Atomistic Processes of Grain Boundary Motion and Annihilation in Graphene

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To realize graphene electronics, it is essential to fabricate high-quality graphene on a large scale. Much effort has been devoted to the development of methods for the mass production of graphene. It has been shown that CVD growth of graphene on Cu [1] foil enables the mass production of high-quality single-layer graphene through the roll-to-roll method [2]. Generally, graphene films fabricated by CVD growth on metal surfaces are polycrystalline, composed of fine grains and grain boundaries (GBs). Although some theoretical studies have suggested that GBs can cause abnormal strength in graphene and open a transport gap, which is desirable for graphene-based nanoelectronic devices, many experimental studies have shown that GBs act as defects in graphene, impeding electronic transport, lowering thermal conductivity, and weakening mechanical properties. To realize the potential of graphene in 'carbon-based' electronics, it is highly desirable to achieve better control over the nucleation of individual graphene grains and to avoid GBs in fabricated graphene devices. Recently, we investigated the motion of GB at high temperature by performing tight binding molecular dynamics (TBMD) simulations and also carried out *ab initio* total energy calculations to verify the TBMD results of the formation energies and energy barriers for important configurations and their reconstructions. The TBMD simulations are performed using a modified environment-dependent tight-binding (EDTB) carbon potential [3], which was modified from the original EDTB carbon potential [4]. This modified EDTB carbon potential has been successfully applied to investigations of various defect structures in graphene and carbon nanotubes [5-7] In this study, we found that meandering structures of GB is energetically more favorable than other structures, in excellent agreement with experiments [8,9]. In TBMD simulation, sequential evaporation of dimers and Stone-Wales (SW) transformations cause the motion of GB toward armchair grain, resulting in the decrease of distance between two GBs. In spite of the evaporation of dimers, the structure of GB keeps the distinctive meandering structure through sequential SW transformations. In the TBMD simulation for the process of dimer evaporation, the erection and evaporation of dimers are observed after the formation of adatoms due to the bond breaking. In TBMD simulation, two approaching GBs also merge and annihilate through sequential evaporation of carbon dimers and SW transformations. It is interesting to note that various GB structures observed in experiments are also found in our TBMD simulation. These results elucidate the atomic scale mechanism for annihilation of GB which would occur in a high temperature annealing experiment. It is also expected to be found in experiments such as TEM studies. These results will shed interesting light on the fabrication of high-quality graphene nanoelectronic devices through the control of GB dynamics.

References

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Figures

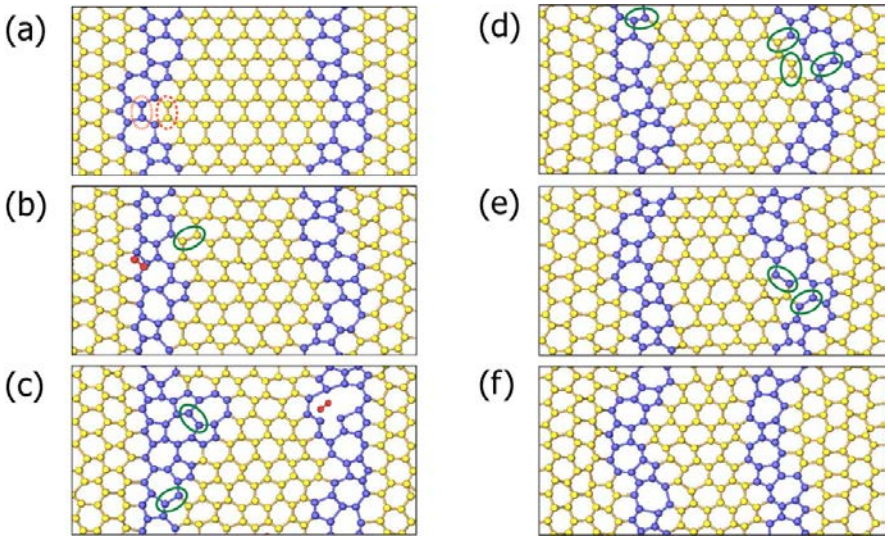

Figure 1: Snapshots from TBMD simulations for graphene GBs. The distance between two GBs become smaller as the simulation goes on. The yellow and blue colors indicate carbon atoms and bonds on hexagonal rings and non-hexagonal rings, respectively. The red colors indicate evaporated dimers. Red solid circles indicate dimers which will undergo SW transformation at next step.

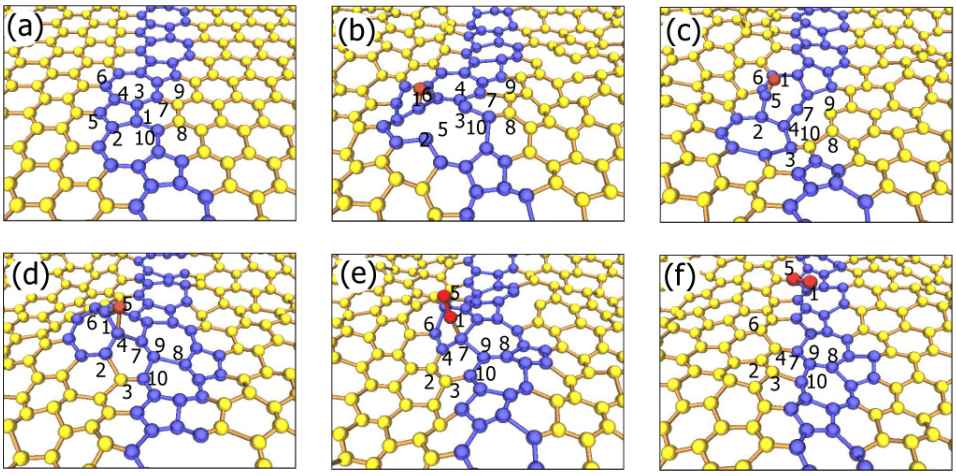

Figure2: Snapshots from TBMD simulations for dimer evaporation. Numbers show the trajectory of identical atoms.