mask is nearly free of etch pits. Quantitatively, the dislocation density in the wing region is about $5\times10^{4}cm^{-2}$, which is two orders of magnitude less than that of the GaN template. In addition, few etch pits, with a density of $1\times10^{4}cm^{-2}$, can be found along the merging fronts.

A 5μm×5μm AFM scan of the typical surface morphology of the GaN template grown by the two-step method is shown in Fig.3(a). The surface

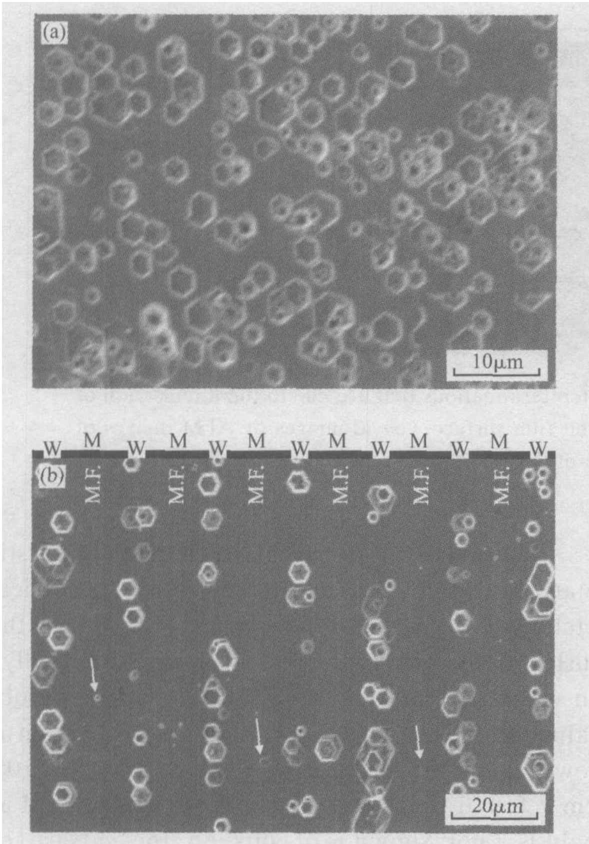

Fig. 2　Surface morphology of GaN template (a) and ELOG GaN (b) after etching in molten KOH, where the symbols M, W, and M. F. indicate the region over the mask, window region, and merging front, respectively

of the GaN template is quite smooth, with a root-mean-square (RMS) roughness of 0.61nm, and consists of well-defined terraces separated by high steps of about 0.3nm. This step height agrees well with a monolayer of (0001) GaN ($c/2 = 0.26$nm). It is generally accepted that the step termination on a single crystal surface results from the intersection of TD of either pure screw or mixed screw-edge character with the free surface [10].

A density of $2.1 \times 10^{8}\mathrm{cm}^{-2}$ of TD with a screw component can be extracted by counting the step terminations in the image. Figure 3(b) shows AFM images of the ELOG GaN. These images are a montage of 4μm ×4μm scans showing TD reduction across the window region. Several step terminations can be found in the GaN grown in the opening, corresponding to a TD density of $2.5 \times 10^{7}\mathrm{cm}^{-2}$ with screw character. In contrast, the laterally overgrown GaN, at least for the surface scanned in this study, is essentially free of step terminations. The absence of such terminations on the wing region indicates that the density of pure screw or mixed character dislocations reaching the surface is much lower in the overgrown GaN than in the coherent GaN.

The right part of Fig. 3(b) reveals that the step structure of the overgrown GaN is dramatically different from that of coherent GaN or template GaN. The atomic steps on the surface of coherent GaN or template GaN are closely associated with the distribution of TDs, as can be seen in Figs. 3(a) and (b). The surface of GaN within the wing region is nearly free of TDs, and thus the steps tend to develop well-defined patterns along crystallographic directions. The steps that are aligned with the $\langle 1\bar{1}00\rangle$ directions exhibit a pairing effect where the width of the terraces alternates between adjacent steps in a given direction, and between adjacent $\langle 1\bar{1}00\rangle$ directions for a given step (see the white arrows in Fig. 3(b)). In some cases the narrow steps seem to disappear completely in a given direction but reappear when the step orientation rotates by 60°. Similar step-edge anisotropy was observed on all samples analyzed here so far and was also found by other groups[11]. The observations of such step-edge anisotropy can be explained by the crystal structure of GaN[12]. For an *hcp* film, the (0001) surface of the terraces consists of Ga atoms with one dangling bond per atom. Note that the GaN bilayers are rotated by 60° for adjacent terraces, as is expected from the symmetry of the wurtzite GaN (6_3 screw axis parallel to the *c* axis). By minimizing the dangling bond density, one can find that the number of dangling bonds per nitrogen atom on successive step edges in the $\langle 11\bar{0}0\rangle$ directions alternates between one and two. Therefore, the termination type alternates between adjacent steps in a given direction and between adjacent $\langle 1\bar{1}00\rangle$ directions for a given step.

TEM analysis was performed to provide further, direct insight into the dislocation behavior in the ELOG sample. Figures 4(a), (b), and (c) show the dislocation arrangement in the wing, window, and merging front regions, respectively. In the wing region (Fig. 4(a)), all the dislocations under the dielectric mask terminate when they encounter the mask. As a result, the wing region is almost free of threading dislocations. In the window region

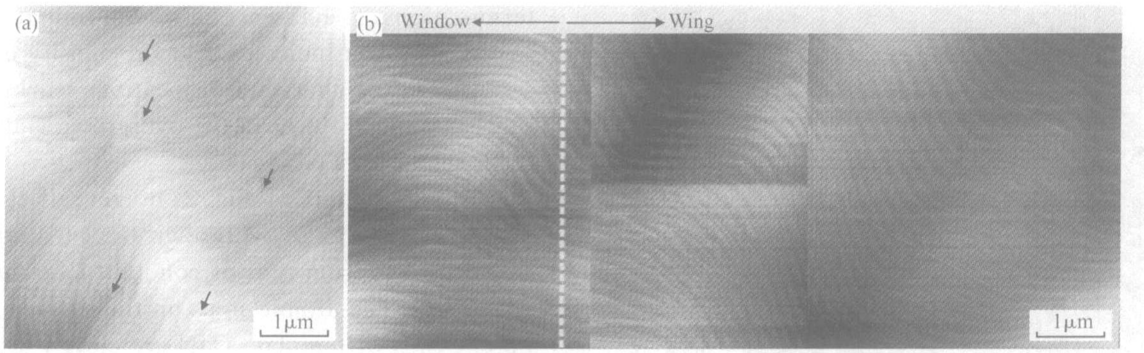

Fig. 3 (a) An AFM image of typical GaN film showing step terminations that are due to the intersection of either a pure screw or mixed threading dislocation with the film surface; (b) Montages of AFM images of successive 4μm ×4μm scans from window to wing, where dark arrows are shown as a guide to the eye to point out the step terminations

(Fig. (4b)), although all the dislocations within the window come from the underlying GaN template, they do not thread through the upper material to the epilayer surface. The dislocation bending behavior within the window may be ascribed to the phenomenon that threading dislocations will change their direction once they intersect the inclined $(11\bar{2}n)$ plane along the growth window edge formed at the end of the first step of the ELOG procedure (the morphology of the ELOG sample at the end of the first growth step is not presented here).

As observed in the cross-sectional SEM image, there is a void between neighboring merging fronts in the central region above the dielectric mask. A coalescence boundary is visible over the void which may have resulted from the coalescence of lateral overgrowth (Fig. 4(c)). Threading dislocations lying in the (0001) plane, which originated from the bending of threading dislocations within the window when encountering the inclined plane in the first growth step, propagated along the $\langle 11\bar{2}0\rangle$ direction and terminated at the coalescence boundary or the side face of the void. A large number of defects were formed at the coalescence boundary, and these defects may result from the coalescence of two neighboring GaN growth fronts which have oblique crystallographic planes with respect to each other[13]. Note also that such a coalescence boundary is accompanied with a high-density of defects that propagate vertically up to the free surface, but the density of these defects is obviously much higher than that observed in wet chemical etching along the merging fronts.

There exists a disagreement between the estimate of dislocation density from wet chemical etching and AFM observations, especially for the dislocations within window regions. Admittedly, in wet chemical etching, the etch pit density obtained is in the range of 10^5 to $10^8 cm^{-2}$, which is lower than the dislocation density of $10^8 \sim 10^{10} cm^{-2}$ found by TEM[14]. In cross sectional TEM analysis (not shown) of pits on the surface of etched GaN, each etch pit corresponds to more than one dislocation, with the result that the dislocation density revealed by wet chemical etching is much lower than that found by AFM and TEM. Therefore, although the etch pit density cannot provide information about the real dislocation density in the epilayer, wet chemical etching is still a simple and efficient way to determine the spatial distribution of dislocations on the surface of ELOG GaN. Additionally, the defects over voids at the coalescence boundary were found in all XTEM images of ELOG GaN analyzed so far, which implies that a grain boundary, instead of small angle grain boundary, exists at the coalescence boundary.

An AFM image scanned across a merging front, shown in Fig. 5, confirms the coalescence boundary observed with XTEM. In Fig. 5, a microscopic step, instead of atomic step, with a fall of tens of nanometers across the neighboring merging front can be found along the merging front. The misorientation between the GaN in the window region and that in the wing region may account for the large height difference of neighboring merging fronts. The defect array formed along the coalescence front is beyond the capability of wet chemical etching, which only reveals a few such defects, as can be seen in Fig. 2(b).

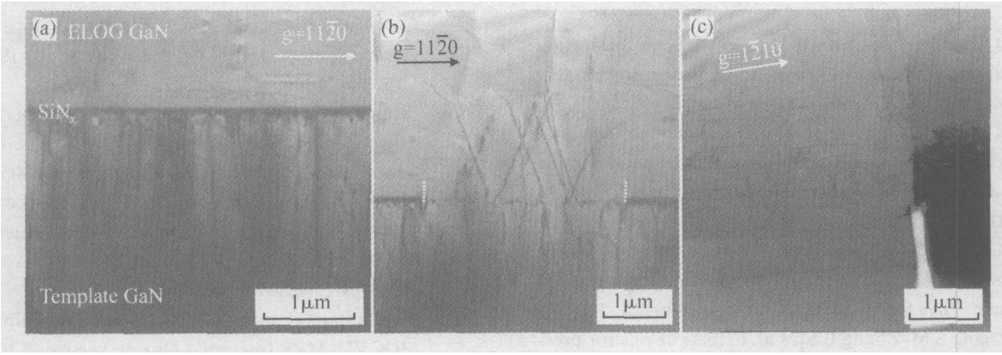

Fig. 4 $(10\bar{1}0)$ cross-sectional TEM images of ELOG sample taken in the wing region (a), window region (b), and the vicinity of the merging front (c)

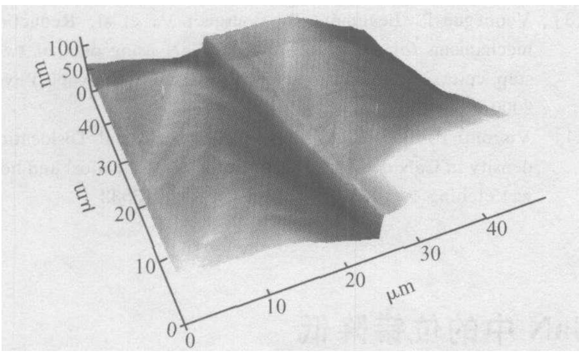

Fig. 5 50μm × 50μm AFM scan of ELOG GaN

Now we discuss the merging front related contrast lines in plane view SEM images as in Figs. 1 and 2(b). The origin of such contrast lines remains unclear. We present two possible explanations for this phenomenon in this paper. The first possible explanation is the enhanced emission of secondary electrons along the merging fronts. It is well known that secondary electron images present a three-dimensional appearance due to the shadowing of emitted electrons by rough surfaces and provide an excellent view of surface topography. The dramatic height difference across the coalescence boundary (see Fig. 5) may lead to the concentration of secondary electrons and consequently the enhanced emission of secondary electrons in the region, which makes the coalescence boundary appear to be a contrast line in the plane view SEM images. The second but no less plausible reason might be the high density of defects formed along the merging fronts, as revealed by the XTEM image in Fig. 4(c). The high density of defects may help to improve the conductivity within the merging front. When struck by the accelerated electron stream in the SEM chamber, these regions tend to emit larger amounts of secondary electrons than those in the adjacent wing regions, and thus contrast lines are visible exactly along the coalescence boundaries.

4 Conclusion

In summary, high quality GaN was grown on GaN substrate with stripe pattern by MOCVD by means of epitaxial lateral overgrowth. The reduction of threading dislocations in the ELOG sample has been studied by AFM, wet chemical etching, and TEM. Experimental results from these characterization methods consistently demonstrate the presence of three distinctive regions in the ELOG GaN sample, namely, window regions with reduced threading dislocations, wing regions with almost no dislocations, and merging fronts with dense arrayed defects. Wet chemical etching cannot exactly determine the dislocation density in GaN, and most of the densely distributed defects along the coalescence front are undetectable in wet chemical etching. However, the extremely low density of threading dislocations within the wing regions makes the ELOG GaN a potential template for the fabrication of nitride-based lasers with improved performance.

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蓝宝石衬底上侧向外延 GaN 中的位错降低*

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摘要: 采用侧向外延(ELOG)方法,在制作了条形掩膜图形的 GaN 衬底上用 MOCVD 生长高质量 GaN. AFM,化学湿法腐蚀及 TEM 分析表明:采用两步法 ELOG 生长的 GaN 中,掩膜下方的缺陷被掩膜所阻挡,窗口区内二次生长的 GaN 的位错也大幅降低;在相邻生长前沿所形成的合并界面处形成晶界;化学湿法腐蚀无法得到关于合并界面处缺陷的信息. 侧翼区域中极低的穿透位错使得 ELOG GaN 适用于在其上制作高性能的氮化物基激光器.

关键词: 金属有机物化学气相沉积; GaN; 侧向外延; 位错

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