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Molecular dynamics simulations for gas cluster ion beam processes

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Abstract

Molecular dynamics (MD) simulations of large argon clusters impacting on silicon targets were performed. The characteristics of crater-formation, a typical collisional effect with large cluster impact were examined from the viewpoint of incident energy, cluster-size and incident angle. The MD simulation results suggested that the condition where an incident cluster penetrates into the solid target and causes a crater is mainly dominated by the incident energy-per-atom rather than total incident energy of the cluster. Additionally, the MD simulations of sequential multiple cluster impacts and grazing-angle cluster irradiation on irregular surface structures were studied to characterize the surface modification effects with large cluster ion beam process.

Introduction

A cluster is an aggregate of atoms or molecules and is of interest from the viewpoint of not only material science, but also, ion beam engineering and physics. In ion beam engineering and physics, the interaction between cluster and solid target is sometimes termed as a 'non-linear' process, which means that the irradiation effects with clusters, such as sputtering and damage formation, are not explained by simple summations of those by individual monomer atoms. Obviously, the non-linear effects are due to the fact that many atoms in the incident cluster hit the target surface in a narrow region simultaneously, which results in high-density multiple collisions between cluster and surface atoms. This characteristic collisional process is considered to depend on cluster size (number of atoms in one cluster) as well as energy, species and other parameters controlled in conventional monomer ion beams. However, the irradiation effects of cluster ion beams are not understood

completely. Computer simulation using the molecular dynamics (MD) technique has played an important role in the analysis of the multiple collision process of clusters in collaboration with fundamental experiments. In this presentation, MD simulations of large gas clusters impacting on solid targets are demonstrated. The characteristics of surface deformation effects depending on incident energy, cluster size and incident angle are discussed. Additionally, surface modifications by multiple impacts and grazing angle irradiation were examined.

MD simulation model

The gas cluster ion beam technique provides high-intensity and large-size cluster ion beams. The gas cluster is generated by injecting source gas through a nozzle under high-pressure, where adiabatic expansion occurs and the source gas is condensed to form a large cluster. The cluster size of the generated gas cluster is very large ranging from several tens to several tens of thousands [1, 2]. The schematic of the MD simulation used in this study is shown in Fig. 1 [3]. The incident cluster imitates the solid state of a h.c.p. structure with an inter-atomic distance of 3.84 Å. The clusters irradiate a Si(100) target surface in the normal direction. A Si(100) target with diamond structure consisting of about 2,000,000 atoms is prepared as the target material. The target has a surface area of about 40 nm × 40 nm and a depth of 20 nm. Periodic boundary conditions are applied in the lateral direction. The atoms in the bottom unit cell layer are fixed in order to keep the target in the crystalline state, and 1/4 of the bottom target is set as a thermal bath using the Langevin dynamics method to absorb the excess irradiation energy propagating as a shockwave. The interatomic potential for Si–Si atoms is described by the Stillinger and Weber model potential [4]. On the other hand, the interactions between Si–Ar and Ar–Ar are described only by the Ziegler, Biersack and Littmark (ZBL) model potential [5], which assumes that attractive interactions of Si–Ar, and Ar–Ar are weaker than the interaction of Si–Si and that radiated energy is given by the cluster impact. In order to retain the cluster structure at the initial stage, 3.06 Å and 3.76 Å of cutoff lengths were applied for the Si–Ar and Ar–Ar interactions, respectively.

Crater formation by single cluster impact

Fig. 2 shows the snapshots of an Ar_{2000} cluster impacting on a Si(100) solid surface. In Fig. 2, the Ar_{2000} cluster is accelerated with a total energy of 20keV, so each constituent atom carries 10eV. Even at this low energy-per-atom, the incident cluster penetrates the target keeping itself as a cluster state because of the proximity effect. In other words, the cluster atom which first contacts with target surface can knock-on the surface target atom and is

implanted into the target. In this case, the following cluster atoms can penetrate deeper without energy loss, which leads the enhancement of the penetration depth of the cluster. The implanted cluster atoms spread under the target surface. A large number of target surface atoms are displaced hemispherically from the impact area due to the multiple collisions between cluster and surface atoms. Almost all of the Ar atoms in the incident cluster leave the target and the crater-like damage is left on the surface. This characteristic crater formation is confirmed by MD simulations of other materials [6, 7, 8] and experiments [9, 10].

Fig. 3 shows the surface profiles, depth and opening radius of craters caused by Ar_{2000} clusters with various incident energies. Fig. 3 shows that, when the total incident energy is higher than several-tens keV, the crater depth and radius increases in proportional to a cubic root of the incident energy. This is because, in this energy regime, almost all the incident kinetic energy is consumed for crater formation, and the crater shape keeps its aspect ratio. On the other hand, the crater depth rapidly decreases at less than 10keV. When the total incident energy is 10keV, each Ar atom in the cluster carries only 5eV which is similar in magnitude to the displacement energy for the Si target atoms. In this energy regime, the first-contacting Ar atom cannot penetrate the surface but is reflected, and it contributes to the corruption of the cluster on the target surface. Thus, the threshold condition for crater formation is considered to be dominated by the incident energy-per-atom rather than the total energy of the cluster [11].

Fig. 4 shows surface profiles and crater depth caused by Ar clusters of various sizes but constant total incident energies of 20keV [12]. Fig. 4 indicates that there is a specific cluster size around 5000 where the crater depth rapidly decreases. When the cluster size is 5000, the incident energy per atom is calculated as 4eV, which supports the above discussion about a threshold condition for crater generation. On the other hand, the change of crater depth is relatively small in spite of changes in cluster size ranging from 20 to 2000. It is considered that, in this cluster size regime, the cluster could be treated as a continuous material with the function of transferring its kinetic energy to surrounding target atoms isotropically.

Fig. 5 shows the surface profile and crater depth caused by 20keV Ar_{2000} impact at various incident angles [13]. As the incident angle increases, the crater shape becomes asymmetric and shallower. When the incident angle is more than 70 degrees from the surface normal, the crater diminishes. The conditions for crater formation can be also explained by considering the vertical component of incident energy-per-atom, which is calculated as 5, 2.5, 1.2 or 0.3 eV/atom for 45, 60, 70 or 80 degrees, respectively. These results on the conditions

to reduce the surface damage in cluster impact are expected to form one of the guidelines for surface processing with less damage but high-efficiency.

Surface morphology evolution by sequential multiple cluster impacts

With the development of computational resources in calculation speed and memory or storage capacity, MD simulation of cluster impacts is expanding from the analysis for a single impact event only to that for complex effects arising in multiple impacts. In order to examine the evolution of surface morphology during the cluster ion beam process directly, MD simulation of multiple cluster impacts iteratively on the same solid target is performed. Each impact iteration is, I) Select impact point randomly II) Perform a MD simulation of cluster impact for 16ps. After the impact simulation, molecules/atom desorbed into the vacuum are removed. III) Cool down the target to 300K for 4ps and remove desorbed materials during the cooling down process [14]. Fig. 6 shows the evolution of surface roughness by Ar_{2000}, Ar_{600} and Ne_{600} clusters. In these simulations, Si target sizes are usually chosen to be 16nm×16nm×16nm with about 260,000 atoms. For Ar_{2000} 20keV, irradiation on a large target of size 40nm×40nm×20nm was also performed. The surface roughness is calculated as root mean square of the highest atomic coordinates for every mesh with 0.5nm interval. As shown in Fig. 6, the surfaces become rough for any cluster irradiation condition. The dependence of the surface roughnesses are well fitted by the curves of exponential convergence indicated as dotted lines, which enables the estimation of the final roughness for each condition. Fig. 7 shows the correlation between the peak-to-valley of a single crater trace and the final roughness by sequential multiple impacts of large clusters estimated in Fig. 6. Fig. 7 suggests that the shape of a crater given by a single impact on a planar surface reflects the final roughness. These results suggest that, by controlling irradiation energy and cluster size, the performance and smoothness of cluster ion beam processing could be optimized depending on application requirements.

Surface modification using grazing incidence angles

As shown in Fig. 5, grazing angle incidence of a large gas cluster is expected to have no effect on a planar surface which is parallel to the incident cluster direction, but to experience significant for a non-parallel or irregular surface structure. Fig. 8 shows the snapshots of a grazing-angle Ar_{2000} cluster impact on a Si target to which a small block is attached [15]. The incident Ar cluster is accelerated at 20keV and impacts at 80 degrees to the surface normal. The small Si_{4096} block was a 4nm cube and was set on the planar Si target surface. As shown

in Fig. 8, the bottom part of the cluster contacts with the target at a very early stage of the simulation, but the target does not deform while the cluster slides on the planar part. The incident cluster collides with the small block and causes multiple collisions in a similar manner as when it collides on a planar surface in the normal direction shown in Fig. 2. The kinetic energy and momentum of the incident cluster is transferred to the small block, which is sputtered as a large chunk.

Conclusions

MD simulations of large gas clusters with various energies, sizes and incident angles were performed. The MD simulations have revealed that the threshold to cause surface damage is mainly dominated by the incident energy per atom rather than total energy. When the incident energy-per-atom is as low as several eV, each constituent cluster atom does not penetrate the target surface even with the help of high-density multiple collision effects. The incident cluster collapses on the target surface without damage. When incident energy-per-atom is more than 10eV, the cluster can be implanted into the target and causes multiple collisions. The multiple collisions between cluster and surface atoms provide isotropic atomic motion and energy propagation at the impact area, which results in spherical crater formation.

The MD simulation of sequential multiple impacts on a single target has suggested that, the final surface roughness obtained which would be realized by a real cluster ion beam process can be estimated from the shape of a single crater trace formed on a planar surface. In addtion, the grazing angle irradiation shows high selectivity against any surface irregular structure and also shows high degrees of smoothing. These MD simulation results are confirmed by experiments and suggest that the method is a high-performance surface modification process [16, 17]. It is expected that the application fields of MD simulations of cluster impacts will expand not only in fundamental analysis but also in practical applications after optimization of the cluster ion beam technique.

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Figures
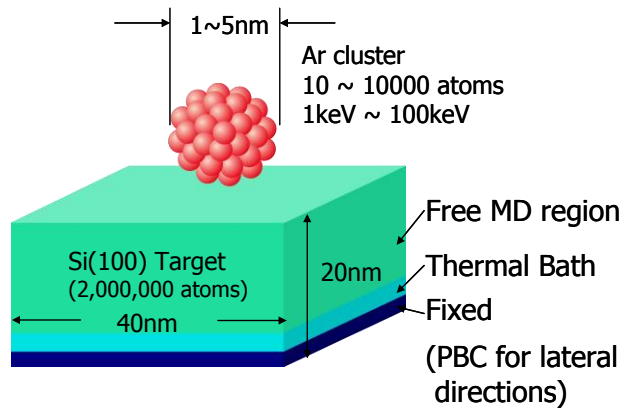
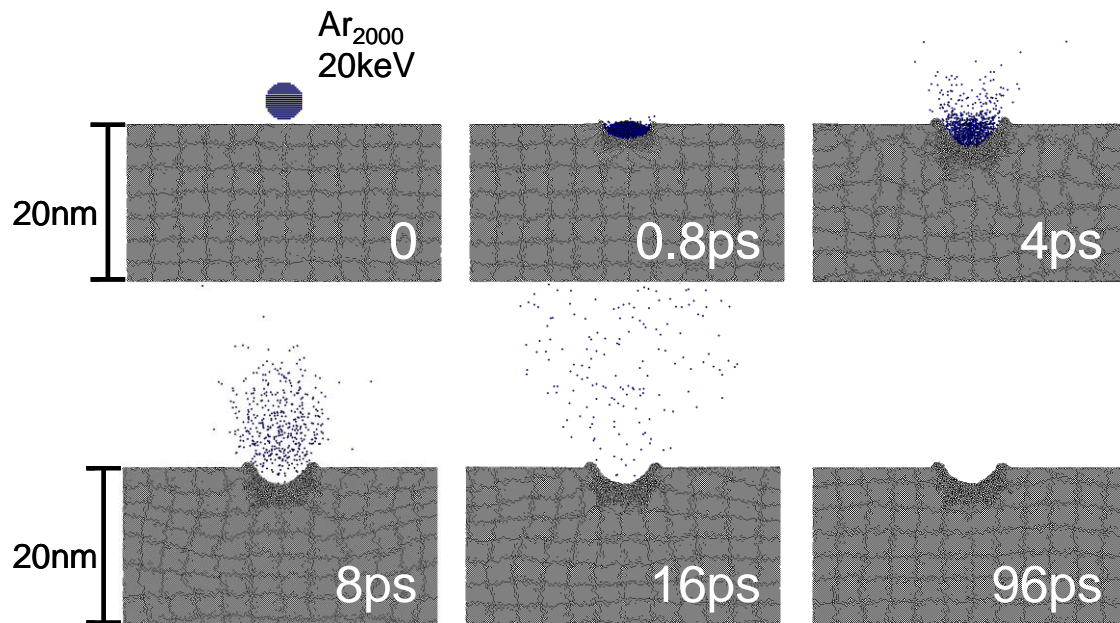Fig. 1 A schematic of MD simulation of cluster impact used in this study.

Fig. 2: Snapshots of MD simulation of Ar_{2000} with 20keV impacting on a Si(100) surface. Each snapshot represents a cross-secsion of 2nm thickness including the impact point.
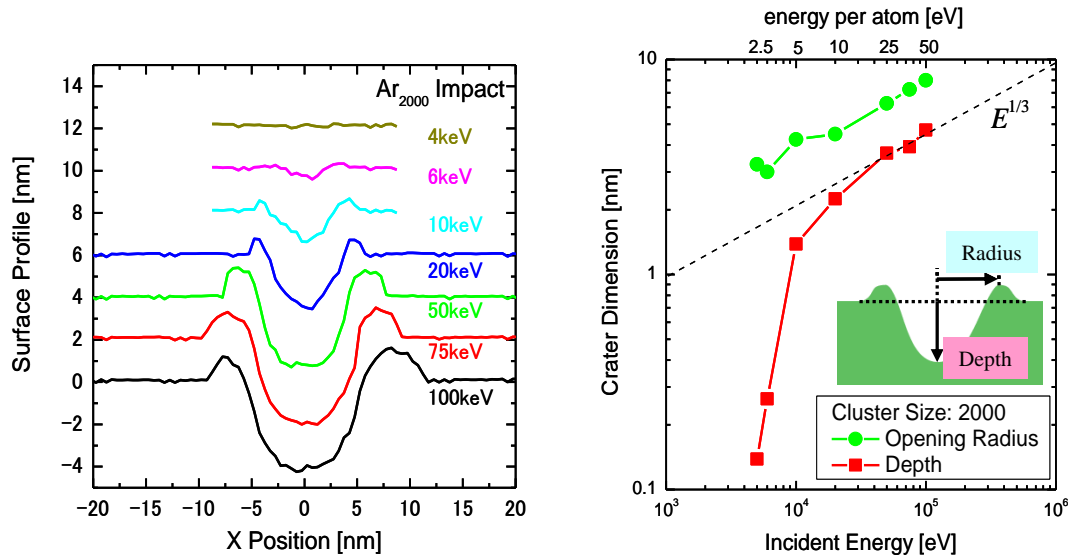
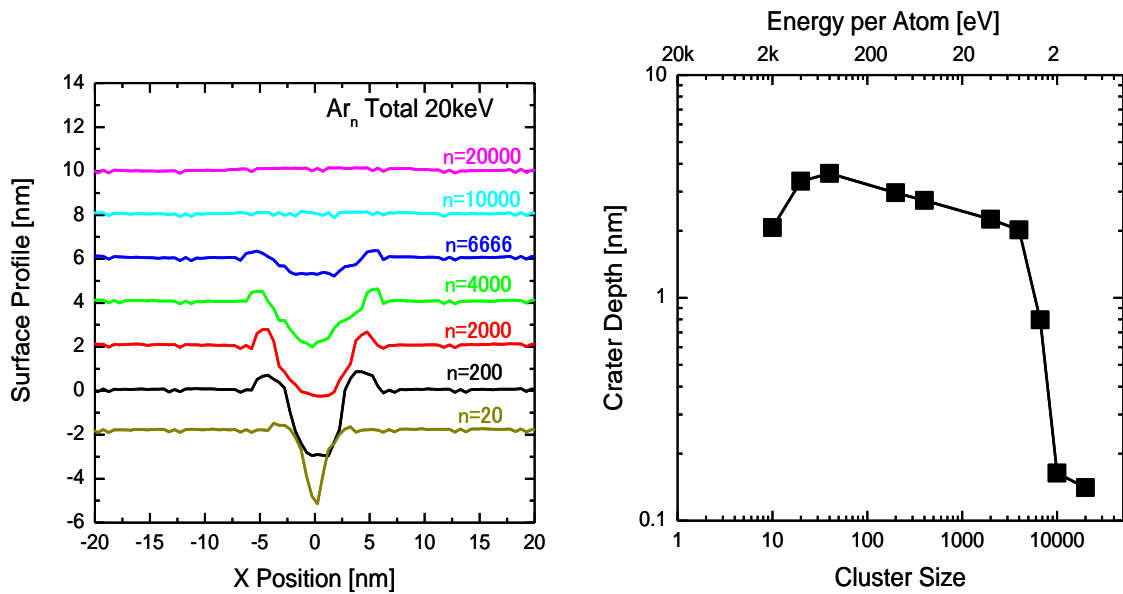Fig. 3: Surface profiles, crater depth and radius caused by Ar_{2000} impacts with various incident energies.

Fig. 4: Surface profiles and crater depth caused by Ar cluster impacts with various cluster sizes. The total incident energy is kept at 20keV.
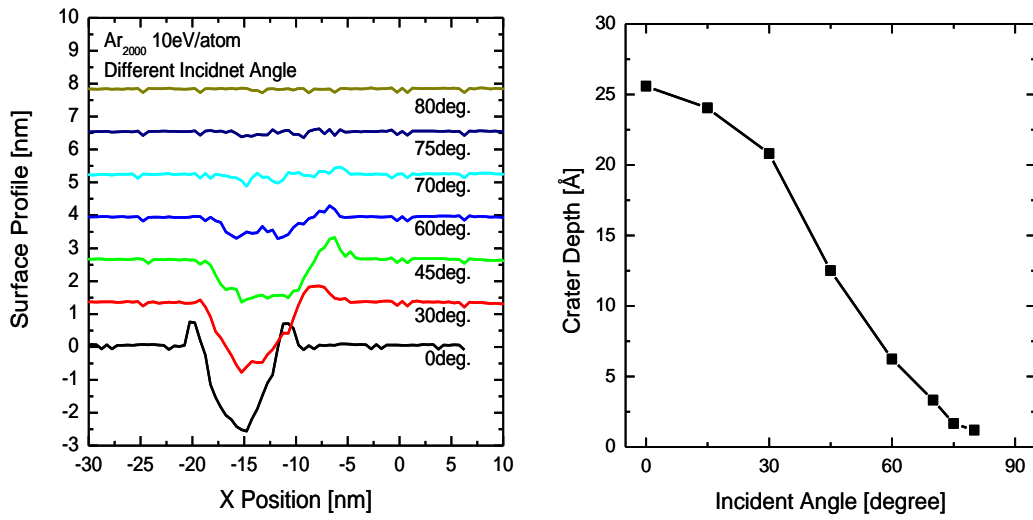
Fig. 5: Surface profiles and crater depth caused by 20keV Ar_{2000} cluster impacts with various incident angles.
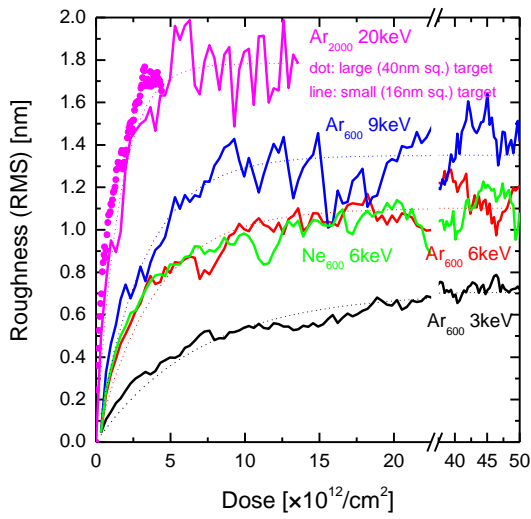

Fig: 6. Dose dependence of surface roughness caused by sequencially multiple impact of large clusters. The fitting by exponential decay are shown by dotted lines.
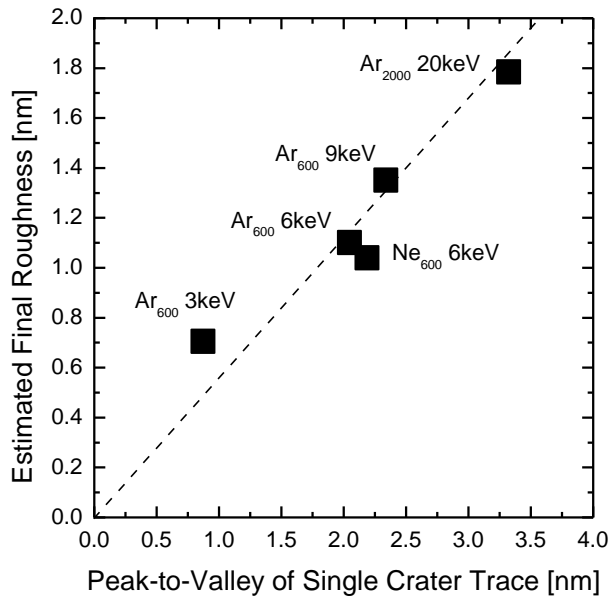
Fig. 7: Correlation between the peak-to-valley (P-V) of single crater trace and the final roughness by seqential multiple impacts of large clusters estimated in Fig. 6.
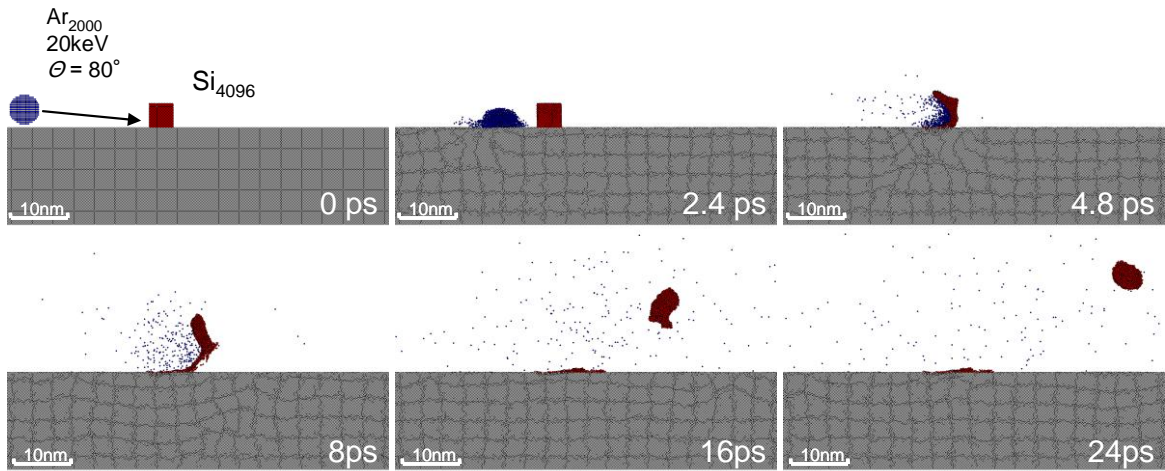

Fig. 8: Snapshots of 20keV an Ar_{2000} cluster impacting on Si target at 80 degrees from the surface normal.