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Surface roughness and high density of cubic twins and hexagonal inclusions in cubic GaN epilayers

QU Bo (渠 波)[1,2], LI Shunfeng (李顺峰)[1], HU Guoxin (胡国新)[1], ZHENG Xinhe (郑新和)[1], WANG Yutian (王玉田)[1], LIN Shiming (林世鸣)[1], YANG Hui (杨 辉)[1] & LIANG Junwu (梁骏吾)[1]

1. Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China;
2. Beijing Laboratory of Electron Microscopy, Institute of Physics and Center of Condense Matter Physics, Chinese Academy of Sciences, Beijing 100081, China

Correspondence should be addressed to Qu Bo (email: qubo@red.semi.ac.cn)

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Abstract Surface roughness and its correlation with the polarity of internal hexagonal inclusions and cubic twins have been investigated by atomic force microscopy (AFM), scanning electron microscopy (SEM), transmission electron microscopy (TEM) and X-ray diffraction (XRD). The surface roughness resulted from large amount of strips, which prolonged in $[1\bar{1}0]$ direction with small size in $[110]$ or $[\bar{1}\bar{1}0]$ direction. The sidestep of each strip is just the top of high density of hexagonal inclusions or cubic microtwins. Moreover, XRD shows that the amount of hexagonal inclusions and cubic microtwins measured in $[110]$ direction are twice or more as much as in $[1\bar{1}0]$ direction. Therefore, it is hexagonal inclusions, cubic twins and their distributive polarity that is responsible to the surface characteristics of cubic GaN epilayers.

Keywords: GaN, polarity, surface roughness.

Cubic GaN (c-GaN) with high crystallographic symmetry has superior electronic properties and is easy to be doped or alloyed. The growth temperature of c-GaN is lower than that of hexagonal GaN (h-GaN). And the cubic wafer can also be cleaved easily in the device processing, which can be compatible with the GaAs-based devices. Recently, cubic GaN LEDs have been reported[1, 2].

Although many attempts have been made to grow c-GaN on (001)GaAs substrate, the quality of c-GaN is still inferior to hexagonal GaN (h-GaN) for its metastability. Hexagonal phases and cubic twins are often unexpectedly introduced in the c-GaN epilayers, which have been focused in many researches[2—4]. However, the epitaxial surface of cubic GaN is always so rough that the optoelectronic properties are affected seriously and the device fabrication becomes more difficult. Unfortunately, until now, the serious surface roughness is still neglected. It will be helpful for the amelioration of growth processes and improvement of epitaxial surface to characterize surface roughness and to analyze its formation mechanism.

In this paper, the surface roughness of cubic GaN/GaAs(001) epilayers grown by metalorganic chemical vapor deposition (MOCVD) was characterized by the atomic force microscopy (AFM) and scanning electron microscopy (SEM). Transmission electron microscopy was used successfully to investigate the formation mechanism of surface roughness. Then the distributive polarity of hexagonal inclusions and cubic twins and its effects on surface roughness were measured by XRD.

1 Experimental and results

The c-GaN was grown on (001)GaAs substrates by metalorganic chemical vapor deposition (MOCVD) using NH_3 and triethylgallium (TEGa) as the N and Ga precursors, respectively. H_2 was used as the carrier gas. A thin GaN buffer layer was first grown at 550℃ for 5—10 min, and then a GaN epilayer was grown at 850℃.

1.1 SEM and AFM analysis

Fig. 1(a) shows the secondary electron graph of cubic GaN epitaxial surface by a JOEL-6301F scanning electron microscope. There are large amounts of strips, alternating with light and dark contrast, which prolonged in $[1\bar{1}0]$ direction. The sizes of these strips in [110] or $[\bar{1}\bar{1}0]$ direction are very small, about 10—200 nm, and different from each other. In principal, the excitation of secondary electrons is sensitive to the surface roughness. The turnout of secondary electrons intensively depends on the angle η between incident electron beam and the normal of sample surface, and is the inverse ratio of $\cos\eta$. Thus the different brightness in the secondary electron graph represents the different η in different regions. The stripes with darker brightness are the smoother portions with smaller η, whereas, the stripes with lighter brightness just are the rough region with variational heights and larger η. So the epitaxial surface of cubic GaN films is much smoother in $[1\bar{1}0]$ direction than in [110] direction. The statistical area of rough region is about 20% of the total epitaxial surface. But the relative height of smooth region cannot be determined by SEM graph.

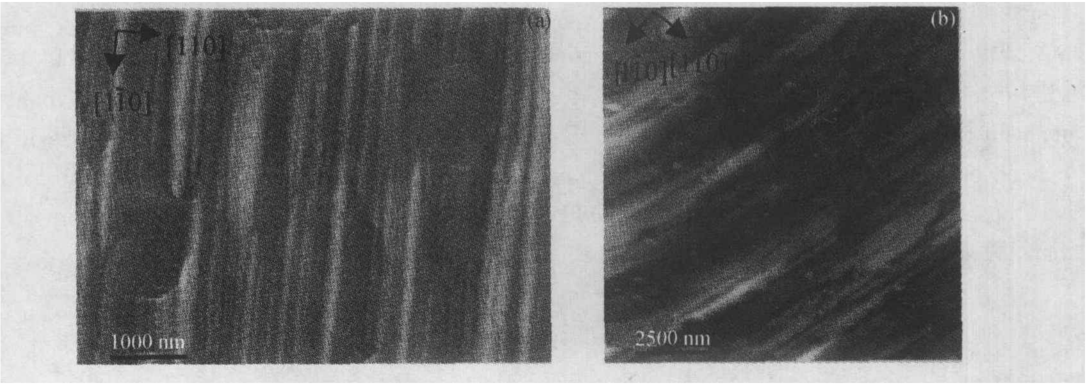

Fig. 1. SEM (a) and AFM (b) graphs of cubic GaN/GaAs (001) epilayers.

Fig. 1(b) shows the atomic force micrograph of cubic GaN epitaxial surface by a CSPM-930 multifunction scanning probe microscope with the light beam reflecting method. Because the invariant force pattern was adopted in this experiment, the different grey scales in atomic force micrograph reveal directly the different epitaxial heights. The smaller the grey scale is, the higher the stripe region is. The maximum value of variational height is about 150 nm. The root of square mean (RSM) of the surface roughness is 14.9 nm. The strips prolonged in $[1\bar{1}0]$ direction have been appeared clearly. And there often is the gradual change for many stripes, especially in [110] direction. However, the grey scale will not be changed distinctly between different regions with variational heights, and these regions cannot be separated easily.

1.2 TEM observation

Fig. 2 shows the microstructure of cubic GaN epilayer by a Philips CM12 transmission electron microscope. High density of disorder dislocations appears near the interface between the GaN epilayer and GaAs substrate. Some straight contrast lines start at some local regions whose density of dislocations is extremely high. These contrast lines are parallel to {111} plane, penetrating into the whole epitaxial layer and result in the local surface rough. It can be concluded that these contrast lines represent the hexagonal inclusions or cubic twins. The higher the density of these contrast lines is, the more serious the epitaxial height changes. There are only tiny height differences in the regions with few contrast lines or without contrast lines.

Fig. 2. TEM microstructures of cubic GaN/GaAs(001) epitaxial films.

For the 700 nm thick epilayer, the epitaxial height is varied about 60 nm. Such rough surface is very disadvantage for the fabrication of cubic GaN-based heterojunction and multilayer structures.

1.3 XRD investigation

The experiment was carried out on a multifunction four-circle X-ray diffractometer with a 4000 W rotating anode Cu target. The CuK_{α} radiation was monochromatized into $CuK_{\alpha 1}$ radiation by Si(111) high-purity crystal and other slits. A transverse and a vertical soller slit were placed in front of the detector so as to improve the monochromatic and parallel quality of incident X-ray. The position precisions used in ω, 2θ, φ and χ movements were as small as 0.01°.

Fig. 3 shows φ scan curves of hexagonal $\{10\bar{1}0\}$ at $\chi = 35.26°$ (a), cubic {111} at $\chi = 15.79°$ (b) and cubic {111} at $\chi = 54.74°$ (c), which were generated from the hexagonal, cubic twin and cubic GaN, respectively[5]. The incident directions of X-ray for the peaks in every φ scan curve are $[110]$, $[1\bar{1}0]$, $[\bar{1}\bar{1}0]$, and $[\bar{1}10]$, respectively. The diffracted intensities of both hexagonal $\{10\bar{1}0\}$ and cubic twin {111} along $[110]$ or $[\bar{1}\bar{1}0]$ direction are more stronger than those along $[1\bar{1}0]$ or $[\bar{1}10]$ direction.

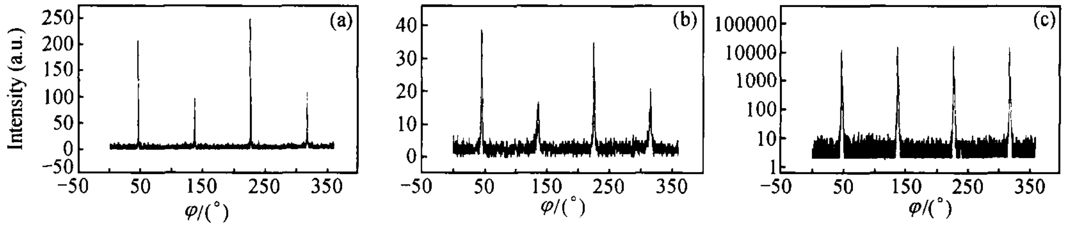

Fig. 3. Hexagonal $\{10\bar{1}0\}$ φ scan curve at $\chi = 35.26°$ (a), cubic {111} φ scan curve at $\chi = 15.79°$ (b) and cubic {111} φ scan curve at $\chi = 54.74°$ (c) of GaN/GaAs (001) epilayers.

Because the hexagonal {0001} planes are parallel to the cubic {111} planes and the twin planes are also {111}, the difference of diffracted intensities in different $\langle 110\rangle$ directions just indicates that the probabilities in respective {111} planes are different to form hexagonal inclusions by stacking faults and cubic twins by unclear mechanism. Thereby, the lamellated hexagonal grains and cubic twins formed in $(111)_{Ga}$ and $(\bar{1}\bar{1}1)_{Ga}$ parallel to $[1\bar{1}0]$ direction are more than those in $(\bar{1}11)_N$ and $(1\bar{1}1)_N$ parallel to $[110]$ direction, which accord with the XRD and RHEED results on cubic GaN epilayers grown by molecular and hydride vapor phase epitaxy by Qin et al.[6] and Tsuchiya et al.[7], respectively.

By calculating the integrated intensities of hexagonal $\{10\bar{1}0\}$ and cubic twin $\{111\}$ in φ scans curves, their contents in $\{111\}$ plane with different polarities can be obtained. The contents of hexagonal inclusions and cubic twins are 2.1% and 0.4%, respectively, in $\{111\}_{Ga}$, and 1.1% and 0.3%, respectively, in $\{111\}_N$. Hence, the contrast lines in TEM graphs are mainly hexagonal inclusions.

2 Discussion

In zinc-blende structure such as cubic GaN without centrosymmetry, the chemical properties are different even for the atomic planes with the same atomic density. The growth rates will be different in $(111)_{Ga}$, $(\bar{1}\bar{1}1)_{Ga}$, $(\bar{1}11)_N$ and $(1\bar{1}1)_N$. And the epitaxial structure will then show some differences in respective directions.

During the early stage of epitaxial growth, the isolated islands of cubic GaN are nucleated and grown. The internal defects are not planar defects, but mainly misfit dislocations in individual nuclei. But the periodicity of the dislocation array will be broken upon their coalescence, and stacking faults and microtwins are generated during this stage[8]. And the local strain concentrations from residual misfit between GaN and GaAs lattice planes are responsible for the generation of these secondary defects such as stacking faults mainly and cubic twins, which are able to fit the coincidence lattice in the region of the coalesced islands. In general, stacking faults will introduce hexagonal phase in the epitaxial films.

Since the growth rates on $(111)_{Ga}$ and $(\bar{1}\bar{1}1)_{Ga}$ planes are often higher than those on $(\bar{1}11)_N$ and $(1\bar{1}1)_N$ planes, the initial islands will be coalesced earlier in $[110]$ or $[\bar{1}\bar{1}0]$ direction and the stacking faults and microtwins then will be formed easily on $(111)_{Ga}$ and $(\bar{1}\bar{1}1)_{Ga}$ planes. Thus the content of hexagonal inclusions and cubic twins on (111)Ga and $(\bar{1}\bar{1}1)_{Ga}$ planes are more than that on $(\bar{1}11)_N$ and $(1\bar{1}1)_N$ planes.

Once the hexagonal phase is nucleated, the respective steps on the growing foreland of stacking faults will provide preferential nucleating position. The cubic twins can also introduce steps to improve the growth rate[9]. Therefore, the foreland of lamellated hexagonal grains and cubic twins are always the region to nucleate easily and grow quickly. These tiny grains can prolong up to the epitaxial surface and result in the serious surface roughness. With the distributive polarity of hexagonal inclusions and cubic twins in different $\langle 110 \rangle$ directions, the variations of epitaxial thickness will then show the similar polarity——strips prolonged in $[1\bar{1}0]$ direction.

3 Conclusion

The correlation between surface roughness and distributive polarity of mixed phases, such as hexagonal inclusions and cubic twins, have been investigated in cubic GaN epitaxial films grown by metalorganic chemical vapor deposition on GaAs(001). SEM and AFM graphs show that there are a lot of strips on the epitaxial surface prolonged in $[1\bar{1}0]$ direction with small size in $[110]$ or $[\bar{1}\bar{1}0]$ direction. TEM microstructure reveals that the lamellated hexagonal inclusions and cubic twins penetrate into the whole epitaxial layer and result in the variational thickness of the epitaxial thickness. XRD shows that the lamellated hexagonal grains and cubic twins formed in $(111)_{Ga}$ and $(\bar{1}\bar{1}1)_{Ga}$ parallel to $[1\bar{1}0]$ direction are more than those in $(\bar{1}11)_N$ and $(1\bar{1}1)_N$ parallel to $[110]$ direction, which is induced by the different growth rates on different $\{111\}$ planes. Therefore, it is hexagonal inclusions, cubic twins and their distributive polarity that is responsible for the surface characterization of cubic GaN epilayers.

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