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Ion impact energy distribution and sputtering of Si and Ge

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The spatial distribution of ion deposited energy is often assumed to linearly relate to the local ion-induced sputtering of atoms from a solid surface. This—along with the assumption of an ellipsoidal region of energy deposition—is the central mechanism used in the Bradley and Harper [J. Vac. Sci. Technol. A 6, 2390 (1988)] explanation of ion-induced surface instabilities, but it has never been assessed directly. To do this, we use molecular dynamics to compute the actual distribution of deposited energy and relate this to the source of sputtered atoms for a range of ion energies (250 eV and 1500 eV), ion species (Ar, Kr, Xe, and Rn), targets (Si and Ge), and incidence angles (0°, 10°, 20°, 30°, 40°, 50°, 60°, 70°, and 80°). It is found that the energy deposition profile is remarkably ellipsoidal but that the relation between local deposited energy and local sputtering is not simple. It depends significantly upon the incidence angle, and the relation between energy and local sputter yield is nonlinear, though with a nearly uniform power-law relation. These results will affect, in particular, surface instability models based upon simpler approximations.

I. INTRODUCTION

Ion bombardment is widely used for implanting atomistic impurities into electronic devices, profiling depths, chemical etching, analyzing composition of solids surfaces, and preparing substrates for various other applications. 1–3 During this process, the incident ion transfers its kinetic energy to the target atoms, causing redistribution of atoms surrounding the point of impact and sputtering or removal of materials from the target. One of the key unknowns is how the deposited ion energy is related to local sputtering of the target material, which can play a critical role in interpreting ion bombardment experiments as well as in theoretical explanations for the evolution of surface patterns. 4–7 An assumed ellipsoidal energy deposition and a hypothesized linear relation to sputtering are the basis of Sigmund’s 4,5 model, which leads to curvature dependent sputtering and within Bradley and Harper’s stability analysis predicts the formation of a rich range of nanometer-scale surface morphologies. 6–8, 10–11

These models are consistent with several erosion related surface behaviors. However, they fail to explain particular observations, including the formation of ripples at lower angles as observed during low-energy ion bombardment. 3,12–14 Models based on simplified descriptions of binding forces 15 and amorphous targets 16 suggest that the energy deposition distribution differs from Sigmund’s ellipsoidal assumption. It has also been argued that the key deficiency of the Bradley-Harper theory, in explaining surface morphologies, is the assumed ellipsoidal shape of energy deposition. 15,16 However, the actual shape is unknown and it is unclear how it might be measured experimentally. Here, to directly quantify the energy deposition and its relation to sputtering, we perform molecular dynamics (MD) simulations of 250 eV and 1500 eV impact energies of four ion species (Ar, Kr, Xe, and Rn) on Si and Ge targets at nine impact angles (0°, 10°, 20°, 30°, 40°, 50°, 60°, 70°, and 80°). Averaging over many simulations shows that energy distributions are indeed approximately elliptical in shape and Gaussian in profile, but the relation to sputtering is more complex.

II. SIMULATION METHODOLOGY

The MD simulations are carried out using the Stillinger-Weber potential 18 to model Si-Si and Ge-Ge interactions, and a Moliere potential for Ar-Si or Ar-Ge interactions. To model a surface that has already been repeatedly bombarded, initial amorphous targets are prepared by 200 normal incidence impacts in random locations over the entire periodic surface (13.0 × 13.0 nm² for Si and 13.6 × 13.6 nm² for Ge). Only 1.2 Si atoms and 1.5 Ge atoms per impact on average are sputtered in this preparation stage. Extensive analysis of targets prepared in this manner has been reported elsewhere. 19–21 The bottom layer of the simulation cell is held fixed, which is deemed acceptable since ions never penetrate to the full depth (10.9 nm for Si; 11.3 for Ge) of this target. The ions are initially positioned 0.7 nm above the surface with the velocity for the impact to be studied. Newton’s equation of motion is then integrated for 1.0 ps, using the velocity-Verlet algorithm. Sputtered atoms are identified by lack of interaction with neighbors and are prohibited from returning to the target. For each set of impact parameters, the average response of the target is computed by averaging over 500 such impacts. To generate a continuous representation of the deposited energy, the target is discretized into a regular mesh with 2.0 Å × 2.0 Å × 2.0 Å cells. The change in energy (kinetic energy plus potential) within each cell is determined by computing the ensemble averaged change in energy within a cube (surrounding the mesh point) with lengths of half of the mesh size along each direction. In this
accounting, potential energies are divided equally amongst participating atoms. The change in energy represents the change in energy before and after an impact.

III. RESULTS AND DISCUSSIONS

A. Time history of sputtering and energy deposition

To examine how the distribution of energy evolves over time, the deposited energy is computed at a series of times between 0.1 ps and 0.9 ps after the first ion-target interaction, as shown in Fig. 1(a). During this period of interaction, the core of the deposited energy that involves a few highly energetic atoms transfers energy to the surrounding atoms and becomes cooler. However, the number of atoms increases with the cubic power of the distance from the deposited energy center. As the atomic interaction progresses, the higher energy from the fewer core atoms is distributed over a large number of surrounding atoms causing a much smaller change in their energy, as shown in Fig. 1(b). Thus, there is no substantial difference in the normalized energy profile, as shown in Fig. 1(c), indicating that the shape of the energy profile is formed within a very short time (0.1 ps).

However, sputtering starts at approximately 0.06 ps and finishes within 0.8 ps of the initial ion-surface interaction, as shown in Fig. 2. Also, the phase space history of the sputtered atoms indicates that the kinetic energy of most sputtered atoms reaches a maximum within 0.2 ps, as shown in Fig. 2, while a few sputtered atoms attain their maximum kinetic energy within 0.2 to 0.6 ps of ion-surface interaction. Thus, most of sputtering takes place after the energy deposition takes its shape. As a result, the relation between sputtering and energy deposition is time dependent.

B. Profiles of deposited energy

According to Sigmund’s model,4,5 sputtering from the point $r$ is proportional to the energy deposited at that point $E_{dep}(r)$. The deposited energy profile along the projected beam direction at a distance of 3 nm below the surface at 0.1 ps and 0.9 ps at normal incidence. The deposited energy at the center of the energy profile is higher at 0.1 ps than at 0.9 ps. Conversely, it is lower in the far field from the center indicating the dissipation of energy from the core of the deposited energy towards its neighbors. Profiles of the normalized energy on the surface along the projected beam direction at 0.10, 0.15, 0.25, 0.4, 0.6, and 0.9 ps of ion interactions.

and this energy is deposited in an ellipsoidal region, as shown in Fig. 3, with a Gaussian distribution in the planes parallel and perpendicular to the beam direction. The cross-sections of the energy deposition in the plane of the initial ion trajectory are shown in Fig. 4 for Kr bombardment of Si at eight different angles of incidence. The energy deposition profiles shown in the figures are evaluated at 1.0 ps of ion-target interaction. The deposited energy profiles for off-normal incidence vary in dimensions along the longitudinal and lateral directions of the ion impact. For glancing angles, the peak energy deposition depth ($a \cos \theta$ in Fig. 4) is close to zero, which might be expected to cause substantial sputtering. For a particular energy-ion pairing, the dependence upon angle appears to be primarily a rotation of the ellipsoid.

However, the basic shape of the energy deposition contours changes as the material or ion type is changed, as shown in Fig. 5. Heavier ions (such as Rn) penetrate deeper into the target, making the axial dimension of the energy profile larger than its lateral dimension. For the same ion energy and ion species, the axial dimension is smaller in the denser Ge target compared to Si. Also, the cross-sectional area of

![Figure 1](https://example.com/figure1.png)

**FIG. 1.** (a) Deposited energy profile for normal incidence 1500 eV Kr impact of Si at 0.10, 0.15, 0.25, and 0.9 ps of ion interactions. (b) Deposited energy profile along the projected beam direction at a distance of 3 nm below the surface at 0.1 ps and 0.9 ps at normal incidence. The deposited energy at the center of the energy profile is higher at 0.1 ps than at 0.9 ps. Conversely, it is lower in the far field from the center indicating the dissipation of energy from the core of the deposited energy towards its neighbors. (c) Profiles of the normalized energy on the surface along the projected beam direction at 0.10, 0.15, 0.25, 0.4, 0.6, and 0.9 ps of ion interactions.

![Figure 2](https://example.com/figure2.png)

**FIG. 2.** Sputter history for bombardment with 1500 eV Xe for Si target. The primary axis shows the number of sputtered atoms reaching their maximum kinetic energy at a particular time. The secondary axis shows sputter yield as a function of time indicating the fraction of sputtered atoms at a particular time.

![Figure 3](https://example.com/figure3.png)

**FIG. 3.** Geometric parameters of an ellipsoidal energy deposition for an off-normal beam angle $\theta$. O is the point of impact, $O'$ is the point of peak energy deposition, and T is surface point with maximum surface deposited energy. The parameters characterizing the profile are the longitudinal extent of the energy profile ($x = 2x_2$), the distance between the point of impact and the maximum of deposited energy ($a$), the lateral extent of the deposited energy profile ($2 = \beta \alpha + \beta_2$), the distance between the point of impact and the maximum of the deposited energy on the surface ($\delta$), and the depth of the peak energy.4

![Figure 4](https://example.com/figure4.png)

**FIG. 4.** Geometric parameters of an ellipsoidal energy deposition for an off-normal beam angle $\theta$. O is the point of impact, $O'$ is the point of peak energy deposition, and T is surface point with maximum surface deposited energy. The parameters characterizing the profile are the longitudinal extent of the energy profile ($x = 2x_2$), the distance between the point of impact and the maximum of deposited energy ($a$), the lateral extent of the deposited energy profile ($2 = \beta \alpha + \beta_2$), the distance between the point of impact and the maximum of the deposited energy on the surface ($\delta$), and the depth of the peak energy.4
FIG. 4. Energy distribution at 1.0 ps for 1500 eV Kr bombardment of Si at \( \theta = 10^\circ, 20^\circ, 30^\circ, 40^\circ, 50^\circ, 60^\circ, 70^\circ, \) and \( 80^\circ \) angles of incidence. Solid-line contours show energy levels 0.75 eV, 0.5 eV, and 0.25 eV, though the 0.75 eV level is not present in \( 80^\circ \). The ion direction is indicated by the black arrow, with its tip showing the point of impact.

The deposited energy at the surface in Ge is higher, irrespective of ion species considered. The width of the energy profiles based on \( \beta \) is 2.5%–6.3% higher for Ge targets. At 1500 eV impact energy, the longitudinal extent of the deposited energy—that is, the parameter \( \alpha \) in Sigmund’s notation—increases in both Si and Ge with heavier ions (11% for Xe impact on Si, 40% for Rn impact on Si, 10% for Xe impact on Ge, and 22% for Rn impact on Ge).

The energy distribution due to 1500 eV impacts, shown in Fig. 5(a), can be compared to those for 250 eV ions shown in Fig. 5(b). The basic shape is insensitive to the energy difference, though the energy contours are expected to be smaller and the peak energy is closer to the surface. The peak energy deposition depth, \( a \cos \theta \), is 1.2 to 2.5 times smaller at this lower impact energy, depending upon the case, with larger variation for lighter ions. This can be seen particularly for Ar impact on Si and Ge at 250 eV or 1500 eV. As might be expected since \( M_{Ge} = 2.66M_{Si} \), the penetration depth in Ge (atomic mass 72.6) is smaller compared to that in Si (28.1) in all cases. The shapes in Si for different ion species Ar (39.9), Kr (83.8), and Xe (131.3) vary, particularly for 1500 eV, from ellipsoidal (for Rn on Si) to tear-drop (for Ar on Si) as the mass of the incident particle increases. On the other hand, in Ge, energy distributions are more spherical.

These observations have some immediate consequences for the application of Bradley-Harper theory to surface patterning. For example, on a Ge target at 250 eV, the longitudinal and lateral dimensions of the energy profiles are comparable (\( \alpha = 1.2 \) nm and \( \beta = 1.0 \) nm), the parameter “\( a \)” is zero, and the peak of the deposited energy lies on the surface. Consequently, BH’s angle dependent sputtering coefficients are \( \Gamma_s(\theta) = (1/2)(3\cos^2\theta - 1) \) and \( \Gamma_\ell(\theta) = (1/2)(\theta^2 - 1) \). With \( \Gamma_s(\theta) > \Gamma_\ell(\theta) \) for all \( \theta \), the BH criterion rules out the possibility of observing parallel ripples. However, rippling is observed in experiments for bombardment in Ge at 250 eV.22 This suggests that although Sigmund’s energy distribution is indeed a good model for our MD results, the BH’s erosion based criterion seems to fail to fully characterize surface instabilities.

C. Relation between sputtering and surface-deposited-energy

The local “surface deposited energy” (the energy deposited on the surface, where most of the sputtering takes place) and local sputtering profiles, for all impact conditions considered, have similar shape and angle dependence, as shown in Fig. 6. Notably, the surface deposited energy and sputter-yield profiles decay at different rates from their corresponding maximum values, which indicate a spatial dependence of their relation and disagree with Sigmund’s linear prediction. However, the Sigmund model agrees with there being more sputtering downstream than upstream with respect to the projected ion direction, especially for higher angles of incidence, confirming the notion that sputtering from an inclined surface will make it steeper. Nonetheless, local sputtering does not vary linearly with local energy deposition on the surface. The deposited energy on the surface increases with higher incidence angles, which is in accord with more
sputtering for glancing-angle bombardment. Nonetheless, at the highest incidence angles, such as 80° in the present work, surface energy deposition drops significantly compared to that near normal incidence, with a concomitant decrease in sputter yield \(^4\). To determine the dependence of sputtering on surface deposited energy, we normalize computed local sputter yield \(S(x, y)\), surface deposited energy \(E(x, y)\), and number density of atoms on the surface \((x, y)\) by their maximum values, \(S_m\), \(E_m\), and \(N_m\); the results are shown in Fig. 7(b). (It should be noted that existing sputtering theories \(^4\) do not include the effect of ion-impact induced number density variation in describing local sputtering.) The maximum for \(S\), \(E\), and \(N\) occur the same point in the downstream direction of the point of impact. To establish relations between \(E(x,y)/E_m\) and \(S(x,y)/S_m\) and \(N(x,y)/N_m\) and \(S(x,y)/S_m\), we seek the values of \(p\) and \(q\) for which

\[
\sum \frac{[E(x,y)]^p}{E_m} = \sum \frac{S_i(x,y)}{S_m}, \quad (1)
\]

\[
\sum \frac{(N_i(x,y) - N(x,\infty, \infty))}{N_m(x,y) - N(x,\infty, \infty)} = \sum \frac{S_i(x,y)}{S_m}, \quad (2)
\]

where \(E_i(x,y), S_i(x,y),\) and \(N_i(x,y)\) are surface deposited energy, sputter yield, and number density, respectively, at the mesh points on the surface \((x, y)\); and \(N(x, \infty, \infty)\) is the number density far from the point of impact. The number density on the surface is determined by dividing the number of atoms in any columnar region (as defined for computing local sputter yield or energy deposition) by the volume of the columnar region. The height of the topmost atom within each columnar region is taken as the height of that columnar region to compute its volume. This procedure for meshing and computing variations in the number of atoms is explained fully in Refs. 19 and 21.

The relation between sputtering and surface energy deposition—that is, the exponent \(p\)—is computed at seven different times to determine the time evolution of the exponent. The exponent \(p\) initially increases with time and then reaches a steady state value of 2.7 at around \(t = 0.25\) ps, as shown in Fig. 7(b). The exponent \(q\), on the other hand, reaches a steady state value of 1.58 at \(t = 0.20\) ps. Sigmund’s conjecture that \(p = 1\) holds at \(t = 0.08\) ps, which is 0.9 – 0.08 = 0.82 ps earlier than the time required for the sputtering process to finish. Therefore, Sigmund’s assumption of the linear energy dependence of local sputter yield—which is used widely to investigate sputtering related mechanisms—is inaccurate for most of the sputtering process.

The difference in decay rates of normalized sputtering and surface energy deposition, as shown in Fig. 7(a), indicates that sputtering from locations away from the point of impact—say, at \((X-x_0) = 1\) nm—is less than what Sigmund’s linear theory would predict. This reduction in sputter-yield can be attributed to the spatial variation in normalized number density of the atoms on the surface. The deposition of energy causes variation in number density near the point of impact. The maximum number density is although within only 0.5% of the far field or bulk number density, the relative measure of the spatial variations, \((N - N_{\infty}))/N_{\infty}\) shows that the normalized quantity is related to normalized sputtering in a way similar to the relation between normalized sputtering and normalized surface energy deposition.

Combining Eqs. (1) and (2), the normalized surface deposition energy, number density, and sputter yield results obtained for 1500 eV Kr ion bombardment of Si at normal incidence yields

\[
\sum \frac{E_i(x,y)}{E_m} \times \left( \frac{N_i(x,y) - N_{\infty}}{N_m(x,y) - N_{\infty}} \right) = \sum \frac{S_i(x,y)}{S_m} \times 1.00328 \approx \sum \frac{S_i(x,y)}{S_m}, \quad (3)
\]

where the exponent 1.00328 resulted from the summation of inverses of the exponents, \(p = 2.7\) and \(q = 1.58\). The values of \(p\) and \(q\) are computed for three ion species, two energies, two targets, and nine angles of incidence. The resulting ensemble (comprising 108 cases) shows no correlation between either of the exponents and the impact condition (ion-target-energy combination). However, the angle dependent variation for the exponent \(p\), for all ion-target-energy combinations considered in this work, is well fitted by \(p(\theta) = 2.7 + 0.5 \sin \theta\); and \(q(\theta)\) lies within 1.58 to 1.5 with no particular trend in its variation. The sum of their inverse falls within 1.007 to 0.975, which can be approximated to be equal to unity

\[
\frac{1}{p} + \frac{1}{q} \approx 1. \quad (4)
\]
For higher angles, sputter yield is more sensitive to incidence angle than the surface deposited energy, as shown in Fig. 8(b). A comparison of the surface deposited energy and sputter yields for off-normal angles, normalized by their values at normal incidence, indicates that the angular variation of sputter yield is higher than that of the surface deposited energy. For example, sputter yield at 70° is six times the sputter yield at normal incidence, whereas the deposited energy at 70° is only twice the deposited energy at normal incidence. This increase in surface energy deposition at higher angles is because the core of the total deposited energy in off-normal incidence lies closer to the surface and the energy deposition area on the surface is higher than that in normal incidence. Moreover, the percentage of ion energy deposited in the target (which is computed by subtracting the energy of the reflected ions and sputtered atoms from the initial ion energy) decreases with an increase of the angle of incidence, as shown in Fig. 9.

A negligible amount of energy is taken up by the sputtered atoms. Also, a significant amount of ions are reflected at higher angles, as shown in Fig. 10, and the ions that are reflected from the target carry a significant amount of their initial kinetic energy with them, especially at higher angles. Nonetheless, the difference in angle dependent variations of sputtering and surface energy deposition is mainly compensated by the angular variation of the normalized atomic number density. This suggests that number density, which has been neglected in existing local sputtering theories, as well as surface energy deposition both influence sputtering in an intricate fashion.

IV. SUMMARY

From a macroscopic point of view, it has been shown that total sputtering from a target depends on atomic masses of projectile and target atoms, number density and binding energy of the target material, and angle of incidence. Here, MD results show that, at the atomic scale, sputtering depends on number density and deposited energy, for a particular ion-target combination where atomic mass and binding energy are fixed. For the theories of surface instabilities which are based on the linear relation between energy and sputtering, it is important to include the effects of both number density and deposited energy on sputtering or nonlinear variation in sputtering with respect to deposited energy, where Eq. (4) together with Eqs. (1) and (2) can provide a guideline for incorporating their combined effects.

To conclude, through the first comprehensive atomistic studies, it is shown that the shape of the deposited energy depends on ion species, target material, and angle of incidence. The deposited energy as well as the local sputter yield dependence on the beam direction can be fitted well by Gaussian functions, as originally predicted by Sigmund. Therefore, the limitations of Bradley-Harper theory in predicting surface morphology can be argued not to arise from Sigmund’s ellipsoidal energy deposition. However, the assumptions involving the linear relation between sputter yield and deposited energy are not supported by MD simulation results. For a range of impact conditions, it is shown that the relation between sputtering and surface deposited energy is nonlinear and time dependent. Additionally, both number density as well as surface deposited energy affect local sputtering and follow a simple relation.

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