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Study on Surface Polarity of GaN by Density Functional Theory and Molecular Dynamics

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1. Introduction

GaN is one of the most attractive materials for optoelectronic devices because of its application in blue light-emitting diodes and lasers [1,2]. The heteroepitaxial growth of highly perfect GaN layers on sapphire substrates by metalorganic vapor phase epitaxy (MOVPE) method has become possible due to the development of the GaN or AlN buffer layer deposition technique [3]. It is reported that the initial treatment of sapphire substrate, such as initial nitridation and low-temperature GaN buffer layer deposition, affects the surface morphology and crystallinity of GaN grown layers [4]. GaN grown planes have a polar configuration, i.e., either Ga or N can occupy the first-atomic layer. The polarity of these surfaces can have important effects in semiconductor interfaces, i.e., InGaN/GaN interfaces. However, as for more relevant (0001) growth surface of hexagonal GaN, its structure is not well known yet. It is important to understand the surface structure of GaN, since this knowledge will achieve high quality heteroepitaxial growth of materials as required for optoelectronic applications.

In this paper, we have studied the stability and structure relaxation of the Ga-terminated (0001) and the N-terminated ($000\bar{1}$) GaN surface by periodic density functional (DFT) method and the effects of surface polarity of GaN on InN/GaN interface (as simple model of InGaN/GaN interface) by molecular dynamics (MD) method.

2. Method and Model

All DFT calculations were performed by solving Kohn-Sham equation self-consistently [5] as implemented in the DSolid program [6] provided by MSI. We employed local density approximation (LDA) with Vosko-Wilk-Nusair (VWN) functional [7] for geometry optimization. In order to calculate surface relaxation energies, we applied the Beck-Lee-Yang-Parr (BLYP) [8,9] nonlocal functional as a correction. Double numerical plus polarization functions (DNP) basis set was used. The GaN surface structure models were constructed by 6 atomic layers (total 24 atoms). The 2 bottom layers were fixed to approximate bulk structure and other layers were fully optimized.

MD calculations were carried out with the RYUDO program developed by Miura for the analysis of InN/GaN heterointerface structure. The two-body central force interatomic potential, as shown in Equation 1, was used. In this equation, the first, second, and third terms refer to Coulomb, exchange repulsion, and Morse interactions, respectively.

$$u(r_{ij}) = Z_i Z_j e^2/r_{ij} + f_0 (b_i + b_j) \exp[(a_i + a_j - r_{ij})/(b_i + b_j)] + D_{ij} \{\exp[-2\beta_{ij} (r_{ij} - r^*_{ij})] - 2\exp[-\beta_{ij} (r_{ij} - r^*_{ij})]\} \quad (1)$$

where Z_i is the atomic charge, e is the elementary electric

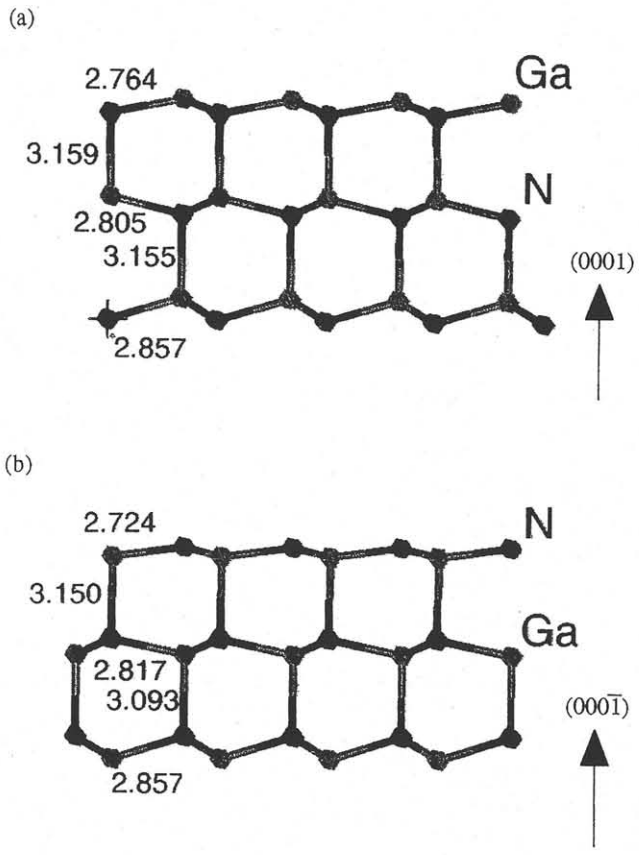
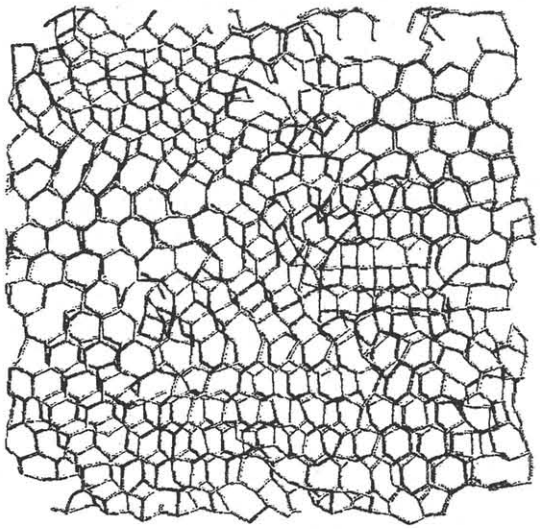


Fig. 1 Optimized structure of GaN surface
(a) (0001) Ga face (b) ($000\bar{1}$) N face

charge, r_{ij} is the interatomic distance, and f_0 is a constant. The parameters a and b represent the size and stiffness, while D_{ij}, r^*_{ij}, and β_{ij} represent bond energy, equilibrium bond distance, and stiffness.

The calculations were performed for 20,000 steps with a time step of 2.5 fs at 900 K. InN thin films were constructed on both GaN surface models and their crystal structures were relaxed. The relaxed structures were analyzed on both GaN surfaces.

(a)



(b)

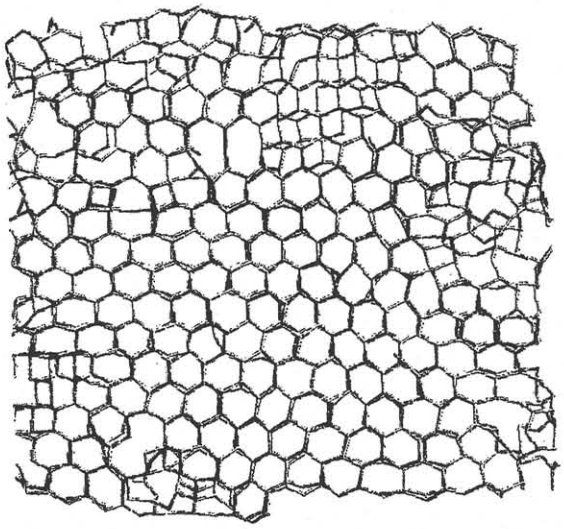


Fig. 2 The relaxation structure of InN thin films (a) on (0001) Ga face (b) on (000$\bar{1}$) N face

3. Results and Discussion

Optimized structure of GaN surface is shown in Figure 1.

Figure 1(a) shows (0001) GaN surface in which the first-atomic layer is Ga. In this figure, the first Ga layer has moved downward and the second N layer has moved upward. This behavior is called "rumpling", Ga and N layers approach each other to stabilize the structure in order to equalize surface charge distribution. Figure 1(b) shows (000$\bar{1}$) N face. A similar rumpling structure is observed on this relaxation structure also. But the degree of rumpling on N face is greater than that on Ga face.

We have also calculated the distribution of HOMO (Highest Occupied Molecular Orbital) on each surface structure. HOMO of Ga face are localized on each atom. On the other hand, HOMO of N face are spread out around the surface of N or on the second Ga atoms. These results suggest that Ga face ionicity is greater than N face.

Furthermore, we have calculated the stabilization energy by surface relaxation. The energy value clearly shows that the stabilization energy of N face is about 12.1 kJ per unit cell smaller than that of Ga face. This result suggests that N face of GaN (000$\bar{1}$) surface is more favorable energetically. Sun et al. studied the thermal stability of GaN (0001) polar planes under the hydrogen atmosphere and reported that the N-terminated (000$\bar{1}$) surface is more stable than the Ga-terminated (0001) surface [10].

Figure 2 shows the relaxation structure of InN thin films on each GaN surface obtained from MD calculations. The InN structure on (0001) Ga face destroys its crystallinity largely, but that on (000$\bar{1}$) N face preserves its crystallinity laterally. This is the expected result as the bonding strength of Ga-N is higher than that of In-N. Thus, it is suggested that InN thin films can relax its structure retaining the crystallinity on the (000$\bar{1}$) N face than on (0001) Ga face, provided each GaN surface grown has equal fine crystal surface.

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