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Effect of kinetic energy and impact angle on carbon ejection from a free-standing graphene bombarded by kilo-electron-volt C_{60}

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Molecular dynamics computer simulations are employed to investigate the effect of the kinetic energy and impact angle on the ejection process from a free-standing graphene of thickness between 1 and 16 layers. The target is bombarded by C_{60} projectiles with kinetic energy between 5 and 40 keV and the impact angle ranging between 0° and 80°. The yields, kinetic energies, and ejection directions of atoms are monitored. Computer simulations are used to point to optimal conditions when a soft ejection of unfragmented molecules may occur, which may be invaluable information for the development of secondary ion mass spectrometry based on a transmission geometry. *Published by the AVS.* https://doi.org/10.1116/1.5019732

I. INTRODUCTION

In recent years, cluster ion beams have attracted increasing experimental and theoretical attention due to their capacity to enhance the ejection of large intact organic molecules in secondary ion mass spectrometry (SIMS).[1,2] One of the most successful clusters used in organic SIMS is C_{60} fullerene.[3] In a typical SIMS geometry, the detector is located on the same side of the target as the ion gun. Usually, metal or semiconductor supports are used to deposit the analyzed material. A novel SIMS configuration, based on ultrathin free-standing graphene substrates and a transmission geometry, was proposed recently by a group from Texas A&M University.[4,5] In this approach, the analyzed organic material is deposited at one side of the ultrathin substrate, while another side is bombarded by cluster projectiles. It is argued that such geometry can be particularly attractive for analysis of small amounts of organic material, molecular nano-objects, and supramolecular assemblies. It has also been reported that the formation of negative ions emitted from ultrathin organic films deposited on a free-standing graphene[4] or covered by a graphene sheet[6] is enhanced.

While the experimental data showing the advantages of graphene application to SIMS are convincing, much less is known about the processes leading to material ejection from this system. Only a few simulations have been performed so far for C_{60} bombardment of a free-standing graphene.[4,5,7–10] Most of the existing simulations modeled projectile impact at graphite.[11–18] Moreover, many of these studies concentrate on defect creation in the bombarded system rather than on material ejection. Theoretical studies of sputtering of graphite by kilo-electron-volt C_{60} projectiles show that the sputtering yield is unexpectedly low.[17,18] Krantzman *et al.* attributed this fact to a low atomic density of graphite,[17] while the effect of the layered structure of graphite was emphasized by Tian *et al.*[18] It has also been shown that the membranelike structure of graphite can vibrate after a low-energy cluster impact.[4,16] Similar vibrations have been observed in a single layer of graphene.[19,20] It has been argued that the energy stored in this process can be sufficient to uplift molecules adsorbed on a graphite.[14,16]

The effect of the graphene substrate thickness on the ejection process has been recently investigated for a single primary kinetic energy and a single impact angle.[8] In that study, free-standing graphene substrates, 2–16 layer thick, were bombarded by 10 keV C_{60} projectiles at normal incidence. It has been shown that the yield depends on the sample thickness in a nonmonotonic way and the shape of this dependence is a consequence of an interplay between the amount of material available for ejection and the energy deposited in the subsurface regions by impinging projectiles. The goal of this paper is to investigate the effect of the kinetic energy and the impact angle on the ejection processes.

II. COMPUTER MODEL

The molecular dynamics (MD) computer simulations were used to model cluster bombardment. Briefly, the movement of particles is determined by integrating Hamilton's equations of motion. The forces among carbon atoms in the system are described by the ReaxFF-lg force field,[21] which allows for the creation and breaking of covalent bonds. This potential is splined at short distances with a Ziegler-Biersack-Littmark potential[22] to properly describe high energy collisions. A detailed description of the MD method can be found elsewhere.[1] The shape and size of the samples are chosen based on visual observations of energy transfer pathways stimulated by impacts of C_{60} projectiles.[8] As a result, cylindrical samples with a diameter of 40 nm are used. Samples with a thickness between 1 (1L) and 16 (16L) graphene layers with a highly oriented pyrolytic graphite structure are bombarded by C_{60} directed at the bottom of the sample. The kinetic energy and the impact angle of the projectile are changed to investigate the effect of these parameters on the particle ejection process. Particles ejected both in the direction of the primary beam (transmission direction) and in the opposite direction (sputtering direction) are collected, as shown schematically in Fig. 1. Rigid and stochastic regions are used to simulate the thermal bath that keeps the sample at required temperature, to prevent

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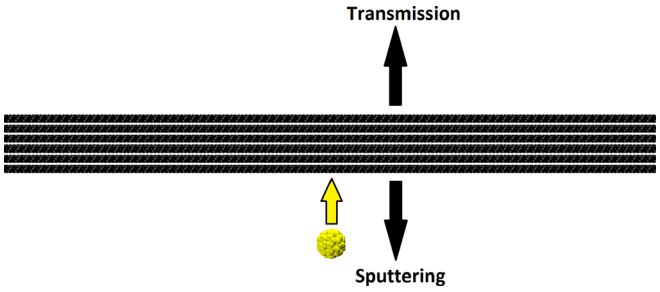

FIG. 1. (Color online) Schematic side view of the modeled system. The bright (yellow) arrow indicates the impact direction. Black arrows show the transmission and sputtering directions mentioned throughout this paper.

reflection of pressure waves from the boundaries of the system, and to maintain the shape of the sample.[1,23] The simulations are run at a target temperature of 0 K in an NVE ensemble and extend up to 10 ps, which is long enough to achieve saturation in the ejection yield versus time dependence. Between 8 and 32 randomly selected impact points located near the center of the sample are chosen to achieve statistically reliable data. Simulations are performed with the large-scale atomic/molecular massively parallel simulator code[24] which was modified to better describe sputtering conditions.

III. RESULTS AND DISCUSSION

The effect of the substrate thickness and primary kinetic energy on the yield of carbon atoms ejected from free-standing graphene systems bombarded by C_{60} projectiles at normal incidence is shown in Fig. 2(a). The yield initially increases with the increase in the surface thickness for substrate atoms ejecting in both transmission and sputtering directions. Subsequently, the yield decreases for atoms ejected in the transmission direction and it saturates for sputtered atoms. The primary kinetic energy does not influence the shape of the yield versus thickness dependence. However, bombardment by a more energetic projectile leads to a stronger emission and shifts the position of the maximum in the yield versus thickness dependence to thicker systems. Our results demonstrate a direct proportionality between the position of the maximum and the projectile kinetic energy.

Variation of the substrate thickness has a different impact on ejection of projectile atoms. The yield decreases monotonically with the sample thickness for atoms ejected in the transmission direction. The yield of backscattered projectile atoms is very low and does not exhibit a consistent dependence on the substrate thickness. These observations indicate that projectile atoms are being trapped inside the graphene substrate. The magnitude of this process increases with the sample thickness. More energetic projectiles are able to perforate thicker substrates. As a result, the yield versus thickness dependence broadens for more energetic projectiles.

Almost all projectile atoms penetrate through a thin substrate, as shown in the inset of Fig. 2(a). However, even in this case, the projectile-graphene interaction is significant, especially for low-energy projectiles. For instance, for 5 keV C_{60},

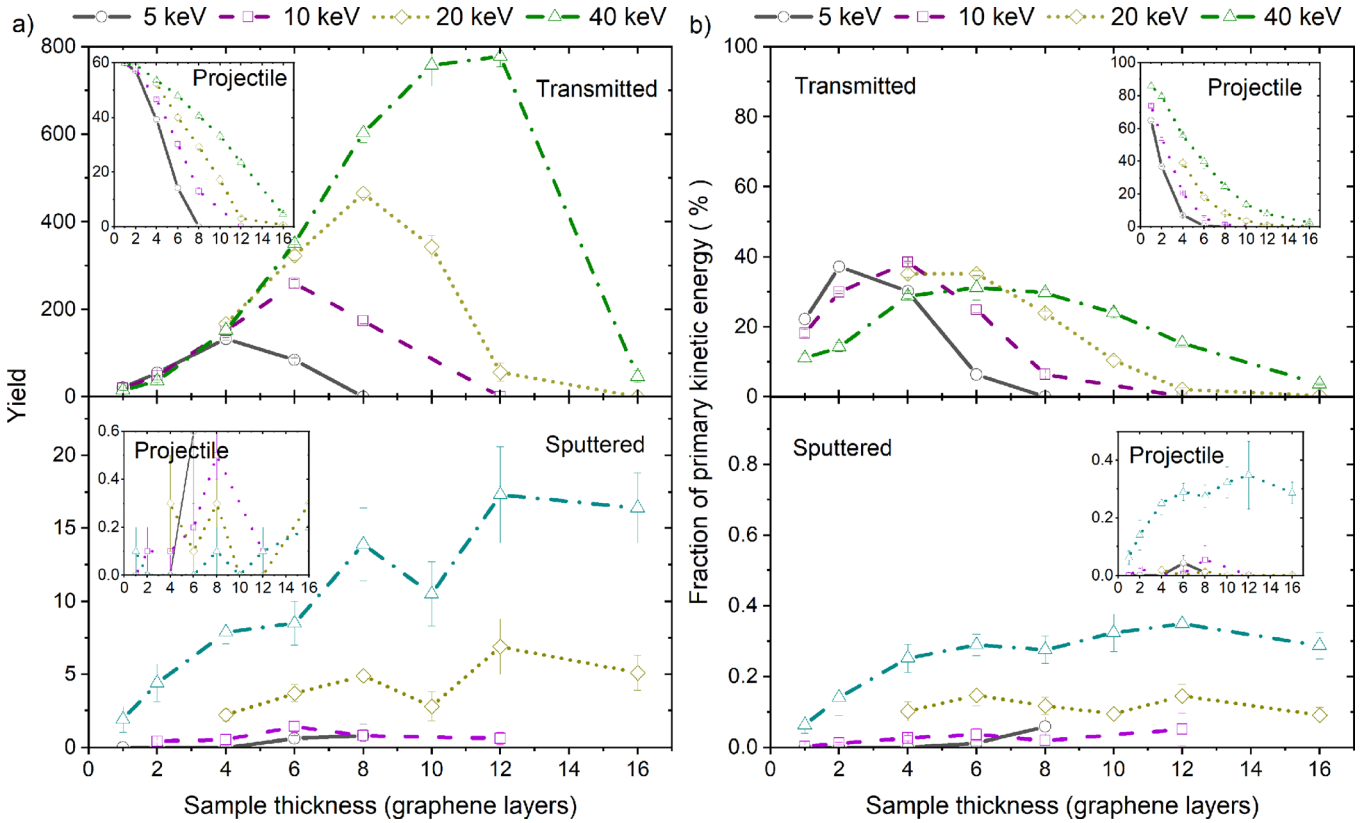

FIG. 2. (Color online) Dependence of (a) the ejection yield and (b) the fraction of primary kinetic energy carried away by particles emitted in the transmission (top) and sputtering (bottom) directions on the thickness of the sample bombarded by 5, 10, 20, and 40 keV C_{60} projectiles at normal incidence. Main graphs represent the atoms ejected from the sample, while the insets depict projectile atoms.

almost 40% of the primary kinetic energy is deposited into the one layer (1L) sample, as shown in the inset of Fig. 2(b). Almost 70% of impact energy is deposited in the 2L system. These numbers drop to 15% and 20% for analogous systems bombarded by 40 keV projectiles. Most of the deposited energy is carried away by substrate atoms emitted in the transmission direction. The energy carried away by sputtered atoms is small and does not exceed 0.5% of the initial kinetic energy. For a given primary kinetic energy, the amount of deposited energy increases with the sample thickness. Nevertheless, there is always an optimum thickness when the largest portion of the primary kinetic energy is carried away by ejecting atoms. This optimum thickness is not equal to the thickness yielding the largest particle emission. For instance, the largest fraction of primary kinetic energy is carried away from a 2L system for 5 keV C_{60} projectiles, whereas the most efficient ejection occurs from the 4L system at this impact energy. The increase in the primary kinetic energy shifts this optimal thickness to a higher value. Interestingly, the maximum fraction of the primary energy carried away by substrate atoms does not depend on the kinetic energy of a projectile. It is approximately 40% regardless of the value of this parameter.

The effect of the impact angle on the ejection yield from the 2L and 8L systems bombarded by 10 and 40 keV C_{60} projectiles is shown in Fig. 3. The impact angle has a similar influence on the yield of substrate atoms emitted in the transmission and sputtering directions for both these systems. First, the yield increases with the impact angle, and then, it decreases. The position of a maximum shifts to a larger impact angle and becomes more pronounced for more energetic projectiles.

The impact angle also has a significant influence on a number of ejected projectile atoms. The functional form of this influence is, however, different from the one observed for substrate atoms. The number of projectile atoms penetrating through the sample decreases monotonically with the impact angle, whereas the yield of backreflected atoms increases for more oblique impacts. For the 2L system, these yields are almost complementary, which indicates that projectile atoms can be either transmitted or backreflected. In other words, projectile atoms cannot be trapped inside such a thin system. The situation is different for the 8L system, especially when it is bombarded by low-energy projectiles. In this case, many projectile atoms are trapped inside the sample.

Cross-sectional views of the temporal evolution of the bombarded systems can be used to gain insight into the mechanism of particle ejection. As SIMS analysis with graphene substrates is performed at high kinetic energy,[4,5] we limit our discussion to a 40 keV bombardment. Cross-sectional views of the 2L and 8L systems bombarded by 40 keV C_{60} projectiles are shown in Figs. 4 and 5, respectively. See supplementary material for animations of the impacts.[29] The plots are made for impact angles which correspond to a normal incidence, the incidence when ejection of substrate atoms is the most efficient, and the impact angle

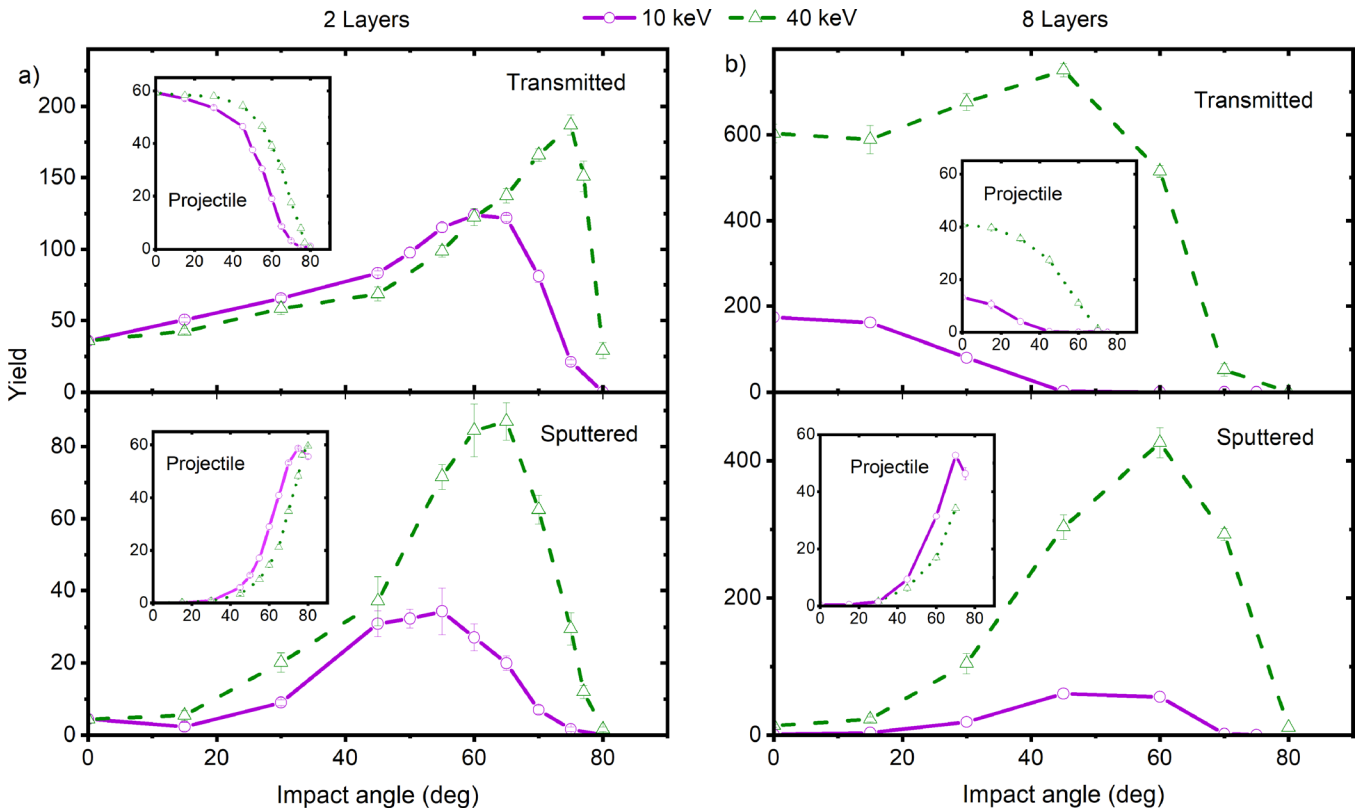

FIG. 3. (Color online) Dependence of the yields of carbon atoms ejected in the transmission (top) and sputtering (bottom) directions from the (a) two and (b) eight layer systems bombarded by ten (solid line) and 40 keV C_{60} (dashed line) projectiles on the impact angle. Main graphs represent the atoms originating from the sample, while the insets depict projectile's atoms.

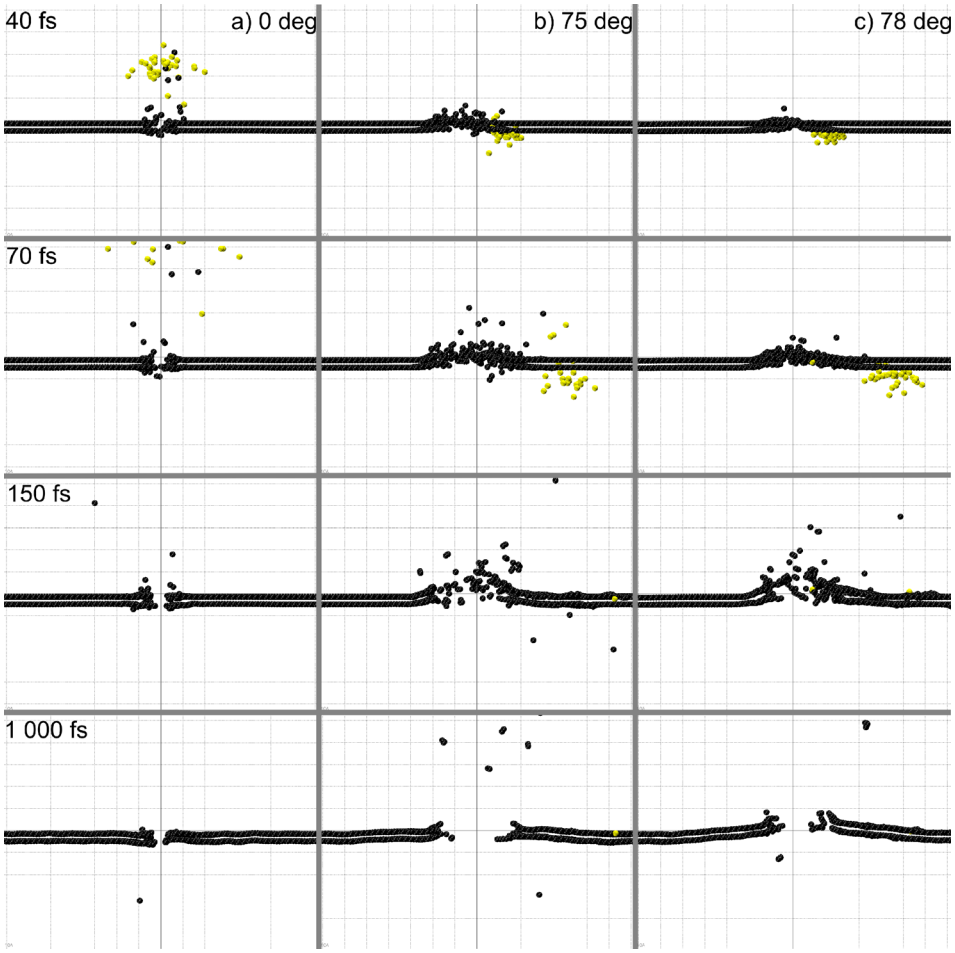

FIG. 4. (Color online) Cross-sectional view of the temporal evolution of a typical collision event leading to ejection of atoms due to 40 keV C_{60} bombardment of a system composed of two graphene layers. Bright (yellow) spheres indicate projectile atoms. A 1 nm slice of the system centered at the impact point is shown. The plots are made for impact angles, which correspond to a normal incidence, the incidence when ejection of substrate atoms is the most efficient, and the impact angle when the yield of these atoms decreases. The dashed lines in the background are separated by 1 nm.

when the yield of these atoms decreases. These impacts should lead to the widest range of phenomena stimulated by projectile impact. From Figs. 4 and 5, it is evident that the integrity of the C_{60} projectile is compromised almost immediately after the impact. However, the projectile atoms remain together and interact collectively with the sample.

For the 2L sample bombarded at normal incidence, all projectile atoms penetrate through the substrate and an almost circular nanopore is created, as shown in Fig. 6. Zhao *et al*. have found that energetic clusters can be used to fabricate nanopores in graphene in a controlled way by varying the properties of the incident projectile.[7] They have found that an impact energy of 11.4 eV/atom is needed to create a nanopore in a single layer of graphene when bombarded with C_{60} projectile at normal incidence.[7] This energy corresponds to approximately 0.68 keV for the entire C_{60} projectile. Assuming that the same energy is necessary to perforate additional layers, one can predict that approximately 5.4 keV is needed to perforate the 8L system. This value is close to the threshold energy observed in our simulations [see the top inset of Fig. 2(a)]. The ejection process is very fast, and most of the atoms are emitted within 200 fs after the projectile impact. Regardless of a high projectile kinetic energy, most of the substrate atoms are ejected from the topmost layer in the transmission direction. In fact, approximately 75% from 42 atoms ejected in the transmission direction originate from the topmost layer. The trend is opposite for substrate atoms ejected in the sputtering direction. In this case, ejection from the bottom layer dominates. Most projectile and substrate atoms emitted in the transmission direction are ejected at off normal angles. With the increase in the impact angle, the nanopore becomes larger. However, its size increases almost entirely along the impact azimuth. As a result, it becomes ellipsoidal. The size of the nanopore increases as the projectile impinging at the off normal angle travels a longer path inside the layer and consequently can interact with the larger amount of substrate material. However, simultaneously, the normal component of the projectile momentum decreases, and it becomes more difficult to perforate the substrate. As a result, a larger number of projectile atoms are backreflected from the sample and less energy becomes available to stimulate ejection of substrate atoms. At a certain moment, this process begins to dominate over the increase in the substrate material excited by the projectile, and both the sputtering yield and the nanopore size decrease. The projectile impact leads to the creation of

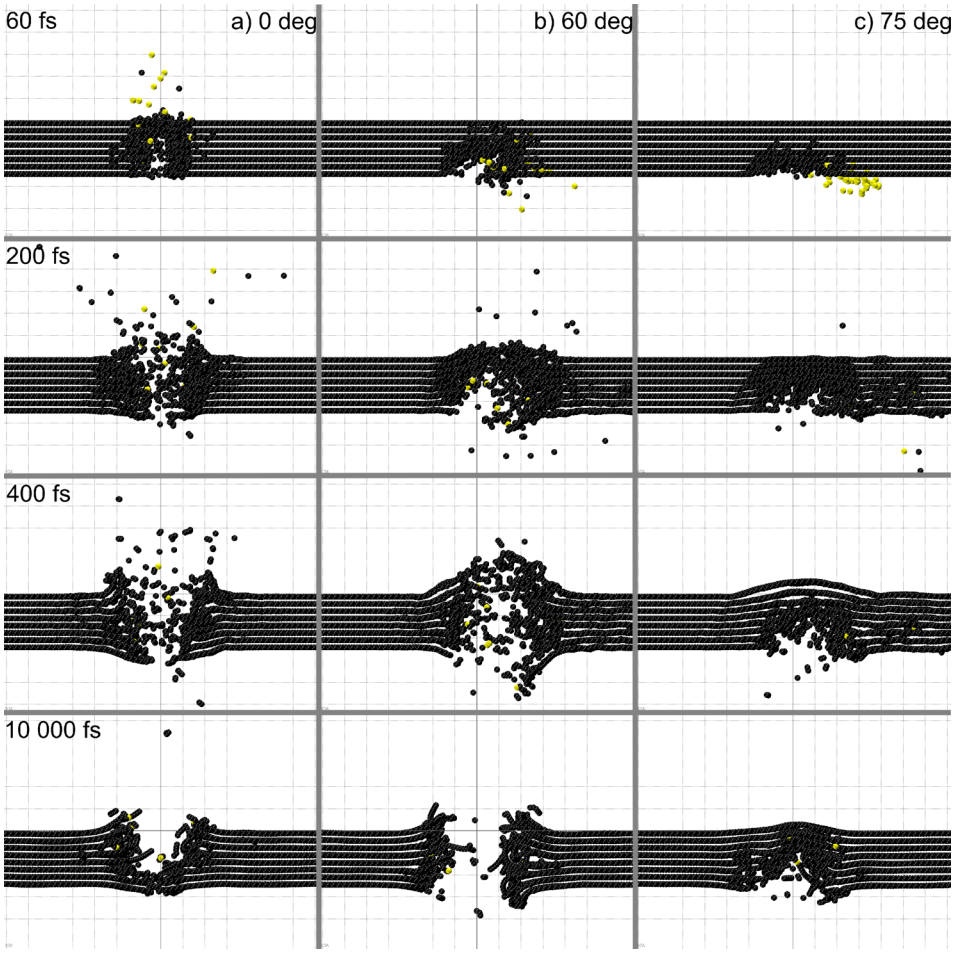

FIG. 5. (Color online) Cross-sectional view of the temporal evolution of a typical collision event leading to ejection of atoms due to 40 keV C_{60} bombardment of a system composed of eight graphene layers. Bright (yellow) spheres indicate projectile atoms. A 1 nm slice of the system centered at the impact point is shown. The plots are made for impact angles, which correspond to a normal incidence, the incidence when ejection of substrate atoms is the most efficient, and the impact angle when the yield of these atoms decreases. The dashed lines in the background are separated by 1 nm.

cylindrical acoustic waves that propagate outward from the point of impact with a maximum amplitude of approximately 0.1 nm.

More dramatic alteration is observed in the 8L system. In this case, the ejection process requires more time to complete. The projectile is more efficiently decelerated, depositing almost all its kinetic energy into the sample. Ejection of substrate atoms in the transmission direction is the main channel of material removal. The original location of ejected substrate atoms is not restricted to the top layers, but it extends deep into the sample. For instance, approximately 40%, 25%, 16%, 10%, and 5% of 603 atoms ejected in the transmission direction originate from the first (topmost), second, third, fourth, and fifth layers, respectively, which explains a conical shape of the evacuated volume. The remaining part of deposited energy is used to deform the substrate. Near the point of impact, for a short time, graphene sheets become separated from each other and bend up in a direction parallel to the movement of incoming projectile. Finally, a circular opening is formed surrounded by the elevated rim at the top surface of the sample. No rim is formed at the bottom surface. Bonds of many carbon atoms located in the energized volume are broken, which means that these atoms become highly reactive. Many of the decelerated projectile atoms bound with these atoms.

The evolution of a system bombarded at an impact angle corresponding to the most efficient ejection of substrate atoms is shown in the second column of Figs. 4 and 5. For a 2L system, this angle is approximately 75°, while an angle of 60° is the most optimal for an 8L system. The 2L system is perforated within 70 fs. The projectile integrity is compromised again, but most of the projectile atoms preserve their original movement direction. There is a lot of movement at the edges of the created nanopore which now is elongated along the impact azimuthal direction, as shown in Fig. 6. Because the movement trajectory is now oblique, a larger volume of the sample is energized. However, the component of the projectile momentum perpendicular to the surface is reduced. As a result, it is easier to reflect projectile atoms.

Again, a more dramatic action is observed for an 8L system. The projectile atoms penetrate along the initial direction, but soon they become decelerated by collisions with substrate atoms. Most of these atoms become trapped inside the sample. First, the opening at a bottom surface is created and the substrate atoms are sputtered at oblique angles. Approximately 400 fs after the projectile impact, the integrity of the upper

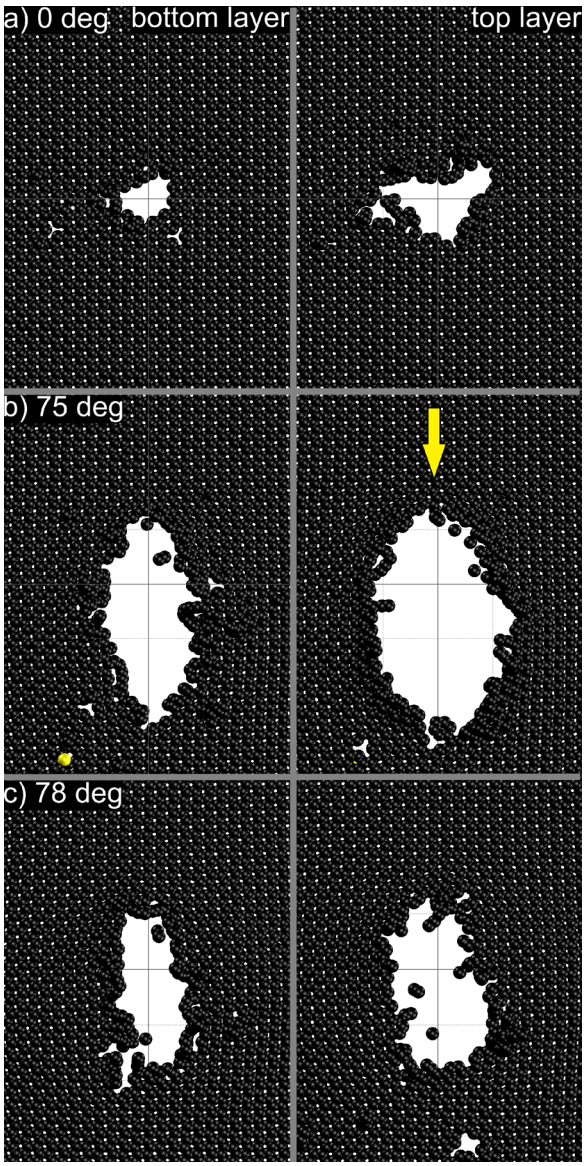

FIG. 6. (Color online) Top view of the 2L system bombarded by 40 keV projectiles at several impact angles. Bright (yellow) balls depict projectile atoms. The image is collected 1 ps after the projectile impact. The dashed lines in the background are separated by 1 nm. Arrow indicates azimuthal direction of the impacting projectile.

part of the sample is compromised, and the sample atoms start to eject in the transmission direction. It is interesting to note that regardless of a very large impact angle, most of these atoms eject in directions close to the surface normal. Finally, a cylindrical nanopore is created with openings of similar dimensions. Strong deformations are observed near the point of impact, which results in a graphene layer unfolding over a larger area.

A further increase in the impact angle leads to a signal decrease as shown in the third column of Figs. 4 and 5 for bombardment at the 78° and 75° impact angles of the 2L and 8L systems, respectively. The normal component of the projectile momentum is now so low that it becomes difficult to perforate even a 2L system. The 8L system is not perforated. Almost all projectile atoms backreflect from the 2L sample, while some of these atoms are trapped inside the 8L system. Although the projectile atoms are not penetrating through the 8L substrate, its upper surface bulges outward during projectile deceleration.

Based on all observations, it can be concluded that the yield of substrate atoms is determined by two factors. The first factor is the amount of material available for sputtering. This quantity will increase with the sample thickness and with the impact angle, as projectile will travel a longer path inside the layer. The second factor is the amount of energy stored near the surface from where the ejection occurs. For ejection in the transmission direction, the upper surface is important. The energy stored near this surface will decrease with the sample thickness and the impact angle as projectile atoms have to sacrifice more energy to penetrate through the layer. For conditions where the substrate is perforated, an increase in the material available for ejection dominates and the yield increases with the substrate thickness or impact angle. However, ultimately less energy becomes available near the upper surface and the yield drops. For ejection in the sputtering direction, a bottom surface is important. The energy deposited near this surface increases with the sample thickness until the layer becomes thicker than a depth of a volume from where particles are ejected. Subsequently, the yield will saturate. The increase in the impact angle has also a positive effect on the amount of energy stored near the bottom surface, as the energy deposition profile is shifted downward. However, the increase in the impact angle also reduces the projectile momentum component perpendicular to the surface. It becomes easier to backreflect the projectile atoms, and more energy is carried away by these particles. As a result, less energy is deposited near the bottom surface and the yield decreases for too oblique impacts.

The yield of the projectile atoms ejected in the transmission direction is determined only by the capability of projectile atoms to perforate the sample. This capability decreases with the increase in both the layer thickness and the impact angle. The yield of the projectile atoms ejected in the sputtering direction will be determined by the capability of the sample to backreflect the projectile atoms. As already discussed, this capability increases with the impact angle.

The increase in the primary kinetic energy leads to the deposition of a larger amount of this energy in the sample and to a larger projectile range. Both these factors lead to a stronger ejection of substrate atoms. A larger penetration range increases the substrate thickness which can be perforated by the projectile. This factor leads to a shift of the emission maximum toward thicker samples for a constant impact angle or toward larger impact angles for a constant thickness when the projectile kinetic energy is increased. The increase in the primary kinetic energy also results in a broader distribution for projectile atoms.

Finally, a few comments can be made about the applicability of ultrathin graphene substrates for SIMS analysis of organic overlayers. It is known that for a standard sputtering geometry, collisions of adsorbed molecules with ejecting substrate atoms or a concerted action of the unfolding of the crater rim are the main processes leading to molecular emission from ultrathin organic layers deposited on solid substrates

bombarded by atomic and cluster projectiles, respectively.[1,25–28] Direct collisions between projectile atoms and adsorbed molecules lead to molecular fragmentation.[27,28] From this point of view, strong ejection of substrate atoms is a preferred experimental condition. The application of graphene and a transmission geometry allows us to satisfy this requirement. As shown in Fig. 2(a), ejection in the transmission direction is much stronger than that in the sputtering direction. However, the energetics of collisions between ejecting substrate atoms and the adsorbed molecules is also important. From the point of view of this factor, the application of a transmission geometry is less beneficial, as the energy of substrate atoms ejecting in the transmission direction is higher than the energy of a typical bond. For instance, in a 2L system bombarded by 5 keV projectile at normal incidence, the molecules located immediately above the point of projectile impact will collide with projectile atoms moving with the average kinetic energy of almost 70 eV per atom. Even for substrate atoms, the average kinetic energy will be close to 14 eV per atom. Collisions with such atoms will certainly lead to molecular fragmentation. Much more promising is a process of unfolding of the topmost graphene layer. In this case, the graphene sheet acts as a catapult that can gently hurl molecules into the vacuum. There is a considerable amount of energy associated with this movement, which means that even very large molecules can be uplifted. In the transmission geometry, this movement extends to a much larger lateral distance from the point of impact, as compared to a similar process present in metals or semiconductors.[23,26] As shown in Figs. 4 and 5, it may be even more advantageous to bombard thicker samples and use the off-normal impact angle to enhance the catapult action. Consequently, a larger number of adsorbed molecules could be ejected by a single projectile impact, making analysis of small amounts of organic material viable. However, it should also be kept in mind that ejection of electrons is necessary to stimulate formation of negative ions,[4–6] which means that a certain amount of kinetic energy must be present near the area of molecular ejection to emit such electrons. From this point of view, the application of thick substrates or bombardment at large impact angles may not be optimal.

IV. SUMMARY

Processes responsible for particle ejection from graphene substrates of various thicknesses bombarded by C_{60} projectiles in a wide range of primary kinetic energies and impact angles were investigated. It has been observed that these quantities have a significant influence on the yield and the dynamics of particle ejection. For a given impact angle and primary kinetic energy, the yield of the substrate atoms ejected in the transmission direction has a nonmonotonic dependence on the sample thickness, with a pronounced maximum. A similar shape of dependence is observed if the impact angle is changed, while the primary kinetic energy and the sample thickness are kept constant. The position of the maximum in these dependencies shifts to thicker samples for a constant impact angle and to a larger impact angle for a constant thickness, if the kinetic energy of a projectile is increased. The yield of sample atoms ejected in the sputtering direction saturates with the sample thickness for a given kinetic energy and impact angle. The number of projectile atoms ejected in the transmission direction decreases monotonically with the increase in the sample thickness and impact angle or with the decrease in the primary kinetic energy. All these changes result in a decrease in energy deposited in the top subsurface region. The yield of projectile atoms backreflected from the sample does not have a visible dependence on the sample thickness, but it increases for more oblique impacts, as it is easier to reflect atoms with a small component of momentum perpendicular to the surface. The width of these dependencies broadens with the increase in a primary kinetic energy. All observed trends can be explained by an interplay between the amount of material available for ejection and the amount of primary kinetic energy being deposited in the top and bottom subsurface regions of the sample.

Our study confirms that graphene supports and a transmission geometry have advantages over traditional metal or semiconductor substrates for analysis of ultrathin materials. First, the extremely small thickness of the support results in small amounts of emitted substrate material. As a result, there is a minimal interference between the substrate and the analyzed signal. A large portion of the primary kinetic energy can be transmitted to the organic overlayer in the direction toward the detector by the collective movement of the topmost layer, increasing a chance that a small amount of analyte can be recorded. Our results confirm that the graphene sheet can act as a catapult, leading to efficient soft ejection of adsorbed organic molecules. However, ejection of secondary electrons is also necessary to stimulate efficient ionization.

ACKNOWLEDGMENTS

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[29]See supplementary material at https://doi.org/10.1116/1.5019732 for aminations of 40 keV C60 impacts at 2 L and 8 L graphene at impact angles corresponding to a normal incidence, the incidence when ejection of substrate atoms is the most efficient, and the impact angle when the yield of these atoms is decreasing.