

Atomically smooth gallium nitride surface prepared by chemical-mechanical polishing with $S_2O_8^{2-}$ - Fe^{2+} based slurry

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Abstract

In this paper, a significant improvement of chemical-mechanical polishing on gallium nitride with $S_2O_8^{2-}$ - Fe^{2+} based slurry is presented in detailed analysis. The results indicate that the $S_2O_8^{2-}$ - Fe^{2+} additives possessed obvious effect to enhance the polishing efficiency of GaN, and successfully achieved good surface quality after polishing. The addition of complexing agent obviously improve the stability of catalytic system. Besides, we also studied the special change rule of atomic step-terrace topography from the surface of GaN to describe the material removal mechanism during CMP process. The results show that the removal of materials by CMP follows rigid rules, which may help to improve the material removal mechanism of CMP.

Keywords: Gallium nitride; Chemical-Mechanical Polishing; $S_2O_8^{2-}$ - Fe^{2+} additive; Step-terrace structure; Material removal mechanism

1. Introduction

In light-emitting diode (LED) manufacturing, Gallium nitride (GaN) has been treated as one of the most potential material for LED substrate, owing to its better crystal lattice matching with epitaxial GaN films, higher thermal conductivity, lower thermal expansion coefficient and higher temperature stability than sapphire substrate [1]. As a result, flat, clean and undamaged surface is essential to form homo-epitaxial GaN layers with a low dislocation density [1]. However, GaN is very difficult to achieve an ideal polishing efficiency because of its high hardness and stable chemical properties [2]. Chemical-mechanical polishing (CMP) is considered to be an effective method to realize atomic-level smooth surface of materials. Many researchers attempted to utilize CMP technique for GaN polishing and obtain a material removal rate (MRR) of 50 nm/min by using alumina-based slurry [3], and the final surfaces were achieved with a roughness range of 0.3~0.6 nm [3-5]. However, these roughness values were too large to meet the requirement of further epitaxy. Then some researchers started to further improve the final surface quality of GaN by using silica-based slurry and obtained a lower roughness of 0.1 nm [6], but the low MRR efficiency (only 17 nm/h) still need further improvement to meet the demand of industrialization.

In our previous work [7-10], a two-step experimental method by using two kinds of abrasives, aluminum oxide (Al_2O_3) and colloidal silica (SiO_2), was put forward for CMP of GaN. The mechanisms of CMP for

GaN (Ga-face) by using Al₂O₃-based and SiO₂-based slurry were studied, and the corresponding removal rates were 594.79 nm/h and 66.88 nm/h respectively. By using Al₂O₃-based slurry, we achieved a high removal rate and reduced the roughness value quickly. By using of SiO₂-based slurry, we removed the scratches created by Al₂O₃-based slurry, and obtained an atomic flat GaN surface with roughness Ra of 0.056 nm, which showed a step-terrace structure. In order to further enhance the efficiency of CMP for GaN and meet the goal of realizing industrial production, new catalytic system, as well as advanced CMP technique are needed.

In this paper, we chose much powerful oxidant and catalytic agent described as S₂O₈²⁻ - Fe²⁺ to further improve the MRR of GaN and the final surface quality of GaN. More advanced AFM technique was introduced to lower the test error of surface characterization. Besides, we also present a detailed change rule of atomic step-terrace topography from the surface of GaN to describe the material removal mechanism of CMP. The periodicity of atomic terraces was also found and explained.

2. Experimental

GaN epilayer (Ga-face) grown on 2-inch-diameter sapphire by HVPE method was used in this study as targeted substrate material need to be polished. The thickness of the epilayer was 50 μm. Al₂O₃-based and SiO₂-based slurries for CMP were prepared. These slurries were synthesized by adding different complexing agents, surfactants and abrasives to deionized water. S₂O₈²⁻ and Fe²⁺ additives were used as oxidizer and catalyst respectively, as well as H₂O₂ - Fe²⁺ treated as control group. Citric acid and sodium dodecyl sulfate (SDS) were used as compound agents and surfactants treated as auxiliary additives respectively. Different polishing pads were used, and the main ingredient on polishing pad is polyurethane.

A two-step experiment method was used in the CMP process of GaN. The Al₂O₃-based slurry was used for the first step and the SiO₂-based slurry was used for the second step. The detailed condition of CMP process was summarized as below: the platen rotation speed was 80 RMP, the carrier rotation speed was 180 RMP, the applied pressure was 0.4 Kg/cm², the diameter of platen was 250 mm, the polishing pad type was non-woven fabric type, the average sizes of colloidal silica particles were 100 nm and 10 nm, the average size of aluminum oxide particles was 1.9 μm, the abrasive concentration was 30%.

GaN substrate was polished by CETR CP-4 machine. The process was carried out by slurry recycling flow. After CMP, the GaN substrate was cleaned by liquid cleaner and deionized water, and then dried off by air spray gun for measurements. The surface topography of GaN was obtained by atomic force microscope (AFM) (Bruker Digital Instruments, Dimension ICON). For AFM, Si probes (SNL-10) were obtained from Bruker, Inc. The super-sharp AFM probes possess a typical tip radius of only 2 nm, which combine the sharpness of a silicon tip with the low spring constants and high sensitivity of a silicon nitride cantilever, for an unprecedented level of high resolution and force control on super smooth surface. The reflective side is Au. The typical resonant frequency is 65 kHz. The typical force constant is 0.35 N/m. The typical aspect

ratio is 3:1. The typical tip cone angle is 22.5° . The scan speed of AFM was 0.8 Hz, and the scan size was 768×768 points. The working temperature was kept at 25°C .

3. Results and discussions

3.1 Improvement of polishing efficiency of GaN by using $\text{S}_2\text{O}_8^{2-} - \text{Fe}^{2+}$ additives with complexing agent

GaN is a kind of super-hard and inert material produced mainly by epitaxy technique [11~24]. In our previous work, in order to improve the removal efficiency of GaN during CMP process, catalytic agents are important ingredients in polishing slurries used to generate hydroxyl free radicals ($\cdot\text{OH}$) [25], which possess strong ability to oxidize the surface of GaN, and then generate a softer layer of gallium oxide, which is much easier to be removed during CMP process. We utilized Fe^{2+} ions as catalytic agents to catalyze H_2O_2 releasing hydroxyl free radicals, and resulted in improving the removal efficiency of GaN by from 16.05 nm/h to 66.88 nm/h, and successfully obtained the atomic step-terrace structure of GaN, which indicated an achievement of atomic-smooth surface quality, as Fig. 1 shows.

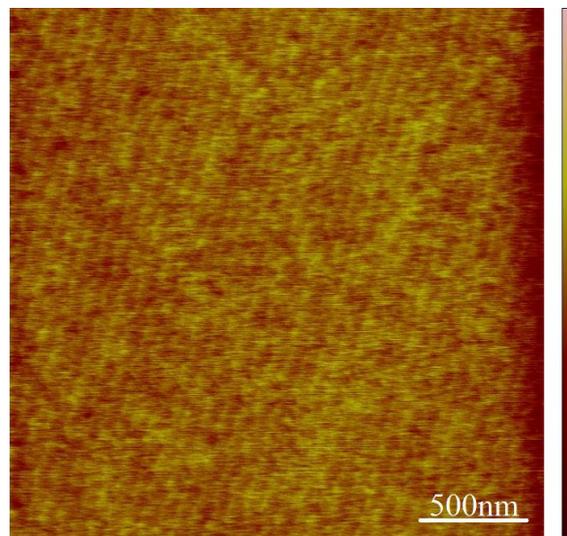


Fig. 1 AFM image of GaN surface after CMP with SiO_2 -based slurry. The surface roughness R_a is 0.056 nm. The scale bar is 1 nm.

However, a removal rate of 66.88 nm/h still not meet the demand of commercialization, and the result shown in Fig. 1 was obtained by a normal scanning AFM probe, which possessed a large tip radius of over 10 nm, resulting in a big error of tested surface roughness R_a and a blur image. By utilizing advanced AFM scanning technique with sharper AFM probe (<2 nm of tip radius), the revised test result of R_a was over 0.09 nm, which was much larger than the original value (0.056 nm).

Due to the instability of $\cdot\text{OH}$, it is easily to lose effectiveness in the slurry during the CMP process, which results in a low polishing efficiency. In order to solve this problem, we chose sulfate radical ($\text{S}_2\text{O}_8^{2-}$) as a new oxidant for enhancing the polishing efficiency of GaN. The $\text{S}_2\text{O}_8^{2-}$ was generated by ionization of persulfate in water. Both H_2O_2 and $\text{S}_2\text{O}_8^{2-}$ possess O – O bond, as Fig. 2 shows.

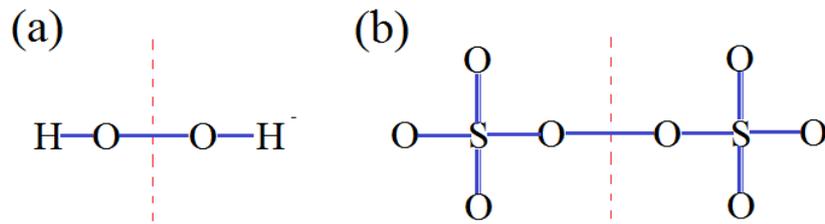


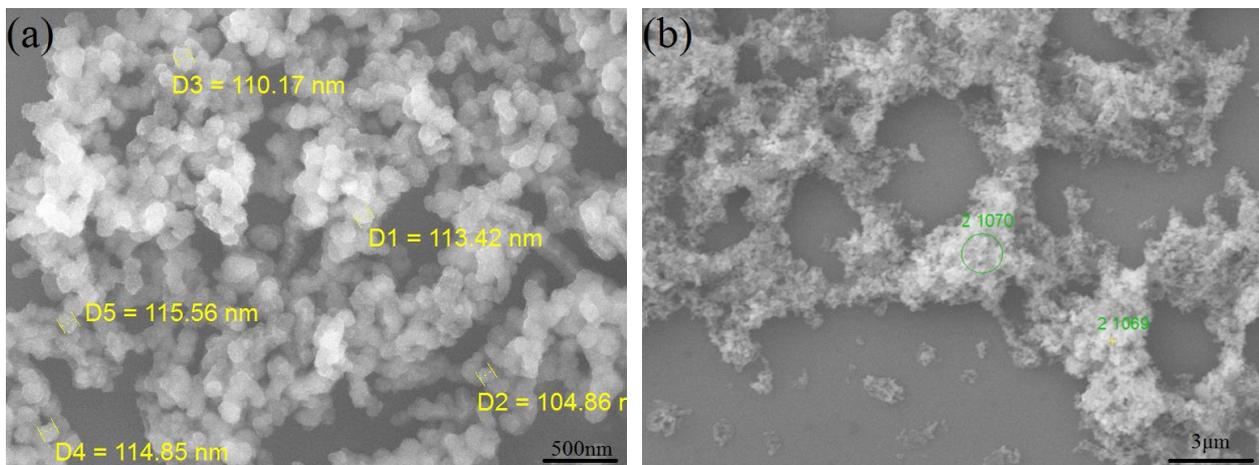
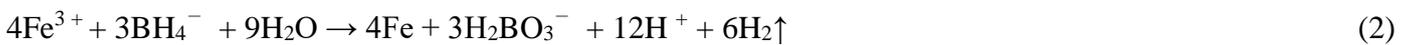
Fig. 2 Structural formulas of (a) H_2O_2 and (b) $\text{S}_2\text{O}_8^{2-}$

At room temperature, both H_2O_2 and persulfate are stable and it needs to add catalytic agent or provide enough energy to break the O – O bond and generate $\cdot\text{OH}$ or $\text{SO}_4^{\cdot-}$. The needed energy or catalytic agent include optical radiation, microwave radiation, heat and transition metal catalysis, such as:



$\text{SO}_4^{\cdot-}$ possess many advantages compared to H_2O_2 . For one thing, $\text{SO}_4^{\cdot-}$ has very high standard electrode potential $E^0 = +2.5 \text{ V} \sim +3.1 \text{ V}$, which is comparable with that of $\cdot\text{OH}$ ($E^0 = +2.7 \text{ V} \sim +2.8 \text{ V}$), indicating that they possess the same ability of oxidization. For another, the half-life period of $\cdot\text{OH}$ is always below $1 \mu\text{s}$, which is much shorter than that of $\text{SO}_4^{\cdot-}$ (about 4 s), indicating that there will be longer time of reaction between $\text{SO}_4^{\cdot-}$ and GaN surface during CMP process. Last but not least, $\text{SO}_4^{\cdot-}$ possess a much wider application range of pH, which is about 2~10.

In terms of the selection of the catalytic agent, compared with heat and ultraviolet light, the reaction of catalyzing persulfate by transition metal ions can be achieved at room temperature and do not need extra energy, so transition metal ions are much more suitable for catalyzing the persulfate during CMP process of GaN. Among all sorts of transition metal ions, Fe^{2+} is chosen as suitable catalytic agent because it is much cheaper and easier to obtain Fe^{2+} , and Fe^{2+} possesses more effectiveness and lower toxicity. The obtaining of Fe^{2+} was realized by the oxidation-reduction reaction of Fe powders and $\text{S}_2\text{O}_8^{2-}$ ions. We used liquid phase reduction method to produce nano-size Fe powders:



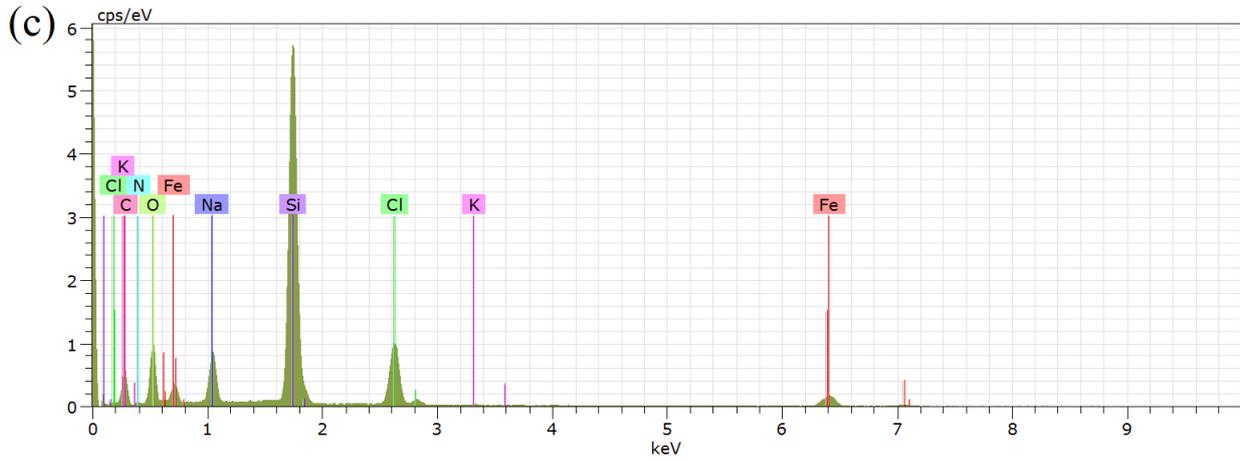


Fig. 3 SEM images of (a) nano-size Fe powders, (b) test spot of EDS (green circle area) and (c) EDS spectrum.

Fig. 3 (a) shows the SEM image of our synthesized nano-size Fe powders. The powders possessed a uniform particle size (110 nm on average) and a good quality of dispersion. This is because that we treated tetraethoxysilane (TEOS) as Si-source to modify the surface of synthesized nano-size Fe powders, which is used to solve the problems of oxidization and aggregation of nano-size Fe powders. EDS result from Fig. 3 (b) and (c) shows that except the element of Fe, there was also content of Si element, the atomic ratio of Si/Fe was $28.89:10.74=2.69$.

Based on the discussion above, we adopted the same CMP technique to compare the polishing efficiency and final surface quality of GaN by using $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system (the oxidant is $\cdot\text{OH}$) and $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system (the oxidant is $\text{SO}_4^{\cdot-}$). For $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system (control team), the polishing slurry contains H_2O_2 , complexing agent, catalytic agent (Fe^{2+}), colloidal silica and deionized water. For $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system, the polishing slurry contains $\text{S}_2\text{O}_8^{2-}$, complexing agent, catalytic agent (Fe^{2+}), colloidal silica and deionized water. The solid contents increased to 30% for both systems, which were much larger than our previous work (5%). The polishing parameters are shown in Table 1.

Table 1 List of Polishing Parameters

Parameters	Values
Polishing Pressure	0.4 Kg/cm ²
Platen Rotation Speed	80 rpm
Carrier Rotation Speed	180 rpm
Velocity of Flow	60 ml/min
pH of Slurry	< 3
Solid Content	30%

Fig. 4 shows the final surface qualities of GaN substrates after CMP by using $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system and $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system based slurries. Both systems could achieve clear and regular atomic step-terrace structure, which indicate that both systems could successfully obtain ultra-smooth surface of GaN. The final roughness Ra of the two substrate were very close, and both of which were much better than our previous work [07], as shown in Fig. 1.

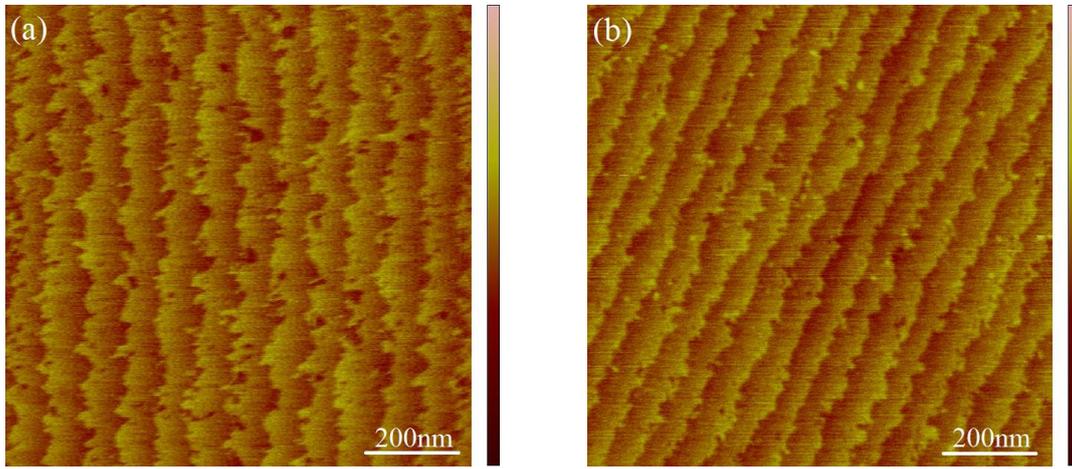
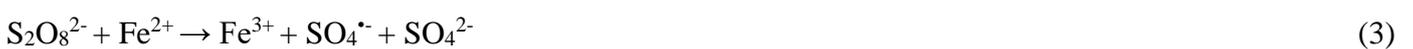


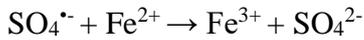
Fig. 4 Atomic step-terrace structure obtained by (a) $\text{S}_2\text{O}_8^{2-}\text{Fe}^{2+}$ system and (b) $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system. The tested roughness R_a was 0.0585 and 0.0565 nm respectively. The scales of images were both 1.2 nm.

However, even though the final surface quality were similar between the two systems, the materials removal rate were obviously different. In terms of $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system, the MRR was 77.5 nm/h on average with a solid content of 30%, which was obvious larger than our previous work (66.88 nm/h on average with a solid content of 5%) [07]. As for $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system, the MRR was 121.1 nm/h on average with a solid content of 30%, which was much larger than that of $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system. The results indicate that with the increasing of solid content, the MRR increased to some extent, and the MRR of using $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system was much larger than that of using $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system.

These results could also be demonstrated by comparison of Fig. 4 (a) and (b). By referred to our previous works we know that the chemical reaction process, as well as the mechanical removal process both always occurred from the edge of terrace in atomic step-terrace structure during CMP [26~28]. Because the MRR of using $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system was much larger, the edge of terrace in Fig. 4 (a) was much rougher than that of Fig. 4 (b), which indicated that the oxidation and removal speed of using $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system were much higher than that of using $\text{H}_2\text{O}_2\text{-Fe}^{2+}$ system (with a much smoother terrace edge).

During CMP process, the formation speed of oxidization layer on GaN substrate surface determine the MRR and final surface quality of GaN. Because the mechanical removal speed was always much faster than the chemical reaction speed during CMP process for these super-hard inert materials [7, 26~35], the oxidization speed mainly determine the MRR. In $\text{S}_2\text{O}_8^{2-}\text{-Fe}^{2+}$ system, the content of catalytic agent Fe^{2+} directly affect the production efficiency of $\text{SO}_4^{\cdot-}$, and further affect the oxidization speed of GaN. Fig. 5 shows the relationship between the content of Fe^{2+} in the slurry and the MRR of GaN during CMP process. It demonstrated that there was a peak MRR value of Fe^{2+} content 2mM. When the content of Fe^{2+} was lower than 2 mM, with the increase of Fe^{2+} , the MRR increased **drastically**. When the content of Fe^{2+} was larger than the peak value, excess Fe^{2+} will react with $\text{SO}_4^{\cdot-}$ and deplete some $\text{SO}_4^{\cdot-}$ in the slurry, resulting in a decline of oxidization speed of GaN, as well as a drop of the MRR. The relevant chemical reaction formulas are listed as:





(5)

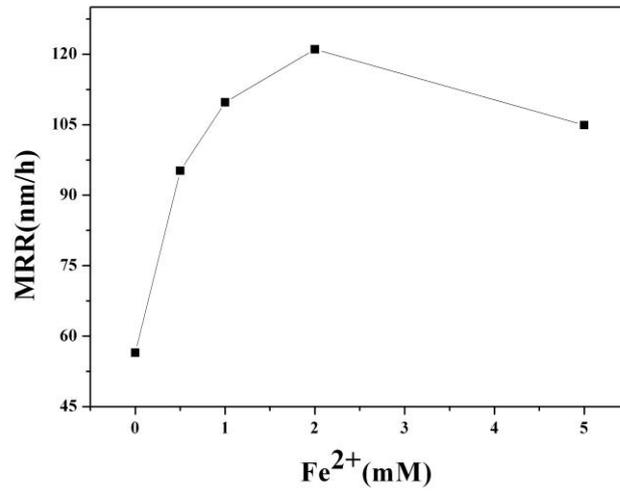


Fig. 5 The relationship between the content of Fe²⁺ in the slurry and the MRR of GaN during CMP process.

In S₂O₈²⁻- Fe²⁺ system, Fe²⁺ will react with SO₄^{·-} and then generate Fe³⁺ deposits such as Fe(OH)₃. These deposits will adhere to the surface of polishing pad and cause the phenomenon of particle aggregation during CMP process, resulting in serious scratches on the surface of GaN substrate, as shown in Fig. 6.

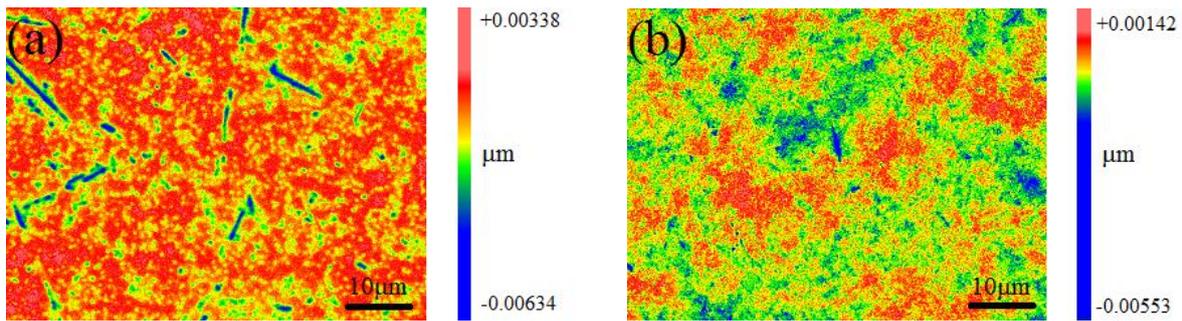


Fig. 6 Optical interferometry profiler images of GaN surface: (a) without complexing agent, Ra=0.708nm, (b) with complexing agent, Ra=0.22 nm.

Fig. 6 demonstrates that the addition of complexing agent could obviously decrease the generation of scratches. However, similar to the situation of Fe²⁺ content, there was also a peak value for the content of complexing agent, as shown in Fig. 7.

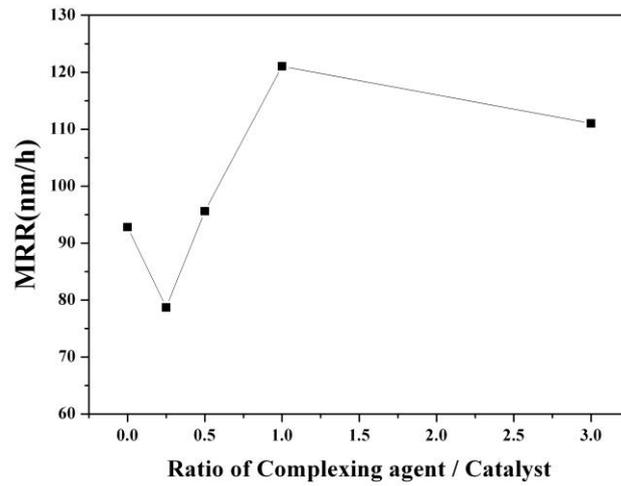
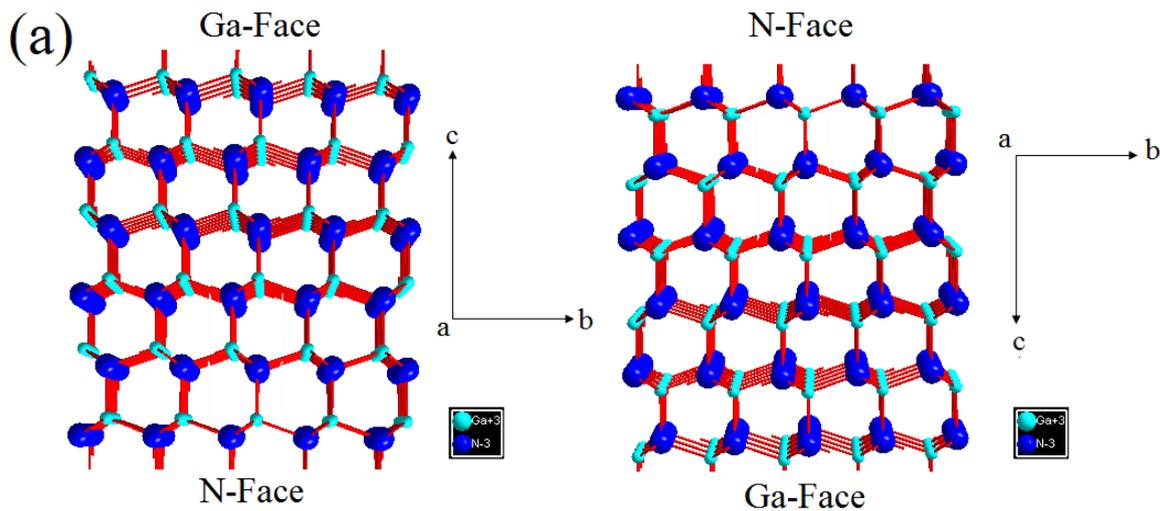


Fig. 7 The relationship between the ratio of complexing agent/catalyst in the slurry and the MRR of GaN during CMP process.

As shown in Fig. 7, the MRR was quite high without complexing agent in the slurry due to the generation of massive scratches. When the complexing agent/catalyst ratio was around 1:1, there was a peak MRR value of 121.1nm/h. If the ratio was smaller than 1:1, excess Fe^{2+} will react with SO_4^{-} and affect the oxidization efficiency of GaN; If the ratio was larger than 1:1, the excess complexing agent will be oxidized by SO_4^{-} and decrease the amount of SO_4^{-} , both result in a decrease of MRR of GaN.

3.2 The study of change rules of atomic step-terrace structure on GaN substrate during CMP process

The ideal crystal structure and the real crystal structure of GaN substrate surface (atomic step-terrace structure) after CMP process are shown in Fig. 8.



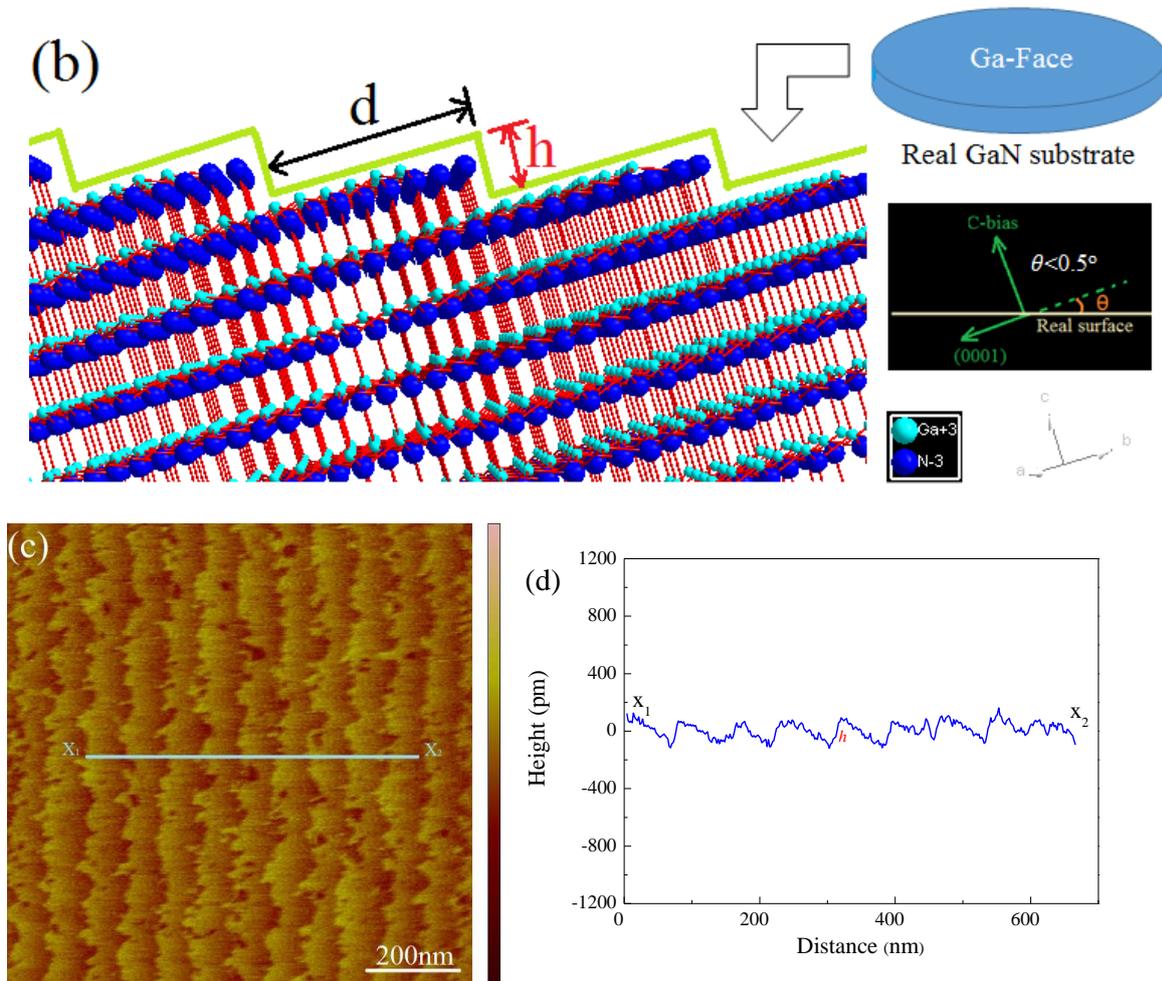


Fig. 8 Ideal crystal structure (a) and the real crystal structure of GaN substrate surface (b) compared to real atomic step-terrace structure (c) and its section line (d) after CMP process. The scale bar of (c) is 1.2 nm.

By comparison of Fig. 8 (b) and (c), it is obvious that the atomic step-terrace structure is determined by the crystal structure of GaN and indicated real crystallographic information of GaN substrate surface. Ga-N layer are periodically assembled by nitrogen atoms and gallium atoms. For the Ga-face of GaN substrate, if the c-axis orientation is absolute 0° -off, we can obtain an ideal smooth surface as Fig. 8 (a) shows. However, there is always an off-angle deflection ($\theta < 0.5^\circ$) with the c-axis orientation due to the impact of dislocations in GaN lattice, as well as the impact of CMP technique [26~28, 33, 35]. This slight deflection of c-axis causes the whole stacking Ga-N layers deflect slightly at a small angle θ . These deflected stacked layers terminate at the surface of GaN substrate, and achieve a step-terrace topography as a real consequence. From the section curve acquired from the atomic step-terrace structure in fig. 8 (d), the average height of terraces was about 0.25 nm on average, which was same as the theoretical thickness of Ga-N layers (0.25 nm). These results demonstrated that the obtaining of atomic step-terrace structure could confirm an achievement of atomic-level smooth surface of GaN due to the reveal of crystal structure information.

Experiments demonstrated that there is directly crystal relationship of the surface topographies of GaN substrate before and after CMP process. Fig. 9 shows the surface topographies of GaN substrate before CMP process.

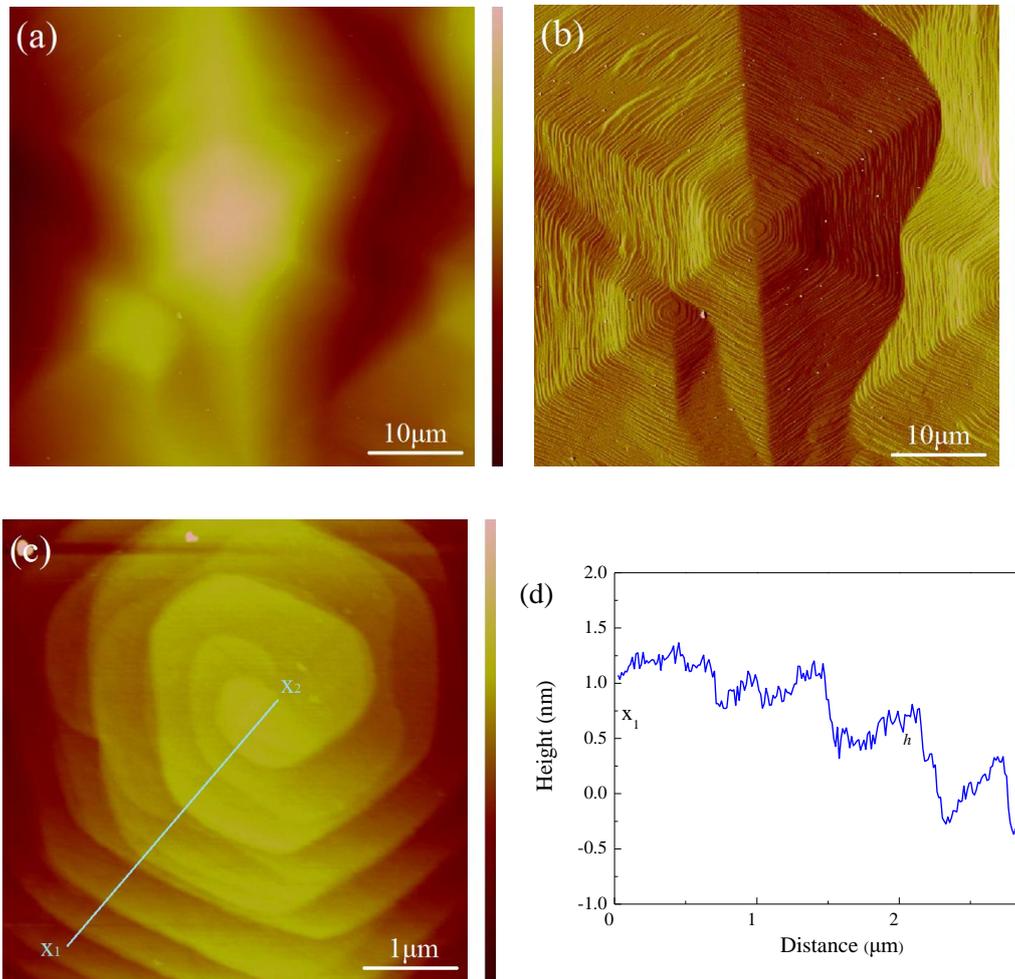
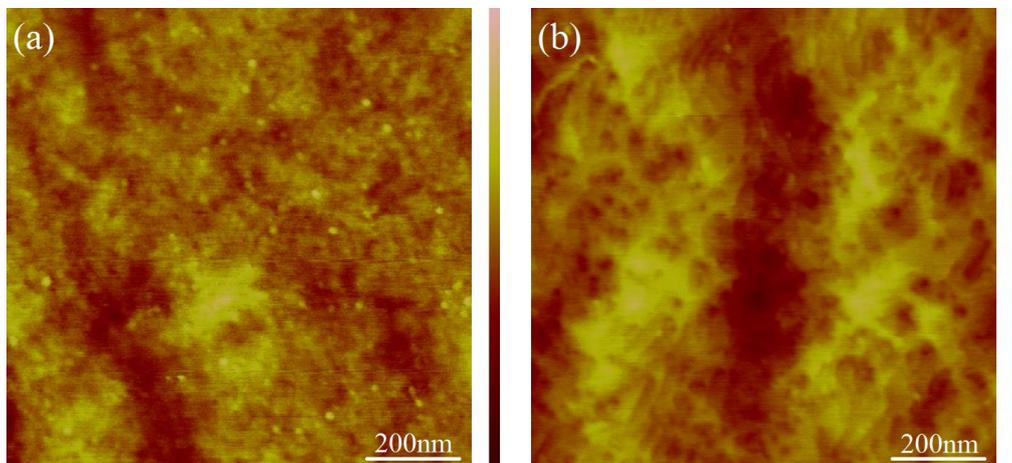


Fig. 9 AFM images of original GaN substrate surface before CMP process: (a) topography image, (b) deflection error image of (a), (c) detailed topography image of conical peak in the center of (a), (d) the section curve of (c). The scale bars are 100 nm, 180 mV and 6 nm respectively.

As shown in Fig. 9 (a) and (b), there were many hexagonal conical protrusions on original GaN substrate surface, which indicated the crystal information of GaN crystal growth. The original surface was very rough, with a huge terrace height of around 1 nm tested in Fig. 9 (c) and (d), and the surface roughness R_a was 13.1 nm. It need appropriate CMP technique to realize atomic-level smooth surface.



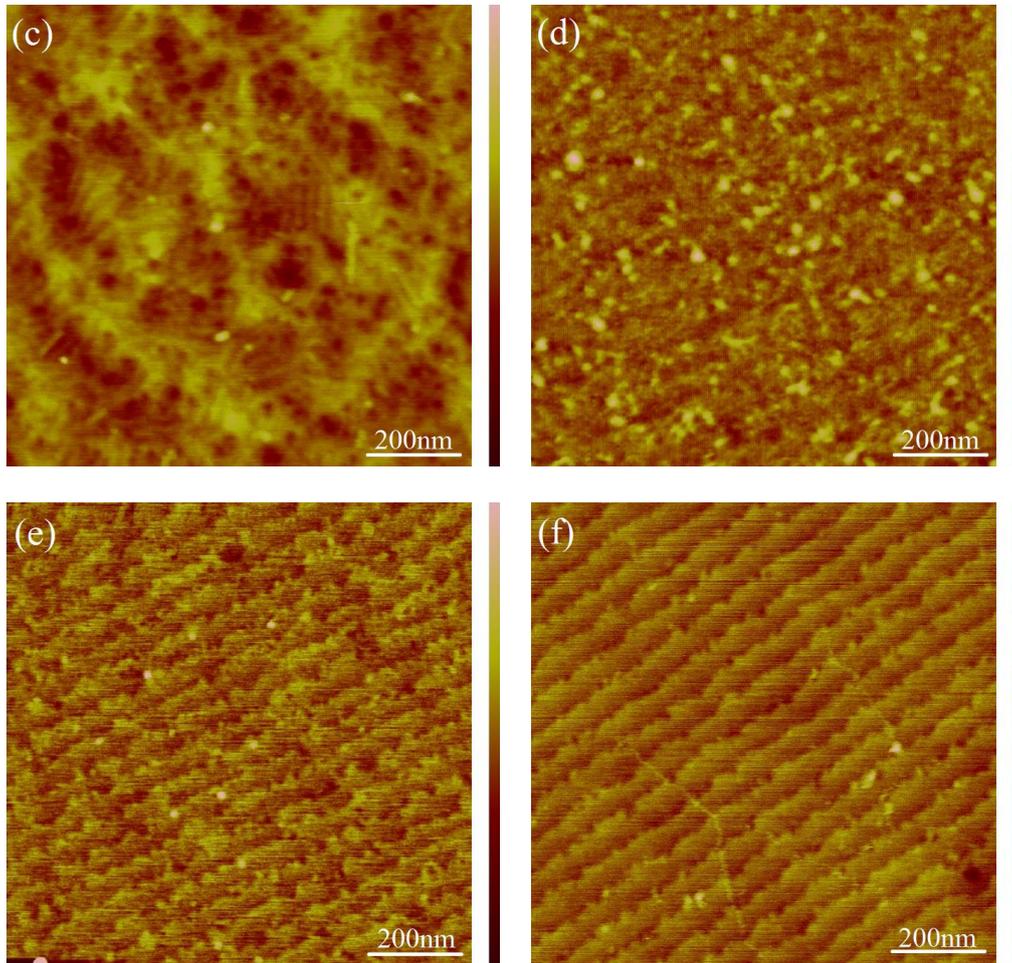


Fig. 10 AFM images of GaN substrate surface topographies during CMP process: (a) after polishing by Al_2O_3 based slurry, (b) polished by 100 nm SiO_2 based slurry for 0.5 h after (a), (c) polished by 100 nm SiO_2 based slurry for 1 h after (a), (d) polished by 100 nm SiO_2 based slurry until regular step-terrace structure appeared, (e) polished by 100 nm SiO_2 based slurry until the step-terrace structure unchanged, (f) polished by 10 nm SiO_2 based slurry until the step-terrace structure unchanged. The scale bars are 4.5 nm, 4.5 nm, 3 nm, 1.5 nm, 1 nm and 1 nm respectively. The surface roughness R_a were 0.430 nm, 0.352 nm, 0.191 nm, 0.0868 nm, 0.0626 nm and 0.0516 nm respectively.

Original GaN substrate surface should firstly be polished by using Al_2O_3 based slurry, which possessed a much larger average abrasive size (about 1 μm) and was used for rapid removal of rough hexagonal conical protrusions on original GaN substrate surface mechanically (rough polishing). At this stage, the MRR was very high (around 600 nm/h). Until the GaN surface roughness did not change anymore (R_a was stable at around 0.5 nm) as shown in Fig. 10 (a), then the Al_2O_3 based slurry was replaced by SiO_2 based slurry for the precise polish. The SiO_2 based slurry possessed a much lower average abrasive size of 100 nm, and the hardness of SiO_2 abrasive was much lower than that of Al_2O_3 abrasives. From Fig. 10 (a) we can see that there was no step-terrace structure at all after rough polishing by Al_2O_3 based slurry. However, after polishing by SiO_2 based slurry for 0.5 hour, the GaN surface began to emerge step-terrace structure, as shown in Fig. 10 (b). At this stage, the newly emerged step-terrace structure was very blur, and the orientation of terraces was also irregular. The surface roughness R_a decreased from 0.43 nm to 0.35 nm. After polishing by SiO_2 based slurry for 1 hour, as shown in Fig. 10 (c), the surface of GaN became much smoother, and the step-terrace structure also became clearer than before, even though the orientation of

terraces was still irregular. At this stage, the surface roughness Ra continued to decrease to 0.19 nm. When the Ra value was below 0.1 nm, the step-terrace structure began to have a regular orientation as shown in Fig. 10 (d), but the terrace structure was still blur under small test scale (1.5 nm). When the surface roughness decreased until the Ra value unchanged (kept at 0.06 nm), the GaN substrate achieved an atomic-level ultra-smooth surface with a clear and regular step-terrace structure, as shown in Fig. 10 (e). If we continued to use nano-size SiO₂ based slurry (with an average abrasive size of only 10 nm [26~27, 29]) to polish this substrate, the edge of terraces would become more smooth (transformed from zig-zag to linear), and the roughness Ra would continue to decrease to 0.05 nm, which was an ultimate value close to the theoretical result. However, the MRR of using 10 nm SiO₂ based slurry was much lower than that of using 100 nm SiO₂ based slurry (about only 1/10 of efficiency of latter one), that is also why the edge of terrace in Fig. 4 (a) was rougher than that of in Fig. 4 (b).

All the results above was obtained from the Ga-Face of GaN substrate. Because the further epitaxy procedure mainly utilizes the Ga-Face of GaN substrate, the N-Face was seldom to study in recent years. However, there was totally different CMP results for different faces, as shown in Fig. 11.

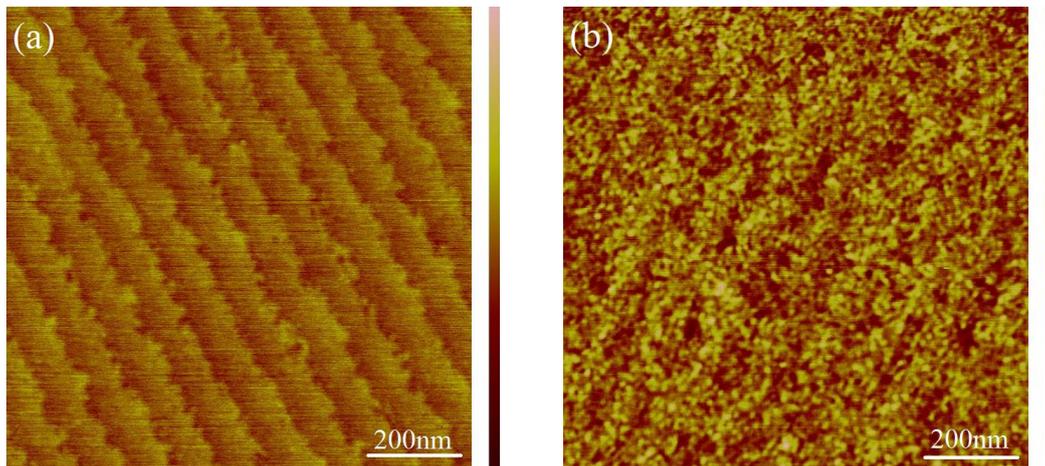


Fig. 11 AFM images of GaN substrate surface topographies after CMP process: (a) Ga-Face (b) N-Face. The scale bars are 1 nm and 2 nm respectively. The roughness Ra are 0.0539 and 0.189 nm respectively.

As shown in Fig. 11, with the same CMP technique, the final surface results of different faces were totally different. Even though both faces had the same terrace width, which indicated that they came from the same substrate, the surface roughness Ra of Ga-Face was much lower than that of N-Face, and the terrace structure of Ga-Face was also much clearer. Besides, the terrace structure of Ga-Face and N-Face possess opposite orientations. This is because that for Ga-Face, the most outside atoms of substrate surface were Ga atoms, as shown in Fig. 8 (a). During CMP process, the chemical reaction (oxidization) on GaN surface was carried out as Ga atoms oxidized by SO₄²⁻, so it was more easily for Ga-Face to react with oxidizing agent (SO₄²⁻) than N-Face, and removed by CMP as a result.

At last, we will discuss the phenomenon of periodicity of atomic step-terrace structure on GaN substrate surface during CMP process, as shown in Fig. 12.

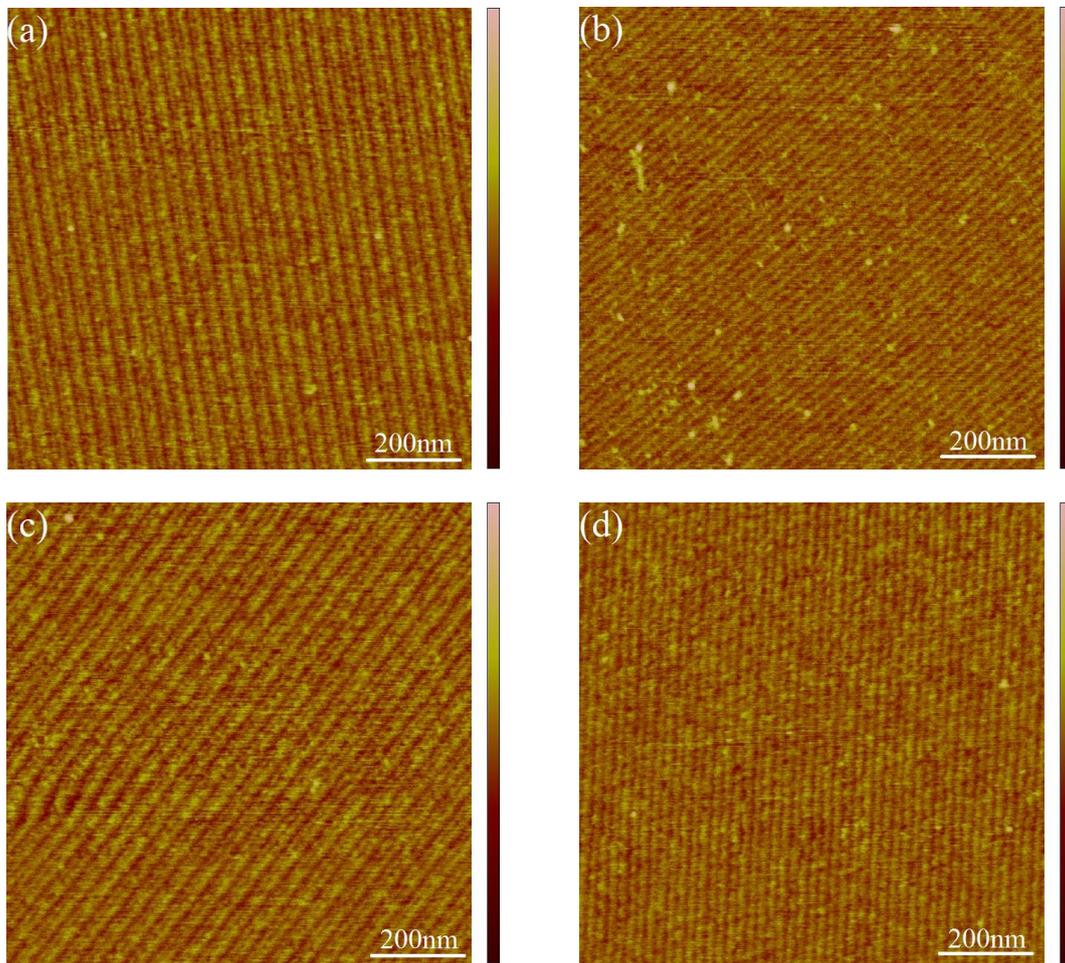


Fig. 12 AFM images of GaN substrate surface topographies after CMP process. (a), (b), (c) and (d) derived from different areas of one GaN substrate. All scale bars are 1 nm.

Similar to the situation of SiC [27~28, 36], GaN substrate can also represent periodicity of atomic step-terrace structure during CMP process, as shown in Fig. 12 (a) and (c). Both the periodicity phenomenon of atomic step-terrace structure on GaN and SiC substrate could be obtained by using nano-size (10 nm) SiO₂ based slurries for ultra-precise polishing. However, in terms of this periodicity, there are still many differences between SiC and GaN. For one thing, GaN substrate should possess a large off-angle deflection θ to obtain periodicity, which caused a narrow-arranged terraces structure with a much smaller terrace width than before, as shown in Fig. 12. For another, not all areas from one GaN substrate could emerge obvious periodicity, the step-terrace structure shown in Fig. 12 (b) and (d) did not show apparent periodicity. This may be caused by the heterogeneity of real CMP process, such as different polishing pressure or different amount of slurry applied in these area. It can be preliminarily concluded that the stacking sequence of Ga-N layers (with a typical sequence of ABABAB...) may cause this periodicity due to that the efficiency of chemical reaction (oxidization, react with oxidizing agent) were different between layer A and layer B. In other words, the bond energy, as well as the bond angle of Ga-N bond in layer A and layer B are slightly different. When the polishing process was as precise as atomic-level, this slight difference may be revealed as the removal priority of layer A and layer B, which results in a periodicity of step-terrace structure.

If there was defect on the GaN Substrate, the periodicity would be more easily to be achieved, as shown in

Fig. 13.

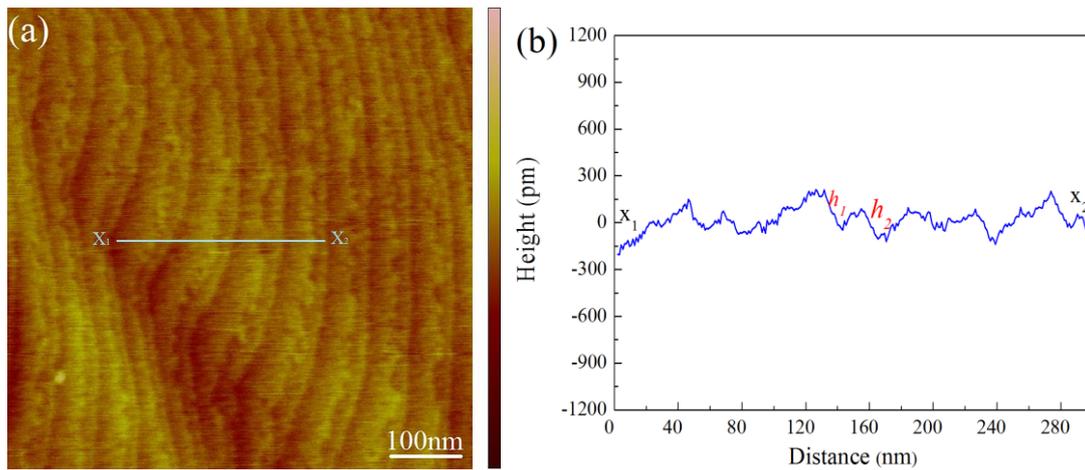


Fig. 13 AFM image of GaN substrate surface near defects after CMP process: (a) topography image, (b) the section curve of (a). The scale bar is 2 nm. $h_1=h_2=0.25$ nm.

In Fig.13 (a), a defect on the surface of GaN made this area become not plain, thus fulfill the potential requirement of periodicity. A regular atomic step-terrace structure, as well as terrace heights $h_1=h_2=0.25$ nm shown in Fig. 13 (b) demonstrated that the surface had achieved atomic-level smooth. The edges of layer A and layer B characterized was totally different as one layer (may be Layer A or B) was with a smooth linear edge, and the next layer (may be Layer B or A) was with a zig-zag edge, periodically. This was a very interesting phenomenon. More experiments need to be undertook in the future to reveal the mechanism of this periodicity.

4. Conclusion

In order to improve the polishing efficiency for GaN substrate by CMP technique, we successfully synthesized nano-size Fe powders as a source of catalytic agent Fe^{2+} , and used $S_2O_8^{2-}$ as a source of oxidizing agent $SO_4^{\cdot-}$. $SO_4^{\cdot-} - Fe^{2+}$ additives based slurry is presented in detailed analysis. The results indicate that the $S_2O_8^{2-} - Fe^{2+}$ additives possessed obvious effect to enhance the polishing efficiency of GaN, and successfully achieved good surface quality after polishing. The addition of complexing agent obviously improve the stability of catalytic system, and the application of advanced AFM technique was introduced to lower the test error of surface characterization.

In terms of the study of the removal mechanism of GaN substrate by CMP technique, we introduced the original surface topography of GaN substrate, studied the special change rule of atomic step-terrace structure to describe the material removal mechanism during CMP process. The results showed that the removal of GaN by CMP followed rigid rules, which may help to improve the material removal mechanism of CMP. Besides, CMP results of both Ga-Face and N-Face were obtained, and the mechanism of terrace periodicity was preliminarily discussed. All of results may provide some ideas and suggestions for the studies of CMP, crystal growth and epitaxy research areas.

Acknowledgements

The authors would like to thank National Key Basic Research Program of China - 973 Program (No. 2011CB013102) for a grant to this research. The support from National Natural Science Foundation of China (No. 91223202) is also gratefully acknowledged. Meanwhile, this research has been supported by a grant from the International Science & Technology Cooperation Program of China (No.2011DFA73410) and the Foundation for Innovative Research Groups of the National Natural Science Foundation of China (No.51321092).

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