

Theoretical approach to oxygen incorporation mechanism in vicinal m-GaN MOVPE

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Abstract

Based on the density functional theory (DFT), we clarify the oxygen incorporation mechanism in vicinal m-GaN Metal-Organic Vapor-Phase Epitaxy (MOVPE). We first identify the surface reconstruction on the vicinal m-GaN surfaces, and then explore various nitrogen sites near step edges for the oxygen substitution. We find that oxygen prefers a nitrogen sites near the step edge on +c 5°off m-GaN substrate compared with that on -c 5°off m-GaN substrate. The tendency agrees well with the recent experimental finding that the oxygen concentration in -c 5°off m-GaN epilayers is one order lower than that in +c 5°off m-GaN epilayers. We newly propose an impurity incorporation model during GaN step-flow growth based on our DFT calculations.

1. Introduction

Gallium nitride (GaN) is one of the most promising materials for the next-generation high-power electronic devices because of its excellent high-voltage and high-current capacity combined with high thermal conductivity. In the power devices, the high breakdown voltage requires the reduction of contamination of epitaxial GaN films and then the control of the lower carrier concentration [1]. An oxygen impurity unintentionally doped in GaN substitutes for the nitrogen site and behaves as a donor, making GaN n-type [2]. Recently, Tanaka et al. [3] reported that oxygen concentration in the epilayers grown on -c 5°off m-GaN vicinal substrates was one order lower than that on +c 5°off m-GaN vicinal substrate. This reduction of the oxygen concentration is crucial in technology and the clarification of its mechanism is important in both science and technology. In this work, we clarify the oxygen incorporation mechanism on the vicinal m-GaN described above.

2. Calculation methods

First, reconstructed structures on vicinal m-GaN surfaces were investigated by ab initio based approach. [4] The surface reconstruction on the m-GaN terrace and modified structures of step edges on +c 5°off m-GaN and -c 5°off m-GaN surfaces were analyzed. Next, the formation energy of the oxygen substituting nitrogen, O_N , near the step edges

was studied. The calculations were performed using the real space density functional theory (RSDFT) as implemented in the RSDFT package. [5, 6] Exchange and correlation energy was treated by the Perdew-Burke-Ernzerhof (PBE) [7] exchange correlation functional using the norm-conserving pseudo-potentials. [8] The Ga3d electrons are treated as core electrons. The mesh spacing is fine enough corresponding to the cutoff energy of 73 Ry in the conventional plane-wave basis. For surface calculation, we used a periodic slab composed of 532 atoms (+ adatoms) (Fig. 1). Ga and N dangling bonds in the back surface of the slab model were terminated by pseudo-hydrogens of charge 1.25e and 0.75e, respectively, to mimic the semi-infinite GaN substrate. The epilayers and adatoms were allowed to relax until the force acting on each atom was less than 5×10^{-4} Hartree/a.u. (atomic units), while the bottom layers and pseudo-hydrogens were kept fixed to mimic bulk-like behavior. Using the slab models, formation energy of O_N near the step edges were calculated.

3. Results and discussions

First, initial structures on +c 5°off m-GaN and -c 5°off m-GaN vicinal substrates were investigated. Fig. 1(b) show the surface phase diagram of m-GaN MOVPE under H_2 carrier gas condition. One can see ideal surface is stable under the typical growth temperature ($\sim 1050^\circ\text{C}$), whereas 4Ga-H and 4N-H step edges are stable on +c 5°off m-GaN and -c 5°off m-GaN, respectively [Figs. 1(a) and (c)]. Consequently, vicinal surface structures on +c 5°off m-GaN and -c 5°off m-GaN were predicted as seen in Fig 1(d) and (e), respectively.

Next, O_N formation energies near the step edges were studied. Fig. 2(a) and (b) show oxygen substituting nitrogen at upper and lower site of the step edge on +c 5°off m-GaN, respectively. Here, O_N formation energy of latter case is 0.73eV lower than that of m-plane terrace. In case of -c 5°off m-GaN, on the other hand, O_N formation energy near the step edge is almost the same with that of m-plane terrace. This theoretical finding has unequivocally clarified that the O incorporation during the step-flow growth is enhanced for the +c 5°off m-GaN vicinal substrate compared with the -5°off m-GaN. The tendency agrees well with experimental results. [2]

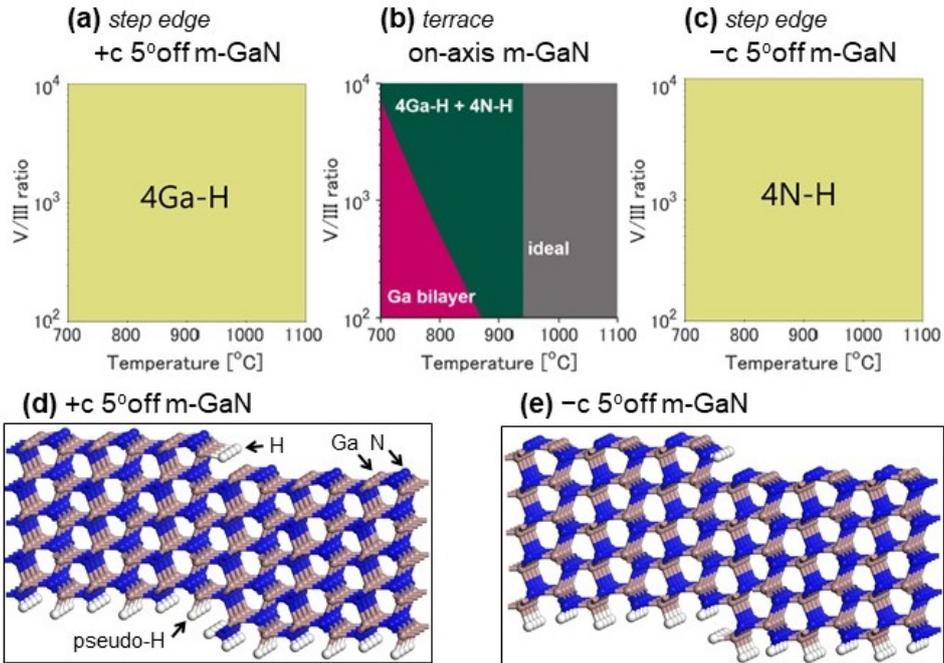


Fig. 1 (a), (b), (c) surface phase diagram of +c 5° off, on-axis and -c 5° off m-GaN, respectively. (c), (d) schematic of +c 5° off and -c 5° off m-GaN vicinal substrates, respectively. Brown, blue, and white circles are Ga, N and H, respectively.

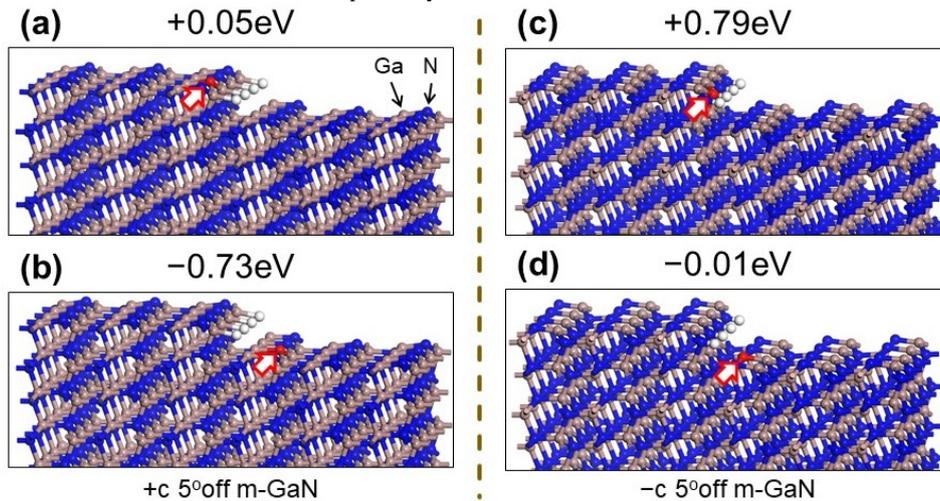


Fig. 2 Oxygen substituting N sites (see red arrows) on (a), (d) +c 5° off and (c), (d) -c 5° off m-GaN vicinal substrates. The energies written in the figures are difference in O_N formation energy between that near the step edge and that on the terrace.

4. Conclusions

Oxygen incorporation mechanism for the vicinal m-GaN MOVPE was investigated based on DFT calculations. First, reconstructed structures on +c 5° off m-GaN and -c 5° off m-GaN were predicted. Next, O_N formation energy near the step edges were investigated. The results suggest that oxygen prefers lower nitrogen site at the step edges on +c 5° off m-GaN. Hence, oxygen incorporation efficiency for the +c 5° off m-GaN growth becomes higher than that of -c 5° off m-GaN growth. The atom-scale model for unintentional doping during GaN step-flow growth was proposed.

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