DFT and Machine Learning Molecular Dynamics Study of Controlled Graphene Self-Assembly on the Diamond (111) Surface

Graduate School of Engineering, Osaka Univ. [1], Advanced Materials Lab., Sumitomo Electric Industries [2], R[3] Institute for Newly-Emerging Science Design, Osaka Univ.[3], Research Center for Precision Engineering, Graduate School of Engineering, Osaka Univ.[4]

◦John Isaac G. Enriquez[1], Takahiro Yamasaki[2], Masato Michiuchi[2], Harry Handoko Halim[1], Kouji Inagaki[1], Masaaki Geshi[3], Ikutaro Hamada[1], Yoshitada Morikawa[1, 4]

E-mail: morikawa@prec.eng.osaka-u.ac.jp

Carbon-based technologies are poised to surpass the limitations of silicon devices, offering superior performance, enhanced biocompatibility, and greater sustainability. Carbon exists in various allotropes, exhibiting distinct electronic, optical, and quantum properties. Combining these allotropes into carbon heterostructures promises to unlock new device innovations with exceptional technological potential. A key step in realizing these advancements is understanding the atomic-level mechanisms of carbon phase transformation on the surface of diamonds. We used density functional theory (DFT) and machine learning molecular dynamics simulations to investigate the thermal degradation of the diamond C(111) and C(100) surfaces. The interatomic potential is based on the graph neural network model, trained using energies and forces obtained through DFT calculations. The C(111) surface degrades by forming graphene, while the C(100) surface forms amorphous carbon. The degradation is promoted at the step edges due to the dangling bonds that enhance sp^3-sp^2 transformation [1]. Terminating the dangling bonds of the C(111) surface with hydrogen atoms restores the surface atoms' bulk-like geometry and electronic properties, strengthening the inter-bilayer bonds and increasing the graphitization resistance (Fig. 1a). Partial hydrogenation followed by controlled heating enables the selective formation of graphene on non-hydrogenated regions, resulting in graphene-on-diamond structures. The edges of the self-assembled graphene remain covalently bonded to the diamond substrate, inhibiting graphitization of the sublayers and making it feasible to limit graphene formation to a single layer (Fig. 1b). This approach presents a novel method for engineering sp^3-sp^2 rehybridization, offering a new pathway for fabricating transistors, circuit elements, data storage, and other devices entirely from carbon materials.

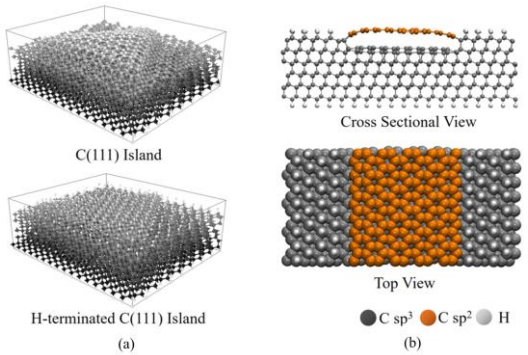

Fig. 1. (a) Graphitization suppression by dangling bond termination and (b) epitaxial self-assembly of graphene on the C(111) surface

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