Observation of ripples under different angles

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The off-normal ion irradiation of semiconductors may induce nanopatterning effects. At ultra-low energy, where the sputtering is negligible, a surface reorganization has been shown to occur under certain circumstances. A new method is applied to test the formation of ripples at different angles, and shown to be in good agreement with experiments.

I. INTRODUCTION

Surface self-organization takes place similarly in different systems and at different length and time scales. This effect e.g. in sand, can develop in ripple formation aligned parallel or perpendicular to the wind direction. The mechanisms behind it was explained by Bagnold, observing that the grains move following the reptation and saltation movements.

The ion-beam induced nanopatterning effect was observed the first time by Cunningham in the 1960’s in Au at 70°. Since then, many theories have been developed in order to explain the formation, focusing on erosion, redistribution and stress build-up. The structures observed in experiments range from ordered nanodots in GaSb and in Si (with sample rotation) to parallel and perpendicular to the ion-beam projection ripples.

Computational methods like Molecular Dynamics (MD) or Binary Collision Approximation (BCA) have been used extensively in order to study this interesting effect. The application of these methods has shed light to the field. Norris et al. introduced the crater function formalism to predict the pattern-formation and calculate the wavelength of such structures. This model was successfully applied to the single-impact simulations, showing that the short induced displacements captured by MD and not by BCA, are crucial to predict correctly the nano-pattern wavelength that can be compared with the experimental results.

A new method of sequential impacts in MD was recently applied to study the reasons behind the surface modification under different energies and angles over different materials. The results show that the contribution of the erosion in counter-position to redistribution, is strongly dependent on the material and the ion energy. These works lead to a different type of methodology to approach the surface modification. Fridlund et al. developed an speed-up method which permits to reach high fluences, and this was applied to the particular case of the nano-patterning effect, in order to provide a direct view of the surface modification. The results were successfully shown to predict the rippiling effect and, at low energy, the different steps until the final creation of the ripples.

In this work, we go further in the application of the model presented in Ref. using it for different angles, in order to predict the surface behavior and test the accuracy of this model. Besides, we test this model against experiments. To do that we need to show that no pattern formation is created below the critical angle (shown to be 55° off-normal) and there is rotation of ripples at grazing incidence.

The different analysis and the follow-up of the evolution of the cell permit to get conclusions on the pattern formation within a certain range of angles.

II. METHODS

A flat-surface cell of amorphous Si containing 73584 atoms was relaxed to 300 K in order to perform the sequential bombardment. The final size of the cell is 16.56 x 16.56 x 5.15 nm³.

The simulations were performed using the PARCAS MD code. The Si-Si interactions are described using the environment-dependent inter-atomic potential (EDIP), complemented at short distances by the purely repulsive ZBL potential. The same type of repulsive potential describes the Ar-Si interaction. The bombardment of the sample is done sequentially, so we used the pair potential with high energy repulsive part from DFT DMol calculations smoothly joined to the LJ equilibrium part at larger distances to described the Ar-Ar interactions.

The irradiation of the sample is done using the speed-up scheme, which allows us to reach high fluences using MD. The 30 eV-Ar atom is located over the surface in a way that, depending on the irradiation angle (θ = {0°, 45°, 85°}), always impacts in the surface in the center of the surface, far from the border to avoid the interactions with periodic boundaries. A fixed azimuthal angle aligned with the x-axis is used. The actual impact point is always random because of the shifting cell prior to every impact. The numbers of impacts for the 0° and 45° cases are 54000 (fluence of 2 x 10¹⁶ ions cm⁻²), enough to reach high fluences. In the case of 85°, since the angle is close to horizontal inclination, we have simulated
up to 160000 Ar impacts (fluence of $5.9 \times 10^{16}$ ions cm$^{-2}$). Experimentally, higher fluences are needed to create an effect on the surface due to the irradiation angle being closer to grazing incidence.\textsuperscript{12}

Every nine impacts, the temperature of the system is restored to 300 K during 30 ps applying the Berendsen thermostat.\textsuperscript{27} Each individual impact lasted 1 ps and a 0.8 nm-thick at 300 K was applied in the x-y direction, leaving out the surface. As was explained in Ref. \textsuperscript{20} the 1 ps time is long enough to collect all the cumulative induced effect, being comparable to those effects observed using longer relaxations time.

We computed the effect of the erosion and redistribution counting the number of sputtered atoms in the system, and the displacement in each component, respectively. Moreover, as in Ref. \textsuperscript{20} the use of MD allows to calculate the accumulation of stress, but as was shown, there is no relation between the formation of ripples and the stress build-up at this low energy.

### III. RESULTS

#### A. Direct observation

In this part we show the evolution of the surfaces at different angles. First we describe the evolution of the surface under normal incidence irradiation,

![Figure 1. Evolution of the a-Si surface under normal incidence 30eV-Ar$^+$ at (a) 4500, (b) 18000, (c) 36000 and (d) 54000 impacts.](image)

We can observe in FIG. 1a that in the early stage of the irradiation, the roughness of the surface starts to increase, leading to the formation of several hills at random positions, as was observed at 70° in the initial stage of the simulation. However in this case as can be observed in FIG. 1b to d, this effect persists until the end of the simulation. In this case we observe no surface patterning consistent with the experimental result.\textsuperscript{12}

Secondly, we study the case at 45° off-normal incidence, and we follow the evolution of the surface,

![Figure 2. Evolution of the a-Si surface under 45° off-normal incidence 30eV-Ar$^+$ at (a) 4500, (b) 18000, (c) 36000 and (d) 54000 impacts. The ions are coming in the positive direction of the x axis.](image)

We can see in FIG. 2 the evolution of the surface under irradiation does not show a clear effect of patterning, as can be seen under 70° off-normal in previous work.\textsuperscript{20} Again, an increase of the roughness occurs, but this is not leading to a following step. We notice that in FIG. 2c and d there is some rising structures, as a product of the local reorganization, but not aligned to any preferential direction. This effect ends up in neither parallel nor perpendicular ripples. This result is consistent with the experimental data shown in Ref. \textsuperscript{14}, since the critical angle is 55°.

Finally, we observe the surface evolution under 85° off-normal incidence,

![Image](image)

We can see in FIG. 3 the evolution of the surface under irradiation does not show a clear effect of patterning, as can be seen under 70° off-normal in previous work.\textsuperscript{20} Again, an increase of the roughness occurs, but this is not leading to a following step. We notice that in FIG. 3c and d there is some rising structures, as a product of the local reorganization, but not aligned to any preferential direction. This effect ends up in neither parallel nor perpendicular ripples. This result is consistent with the experimental data shown in Ref. 14 since the critical angle is 55°.

In FIG. 5 we observe that the fluence needed to reach a notable effect is larger than in other cases, as was reported in Ref. \textsuperscript{14}. We observe how, despite the effect being much shallower, after 18000 impacts (FIG. 5b), we see that the first mounds have developed. As the fluence increases (FIGS. 5c and d), it is noticed that these mounds merge with others and some thin ripple-like structures start to appear. That surface reorganization becomes fully clear at the last stages of the simulation (FIGS. 5e and f), where the alignment of the structures in the perpendicular direction to the projected ion beam is developed. In Ref. 14 the rotation of the ripples is observed at 85°, in the same way as in this work.
FIG. 3. Evolution of the a-Si surface under 85° off-normal incidence 30eV-Ar⁺ at (a) 4500, (b) 18000, (c) 72000, (d) 117000, (e) 135000 and (f) 160200 impacts. The ions are coming in the positive direction of the x axis.

B. Analysis

We analyze the data extracted from the performed simulation in terms of the displacement and sputtered atoms.

In FIG. 4 a we can see how, as can be expected, the highest accumulation of displacements in the x direction takes place at 45° (consistent with the single-impact results shown in Ref. 14). However, that higher induced displacement is not directly converted in the formation of ripples for 45° (see FIG. 2). Moreover, the single-irradiation model from Ref. 14 was not in good agreement with the experimental measurements, since the critical angle was predicted to be 45° (experimentally \( \theta_c = 55° \)).

On the other hand, the simulations presented in this work show that there is no ripple formation, only increase of the roughness leading to a not aligned merging of structures. In this direction we see how, even both \( \delta_x \) and \( \delta_z \) (FIG. 4 b) are higher, it is not translated into a surface re-organization in ripples.

We notice that at 0°, the accumulation of displacement in the x direction is fluctuating around 0 for the entire simulation. On the other hand, \( \delta_z \) is rapidly increasing from the beginning of the simulation, reaching the highest value among all the angles.

In FIG. 4 a, the displacement accumulated in the 70° case lead to formation of ripples in the surface20. The y component is not included because in all the cases, it oscillates around zero.

Regarding the results shown in FIG. 4 on 85°, we notice that the contribution in x direction fluctuates around zero. We can see how either the accumulation of \( \delta_x \) or the sputtered atoms cannot directly explain the formation of the ripples over the surface, and, neither, the orientation of these. We have observed that the process starts with small displacements of the atoms, piling up and, after that, these structures merge and build the ripples. The
displacements in general are short over the surface \((x - y)\) plane. However, the small displacements outward the surface \((\delta z)\) seem to explain the formation of ridges. Due to the grazing incidence, the incoming ions induce shallow effects in the surface, but the high fluence enable to create these small structures aligned to the perpendicular direction. The height of these structures is low, as was tested experimentally.\(^1\)

We can also calculate the root-mean-square (RMS) roughness of the surface, according to the method used in Ref. 28 in the following way,

\[
<R_q> = \sqrt{\frac{\sum_{i=1}^{N} (z_i - \bar{z})^2}{N}},
\]

(1)

where \(z_i\) is the height of the exposed atom in the line of the surface, \(\bar{z}\) is the mean height of all the atoms in the line and \(N\) is the number of atoms in the line.

We can plot this the x and y directions at different steps of the simulations.

![FIG. 5. RMS using all the atoms above 5 Å depth from the initial surface dividing the desired direction in 30 bins: after 27000 Ar\(^+\) impacts in the (a) x and (b) y direction, and at the end of the sequential simulations under different angles in (c) x and (d) y direction.](image)

In FIG. 5a, we observe a clear formation of the surface re-organization in the case of 70°, but not in the rest of cases. On the other hand, in FIG. 5b, there is no noticeable effect. In FIG. 5c, we observe how the situations of 0° and 45° have not changed at the end of the simulation from FIG. 5a, but in the case of 70° we see how the RMS has increased along this direction. In FIG. 5d, the RMS is higher in the 70° case in some areas, but not as in the x-direction case. In all the directions and at different fluences, in the 85° case, the value is the lowest. Only at the end (FIG. 5d) we notice small variations in the y direction.

IV. DISCUSSION

We can observe at normal incidence (FIG. 1), there is no clear atomic orientation, otherwise an increase of the roughness on the surface takes place. As experimentally, also an increase of the roughness is observed, but not a further development of ripples. The momenta induced by the incoming ions mostly in the z direction, translated into the highest accumulation of displacement in the z-direction (FIG. 4 b), suppress the further formation of aligned structures.

We can see in the evolution of the surface under 45° (FIG. 2) that there is not a clear direction of pattern formation, which is in good agreement with the experimental result,\(^2\) which a critical angle of 55° was obtained. In this case, the accumulation of displacement in the x-direction (FIG. 4 a) is the highest, but there is not a clear orientation or reorganization of the structures. The formation of ripples needs of the hills formation and a local incidence angle suitable to develop the ripples oriented in a certain way. The incidence ions remove the possibility of merging-up step as was reported in Ref. 20, this is the reason of the absence of oriented ripples.

In the case of 85° (FIG. 3), we observe a clear orientation of the created ridges in the perpendicular direction of the ion beam projection. These structures start to be discernible at higher fluences, and however the displacement is higher is the x direction, the alignment of the structures is parallel to y-direction. Taking a look to the formation of any of the created structures aligned with the y-direction, we can observe how the small atomic displacements drive the formation of those structures. These irradiation-induced small displacements make the structure to develop after higher fluences than in the case of 70°, where the difference between the two steps, i.e. pile-up and merge, is more clear.\(^2\)

The formation of the perpendicular ripples is described as follows: the grazing incidence induces mostly small displacements in the regions that the final ripples are formed. Longer displacements may occur, but are not determinant to the eventual ripple formation. The sputtering plays an important role for higher energies at grazing incidence, but in this case we can assure that the role is none. The formation is also due to the radiation-driven random walk displacements introduced in Ref. 20, but, in this case, the deposition of energy in the surface is quite limited and takes more time/fluence to develop the final re-organization. Summarizing, the atoms that form the final ripples come from the vicinity of them. The incoming ions dig in the surface superficially, displacing laterally the atoms and as the fluence increase, the ripples start to be more apparent.

V. CONCLUSIONS

In conclusion, we have shown in this work that this model is suitable to describe the surface modification on
Si under different irradiation angles. This method could be applied for higher energies, defining a suitable time for the impacts and the induced cascade for the incoming ions. At higher energies, the erosion takes more importance and in order to reach that high fluences, a larger cell (in the z direction at least) and higher times would be needed to reproduce the ripple formation. However, this method is the only way to study directly the pattern formation.

The simulations are able to reproduce whether the surface modification can eventually developed in parallel, as was shown in Ref. [20] or perpendicular ripples as in the case of 85°. We also observed that at normal incidence and 45° we do not observed pattern formation.

For the first time, the rotation of ripples has been shown directly, as in the previous work [20] was shown the direct formation and propagation, in this work we set this method as a suitable one to study largely the pattern formation at different angles.

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