Theoretical study on hydrogen atom behavior under graphene buffer layer grown on SiC substrate

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Graphene is known to have high electron mobility due to its characteristic electronic properties such as Dirac cone and then expected as a future electronic device material to contribute to the low energy consumption, which could help combat the global warming. Among graphene fabrication methods, thermal decomposition of SiC substrate, in which graphene sheets are formed after Si atom sublimation from SiC surface at high temperature, has an advantage in the sense that the graphene transfer process which would degrade the graphene sheets is not necessary for the device construction, and then has been intensively studied. The first C-atom layer grown on SiC substrate is not graphene but so-called buffer layer (BL), which has similar honeycomb structure to graphene but does not have the graphene's characteristic electronic properties due to the lack of the Dirac cone, because BL is covalently bonded to the SiC surface. To utilize BL grown on SiC substrate as graphene, it is necessary to anneal it under hydrogen (H) ambient to intercalate H atoms between BL and SiC substrate to break the covalent bond between BL and SiC surface. However, the lack of knowledge on the intercalation mechanism make it difficult to control this process to obtain high quality graphene. However there is no studies on the H atoms on the buffer layer with the experimentally observed periodicity of $(6\sqrt{3}x6\sqrt{3})R30^{\circ}$, while there are some studies on the much smaller one with the periodicity of $(\sqrt{3}x\sqrt{3})R30^{\circ}$, in which graphene is largely expanded by about 8%, and then the energetic of H adsorption is not clear on the buffer layer with the periodicity of $(6\sqrt{3}x6\sqrt{3})R30^{\circ}$. So far we have studied the dissociative adsorption of H molecules onto BL and the diffusion and the penetration processes of H atoms and obtained the result that H molecules can dissociatively adsorb onto BL, but before the penetration below BL, they would desorb from the surface due to the much smaller activation barrier of the desorption process than that of the penetration process [1]

In this paper, we report the result of the further study on the interaction between H atoms and BL with the periodicity of $(6\sqrt{3}x6\sqrt{3})R30^{\circ}$ by first-principles density functional calculation [2], especially on the H atoms or molecules between BL and SiC surface. It is found that H molecules put between BL and SiC surfaces can easily dissociate into two H atoms, which make covalent bonds to Si dangling bonds. Surprisingly, there is no energy barrier for the adsorption of H molecule onto Si dangling bonds on a SiC(0001) surface. This is quite contrastive to the adsorption of H molecule onto Si(001) surface, which has an activation energy barrier. To investigate the diffusion of H atoms between BL and SiC surface, we performed first-principles molecular dynamics simulation in NVT condition (T=1500K). Even in a short simulation time of three picoseconds, we observed several H atom hoppings from one Si dangling bond to another, which means that H atom can easily diffuse between BL and SiC surface.

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References:
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