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T. Aoki et al. Nulc. Instr. & Meth. B 228 (2005) pp.46-50

# Molecular Dynamics Simulations of Sequential Cluster Ion Impacts

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## ABSTRACT

Molecular dynamics (MD) simulations of cluster and solid target collisions were performed in order to understand the relationship between surface deformation processes and cluster sizes. MD simulations of single impacts of clusters with various sizes showed that, when a cluster size is less than 10000 atoms, a crater-like damage could be caused at 20 keV of total energy, while no damage was observed at $Ar_{10000}$ and $Ar_{20000}$ clusters. The surface morphology change was examined by MD of sequential irradiation of Ar clusters under various conditions. When the initial roughness of the surface was about 10 Å in r.m.s. and the total incident energy was 20 keV, the surface roughness was reduced with the impact of $Ar_{13333}$ or larger cluster. The MD results give information about the final value and decay speed of surface roughness, which are required to optimize the cluster ion irradiation condition for various nano-scale modification processes.

PACS: 36.40.-c Atomic and molecular clusters, 83.10.Rs (Computer simulations of molecular and particle dynamics)

KEYWORDS: gas cluster ion beam, surface smoothing, molecular dynamics, surface morphology

## INTRODUCTION

Gas cluster ion beam technique is a promising tool for nano-scale surface modification process such as ultra smoothing, high-speed etching and thin film formation [1,2,3]. As the cluster ion beam technique spreads into industrial applications widely, the study about cluster impact process becomes more important in order to achieve precise design and higher performance. Molecular dynamics (MD) simulation of cluster impact on the surface is one of the useful methods to understand the interaction between cluster and solid surface. Various MD simulations of cluster impact have been performed and damage formation process by cluster ion impact was examined [4]. As for surface modification process, it was shown that [5], at the point of cluster impact, the surface atoms move along the lateral direction of the surface, which contributes to the reduction of hill-and-valley structure of the surface. Another group proposed the analytical formula to describe the smoothing effect during metal cluster ion deposition [6,7]. However, quantitative study, such as smoothing effects dependence on surface structure, material, cluster size, etc., is still unknown. In this paper, MD simulations of Ar clusters sequentially impacts on Si target were performed for various cluster sizes. The evolution of surface morphology was directly measured. Cluster size effect on surface smoothing process was discussed comparing with the results from the single impact processes of clusters on solid targets.

## SIMULATION MODEL

Molecular dynamics simulations of Ar clusters and Si(100) surfaces were performed. The interatomic potentials were described with Stillinger and Weber model [8] among Si atoms. For

Ar-Si and Ar-Ar interactions, only ZBL (Ziegler, Biersack, and Littmark) model [9] was used. In this study, Ar clusters with various sizes ranging from 200 to 20000 atoms were used as projectiles. The total incident energy of clusters was fixed at 20 keV, so each cluster has a different energy per atom. Si(100) substrates with the dimension of 345Å×345Å×173Å were prepared as the target. In order to study surface-smoothing mechanism by cluster impact, the target initially has a 2 dimensional sine-curved surface structure with 21.6Å and 86.4Å of amplitude and period, respectively. The MD simulation of sequential cluster impacts was performed by the iterating following process; I) Select impact point randomly from central 160Å×160Å area of the target. II) Perform MD simulation of $Ar_{2000}$ impact in 20keV for 16ps. III) Cool down the target to 300K for 4ps and remove all incident Ar and sputtered Si atoms. In this study, 20 impacts were simulated for each cluster size, which corresponds to $0.75\times10^{13}/cm^2$ impacts.

## RESULTS AND DISCUSSION

The surface morphology change by single cluster impacts was examined. Fig. 1 shows the surface profiles at 16ps after impact of clusters of various sizes. In those simulations, the initial targets had flat surfaces, but the target is largely deformed at several conditions. As shown in Fig. 1, surface deformation process strongly depends on the cluster size. When a cluster size ranging from several hundreds to several thousands of atoms impacts on a solid surface, cluster penetrates the surface and causes multiple collisions in the target. During the multiple collisions, most of kinetic energy of the cluster is transferred to the target isotropically and a crater-like structure with hemispherical hole and round rim is formed. As the cluster size increases and the incident energy per atom decreases, the cluster cannot penetrate the surface and breaks-up on the

target. Therefore, the depth of crater decreases as the cluster size increases and surface deformation does not occur at the cluster size more than 10000 atoms. In this case, the incident energy per atom is 2eV/atom, which is very similar to the binding energy between Si atoms.

Fig. 2 shows the evolution of surface morphology by sequential $Ar_{2000}$ cluster impacts at 20keV of total incident energy. As shown in Fig. 1, $Ar_{2000}$ cluster impact can cause crater-like damage with only one impact. Fig. 2 shows that, as the number of impacts increases, the initial hill and valley structure is largely disturbed and some hills merge with each other to make one larger hill. After 10 impacts, a deep hole can be found because of concentration of impact points, but it is buried by following impacts, this phenomenon means that surface-smoothing and roughening effects can balance with each other. After 20 impacts, the hill diminishes at the central point of irradiated area. This result indicates that cluster ion impacts have a great advantage in removing small tips on the surface.

On the other hand, the change of surface morphology by large $Ar_{13333}$ cluster with 1.5eV/atom is shown in Fig. 3. From the result of Fig. 1, this irradiation condition is not suggested to cause surface deformation for a completely flat surface. However, if the surface is initially rough, the surface structure can be deformed as well as with $Ar_{2000}$ cluster impact. This is because that, when the local structure of impact area is not uniform, momentum and kinetic energy transfer by multiple collisions are not isotropic but rathers concentrated to make the target atoms at hill side moved to bury the valley side [5]. Fig. 3 also shows that the formation of an extremely deep hole is not observed as shown at $Ar_{2000}$ impact because $Ar_{13333}$ carries very low energy per atom and provides very soft impact on the target.

Cluster size dependence on the surface smoothing effect was shown in Fig. 4. In Fig. 4, the surface roughness is calculated as the root mean square of the surface height within the 160$Å^2$

square of irradiated area. Fig. 4 demonstrates that the surface roughness can be increased or decreased by changing cluster size. When the cluster size is about several thousands or less, surface roughness increases as number of impacts and converges at about 20Å, This saturated surface roughness shows a good agreement with the value from a single impact shown in Fig. 1. On the other hand, when a cluster size is larger than 10000, the surface roughness decreases and a smooth surface can be obtained. Additionally, Fig. 4 indicates the speed of surface smoothing process. $Ar_{20000}$ is the largest cluster in those simulations and expected to provide the smoothest surface, but the decay of surface roughness is very slow because incident energy per atom is very low and momentum and kinetic energy transfer is not enough.

## SUMMARY

Molecular dynamics simulations were performed in order to understand the relationship between surface deformation process and cluster size. MD simulations of single impact of clusters of various sizes showed that, when incident energy per atom is more than several eV/atom, a crater-like damage is formed. The depth of crater decreases as the cluster size increases (= energy per atom decreases) and no crater formed at $Ar_{10000}$ at 2eV/atom. The MD simulation of sequential irradiation of clusters showed that the small tip structure diminished and locally smooth structure could be obtained. When the initial roughness of the surface is about 10Å in r.m.s., the roughness was reduced with the impact of $Ar_{13333}$ or larger cluster at 20keV. These results from molecular dynamics simulations give information how to design the cluster ion irradiation condition according to required surface roughness and throughput.

**ACKNOWLEDGEMENT**

This study is supported by Ministry of Economy, Trade and Industry (METI) and New Energy and Industrial Development Organization (NEDO) in Japan.

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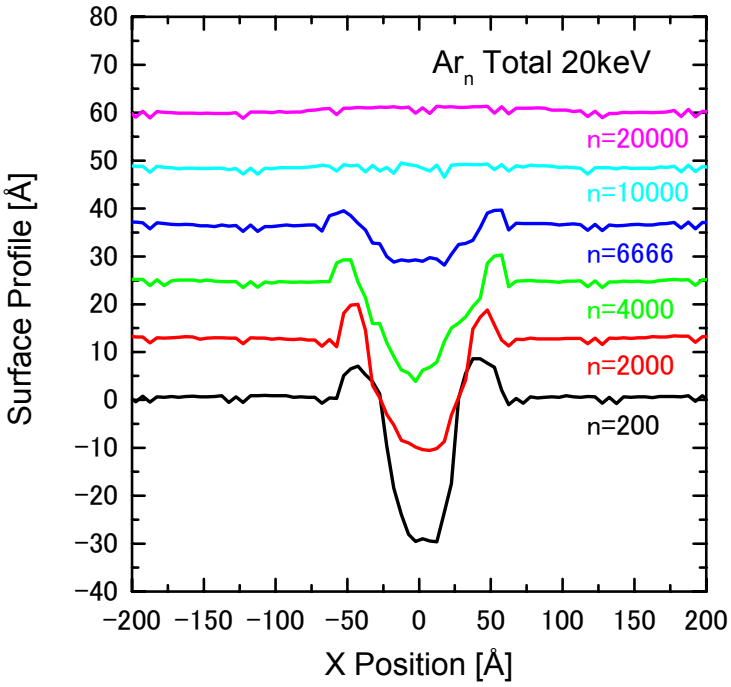


Fig. 1: Profile of Si surfaces after single impact of Ar clusters with various sizes (Total 20keV, 16ps after impact).

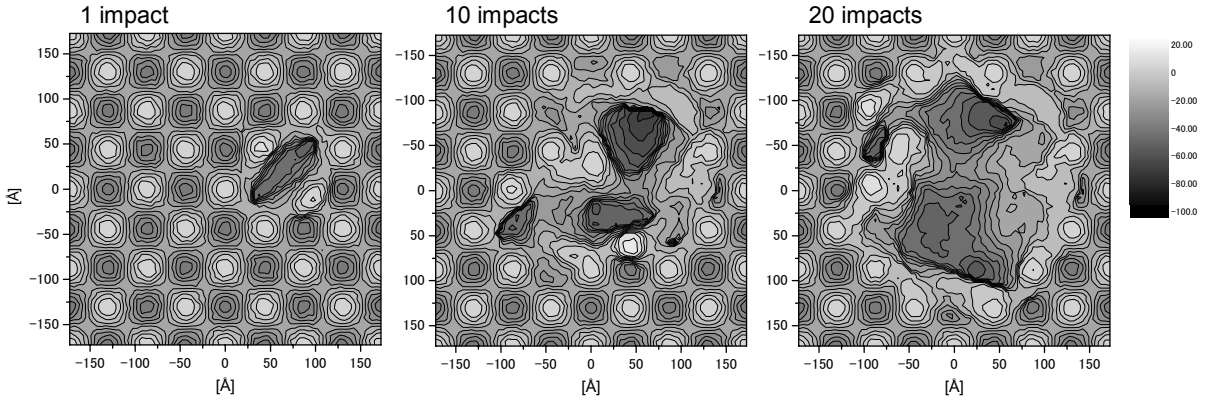


Fig. 2: Contour map of Si surface after 1, 10 and 20 impacts of $Ar_{2000}$ 20keV. Initial surface has 2-D sine curved profile with 40Å of peak-to-valley. Lines are drawn on every 5Å.

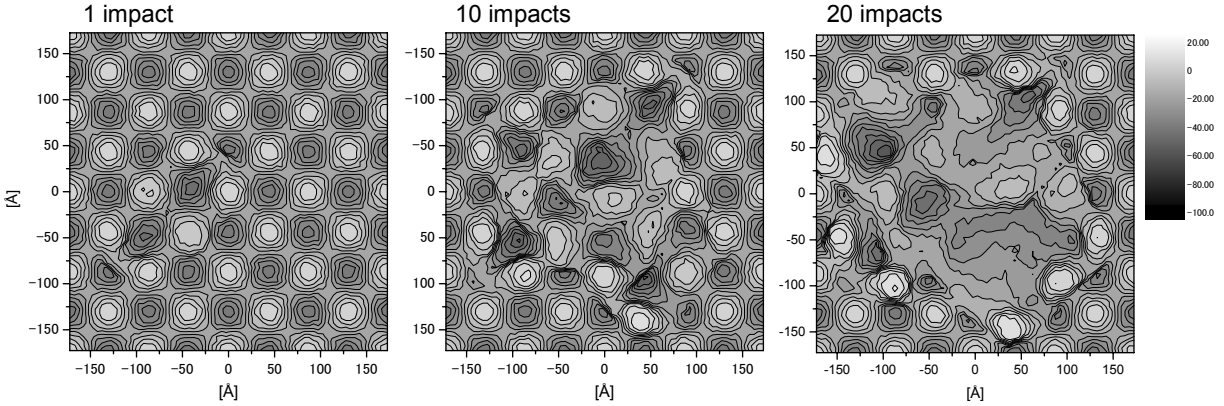


Fig. 3: Contour map of surface profile after 1, 10 and 20 impacts of $Ar_{13333}$ 20keV.

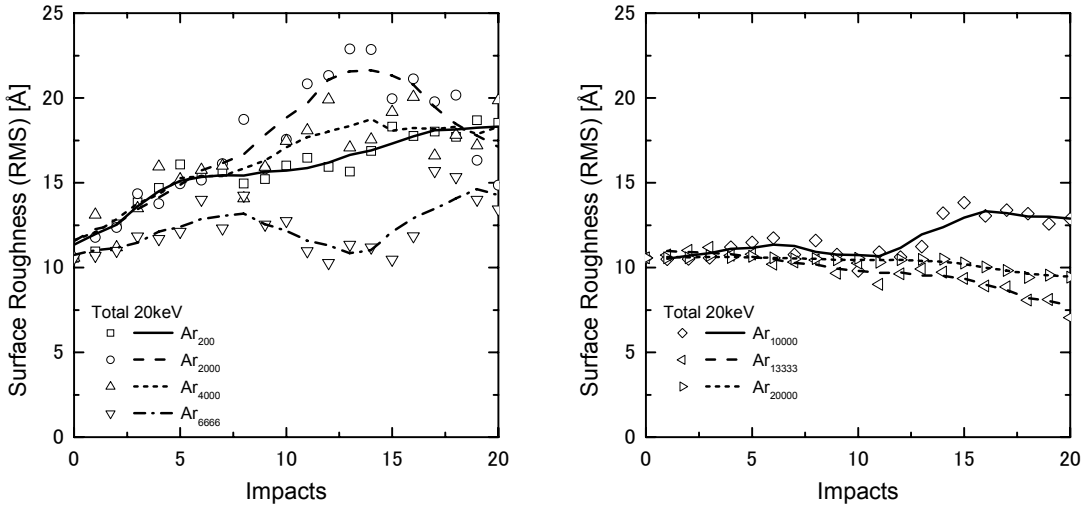


Fig.4: Evolution of surface roughness by sequential cluster impacts with various sizes. Roughness was calculated as the root mean square of the surface height as shown in Figs. 2 and 3. 20 impacts corresponds to $0.75\times10^{13}/cm^2$ of cluster dose.