Nuclear Instruments and Methods in Physics Research B xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Glancing ion incidence on a graphite-supported graphene flake: Lift-off vs welding

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ARTICLE INFO

Article history:
Received 5 December 2016
Received in revised form 18 February 2017
Accepted 3 April 2017
Available online xxxx

Keywords:
Graphene
Graphite
Ion irradiation
Sputtering
Defect formation
Channeling

ABSTRACT

Using molecular-dynamics simulation, we study the impact of 0.5 keV Xe ions at glancing incidence (80° from the surface normal) on a graphene flake supported on a (0001) graphite substrate. The step forming at the ascending edge of the flake allows the entrance of glancing-incidence ions into a subsurface channel between graphene layers. We find that subsurface-channeled ions have a high probability to lift the flake off the substrate, while non-channeled projectiles rather damage the flake leading eventually to welding the flake to the substrate by the formation of sp^3 bonds.

1. Introduction

The interaction of ion irradiation with graphene sheets is of considerable interest, and has recently been reviewed by Krasheninnikov and Nordlund [1]. More recently, oblique and glancing ion incidence on graphene layers was studied; the graphene layers were supported by a metal [2,3] or a semiconducting substrate [4]. These studies provided information about defect formation, ion trapping, and the phenomena of interface and subsurface channeling.

Glancing ion incidence on metals has been studied in considerably greater detail, both by experiment and by simulation [5–11]. Such glancing impact gives rise to the phenomenon of subsurface-channeling where the projectile is channeled immediately under the surface. In this channeling mode the ion creates characteristic damage, such as vacancy islands aligned with the ion incidence direction (projected onto the surface) and even nano-grooves. This ample body of evidence for metallic targets is supplemented by more scarce studies of semiconductors [12,13] and ionic surfaces [14].

Besides metallic or semiconducting substrates, also a graphitic substrate may be used. Indeed a graphene flake on a graphitic substrate naturally appears as an adatom island there. The interaction of ion irradiation with flat – but not with graphene-flake-covered – graphite surfaces has been studied intensely in the past. Experimental studies of ion irradiation on graphite focused on defect generation [15], but also on ion penetration and the sputtering induced [16]. Defect formation under ion irradiation of graphite was extensively studied by Nordlund et al. [17–19], and these results were also extended to cluster impact [20–22]. Radiation effects in graphite are reviewed by Burchell [23]. Most recently, Christie et al. [24] published a detailed study of the evolution of collision cascades in graphite. Many studies were devoted to cluster impact. Webb et al. [25] studied fullerene impacts into graphite and compared its comparison to experimental data obtained by an oxidation technique. Reimann et al. [26] studied experimentally damage creation by metal cluster impact into graphite, and Henkel and Urbassek [27] analyzed this scenario theoretically. These studies were later followed by investigations of fullerene and gas-cluster impacts [28,29].

Desorption from a graphite substrate induced by cluster impact was studied, but usually cluster projectiles were used. Webb [30] used fullerene projectiles to desorb molecules adsorbed on the surface, while Baranov et al. and Anders et al. use large metal cluster projectiles to desorb Au clusters [30–32]. Kornich et al. [33,34]

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http://dx.doi.org/10.1016/j.nimb.2017.04.011
0168-583X/

studied the desorption of small Cu clusters off a graphite surface by single ion impact.

Here we use molecular dynamics (MD) simulation to study ion incidence on a graphene flake adsorbed on a graphite substrate. By focusing on glancing incidence angles and low ion energies, we can study the competition of defect formation in the flake, which may eventually lead to welding of the flake to the substrate by the formation of an sp^3 bond, and desorption of the flake off the surface. The manipulation of graphene flakes by ion irradiation appears to be a promising tool in the light of recent technological applications of graphite [35–37].

2. Method

The graphite substrate consists of 8 graphite (0001) layers and has a thickness of 23.5 Å. The area of the target surface amounts to 113.7×73.8 Å^2. We note that the graphene interlayer distance amounts to $\Delta h = 3.354$ Å; the nearest-neighbor distance amounts to 1.42 Å.

We place on this substrate a graphene flake of hexagonal shape, see Fig. 1. It consists of 150 atoms and constitutes an adatom island on the substrate.

The interaction of carbon atoms is modeled by the AIREBO potential [38].

Before starting the bombardment simulation, the system is relaxed to zero pressure and temperature by applying damping boundary conditions on the lateral sides of the target. The graphene surface is left free, while the bottommost substrate layer is kept fixed. These boundary conditions are also used during the simulation. These conditions are analogous to those used by us in previous surface channeling simulations [4,12,39,40].

As projectiles, we use Xe^+ ions with an impact energy of 0.5 keV. The ion incidence angle is fixed to 80° from the surface normal, since this glancing angle allows for subsurface channeling events leading to flake lift-off. As ion azimuth, we choose the $\langle 10\bar{1}0 \rangle$ direction, since this azimuth has already previously been studied for impacts on large flakes [40]. It is perpendicular to the step bounding the flake, see Fig. 1. Due to the small projectile energy, we disregard electronic stopping. Ions impact in the impact zone shown in Fig. 1. It is built on a unit surface mesh of width $\Delta\xi = 4.26$ Å and breadth $\Delta\eta = 12.35$ Å. The impact zone consists of 13 meshes in total. 10 impact points are chosen randomly inside each surface mesh, such that the total number of ion impacts simulated amounts to 130. We follow the trajectories up to 5 ps simulation time.

We analyze the damage created in the graphene layers by calculating the coordination number of each atom after irradiation using OVITO [41]. The coordination number of an atom is determined as the number of its C neighbors within a sphere centered at the atom of radius $r_c = 1.8$ Å; this value is chosen in between the distance of nearest neighbors (1.42 Å) and second-nearest neighbors (2.47 Å) in graphene. Note that unstrained sp^3-bonds such as in diamond have a length of 1.54 Å, much less than r_c. In an intact graphene sheet, all atoms have three nearest neighbors (sp^2-bonded atoms). We shall call atoms with 2 – or fewer – nearest neighbors *under-coordinated* atoms; they will typically be bonded by sp^1-bonds. Atoms with 4 nearest neighbors will be denoted as *over-coordinated* atoms, bonded by sp^3-bonds.

Atoms are considered to be sputtered if they have moved away from the target by a distance of more than 4.5 Å. Technically speaking, we use the cluster detector built into OVITO [41] to identify all atoms that are still within the interaction range of the target (graphite substrate); these are connected by covalent or van-der-Waals bonds as a large 'cluster' of atoms. All atoms that left this 'cluster' – either as single atoms or since they belong to the flake that has lifted off the substrate – are considered sputtered.

The coordination numbers are calculated at the end of the impact simulation, at 5 ps after ion impact. For several selected events we analyzed the defect structures further by slowly quenching them within 25 ps to 0 K. The differences observed are minor, of the order of only 3%; we therefore presume that we can take our analysis of the as-irradiated samples to describe the trends of damage formation correctly.

3. Results

At 80° incidence angle, a 500 eV projectile has a kinetic energy $E_\perp = 15.08$ eV perpendicular to the surface. Since the threshold displacement energy of graphene is around 15–20 eV [1] and the cohesive energy of graphite (and also graphene) amounts to 7.349 eV [42], this energy is just sufficient to induce some local defects on a flat terrace, leading to local amorphization; however, we did not observe any sputtering on a flat terrace.

When the projectile hits the surface it may induce a number of different effects. We visualize the most important of these in Fig. 2. Under certain conditions, the entire graphene flake is separated from the surface, see Fig. 2a. Energetically, this lift-off process is easily possible, since the interlayer attraction between graphene and the graphite surface is mediated by van-der-Waals forces and hence small. Indeed, the van-der-Waals interaction energy between basal planes in graphite amounts to around 23 meV/atom [43–45]; hence to lift off the entire 150-atom flake requires only 3.45 eV. A more recent compilation [46] gives by a factor of 2

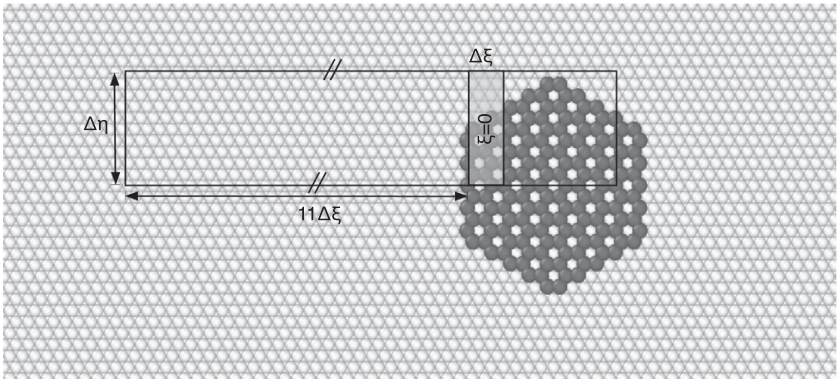

Fig. 1. Top view sketch of the target surface. The graphite substrate is shown in light gray and the graphene flake in dark gray. The impact area is outlined in black; its lateral size $\Delta\eta$ covers the upper half of the flake. The ion impacts from the left along a $\langle 10\bar{1}0 \rangle$ azimuth, which is denoted as the ξ direction. The ascending front edge of the flake is positioned at $\xi = 0$. $\Delta\xi$ denotes the width of an impact cell.

Please cite this article in press as: Y. Rosandi, H.M. Urbassek, Glancing ion incidence on a graphite-supported graphene flake: Lift-off vs welding, Nucl. Instr. Meth. B (2017), http://dx.doi.org/10.1016/j.nimb.2017.04.011