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Computics approach toward clarification of microscopic mechanisms of GaN Metalorganic Vapor Phase Epitaxy*

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Computational approach based on the quantum theory is recognized as a poweful tool to claryfy underlyng physics and chemistry in nature. Performance of supercomputers increases exponentially and now it reaches 187 PFlops (Summit by IBM) as its theoretical peak performance [1]. This development of the computers seems to reinforce the computational approach. However, it is not necessarily true. The performance of a single compute node is already saturated, corresponding to the collapse of the scaling law. Hence the only way to make a high-performance computer is to gather a huge number of compute nodes and connect them with high-speed network (massively parallel architecture). Without the cross-disciplinary collaboration among the fields of materials science, computer science and applied mathematics, it would be impossible to exploit the tremendous performance of the supercomputers. In the collaboration, new algorisms and mathematical schemes suitable for current and future architecture of computers should be developed. We call this new stage of the collaboration *computics* [2]. In this talk, after brief explanation of *computics*, I will present our efforts to clarify microscopic mechanisms of MOVPE of GaN.

I will discuss three issues: (i) surface–catalyzed NH_3 decomposition and N incorporation on GaN(0001) [3], (ii) structural characteristics and energetics of surface steps on GaN(0001) [4], and (iii) dynamical behavior of Ga adatoms on GaN(0001) at growth temperature [5].

In the first issue, we explore reaction pathways for NH_3 decomposition and subsequent N incorporation on GaN(0001) on the basis of the first-principles density-functional calculations. We find that the Ga-Ga weak bond on the Ga-rich surface is attacked by an ammonia, and then the ammonia is easily decomposed with the resulting NH unit being incorporated into the surface Ga-Ga bond. The obtained energy barrier is relatively small, 0.6 eV. In the second issue, considering that the surface steps are stages of the step-flow epitaxial growth, we try to identify atomic structures and energetics of single-bilayer atomic steps. Multi-layer atomic steps are found to be energetically unfavorable. For GaN(0001) vicinal surfaces inclined toward either $[1\bar{1}00]$ or $[11\bar{2}0]$, we consider all the possible atomic steps (5 types in total) and determine their atomic structures and formation energies. The obtained step stuctures show vartiability depending on the growth condition. One general trend is that Ga atoms are likely to stick to the step edges, providing Ga-Ga weak bonds, possible hot spots for the N incorporation. In the third issue, I report first-principles Car-Parrinello molecular dyanmics (CPMD) simulations for the dynamical aspect of Ga-rich GaN(0001). We find that, at the growth temperature (1300K), Ga adatoms suddenly move from their stable positions. Analyses using radial distribution functions and diffusion constants unequivocally clarify that a two-dimensional liquid phase appears on GaN (0001). Its implication for the epitaxial growth will be discussed.

References:

[1] TOP 500 supercomputer sites: http://www.top500.org/
[2] Materials Design through Computics: http://computics-material.jp/
[3] K. M. Bui *et al.*, J. Phys Chem. C 122, 24665 (2018).
[4] K. M. Bui *et al.*, submitted to ICMOVPE XX
[5] K. M. Bui *et al.*, Jpn. J. Appl. Phys. 59, SGGK04 (2020), and unpublished results.

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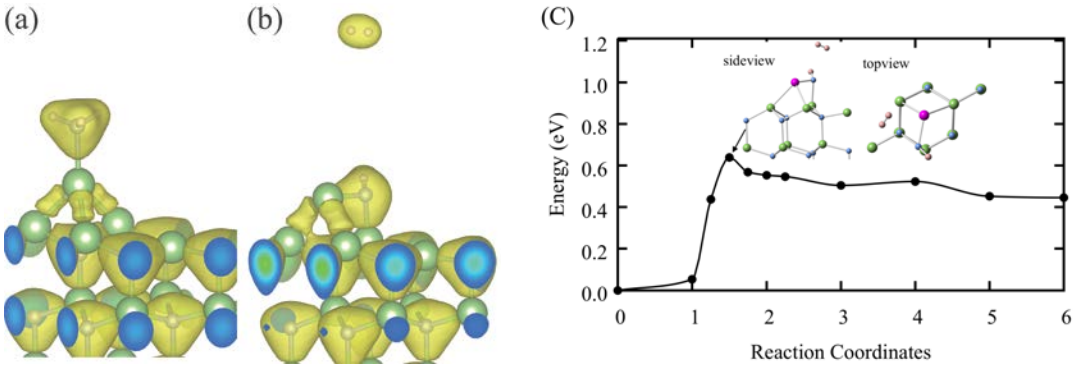

Figure 1: ***Ammonia decomposition and the subsequent NH incorporation on Ga-rich GaN(0001) surface.*** *The initial (a) and the final (b) atomic structures of the decomposition reaction of NH_3 and the corresponding energy profile (c). Large and small balls depict Ga and N atoms, respectively. The electron density in each structure is shown as yellow iso-value surface. Adsorbed NH_3, surrounded by upper yellow surface in (a), is decomposed and –Ga-N(H)-Ga unit is formed in the subsurface region and an H_2 is desorbed (b). The calculated energy barrier for this reaction is 0.6 eV(c).*

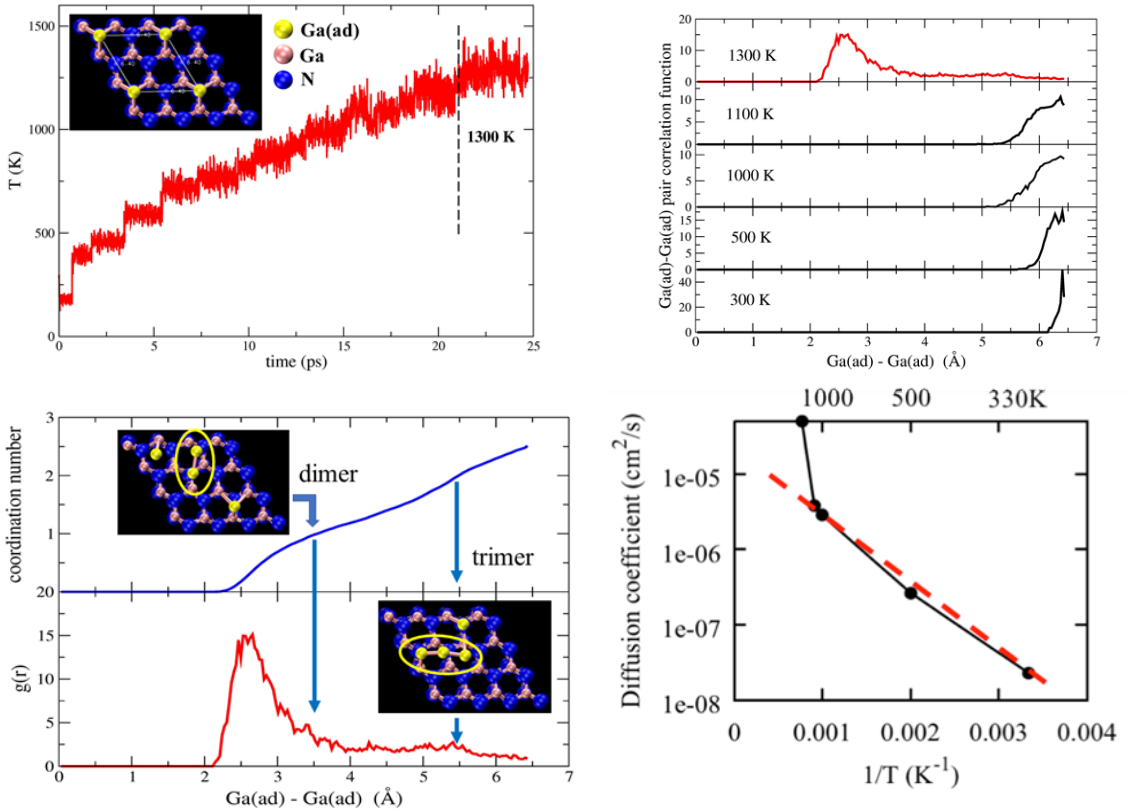

Figure 2: ***Two dimensional liquid phase on Ga-rich GaN (0001) surface unveiled by Car-Parrinello molecular dynamics simulations. Top-left:*** *Temperature increase during the heating phase. The inset shows the surface of the GaN system simulated. Ga and N atoms of the substrate are colored in pink and blue, respectively, whereas the Ga adatom [Ga(ad)] exposed at the surface are evidenced in yellow.* ***Top-right:*** *Evolution of the Ga(ad)-Ga(ad) pair correlation function during the heating process. The initial distance of 6.4 Å corresponds to the relaxed system from which dynamical simulations were started.* ***Bottom-left:*** *Pair correlation function (PCF) for the Ga(ad)-Ga(ad) distribution at 1300 K (lower panel) and corresponding coordination number as obtained upon integration of this PCF. The insets show the structures corresponding to a dimer or a trimer of Ga atoms diffusing on the GaN (0001) surface.* ***Bottom-right:*** *Diffusion coefficient of a Ga adatom as a function of the inverse temperature 1/T for the temperature range (300-1300 K). The dashed red line shows the Arrhenius plot obtained by excluding the point at 1300 K, where the system deviates from the standard trend, showing the emergence of the liquid phase. .*