Ising model-based analysis of the GaN(0001) surface reconstructed structures sampled from Bayesian optimization

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Identifying the realistic surface reconstruction during vapor phase epitaxy is essential for correct understanding of growth mechanism and for quantitative modeling. In GaN(0001) MOVPE, according to ab initio calculations and statistical thermodynamics, the Ga-adsorbed surface ((2×2) Ga_{ad}) or H-adsorbed surface ((2×2) 3Ga-H) are thermodynamically most stable under a typical range of conditions [1]. Since the free energies of these most stable and metastable structures are close to each other, it is considered that the mixed states of these structures actually emerge. However, the number of candidate structures for the mixed states is quite large, so they are studied by a limited number of samples from Bayesian optimization [2]. In this study, two types of analyses were performed on the sampled data.

First, we examined whether electron counting (EC) rule was locally and completely satisfied. This analysis was performed by pattern matching operations of the (2×2) motifs. As a result, we found that some stable structures satisfy the local EC rule in more than one way. We refer to this number as interpretation number. The correlation between mixing enthalpy and the number of interpretations shows that the structures with multiple interpretations are stable, but not all stable structures have multiple interpretations. That is, it is indicated that even soft satisfaction should be considered, rather than the complete (strict) satisfaction of the local EC rule.

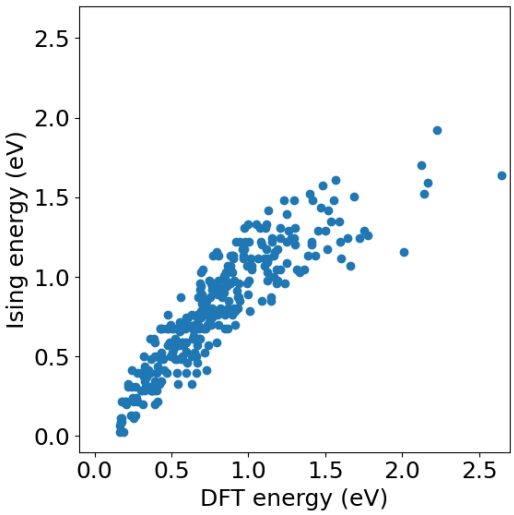

Fig. 1 Performance of data-driven Ising model.

Second, the analysis using an Ising model was performed to take into account the soft satisfaction of the local EC rule. In the Ising model, the local EC rule is expressed by the attraction and repulsion between the Ga-adsorbed, H-adsorbed, and non-adsorbed surface sites. That model with parameters was trained using the data sampled from Bayesian optimization. Figure 1 shows the relationship between the mixing enthalpies calculated by the Ising model and those calculated by DFT calculations. In particular, we found that the Ising model works quantitatively well in the region of low mixing enthalpies.

By utilizing this quantitative Ising model, it is possible to exhaustively calculate a huge number of all candidate structures, which was not possible with DFT calculations. In other words, we have discovered the possibility of revealing a realistic surface structure by efficient data collection using Bayesian optimization and a data-driven Ising model.

References

[1] Kusaba A et al. Thermodynamic analysis of (0001) and (000-1) GaN metalorganic vapor phase epitaxy. Jpn J Appl Phys. 2017;56(7):070304.
[2] Kusaba A et al. Exploration of a large-scale reconstructed structure on GaN(0001) surface by Bayesian optimization. Appl Phys Lett. 2022;120(2):021602.