Molecular dynamics simulation of latent track formation in bilayer graphene

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\textbf{Abstract:} The damage production induced by swift heavy ion irradiation in bilayer graphene (BLG) is investigated by molecular dynamics method. By given energy to a cylindrical region, the carbon chains even nanoholes can be produced, which depends on the electronic energy loss (dE/dx). For BLG, the minimum value needed to generate defects lies in 5-7 keV/nm. A low density core and a high density shell structure can be seen while the radii of tracks are obtained. With increasing the values of dE/dx, the track radius is first increasing and then saturates. The analysis of defects indicates that only a small part of the defects can be recombined and the radiation damage in BLG is less severe than in single layer graphene.

\textbf{Keywords:} bilayer graphene, swift heavy ion, molecular dynamics

\textbf{Classification:} Electronic materials

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1 Introduction

Graphene has recently attracted great attention for a variety of novel applications since its experimental isolation in 2004 [1]. However, the single layer graphene (SLG) suffers from a major drawback of zero energy gaps, which hinders its application as electronic material [2]. The unique ability of controlling the band gap in bilayer graphene (BLG) may lead to new opportunities of electronic devices [3]. Ion irradiation has been shown to be promising in doping graphene with foreign atoms and manipulating its structure [4]. On the other hand, graphene is naturally very sensitive to defects induced by ion irradiation. So, the studies on defect control and behavior of graphene in defect environment play significant role in the proper use of graphene-based devices.

Until now, most of the studies are focused on swift heavy ion (SHI) irradiation effects on SLG [5-7], and none report available for BLG. The study of SHI irradiation is important due to the potential use of BLG devices in space application, especially field effect transistors [8]. Computer simulations can provide an atomistic explanation for the damage production in BLG under SHI irradiation. To open a direct view to the track formation through electron-phonon coupling and defects production in the track, the molecular dynamics (MD) method has been extensively used [9-11]. The goal of this paper is to give insight into SHI irradiation effects on BLG and evaluate the performance of BLG irradiated under SHI.

2 Simulation method

The formation of latent ion tracks in BLG is modeled computationally at the atomic level, using LAMMPS and the structures are visualized with Atomeye. The atomic interactions are calculated using the adaptive intermolecular reactive empirical bond-order (AIREBO) potential function. The BLG used in the simulation is 20.5 nm × 20.2 nm × 0.67 nm and contains 31488 atoms. Periodic boundary conditions are used in the three dimensions. The BLG is equilibrated in the isothermal isobaric ensemble at 300 K for 10 ps. A latent ion track is subsequently created along the z direction by depositing the electron energy to a cylindrical region with a radius of 3 nm, and every atom in this region obtains the same kinetic energy in a random direction. The last 0.5 nm at the borders of the computation cell in the x and y directions are controlled at 300 K by Berendsen temperature control. A variable time step from 0.00001 ps to 0.001 ps is used in the calculation. The simulation is terminated until the cell temperature has dropped to below 500 K, and no further changes could be seen. The defects in the irradiated material are identified using the Voronoi tessellation method.
3 Results and discussion
Swift heavy ions lose energy predominantly through inelastic interactions with target electrons, called electronic energy loss (dE/dx). Through electron-phonon coupling, the excited electrons transfer their energy to atoms, which lead to a rapid local heating of the lattice close to ion path, resulting in local melting and vaporization. During melt quenching, recrystallization and defects recovery occur over nanoscale dimensions. Fig. 1 illustrates the atomistic images of latent tracks at the end of simulations for different values of dE/dx in Atomeye while the ripples of BLG can be clearly seen. In the range of dE/dx used in this work, the track radius generally increases with the increase of dE/dx. For dE/dx = 5 keV/nm (shown in Fig. 1(a)), no obvious amorphous region can be seen. When dE/dx increases to 7 keV/nm, a small amorphous region is observed, which means the threshold electronic energy loss ((dE/dx)\text{th}) below which no damage could be found in SLG lies in 5-7 keV/nm. For SLG, the (dE/dx)\text{th} is determined to be 8 keV/nm [5].

![Fig. 1. Atomistic images of ion tracks in BLG at the end of simulations for different values of dE/dx. The tracks orient perpendicular to the plane of the figure. The plane size is 20.5 nm $\times$ 20.2 nm.](image)

For dE/dx = 19 keV/nm, a relatively small central pore and surrounding mesh-like structure with lots of carbon chains can be seen (shown in Fig. 1(f)). This defects structure is created due to the loss of carbon atoms. As shown in Fig. 1(b)-(e), more carbon chains are formed with increasing dE/dx. This is because more carbon atoms can be displaced far away from the BLG plane, which reduces the probability of atomic reconstructions.

As a function of distance from the track center, the computation density profiles of latent track in BLG are shown in Fig. 2. A low density core and a high density shell fine structure can be seen. The normalized densities are calculated at each cylindrical shell with the thickness of 0.5 nm. When the radii are less than 1 nm, the density fluctuations are mainly because of the carbon chains located around the track center. For dE/dx = 19 keV/nm, the radius of central pore is about 0.45 nm, so the density of central cylinder with a radius of 0.5 nm does not fall to zero.
Fig. 2. Density as a function of distance from the track center in BLG.

The latent track radii determined as the radial distance where the density of the shell falls to that of the unirradiated material are plotted in Fig. 3. In general, the track radius increases with increasing values of dE/dx. The more energy atoms obtain, the more energy atoms deposit. The radius becomes saturated as the initial track radius is set to 3 nm in this work. This simple homogenous energy deposition model is widely used in ion track simulations, although it is not physically correct as it neglects the radial dose distributions of electrons and the velocity effect. However, it is found that the defect structures formed by different energy deposition model are essentially the same [5].

Fig. 3. The latent track radius as a function of dE/dx. The simulation results of BLG in this work (square) are compared with the previous MD simulations results of graphite thin film in Ref. [12] (triangles).

For the same value of dE/dx, the size of latent track in BLG is larger than in graphite thin film. It indicates that radiation damage relates to the thickness of material. The thinner the material is, the more severe the radiation damage is. The differences between their atomic structures are the principle reason. With more and more layer of atoms, there are more and more interstitial atoms produced by irradiation in the space between layers, which makes the opportunities of defects
annealing and recrystallization more and more.

The redundant energy deposited in the thermal spike is dissipated by energy transfer to the surrounding lattice and creation of defects. Fig. 4 shows the numbers of defects in the latent track as a function of time. It is clear that all simulations are run sufficiently long for the numbers of defects to reach steady values. Take $dE/dx = 19$ keV/nm as an example: the number of defects increases rapidly at the beginning, and reaches its peak of 1648 at about 0.7 ps. Then this number decreases and becomes stable to 1310 after 21.3 ps. The rise and fall of the curve respective correspond to the defect producing and defect annealing processes in the track formation. For higher values of $dE/dx$, the time for both defect producing and annealing processes is greatly lengthened while the maximum and steady numbers of defects increase. At the end of the simulations, less than half of the defects can be recombined.

![Fig. 4. Evolution of the defects number with time for different values of dE/dx.](image)

After reaching steady values, the number of defects per unit volume known as density of defects both in BLG and SLG are plotted in Fig. 5. When $dE/dx$ is higher than about 9 keV/nm, the radiation damage in SLG is more severe than in BLG. SLG is a single atomic layer structure, which means each atoms is the surface atom. As a result, there is no interstitial atom, which makes the defects recovery impossible. In a word, at the same $dE/dx$ which is high enough to generate defects both in BLG and SLG, the radiation damage is more severe in SLG than in BLG.
6 Conclusion

The dependences of the latent track size and defects in latent track on the dE/dx are explored. The (dE/dx)th of BLG is lower than SLG of 8 keV/nm. The latent track radius is obtained through the calculation of the density change between irradiated structure and unirradiated lattice while a low density core and a high density shell fine structure can be seen. Higher values of dE/dx induce larger latent track radius. When the dE/dx is equal to or higher than 19 keV/nm, a small central nanopore can be produced. The radius becomes saturated when the dE/dx is high enough, which depends on the energy deposition model. In addition, the defects produced in the latent track are analyzed. The maximum and steady numbers of defects increase with increasing dE/dx while just a small part of defects can be recombined at the end of the simulations. As long as the dE/dx high enough to induce defects both in BLG and SLG, the radiation damage in SLG is more severe.

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