M miscut dependent surface evolution in the process of N-polar GaN(0001) growth under N-rich condition

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A B S T R A C T

The evolution of surface morphology during the growth of N-polar (0001) GaN under N-rich conditions is studied by kinetic Monte Carlo (kMC) simulations for two substrates miscuts 2° and 4°. The results are compared with experimentally observed surface morphologies of (0001) GaN layers grown by plasma-assisted molecular beam epitaxy. The proposed kMC two-component model of GaN(0001) surface where both types of atoms, nitrogen and gallium, attach to the surface and diffuse independently shows that at relatively high rates of the step flow (miscut angle <2°) the low mobility of gallium adatoms causes surface instabilities and leads to experimentally observed roughening while for low rates of the step flow (miscut 4°), smooth surface can be obtained. In the presence of almost immobile nitrogen atoms under N-rich conditions crystal growth is realized by the process of two-dimensional island nucleation and coalescence. Larger crystal miscut, lower growth rate or higher temperature results in similar effect of the surface smoothening. We show that the surface also smoothen for the growth conditions with very high N-excess. In the presence of large number of nitrogen atoms the mobility of gallium atoms changes locally thus providing easier coalescence of separated island.

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1. Introduction

The fact that in wurtzite N-polar GaN, the direction of the polarization fields is inverse to the fields direction in the Ga-polar orientation, makes the N-polar GaN of interest for a variety of device applications such as high electron mobility transistors, solar cells, sensors and light emitting diodes [1–3]. Identification of the optimum growth conditions providing smooth surface morphology N-polar (0001) GaN is one of the key issues to enhance the development of these emerging applications. The growth of atomically flat N-polar (0001) GaN layers by plasma-assisted molecular beam epitaxy (PAMBE) has been proven difficult due to the high adatom diffusion barriers because it is carried out at relatively low temperatures (750 °C). Theoretical works showed that these diffusion barriers are substantially reduced when gallium adlayer is present on the surface during the growth [4–7]. However, previous experimental studies showed that the growth of the N-face GaN must be performed with less than one monolayer (ML) of excess Ga on the growing surface, in contrast to Ga-polarity, when more than 2ML can be stabilized. Therefore, a precise control of the growth conditions is needed to prevent the accumulation of Ga droplets [8]. The morphologies reported for N-polar GaN layers grown by PAMBE under N-rich conditions on low miscut substrates were relatively rough either due to the hillocks or pattern formed by interlacing stripes covered by doubly bunched atomic steps [9]. It is important to note that in case of metal-organic vapor phase epitaxy (MOVPE), the N-polar (0001) GaN films for many years suffered from high surface roughness resulting from the formation of large hexagonal hillocks [10]. The use of 4° miscut N-polar GaN substrates has proven to be effective method of improving surface morphology [3]. In search for the optimum growth conditions of the device-quality N-polar GaN layers it is essential to understand the role of surface miscut, growth temperature, growth rate and III/V ratio (N-excess) that may improve surface morphology.

In this work the growth on the N-polar GaN (0001) wurtzite surface under N-rich conditions is investigated theoretically by kinetic Monte Carlo (KMC) method. KMC is a popular method of study evolution of the surface during crystal growth at different modes [11–15]. Simulations are compared to the morphologies of N-polar GaN (0001) layers grown by PAMBE under N-rich conditions in order to identify the optimum growth conditions. We built

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two-component model of GaN (0001) surface of wurtzite crystal. We use the model that is based on the one previously implemented to study growth and sublimation of GaN(0001) surface [16–19]. The model was changed in such a way that both types of atoms building crystal: nitrogen and gallium are controlled, similarly as in the one applied for SiC crystal in Ref. [20]. Both crystal components attach to the surface, diffuse and detach independently. They can be adsorbed at the steps or form islands by nucleation at the terraces. In our study we apply 1 eV barrier for Ga diffusion as evaluated for N-terminated GaN(0001) surface in ab initio calculations [4] and 1.8 eV high barrier for diffusion of N adatoms. With such slow surface dynamics we model the crystal growth under large nitrogen flux at relatively high surface miscut angles. We choose external fluxes and desorption rates in such a way that simulations follow experimental crystal growth rates. As a result the experimentally observed morphology patterns are well reproduced.

We find that low miscut angle (≤ 2°) results in characteristic surface instabilities (and subsequent 3D growth mode), while for steeper cuts step flow growth mode resulting in double-step surface pattern is observed. Other methods of surface smoothening lead to slightly different surface morphologies. For instance higher temperature smoothness the surface, but contrary to the high miscut case no double step structure can be seen. Distances between all steps are equal suggesting that at higher temperature all steps move with the same rate at low external particle flux (slow crystal growth rate). Differences can be seen when appropriate correlation functions are compared. It is shown that step roughness can be smoothened also using increased nitrogen flux. As nitrogen practically does not move along the surface at studied temperatures and the rate of gallium adatoms diffusion is also below limiting value, the character of this process is different than that discussed above. 2D growth mode is observed. Islands that grow along step-edges and finally attach to the step. Resulting smooth crystal growth is realized by two dimensional island nucleation. We show that a flat surface is developed at relatively low miscut at high nitrogen rates. We discuss numerical data as a function of the growth temperature, growth rate defined by Ga flux and substrate miscut angle. The kMC results are compared with experimental data.

First in Section 2 the model is formulated, then in Section 3 we show how the surface structure changes with miscut, temperature, gallium and nitrogen flux. Surface patterns obtained for the chosen parameters are compared to the experimental data. Finally on the basis of the surface dependence on the nitrogen flux we consider possibility of the surface smoothening via use of higher nitrogen fluxes.

2. Kinetic Monte Carlo model

Numerical simulations of the system were performed with the use of two-component kinetic Monte Carlo (kMC) model of wurtzite GaN crystals. It consists of gallium and nitrogen atoms arranged in a crystallographic lattice of N-face (0001) of gallium nitride. Two species interact via nearest neighbor (NN) and next nearest neighbor (NNN) forces which form N–N, Ga–Ga, and Ga–N bonds. Total energy of an adatom occupying one of the lattice sites is given by the sum over all NNN and NNNN that lie in the same layer or within the layers below and sometimes above:

\[ E_X = E_{GaN} \sum_{NN} n_i + E_{NNN} \sum_{NNN} n_i \]  

(1)

where \( X \) corresponds to Ga or N atom and the value \( n_i \) depends on the state of the neighboring site. If the site is occupied \( n_i = 1 \) otherwise \( n_i = 0 \). Sites occupied by gallium atoms are different from those that are occupied by nitrogen, hence Ga can be adsorbed at Ga covered layer, but with nitrogen layer missed below it will be very weakly attached to it. The same happens for N adatoms at N-covered surface. Energies associated with interactions are \( E_{NN} = 0.3 \) eV, \( E_{NNN} = 0.35 \) eV and \( E_{GaN} = 1.6 \) eV respectively. They were chosen in such a way that total energy of Ga atom built in the crystal is 10.6 eV. This value is in agreement with Ref. [21].

Single simulation consists of several Monte Carlo steps performed by Metropolis algorithm. Each of them starts with particle adsorption which occurs with probability equal to the external flux \( F \) at every site of the system [19]. At each MC step particles are effectively deposited at randomly chosen sites. Adsorbed particles are allowed to diffuse at the surface. That process is modeled in three stages. At first one of the neighboring sites is randomly chosen to be the target of a jumping particle. Next initial \( (E_i) \) and final \( (E_f) \) energies of jumping atom are calculated using Eq. (1). In the third stage of diffusion process particles jump with probabilities

\[ P_{ij} = \begin{cases} e^{\beta(E_f - E_i - \Delta E_{ij})} & \text{when } E_i \geq E_f \\ e^{-\beta \Delta E_{ij}} & \text{otherwise} \end{cases} \]  

(2)

where \( \beta = (k_B T)^{-1} \) and \( \nu = 10^{11} s^{-1} \) sets the time scale. \( B_{ij} \) denotes the diffusion barrier that is different for \( X = \text{Ga, N} \) adatoms. It is known that diffusion barriers for both components under N-rich conditions on GaN(0001) surface are high [4–7]. We use barriers calculated in Ref. [4] for Ga adatoms diffusing over N-terminated (0001) surface equal to \( B^\text{GaG} = 1.8 \) eV. Ga adatoms can diffuse over N covered layer as well as over Ga covered layer, but in this last case the binding energy of Ga adatom is much lower because of lack of the N layer below. The same situation is true for diffusing N adatoms. According to Eq. (2) particle, which is strongly binded in its initial state, resides in a deep potential well and probability of the jump out is low. However when final state has low energy, i.e. particle tries to move to the shallow well the energy barrier between these states decreases and so is jump rate. Finally jumps towards step from above occur with the same probability like those far from the step in such a way that effectively no additional barrier for jumps across step (Schwoebel barrier) is present in the studied systems.

During the last stage of a single MC step, the evaporation phenomenon is simulated. An assumption that incoming atoms can evaporate is necessary in order to reach equal number of Ga and N atoms within the growing crystal. It is known that desorption of N adatoms happens via creating \( N_2 \) molecule first. We do not take into account this process. For high values of \( N \) fluxes, when almost each particle has at least one neighboring particle we do not expect large difference in results in the case when single particle or molecule \( N_2 \) desorption process is studied. We set the following rate of particle desorption:

\[ D_X = \frac{1}{\mathfrak{D} - \beta E_i + \Delta E_v} \]  

(3)

where \( \mathfrak{D}_X \) is the desorption potential, different for Ga and N adatoms. We set \( \mathfrak{D}_N = -0.7 \) eV taking into account that binding energy of single N atom at the surface \( E_N = 5.7 \) eV while the same atom above the (0001) GaN surface covered by nitrogen \( E_i = 0.9 \) eV. This value of desorption barrier ensures that most of N adatoms that are not bonded to gallium atoms in the layer below, desorb from the surface, whereas the ones that have fallen onto the gallium layer stick to the surface. For gallium we set \( \mathfrak{D}_Ga = 0 \). Such choice of desorption potentials provides that the number of adsorbed atoms of both types equalizes at the surface. After evaporation stage new MC step begins and the above procedure
repeats.
At the beginning of each simulated process, the system consists of \( N_t \) straight steps. Helical boundary conditions in one direction and periodic in the second one are posed. Gallium nitride elementary cell consists of four mono-atomic layers, hence in order to close helical boundary condition properly, \( N_t \) is always divisible by 4. Additionally, because N-rich conditions are studied, we start the simulation with total N coverage of the surface that changes during the growth process by evaporation of adatoms.

Summarizing, the MC model used to describe N-polar GaN(0001) growth under N-rich conditions is built of two types of atoms: Ga adatoms that move slowly but are able to diffuse across terraces and reach steps and N adatoms that are almost immobile, but can desorb from the surface and are frequently adsorbed because of the relatively high flux of N atoms. The consequence of such assumptions is that both atomic monolayers (nitrogen and gallium) can grow smoothly within some range of parameters, but due to totally different mechanisms. Gallium adatoms diffuse over the surface, and they start to build islands when incoming flux is too high, whereas nitrogen adatoms being practically immobile build islands all the time. Nitrogen islands have two different roles in the growth process: they build in the step thus completing the layer formation, but they also change diffusion of Ga adatoms. Below we analyze consequences of such adatom dynamics for the evolution of stepped surfaces.

3. MBE experimental details

In parallel we carried out the growth of 200 nm thick N-polar GaN layers by PAMBE custom design Gen20A MBE reactor where active nitrogen was supplied from Veeco RF plasma source. We used bulk, commercially available GaN substrates from Saint Go-bain with threading dislocation density around 5 \( \times 10^7 \) cm\(^{-2} \). Prior to growth, substrates were mechanically polished to get miscut angles of 2° and 4° towards [1010] direction and chemomechanically polished to obtain atomically smooth surface. Epi-ready substrates were then mounted by gallium to single 2 in wafer close to its center to ensure the same growth conditions at both crystals. Growth of 200 nm thick GaN was carried out at 750 °C out under nitrogen rich conditions using nitrogen and gallium fluxes of \( F_N = 16 \) nm/min and \( F_G = 4 \) nm/min (GaN equivalent growth rate) respectively. After the growth samples morphology was examined by atomic force microscope (AFM).

4. Dependence of the surface ordering on the growth parameters

4.1. Miscut

Fig. 1 presents the numerical (a) and experimental (b) results of N-polar GaN layer morphology for the case of 2° miscut surface. For both cases (experimental and calculations) growth parameters like temperature and atomic fluxes were the same. The size of calculated system is 105 nm \( \times \) 105 nm, smaller than the area of AFM scan which is 1 \( \mu m \times 1 \mu m \). Despite this it can be seen that in both cases crystal does not grow smoothly. In Fig. 1a several cavities can be easily seen, steps bend and become wavy leaving deep cavities on their way forward. In the process of further growth more profound structures can build around them. In Fig. 1b we can see that the surface is rough, dominated by islands. This is typical pattern expected for systems with slow adatom diffusion.

The situation changes diametrically when growth is performed under the same conditions (impinging fluxes and temperature), but at the substrate cut 4°. Fig. 2a presents the simulated surface pattern for 4° miscut. Step flow growth mode with double-steps and relatively straight edges are seen. Fig. 2b presents the AFM scan of N-polar GaN layer grown by PAMBE. We can see smooth, stepped surface. The double step structure however is not so clearly visible like in the simulated plot above. The double step structure is a consequence of GaN lattice geometry according to which every second step perpendicular to [1010] direction has different bond arrangements. Thus the rate of adatom adsorption at the step and resulting velocity of every second step is different.

In order to analyze and compare surface parameters more precisely we have calculated root mean square (RMS) roughness

\[
RMS = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (h_i - \mu_h)^2}
\]

where \( h_i \) is the z coordinate (height) of i-th site over leveled surface and \( \mu_h \) is mean system height. RMS parameters for numerical results presented in Figs. 1a and 2a are 0.38 nm and 0.15 nm respectively. The same parameters calculated for corresponding experimental data (Fig. 1b and 2b) are 6.11 nm and 0.49 nm.

Fig. 1. (a) Surface pattern of N-polar GaN(0001) layer simulated by MC on 2° miscut substrate. (b) Morphology of 200 nm N-polar GaN layer grown by PAMBE under the condition used in the KMC simulations on 2° miscut substrate. Fluxes of atoms are \( F_G = 4 \) nm/min and \( F_N = 16 \) nm/min for both experiment and simulations. Temperature \( T = 750 \) °C in both pictures.
Roughness measured for experimental surfaces is obviously higher due to the surface distortion or local defects, however one can notice that RMS parameters for lower miscut are higher both in the experiment and simulations as well. Simulated surfaces are smoother also due to the shorter time of system evolution, which for experiments was about 40 min and 2–3 min for simulations as well as due to different sizes of the surface area.

In order to illustrate and analyze surface features, in particular their periodicity and amplitude we calculated correlation functions along and across steps according to the formula:

$$C(r) = \sum_i [h(t_i) - h_0] [h(t_i - h_0)]$$

where the difference in position $r = (x, y)$ is calculated along $x$ or $y$-axis, i.e. across $(r=x)$ or along $(r=y)$ steps. Fig. 3 presents the correlation functions calculated for experimentally measured surface morphology of N-polar GaN(0001) layers grown by PAMBE on (a) 2° miscut and (b) 4° miscut GaN substrate. We can see that the scale of variability of the 2° curve is much larger than that for 4° surface, i.e. ±30 nm² vs. ±0.2 nm² respectively. Fig. 3a shows that the correlation function calculated for the surface of 2° oscillates in both directions. The length scale of its changes along and across steps is much longer than bilayer step distance ($\approx 10$ nm). We can see that the characteristic correlation length is shorter across than along steps. The correlation function across steps of 4° miscut surface (Fig. 3b) shows periodicity of around 10 nm length denoting bi-step surface structure. The function along step is flat and close to zero describing smooth surface.

Fig. 2. (a) Surface pattern of N-polar GaN(0001) layer simulated by MC on 4° miscut substrate. (b) Morphology of 200 nm N-polar GaN layer grown by PAMBE under the condition used in the KMC simulations on 4° miscut substrate. Fluxes of atoms are $F_G = 4$ nm/min and $F_N = 16$ nm/min for both experiment and simulations. Temperature $T = 750$ °C in both pictures.

Fig. 3. Correlation functions for experimental surfaces for (a) 2° miscut and (b) 4° miscut presented in Figs. 1 and 2 respectively. Solid lines denote correlation calculated perpendicularly to the initial step orientation – along $x$-axis and dashed lines denote correlations parallel to steps – along $y$-axis.
because there are enough of them all around. Hence system grows in 2D island mode and finally forms steps straight and distributed regularly in space. When miscut angle is low, terraces are wider and particles need more time to reach the step. In such case the probability of nucleation is higher. As a result of this process islands would appear at terraces. They are partly adsorbed by steps, leaving some cavities beside them and irregular wavy 3D pattern builds up (Fig. 5a). In this case less mobile nitrogen adatoms seem to play a role in building 3D islands. However for much higher nitrogen flux this reasoning does not work any more. In Fig. 5c we can see the same 2° surface but grown with much higher flux of nitrogen adatoms. In this case their presence has rather an effect of smoothening the surface. We will analyze this case later on.

4.2. Temperature

Higher miscut successfully flattens the surface by shortening the time that adatom needs to reach the step. The same effect should be obtained for 2° miscut by applying a higher growth temperature that increases surface diffusion rate. In Fig. 6 we show the simulated surface morphology for (0001) GaN layer on 2° miscut surface obtained at the temperature 850 °C, that is 100 °C higher than the reference (cf. Fig. 1). In such case diffusion is 4 times faster than in the reference example, which means effectively 2 times larger than the diffusion length. Comparing the surface morphologies in Figs. 1a and 6a we see the positive effect of increased temperature on the surface smoothness. Calculated RMS value for the N-polar (0001) GaN surface simulated at 850 °C is 0.15 nm, that is comparable with RMS for 4° miscut at lower temperature. However the morphologies obtained at raised temperature on 2° miscut (Fig. 6) and at reference temperature on 4° miscut (Fig. 2) are different as can be concluded from the correlation functions, plotted in Figs. 4b and 6b. Regular pattern across steps in Fig. 6b is clearly visible, but with no sign of double step structure because the periodicity of the correlation function is around 10 nm. When we look closer at the surface morphology in Fig. 6 we can see that gallium steps have tendency to bend at characteristic length related to the diffusion length while island-like structures are built by nitrogen layer. All together is different pattern than that seen in Fig. 4b.

4.3. Growth rate

The other method of surface smoothening is to use a lower flux $F$ of incoming particles, i.e. lower growth rate. In such a way diffusing adatom has more time to wander around on the surface and as a result again the steps should become more straight for lower miscuts. In Fig. 7a we see an effect of the slow growth rate ($F$ are 4 times slower than in the reference surface in Fig. 1). Again we can see a smooth surface. It differs from that obtained by increased temperature. We can see bi-step structure, like in the case of 4° miscut and higher flux. Such structure is reflected in the shape of the correlation functions presented in Fig. 7b. Correlation in the direction perpendicular to steps is very similar to this in Fig. 2, but in two times larger distance scale. At the same time correlation along step quickly decreases to zero. However RMS = 0.27 nm and is larger than in the case of surface of crystal grown at higher temperature (Fig. 6).
4.4. N-excess

In Fig. 5c the surface smoothening due to the increased nitrogen flux has been shown. Below we analyze this case in more detail. In Fig. 8a we show the surface of GaN(0001) on 2° miscut obtained under the growth conditions of high N excess, i.e. the flux of incoming nitrogen is 30 times higher than gallium flux. It can be seen that the surface is much smoother than that in Fig. 2, however not in the same manner as 4° miscut case as seen in Fig. 2a. It is easy to note that the increase of nitrogen amount in the system leads to the improvement of surface quality. There are no cavities seen in Fig. 8 and the height differences are not so large as in the reference GaN surface (Fig. 1). However, the overall surface structure is not so regular as in the cases studied before. The calculated value of RMS = 0.28 nm is lower than the reference 2° miscut GaN (0001) layer and higher than that for 4° miscut. When we look at the correlation function along steps presented in Fig. 8b we see that it decreases very quickly, like for smooth structures. The correlation function across the steps shows oscillations of characteristic length for double step structure, i.e. 20 nm but they are not so regular like in previously discussed examples (Figs. 3b, 6 or 7) and evidently decay.

In order to compare these cases let us analyze the evolution of surface RMS parameter as a function of surface coverage, counted in the numbers of grown layers for three cases of GaN growth: high miscut, high temperature, and high N flux. The result is shown in Fig. 9. For the case of layer grown at the 4° miscut and for high temperature 2° miscut growth processes are evidently periodic. One can see oscillations in RMS when the growth proceeds. It can be interpreted as layer by layer growth via creation of islands, then absorption of these islands by approaching steps, and the cycle ends up by next layer to be completed [22,23]. We can
clearly see periodicity of the length of one monolayer in the plotted curves. Typically, the higher is the temperature the smoother is the process, but it has the same period related to the number of layers. Third curve is plotted for high N-excess. In this case RMS value stays the same, relatively high level, showing that the growth process is realized in a different way. We attribute it to the 2D island growth mode that results in a stable RMS value during the growth process.

In general the surface instability appears due to the too slow diffusion of gallium atoms at relatively high flux of incoming atoms and because of the large terraces width as compared to the diffusion length of Ga adatoms [24–27]. However if steps are located more densely, crystal surface becomes smoother under the same growth conditions. In two component system of GaN, at miscut of 4°, diffusion rate of N adatoms is below and of Ga adatoms close to the limiting value for the domain formation on the terrace [25–30]. If the diffusing particle has enough time to find a step edge before the next one attaches to it the island nucleation process is suppressed. When this condition is fulfilled we observe smooth crystal growth realized either by step flow or mixed mode of step flow and two dimensional nucleation process [31]. It is interesting that we observe such growth mode for two component system, when one component is slower and the second moves with rate close to the expected limiting diffusion value. This limiting value can be decreased by higher surface miscut, lower crystal growth rate or higher temperature. Our MC simulations confirmed that all these methods lead to lower surface roughness. Thus we can explain experimentally observed differences in surface morphology of N-polar GaN (0001) grown on substrates with 2° and 4° surface miscuts. However, due to heterogeneous growth and immobility of nitrogen adatoms the character of stable and unstable surface evolution is a mixture of step flow and 2-D or 3-D island growth [25–28].

5. Conclusions

Kinetic Monte Carlo simulations have been employed to study the surface evolution of N-polar GaN(0001) layers grown on the substrates of 2° and 4° miscuts. The numerical results are compared with experimentally observed surface morphologies of GaN layers grown by MBE. Due to the high diffusion barriers during the growth of (0001) GaN under N-rich conditions the resulting surface is irregular and rough. Such surface can be observed for the crystal grown at 2° surface miscut at temperature around 750 °C. Four different methods of smoothening the surface were investigated using kinetic Monte Carlo method: (a) growth of GaN(0001) on high miscut substrates, (b) at higher temperature, (c) at low growth rate and (d) using high N-excess. All of them lead to smoother crystal surfaces. The surface obtained on 4° miscut substrate seems to be the most regular one, that has been also confirmed by experimental results. Other methods like: higher temperature or lower growth rate also work as smoothening factor, but the character of the surface morphology is different than that for the high miscut surface. Other approaches provoke steps meandering and formation of cavities due to nucleation at terraces. The surface that emerged during the growth at high N-excess using very high nitrogen flux is quite unusual, because microscopically it seems to be irregular, but correlation functions exhibit rather regular structure. Thus, we conclude that under high N-excess GaN(0001) growth happens regularly and the surface morphology remains smooth due to stable 2D island growth mode. Therefore the use of high nitrogen flux for the growth of
GaN(0001) under N-rich conditions and relatively high miscuts seems to be a good method to obtain smooth surface morphology.

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