Supported Two-Dimensional Materials under Ion Irradiation: The Substrate Governs Defect Production

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ABSTRACT: Focused ion beams perfectly suit for patterning two-dimensional (2D) materials, but the optimization of irradiation parameters requires full microscopic understanding of defect production mechanisms. In contrast to freestanding 2D systems, the details of damage creation in supported 2D materials are not fully understood, whereas the majority of experiments have been carried out for 2D targets deposited on substrates. Here, we suggest a universal and computationally efficient scheme to model the irradiation of supported 2D materials, which combines analytical potential molecular dynamics with Monte Carlo simulations and makes it possible to independently assess the contributions to the damage from backscattered ions and atoms sputtered from the substrate. Using the scheme, we study the defect production in graphene and MoS2 sheets, which are the two most important and wide-spread 2D materials, deposited on a SiO2 substrate. For helium and neon ions with a wide range of initial ion energies including those used in a commercial helium ion microscope (HIM), we demonstrate that depending on the ion energy and mass, the defect production in 2D systems can be dominated by backscattered ions and sputtered substrate atoms rather than by the direct ion impacts and that the amount of damage in 2D materials heavily depends on whether a substrate is present or not. We also study the factors which limit the spatial resolution of the patterning process. Our results, which agree well with the available experimental data, provide not only insights into defect production but also quantitative information, which can be used for the minimization of damage during imaging in HIM or optimization of the patterning process.

KEYWORDS: two-dimensional materials, ion irradiation, atomistic simulations, defects, sputtering, He ion microscopy

INTRODUCTION

Ion irradiation1–7 is one of the most powerful tools to change the atomic structure and properties of the materials through controllable introduction of impurities and defects. The technique suits particularly well for the processing of two-dimensional (2D) materials because of the small thickness of the system achieving a uniform distribution of dopants and defects with regard to their depth is not an issue, as evident from numerous examples for graphene9–11 or transition-metal dichalcogenides (TMDs)10–12. Moreover, exposure to energetic ions can be combined with in situ or postirradiation chemical treatment of these atomically thin targets13 to incorporate foreign atoms into the atomic network due to chemically reactive vacancies.

Focused ion beams can be used to produce defects with high spatial resolution. They can also be employed for cutting and patterning 2D materials14. In this context, the He ion microscope (HIM),15 where He or Ne ion beams have subnanometer diameters, has been demonstrated to be a perfect tool for not only getting insights into sample morphology but also altering its structure and geometry at the nanoscale. Specifically, structural defects were controllably introduced in a few-layer MoS2 sample16 so that its stoichiometry and electronic transport properties could be tuned. Nanoribbons with widths as small as 1 nm were reproducibly fabricated in graphene17–21 and MoS2 sheets,22 and subnanometer nanopores were produced in MoS2 monolayers23 which were used to study ionic transport through the sheets. An increase in Young’s modulus of MoSe2 samples irradiated in HIM was reported, along with the possibility to tune optical properties of TMDs.24 Structural changes in freestanding graphene and that encapsulated between the sheets of hexagonal boron nitride under focused helium ion beam irradiation have been studied25 in the HIM and showed the benefits of graphene encapsulation for postsynthesis doping and self-healing of the beam-induced lattice damage.
In the typical experimental setup used for ion bombardment, the 2D target is placed on a substrate, normally a Si/SiO₂ slab. However, the experimental data are frequently rationalized using atomistic computer simulations carried out for freestanding systems. It is tacitly assumed that most of the defects are produced from direct interaction of the primary beam with the target, which, as we show below, is not generally true, especially for the case of light ion irradiation in the HIM. Indeed, the experiments carried out for supported and freestanding 2D MoS₂ and graphene clearly indicate that there are substantial differences in the amount of beam-induced damage and the properties of irradiated samples.

In contrast to freestanding targets (e.g., graphene or MoS₂ suspended on a transmission electron microscopy (TEM) grid), where all the defects are created by the impinging ions, the substrate can affect defect production in several ways. Putting aside the case of highly charged ions and swift heavy ions which literally “blow-up” the substrate so that the 2D material on top is torn apart by the atoms coming from the substrate, defects in the 2D system can be produced at moderate ion energies by two other mechanisms, in addition to (i) direct ion impacts, (ii) backscattered ions, and (iii) atoms sputtered from the substrate, as schematically illustrated in Figure 1. The latter two mechanisms are indirect defect production channels. Moreover, experimental investigations using state-of-the-art focused ion beam technology indicate that the substrate hinders higher patterning resolutions, so that a detailed microscopic understanding of the role of the substrate and its effects on defect production under ion irradiation is required. The substrate also plays the key role in the evolution and annealing of defects. Several attempts to account for the effects of the substrate have been carried out for high-energy (MeV) heavy ion and 30 keV He ion irradiation of supported graphene, but the trends and physical processes involved were not systematically analyzed. Computationally, this is a very challenging task, as first-principles approaches cannot be used because of the high computational costs required to collect representative statistics for systems composed of a few thousand atoms. Furthermore, analytical potentials developed for multiatomic systems are either of limited accuracy or are still computationally too expensive for adequate modeling of the system.

In this work, we suggest an efficient scheme to model the irradiation of supported 2D materials, which combines analytical potential molecular dynamics (MD) simulations for the 2D material, augmented with a universal repulsive potential to account for the effects of the substrate on sputtered atoms and a Monte Carlo (MC) method. This allows to independently assess the contributions to the damage from backscattered ions and atoms sputtered from the substrate and hitting the supported 2D system. Using the scheme, we study defect production in graphene and MoS₂ sheets, which are the two most widely used 2D materials, deposited on a Si/SiO₂ substrate. We pay particular attention to helium and neon ions because they are used in HIMs for material modification, nanopatterning, and imaging purposes.

Although for the patterning processes the controllable production of defects in a narrow region is desirable, minimal defect production should be aimed at for nondestructive imaging. Using our approach, we access the defect production rates of He, Ne, and Ar ions for a wide range of initial ion energies including the HIM typical energy interval from 1 to 30 keV. Our results indicate that in this energy range, defect production in graphene and MoS₂ is dominated by backscattered ions and sputtered substrate atoms rather than by the direct ion impacts. Consequently, the area in which defects are introduced is dramatically enlarged as compared to that in the freestanding irradiated material, and in the case of HIM-based irradiation, to the beam diameter (typically 0.5 and 1.8 nm for He and Ne, respectively).

**COMPUTATIONAL METHODS**

**MD Simulations.** MD simulations were carried out using the LAMMPS package to extract the statistics of defects produced by the impacts of the projectiles (He, Ne, and Ar ions, and Si and O atoms) onto the 2D materials (graphene and MoS₂ monolayers). The MD calculations of ion impacts onto targets were performed as described previously. At least 320 impact points per energy and angle configuration were chosen. A modified Stillinger–Weber potential with a smooth transition to the Ziegler–Biersack–Littmark (ZBL) potential for small distances was used. For collisions of ions with the target atoms, the ZBL part dominates at high energies, so that the ZBL potentials were also used to describe the impacts of atoms sputtered from the substrate. This approximation is further validated by a rather weak interaction of O and Si adatoms with pristine graphene and MoS₂: the adatoms are mobile on graphene and MoS₂ surfaces, so that they should form compounds and desorb from the system. The MD calculations for ion bombardment of graphene were carried out using the combined Tersoff and ZBL potential.

The simulation setup is presented in Figure 2. As we are aiming at assessing the effects of low-dose irradiation, we assumed that the ions always impinge into pristine material. To collect representative statistics for different impact points, an irreducible area was chosen as depicted in Figure 2c. The average number of defects produced by impinging and backscattered particles, along with those produced by sputtered substrate atoms, was obtained from MD simulations as functions of ion energy and angle.

The effects of the SiO₂ substrate were modeled as an external repulsive potential acting on the target atoms (Mo, S, or C) in the corresponding region (see Figure 2). The choice of the potential was motivated by the confining effect of the underlying substrate, which makes the presence of sputtered target atoms in the substrate region energetically unfavorable. The potential in this approximation can be expressed as:

\[ U(z) = A(e^{\beta(c-z)} - 1) \]
where the coefficients are determined by the boundary of the potential region $z_0 = 2$ Å, the surface approximation energy—the energy required to approach a surface located at $d = 3$ Å (distance from the monolayer derived from first-principles calculations) from infinity $U(z = d) = 10$ eV, and a defined kinetic energy loss of the incident particle with $U(z = 4$ Å) = 50 eV. This yields the coefficients $A = 10/3$ eV $\approx 3.33$ eV and $\beta = 2 \ln 2$ Å$^{-1} \approx 1.38$ Å$^{-1}$. The choice of the parameters is not unique and depends on the target material—substrate pair, but our calculations with slightly different parameters gave qualitatively similar results. As for other simplifications, for sputtered target atoms (e.g., S) with high energies, our approach disregards the fact that the atoms may be captured by the substrate: instead in the current model, it will be reflected and may act as a secondary projectile and create defects, but the probability for this process is very small. Another possible artifact of our approach is that adsorption on the substrate is not taken properly into account. However, in this case, the atoms are trapped between the 2D material and the substrate, and this does not influence defect production.

The potential has no effect on the impinging ions, whereas backscattered ions were treated using a statistical approach with the data collected from the MC simulations. Splitting defect production into two different channels is possible because of a very small probability for the backscattered ions and sputtered atoms to hit the area where defects were already produced by that same ion. Because of the geometry of the system, we independently considered defect production by the ions coming from above the 2D target, with ion velocity vectors pointing toward the substrate (direct impacts), and from below, with ion velocity vectors pointing away from the substrate (the case for backscattered ions and sputtered substrate atoms).

**MC Simulations.** The MD approach was combined with the MC method implemented in the TRIDYN code$^{48,49}$ to account for projectile properties and their statistics (e.g., backscattering

![Figure 2](image_url) Simulation setup. (a) Atomistic model of MoS$_2$ on a SiO$_2$ substrate. (b) Corresponding system where the substrate is modeled using an external potential. (c) Definition of minimal irreducible area used for choosing the ion impact points. (d) External potential acting on the atoms of the 2D target. The MoS$_2$ is positioned at zero $z$ coordinate.

![Figure 3](image_url) Number of atoms sputtered from the supported monolayer MoS$_2$ and graphene by energetic He ions as obtained from MD simulations. Impacts of He atoms onto MoS$_2$ from above (a) and below (b). Impacts of He atoms into graphene from above (c) and below (d). Angles $\theta_0$ and $\theta_1$ are defined in Figure 1.
probability, energy, and angle distributions). Simulation runs are performed for normal ion incident on a SiO\textsubscript{2} layer with a thickness of 1 \textmu m, and 5 million ions per incident energy. We performed the calculations with the TRIDYN code because it gives more accurate results with respect to the sputtering yield and the angular and energy distribution of the recoils, as compared to the more popular SRIM code.\textsuperscript{50} The dependence of the sputtering yield on the surface binding energy was carefully tested, see Figure S2. A value of $E_{\text{surf,oxygen}} = 2.0$ eV is picked, which is the tabulated value for SiO\textsubscript{2} in SRIM\textsuperscript{50} and known to produce results in accordance with experimental findings.

The MD and MC approaches were combined by treating the impacts of the projectiles on the 2D target material in detail with MD (accessing small length scales), whereas the projectile statistics was sampled according to the results of the MC calculations (covering larger length scales).

\section*{RESULTS AND DISCUSSION}

\textbf{He Ion Irradiation.} The results for He ions are presented in Figure 3. It is evident that the number of sputtered S and C atoms increases with ion energy and then quickly drops because of a decrease in the cross section to displace an atom from the 2D target. This is in agreement with the results of previous calculations.\textsuperscript{28,29} The atoms sputtered from the substrate are more abundant than backscattered ions for typical HIM energies. Detailed statistics data of projectile characteristics obtained from the MC data, such as energy and angle probability distributions, can be found in the supplementary material. The number of sputtered atoms also depends on the incidence angle, with the maximum in defect production being shifted toward higher ion energies for off-normal incidence.

We performed similar MD calculations for the impacts of O and Si projectiles onto MoS\textsubscript{2} and graphene sheets. Only impacts from “below” were considered. The results are presented in Figure 4. The production of defects shows similar trends as for He ions, but the number of sputtered atoms is considerably higher because of larger atomic masses of O and Si atoms as compared to He.

On the basis of the calculated number of atoms sputtered from MoS\textsubscript{2} and graphene sheets per single impact of He, O, and Si projectiles, we evaluated the average number of defects produced in the system by He ion irradiation. To match the usual experimental geometry, normal incidence of He ions was assumed. Using the MC approach, we obtained the number of backscattered He ions as well as sputtered O and Si atoms from the substrate as functions of initial energy of He ions. Using MD simulations, we also assessed the number of ions which have passed through the sheet, which is different from unity at energies below 1 keV. The results for MoS\textsubscript{2} sheet are shown in Figure 5.

By combining the MD and MC data, the average number of sputtered target atoms $\langle N_{\text{total}} \rangle$ (Mo and S for MoS\textsubscript{2} and C for graphene) can now be calculated as

$$\langle N_{\text{total}} \rangle = \langle N_{\text{direct}}(\text{He}) \rangle + \langle N_{\text{BS}}(\text{He}) \rangle + \langle N_{\text{SP}}(\text{Si, O}) \rangle$$

Figure 4. Number of atoms sputtered from the supported monolayer MoS\textsubscript{2} and graphene by O and Si projectiles hitting the sheets from below as obtained from MD simulations. Impacts of O (a) and Si (b) projectiles onto MoS\textsubscript{2}. Impacts of O (c) and Si (d) projectiles onto graphene.
where \( \langle N_{\text{direct}}(\text{He}) \rangle \) is the average number of directly sputtered atoms per He ion and \( \langle N_{\text{BS}}(\text{He}) \rangle \) and \( \langle N_{\text{SP}}(\text{Si}, \text{O}) \rangle \) are the contributions from the backscattered ions and sputtered substrate atoms, respectively. The average number of created S-vacancies from direct impacts is given by \( \langle N_{\text{direct}} \rangle = \langle N_{\text{above}}(E_0, \theta_i) = 0 \rangle \), where \( \langle N_{\text{above}} \rangle \) is derived from MD simulations with an impacting ion starting in the upper half plane with the velocity pointing toward the substrate (see the blue trajectory in Figure 1). The average number of S-vacancies created by backscattered ions \( \langle N_{\text{BS}}(E_0) \rangle \) in turn is estimated by averaging the overall possible trajectories (defined by energies and angles \( E_y, \theta_i \)) of the backscattered ions. This is realized by performing the probability distribution-weighted integration of the average number of defects \( \langle N_{\text{below}}(E_i, \theta_i) \rangle \) (see green trajectory in Figure 1) over the energies and angles of the backscattered particles. The integral is further multiplied by the probability \( P_{\text{BS}}(E_0) \) for the ion to be backscattered and hit the 2D target again and the transmission probability \( T(E_0) \) to obtain the average number of sputtered S atoms

\[
\langle N_{\text{BS}}(E_0) \rangle = T(E_0) \cdot P_{\text{BS}}(E_0) \cdot \int dE p_{\text{BS}}(E_1) \langle N_{\text{below}}(E_1, \theta_1) \rangle
\]

where \( p_{\text{BS}}(\theta_1) \) and \( p_{\text{BS}}(E_1) \) are the angular and energy probability distributions of the backscattered ions, respectively. The latter depends on the incident ion energy \( E_i \) whereas the former is universal.

A similar expression can be used to evaluate the effects of sputtered atoms \( \langle N_{\text{SP}}(E_0) \rangle \) on vacancy production with the backscattering probability replaced by the average number of sputtered atoms and summing over the contributions from Si and O atoms (gray and red trajectories in Figure 1).

The results obtained with the combined MD and MC approaches are presented in Figure 6. The data for damage production by direct impacts and the data for the freestanding materials are obtained directly from MD simulations carried out for the same setup. The results for freestanding systems agree well with the previously published data.

Figure 6 illustrates the defect production in freestanding and supported graphene. It is evident that as in MoS\(_2\), the substrate impedes the production of defects at very low energies, but the effect is stronger than that in MoS\(_2\), as graphene has only one layer of atoms. At high ion energies, more defects are produced in the supported system with a substantial contribution from backscattered ions. The bird’s-
eye view on the defect production mechanisms of He-irradiated MoS2 reveals that the substrate has a dramatic influence on the atom-sputtering rate. For MoS2, the sputtering rate is approximately 5 times larger than without the substrate in the HIM energy interval. A similar behavior is observed for graphene. Such an increase in the damage rate was also found experimentally for He ion bombardment of graphene supported by a Si/SiO2 substrate.33

**Ne Ion and Ar Ion Irradiation.** Similar to the modeling of He ion irradiation, we carried out simulations of the impacts of heavier Ne and Ar ions onto a supported MoS2 sheet. We found that the number of backscattered ions decreases dramatically as compared to helium, whereas the average number of sputtered substrate atoms increases, see Figure 7. In particular, the probability for Ne ions to be backscattered from the SiO2 substrate is less than 1% for all the energies and is zero for Ar. Both observations can be understood within binary collision approximation considering the projectile-to-target mass ratio, as backscattering is more likely for lighter ions, and the momentum transfer to secondary projectiles is more efficient for a mass ratio close to unity ($m_{\text{Ne}}/m_{\text{Si}} = 0.71$, and $m_{\text{Ar}}/m_{\text{Si}} = 1.42$). As expected, for an increasing ion mass, the maximum of the sputtering yield shifts to higher energies.

**Figure 8** illustrates the effects of the substrate on the defect production for MoS2 and graphene. The results for freestanding monolayers are also presented for the sake of comparison. Following from the data on the abundance of the available projectiles, Figure 7, the contribution to the damage in the 2D target from

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**Figure 7.** Transmitted and backscattered ions along with the sputtered substrate atoms as functions of ion energies for Ne (a) and Ar (b). Note that the average number of backscattered ions for Ne is scaled by a factor of 50.

**Figure 8.** Average number of sputtered atoms from MoS2 (left) and graphene (right panels) for Ne (a–c) and Ar (d–f) impacts. The corresponding numbers for freestanding monolayers (grey) are shown for comparison.
backscattered particles is negligible, and defect production process is governed by direct impacts of ions and by atoms sputtered from the substrate.

A salient feature is the suppression of defect production at low ion energies, as the substrate "stops", the atoms sputtered away from the target and increases the probability for immediate recombination of the vacancy–interstitial pair. It is also evident from Figure 8 that although the total sputtering yield increases because of the higher ion mass, the enhancement in the indirect sputtering of target atoms is less pronounced than the boost in direct sputtering. This means that for heavier ions, the effect of the substrate on defect production is smaller. Nevertheless, as panels (a−c) in Figure 8—illustrating sputtering of S, Mo, and C atoms by Ne—demonstrate, the combined effect of direct and indirect sputtering is twice as large as the sputtering yield without the substrate in the energy range relevant for HIM (from 1 to 30 keV). This indicates that for neon in the HIM, the substrate still plays an important role, whereas the processes which enhance or diminish sputtering compensate each other in the case of argon irradiation, as panels (d−f) in Figure 8 illustrate. For the latter, a substantial increase in the sputtering yield because of the presence of a substrate cannot be observed.

Spatial Distribution of Defects. The substrate not only influences the number of defects produced under ion bombardment but also has an important impact on the spatial distribution of defects. From the application point of view, the spatial extension of the defect region in supported 2D materials is of paramount importance, as it determines the resolution of the HIM during patterning of 2D materials. Our combined MC/MD simulations provide direct access to the spatial distribution of defects with respect to the impact point of the ion. Although the impinging ions produce defects only in the immediate vicinity of the impact point (1 nm range; see the inset in panel (c) of Figure 9), the backscattered ions and atoms sputtered from the substrate give rise to the production of defects in a wider region, with the average extension of the region being dependent on initial energy of He (or Ne) ions.

For both He and Ne ions with energies below 1 keV, almost all sulfur vacancies are produced within an 8 nm radius from the impact point. However, for typical HIM energies, the defective region is more extended: up to two-thirds of the defects are produced outside the 8 nm region, as is evident from Figure 9, panels (a) and (b). Although the defect density decays rapidly with an increasing distance R from the impact point, defects can be created at typical HIM energies even outside the 10 nm range. As compared to the freestanding material, where defects are expected only in the close vicinity of the impact point, this result agrees well with the previous experimental observations and indicates that backscattered ions and sputtered substrate atoms are the main reason for resolution limitations in patterning of supported 2D materials.9,37,39 It is interesting to note that spatial extend of the damage also depends on ion mass. As evident from our results (see Figure 9), higher energies (e.g., 30 keV) would give a better spatial resolution for He, whereas it is the other way around for Ne. For the latter, the lowest energies still attainable in HIM (e.g., 5 keV) are preferable.

Comparison to Experimental Results. To validate our approach, we analyzed the available experimental data on the amount of damage produced in the supported graphene and MoS2 under ion irradiation. It should be pointed out that the direct comparison of the theoretical and experimental defect densities is not straightforward. On the one hand, in situ annealing of defects even at room temperature is possible, and on the other hand, at high irradiation doses, more defects can
be produced, when defects already exist in the area where the ion hits the sample. Neither of these effects can easily be accounted for in simulations, but one can assume that they cancel each other to the first order. The accurate determination of defect concentrations in the experiments from Raman spectra or energy-dispersive X-ray spectroscopy data is also a challenge.

Irradiation of freestanding MoS$_2$ flakes by 30 keV He ions gave rise to the loss of about 50% of the S atoms at a dose of $10^{18}$ He$^+)/cm$², whereas an order of magnitude smaller dose was required for the sample on a substrate. ¹³ Our results (Figure 6) indicate that $Y \approx 0.008$ S atoms are on average sputtered away by a 30 keV He ion. A dose of $10^{17}$ He$^+)/cm$² corresponds to $N_\text{S} \approx 90$ ions hitting the primitive cell area. Correspondingly, the relative number of atoms which should still remain in the system is $(2 - Y \cdot N_\text{S})/2 \approx 0.6$, which is in very good agreement with the experimental data.

As for He ion irradiation of graphene in HIM, our simulations indicated that the number of defects should be larger in the supported samples (as compared to freestanding) by a factor of about 3, which qualitatively agrees with the experimental ratio ¹⁵ for doses below $10^{16}$ He$^+)/cm$². Our results for 500 eV Ar irradiation on MoS$_2$ were also in the order-of-magnitude agreement with the dose (about $5 \times 10^{18}$ Ar$^+)/cm$²) required to sputter ca. 25% of the S atoms.⁺¹⁷ The agreement with the published experimental values for higher Ar energies is even more striking: although for bilayer graphene on SiO$_2$ with the published experimental values for higher Ar energies is freestanding material, which is deposited on top of a trench in the substrate or on a TEM grid. For helium and neon ions with a wide range of initial ion energies including those used in HIM, we showed that the energy defect production in 2D systems can be dominated by backscattered ions and sputtered substrate atoms rather than by the direct ion impacts. The last statement is especially true for light ions. In particular, this is the case for 30 keV He ions, most widely used for imaging and patterning of 2D targets using HIM. We also studied the factors which limit the spatial resolution of the patterning process. Our results provide microscopic insights into defect production mechanism, along with the quantitative information, which can be used for the minimization of damage during imaging or optimization of the patterning process.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.8b08471.

Energy and angular distribution from MC (TRIDYN) simulations, influence of surface binding energy on the sputtering yield, and influence of substrate thickness on the sputtering yield (PDF)

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#### Notes

The authors declare no competing financial interest.

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#### ADDITIONAL NOTE

“The charge state of the projectile cannot be accounted for in the classical MD simulations, and its value at each moment of time is generally unknown. Therefore, just to differentiate between the target/substrate atoms and primary projectiles, we refer to the former as “atoms” and the latter as “ions”.

#### REFERENCES

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