Ab-initio study on stabilization and intrinsic properties of bilayer graphene forming sp^3 bonding

T. Pakornchote[a,b], A. Ektarawong[a,c], B. Alling[c,d], U. Pinsook[a,b], S. Tancharakorn[e], W. Busayaporn[e], T. Bovornratanaraks[a,b]

[a] Extreme Conditions Physics Research Laboratory, Physics of Energy Materials Research Unit, Department of Physics, Faculty of Science, Chulalongkorn University, Bangkok 10330, Thailand
[b] Thailand Center of Excellence in Physics, Commission on Higher Education, 328 Si Ayutthaya Road, Bangkok 10400, Thailand
[c] Theoretical Physics Division, Department of Physics, Chemistry and Biology (IFM), Linköping University, SE-581 83 Linköping, Sweden
[d] Max-Planck-Institut für Eisenforschung GmbH, Max-Planck Strasse 1, 40237 Düsseldorf, Germany
[e] Synchrotron Research Light Institute (Public Organization), Nakhon Ratchasima 30000, Thailand

Bilayer graphene (BLG) indented by a tip of either the scanning tunneling microscope or the atomic force microscope [1,2] has recently been experimentally demonstrated to exhibit the superhard property. The origin of superhard property has been attributed to the diamondization of BLG, leading to the sp^3 hybridization of carbon atoms between the two graphene layers being pushed towards each other during the indentation process [1,2]. In the present work, we employ the *ab-initio* method to investigate the phase stabilities of diamondene and lonsdaleitene, referred to as AB- and AA-stacking diamondized BLGs, respectively. Our results demonstrate that diamondene and lonsdaleitene are not metastable. However, introducing H atoms to diamondene and lonsdaleitene in order to fully passivate all carbon atoms on their surfaces can result in the thermodynamics stabilization as well as the metallic-to-semiconducting transition of the materials. Our simulations further reveal that both hydrogenated diamondized BLGs are dynamically stable up to at least 1000 K, while the mechanical stability of the materials is confirmed by their elastic constants, satisfying the Born's stability criteria. Details of vibrational analysis of the two hydrogenated diamondized BLGs indicating their possible Raman and IR active/inactive modes, as a fingerprint for future characterization of the materials, will be presented and discussed.

References

[1] Y. Gao, T. Cao, F. C. ad C. Berger,W. A. de Heer, E. Tosatti, et al., Ultrahard carbon film from epitaxial two-layer graphene, Nat. Nanotech. 13 (2018) 133–138.
[2] A. P. M. Barboza, M. H. D. Guimaraes, D. V. P. Massote, L. C. Campos, N. M. B. Neto, L. G. Cancado, R. G. Lacerda, et al., Room-temperature compression-induced diamondization of few-layer graphene, Advance Mat. 23 (2011) 3014–3017.