Surface Deformation of Ion Collision Process via Molecular Dynamics Simulation

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Abstract. Molecular dynamics simulations of Ga ion collision on a Si surface using an optimized potential function were carried out for acceleration voltages of 1, 40, 100 keV for 36, 50, 100 Ga ions. A hillock structure was formed by the Ga ion impact. The height of the structure calculated by the simulations corresponded to experimental values. The sputtering yield was found to be proportional to the acceleration energy and for a high acceleration energy of 100 keV Si atoms deep within the impact surface were sputtered.

1. Introduction
Focused ion beams (FIBs) are a very useful tool in lithography, doping, deposition, and etching. In particular, FIB etching and deposition techniques have been widely used for mask repair and for sample preparation for transmission electron microscopes. Recently, these techniques have been extended to the fabrication of three-dimensional (3D) nanostructures. FIB chemical vapor deposition (CVD) is used to fabricate 3D nanostructures and aerial wiring applied to photonic crystals and nanoscale tweezers. FIB etching, however, has had very limited use in the fabrication and formation of 3D structures, such as predetermined curved shapes, microlens components, and diffractive optical elements, because of the very demanding specifications for etching tasks.

Recently, nanometer-order surface deformation on a Si surface was systematically achieved in experiments with a Ga ion beam collision process after an etch pit was created on the Si surface [1]. The height of the deformation was proportional to the ion beam dose. These experiments demonstrate the requirement for a high-accuracy process technology for nano-masks. The SRIM code [2] has generally been used as a reliable model for ion doping. Although SRIM results are generally highly consistent with experimental data, the detail of sputtered atoms and atomic-order deformation of a surface cannot be tracked dynamically. On the other hand, a molecular dynamics (MD) simulation can track such phenomena, because it can track each ion and atom dynamically. Thus, MD simulation is a powerful and effective tool for analyzing material deformation by ion beam processing.

Previous MD simulations of ion bombardment have studied low acceleration energy [3] and two-dimensional simulations with a two-body potential function [4]. Simulations that assume an actual
material through a multi-body potential have been done. However, using time integration with a real radiation time is impossible except for MD simulations of cluster ion irradiation[5] [6][7].

The present study considers nano-order deformation of a surface when the dose value of the irradiation parameter for the calculations is set to be a realistic value for an FIB, while neglecting the time integration problem. Estimations of the deformation height from the simulation results will be important information for the crafting of nano masks by ion beam processing.

Performing this MD simulation is difficult because the interval of the irradiation ions calculated for an actual ion beam facility is of the order of nanoseconds. Therefore, in the simulation the interval of the irradiation ion was compressed and all the ions were assumed to simultaneously collide at the surface. Large-scale MD simulations with the simultaneity condition were performed for acceleration voltages of 1, 40, 100 keV for 36, 50, and 100 Ga ions. The deformation on the surface caused was analyzed after the ions collided on the Si surface.

2. Simulation methods

We performed MD simulations to investigate the Ga ion collision process on a Si target. We have previously found an optimal combination of potential functions for the target material and the impacting ions [8]. The optimal potential combination is found for Silicon and Ga ion: Silicon is Tersoff potential [9], and Ga ion is Ziegler, Biersack, and Littmark (ZBL) potential [10]. The combination indicated the depth being comparable to previous value such as SRIM. We used our previous MD code [8]. In the present MD calculations, the Verlet algorithm was used for time integration.

2.1. Potential function for Si crystal

For the simulation with a Si target, the Tersoff potential function [9] was employed:

\[ \phi = \frac{1}{2} \sum_i \sum_{j \neq i} f_i(r_{ij}) \left( f_R(r_{ij}) + b_{ij} f_A(r_{ij}) \right) \]

Here \( f_k(r) = A \exp(-\lambda r) \)

\[ f_i(r) = \begin{cases} 1, & r < R \\ 1 + \frac{1}{2} \cos \pi (r-R)/S-R, & R < r < S \\ 0, & r > S \end{cases} \]

\[ b_{ij} = (1 + \beta \xi_{ij})^{-1/\alpha}, \quad \xi_{ij} = \sum_{k \neq i,j} f_k(r_{ik}) g(\theta_{ijk}) \]

Here \( i, j, \) and \( k \) label the atoms of the system, \( r_{ij} \) is the length of the \( ij \) bond, and \( \theta_{ijk} \) is the bond angle between bounds \( ij \) and \( jk \). The parameters in Eq. (1) are summarized in Table 1.

2.2. Ga ion for Ziegler, Biersack, and Littmark (ZBL) potential

The Ga ion potential is represented by the Ziegler, Biersack, and Littmark (ZBL) potential [10]:

\[ \phi(r) = \frac{Z_i Z_j e^2}{4 \pi \epsilon r_{ij}} \Phi(r) \]

\[ \Phi(r) = \sum_{k=1}^4 c_k \exp \left( -d_k \frac{r_{ij}}{a_k} \right), a_n = \frac{0.8854a_0}{Z_i^{0.23} + Z_j^{0.23}}, a_0 = 0.529 \text{Å} \]

Here \( Z_i \) and \( Z_j \) are atomic numbers. The detailed parameters are shown in Table 2.
Table 1. Parameter for Silicon.

|          | Si                        |
|----------|---------------------------|
| $A$ [eV] | 1.8308×10^3              |
| $B$ [eV] | 4.7118×10^2              |
| $\lambda$ [Å⁻¹] | 2.4799            |
| $\mu$ [Å⁻¹] | 1.7322             |
| $n$      | 7.8734×10⁻¹              |
| $\beta$  | 1.1000×10⁻⁶              |
| $c$      | 1.0039×10⁵               |
| $d$      | 1.6217×10¹               |
| $h$      | -5.9825×10⁻¹             |
| $R$ [Å]  | 2.7                      |
| $S$ [Å]  | 3.0                      |

Table 2. Parameter for ZBL potential.

| $c_i$  | $d_i$  |
|--------|--------|
| 1      | 0.028171 0.20162 |
| 2      | 0.28022 0.40290 |
| 3      | 0.50986 0.94299 |
| 4      | 0.18175 3.1998 |

3. Computational parameters

The computational domain for the large-scale calculation is shown in Fig. 1. The target domain is 25.3 nm × 25.3 nm × 8.55 nm, consisting of 282,752 Si atoms. The MD simulations under the simultaneity condition were performed for acceleration voltages of 1, 40, 100 keV for 36, 50, and 100 GA ions. Periodic boundary conditions were applied in the $x$- and $y$-directions. The top boundary was represented by a free boundary condition. The bottom boundary consists of a fixed heating bath layer. Table 3 shows the irradiation areas and ion numbers. Each colored slice in Fig. 2 corresponds to one of sixteen parallel partitions from the surface to the bottom of the Si target, referred to as regions.

![Fig. 1 Computational domain.](image-url)
4. Results and discussion

Figure 3 shows the height against ion irradiation. The height is determined from the extracted atoms with the kinetic energy calculated for all particles and the binding energy at the Si surface being less than 4.7 eV. Moreover, the height at \( t = 1.5 \) ps is expressed as the distance from the coordinate before the collision. Figure 3 (a) shows the height versus acceleration energy for various doses corresponding to the irradiation densities in Table 3. The experimental value by Kawasegi et al. (2005)[1] is also shown in Fig. 3, showing the height irradiated by Ga ions when the acceleration is 30 keV for a dose of \( 4.2 \times 10^{14} \) ions/cm\(^2\). In the present simulation, when the acceleration energy is less than 100 keV, the height ranged from 1 nm to 5 nm. In Fig. 3(b), the height is shown versus the dose value. The figure shows the height of the resultant hillock structure, exhibiting an increasing height with increasing dose. Although the time interval of an irradiation ion is compressed, the height of the hillock structure in the simulation corresponds to the same order as the experimental value. However, the height of the simulation is higher than that of the experiment. A time interval of the irradiation ions in the experiment is of the order of nanoseconds. This time interval is sufficiently long to be cooling at the collision surface. On the other hand, the surface temperature of the simulation can not recover owing to the collision of all the ions simultaneously, and the thermal expansion near the surface remained. Therefore, the simulation value has a higher height than the experiment value.

To examine the effect of acceleration energy, the initial ion velocity is changed for the case of 100 ions. The time development at the center surface is investigated by visualizing a 1-nm depth in the z-direction, shown in Fig. 4. In the figure, the computational region is divided by color for the parallel CPUs as in Fig. 2. The each colored layers are destructed by the ion collision, the layers are expanded, and it became the hillocks structure. At \( t \approx 0.3 \) ps for 1 keV, after a low density space is formed at Region 4 or Region 5, the upper layer at the surface appears to sputter. The case for 40 keV is similar to that for 100 keV. Figure 5 shows the probability density function, indicating that the Si atoms are further than 20 nm from the initial target height. The highest layer at the initial target height is the
most greatly enhanced for all cases; the deeper atoms are not sputtered. However, at 100 keV, atoms in Regions 13 to 16 are sputtered, whereas this is not observed at lower acceleration energies.

Fig. 3 Height of hillock; (a) Height versus acceleration energy, (b) Height versus dose

(a)

(b)
Fig. 4 Snap shot for 0.1, 0.3, 0.9, 1.5ps and the layers are divided by color for the parallel CPUs as in Fig. 2; (a)1keV, (b)40keV, (c)100keV

Fig. 5 Number of sputtered atoms and the Regions are divided by color for the parallel CPUs as in Fig. 2; (a) 1keV, (b) 40keV, (c) 100keV
5. Conclusions
Large-scale MD simulations were carried out to understand the influence of dose value and acceleration energy on surface deformation during ion collision. A hillock structure was observed and the height of the structure was analyzed and compared to experimental results.
1. The computation adopted a compressed time interval of ion irradiation because it was impossible to use a real irradiation time. Despite this approximation, the height of the hillock structure is of the same order as the experimental value.
2. The height of the hillock structure is 2 nm when the dose value is $10^{14}$ ion/cm$^2$.
3. While the hillock structure is formed, sputtering also occurs. The sputtering yield is proportional to the acceleration energy.
4. For a large acceleration energy, Si atoms deep within the impact surface were also sputtered.

Acknowledgement
This study was supported by "Academic Frontier" Project for Private Universities: matching fund subsidy from MEXT (Ministry of Education, Culture, Sports, Science and Technology), 2006-2010.

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