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Molecular dynamics simulation for the sputtering of an Al_2O_3 sample bombarded with MeV Si ions*

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Sputtering yield and kinetic energy distribution (KED) of Al particles from an Al_2O_3 sample bombarded with 1–5 MeV Si ions have been simulated using the molecular dynamics method. These have also been measured experimentally with a conventional time-of-flight facility. In the simulation, a new interatomic potential specific to the Al_2O_3 target was developed, and both the nuclear energy loss S_n and electronic energy loss S_e were taken into consideration. By carefully adjusting the simulation parameters, the simulated sputtering yields fit well with the experimental results, and the simulated KED of Al particles also fits roughly with the experimental KED after being modified theoretically.

Keywords: sputtering, MD simulation, kinetic energy distribution, sputtering yield
PACC: 3450D, 7115Q

1. Introduction

When an energetic ion impinges on a solid, its energy can be transferred to the target nuclei and electrons, and both processes can cause the target atom to be sputtered out; these are called nuclear sputtering and electronic sputtering, respectively.

Insulators have a very wide range of applications in modern industry due to their novel properties, and their sputtering behaviour is a very interesting topic both for theoretical and practical purposes. However, the sputtering process of insulator materials induced by heavy ions is very complicated. In general, the nuclear sputtering process is dominant when ion energy is low (keV order), and it can be accurately described by collision theory.[1]

When the incident ion energy is very high, the electron energy loss S_e becomes much higher than the unclear energy loss S_n, the electronic sputtering becomes significant and the nuclear sputtering can be neglected. Unlike the nuclear sputtering, the mechanism of electronic sputtering is still not clear. Several models have been set up to explain this process, such as the thermal spike model,[2,3] the ionic model[4,5] and the pressure model.[6,7] Among these models, the thermal spike model is the most popular, and it has successfully predicted the registration of the ion track in metal and isolator materials induced by fast ions.[8]

For MeV heavy ions, S_e is usually higher than S_n, but not high enough for the contribution of the nuclear sputtering to be neglected. It has been found that the sputtering of SiO_2 and LiF bombarded with 1–8 MeV heavy ions is the cooperation of these two mechanisms.[9,10]

The molecular dynamics (MD) simulation method is a very powerful tool to investigate the ion sputtering process. Many MD simulations have been carried out to simulate the nuclear sputtering process,[11–12] and some works have been performed for the pure electronic sputtering of weakly bonded materials, such as solid gas samples and biological samples.[13–15] But no simulation on oxide isolators bombarded with MeV heavy ion has been performed. Moreover, the above simulations only consider either the nuclear sputtering process or the electronic sputtering process, but not both.

In this paper, we present the primary MD simulation results on the sputtering process for Al_2O_3 bombarded with MeV Si ions. Both nuclear and electronic sputtering have been taken into consideration. The simulated sputtering yields (Y) and kinetic energy distribution (KED) are compared with the experimental results.

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2.Simulation method

The code used in the simulation was developed by our group on a PC. The average force method[16] was used to calculate the target atom movement. A Hookean-like force was applied to the boundary atoms to keep the sample stable,[11] except those in the sample's free surface on which the incident ions hit. The details of the simulation are as follows.

2.1. Sampling the Al_2O_3 target and the interatomic potential

Alumina has several types of structure, namely the α, β and γ phases. α-Al_2O_3 is used in this simulation because the sample used in our experiment is in the α phase. It is composed of six oxygen planes in the hcp stacking sequence, while the Al cations occupy 2/3 of the octahedral interstitial sites. Details of this structure can be found in Refs.[17–19]. In the simulation, 12×12×10 atoms were arranged according to the above structure, and O atoms were set in the first layer.

Several types of potential have been developed for Al_2O_3 for the purpose of simulating its physical properties.[18−22] Among these potentials, the so-called transferable potential developed by Matsui has been adopted in this study as shown in Eq.(1). This is because it has a simple form and is accurate enough to describe the Al_2O_3 physical property[18,19]

$$V(r_{jk}) = Q_j Q_k / r_{jk} - C_j C_k / r_{jk}^6 + D(B_j + B_k) \times \exp((A_j + A_k - r_{jk})/(B_j + B_k)). \quad (1)$$

Here r_{jk} is the distance between atom j and atom k; Q_j and Q_k are the effective charges of atoms j and k, respectively; and D is the standard force constant of 41.8kJ/nm. By fitting Al_2O_3 physical properties we can derive the parameters of A, B and C, and their values used in the simulation are shown in Table 1.

Table 1. Parameters used in the transferable potential for α-Al_2O_3 from Refs.[18,19]).

	Q	A/nm	B/nm	$C/(\mathrm{nm^3\ kJ^{1/2}mol^{-1/2}})$
Al	1.4175	0.07852	0.0034	0.03682
O	−0.9450	1.18215	0.0138	0.09061

However, this potential cannot be used directly in the MD simulation of the sputtering process, since it is not valid when r is very small. For example, when r is less than 0.05nm, the interatomic force will be attractive. This potential has been developed for simulating physical properties of materials where two atoms cannot come too close. But in the simulation of the sputtering process, the short-range interaction event is very popular and important.

To avoid this difficulty, an exponential equation of $A \times \exp(-B \times r)$ is used instead of the transferable potential at short distances. The parameters A and B for different atom pairs are shown in Table 2.[11]

Table 2. Parameters for different atom pairs in Eq.(2) (from Ref.[11]).

	A/eV	$B/(1/\mathrm{nm})$
Al-Al	6285.1	2.5
Al-O	5460	2.5
O-O	8460	4.0

Then the final potential can be described as

$$V(r) = \begin{cases} A \times \exp(-B \times r), & r < R_1; \quad (2) \\ \text{cubic polynomial}, & R_1 \le r < R_2; \quad (3) \\ \text{Transferable potential}, & R_2 \le r < R_3; \quad (4) \\ 0, & r \ge R_3. \quad (5) \end{cases}$$

R_1, R_2 and R_3 are the characteristic distances; R_1=0.07nm, R_2=0.11nm and R_3=0.45nm were used in the simulation. The cubic polynomial of Eq.(3) is used to connect the two potential equations smoothly in order to make sure the simulation system obeys the energy conservation law.

2.2. Physics model

The nuclear energy loss process can be simulated directly with the MD method by applying Coulomb repulsive force between the incident ion and target nuclei. In this simulation, the screened Coulomb potential with ZBL parameters given in Ref.[23] was used to describe the repulsive interaction.

As to the electronic energy loss, it is impossible to simulate the S_e–lattice energy transfer process with the MD method since the mechanism is not clear. According to recent works and the thermal spike model,[2,14,15] it can be assumed that a certain amount of energy will be transferred to sample atoms immediately after the ion penetrates the target. Only atoms within a cylindrical region around the ion track can get kinetic energy from the electronic energy loss process while others cannot, and the energy one atom can get depends on its distance from the ion track:

$$E_{\text{atom}} = R_{\text{couping}} \times S_e/r \qquad r < R_t. \tag{6}$$

Here R_t is the ion track radius, and we use R_t=0.17nm in this simulation. R_{coupling} is a free parameter related to the coefficient for S_e to be transferred into the atom's kinetic energy. The values of S_e for the heavy ions in the Al_2O_3 target were calculated with a TRIM code.

In the simulation, silicon ions with energies ranging from 1 to 5 MeV were used to bombard the Al_2O_3 target, and R_{coupling}=0.05, 0.016 and 0 was tested, respectively. In order to be able to compare with the experimental results directly, the ions impinge the target surface at an angle of 60° to the surface normal. The incident ion number was 100 for every type of ion, and the total simulation time was 5 ps. Finally, those atoms that went beyond 0.8nm from the sample surface were picked out as sputtered particles.

3. Experimental details

The energy spectra of sputtered secondary ions were measured with a conventional time-of-flight (TOF) facility at Kyoto University. The silicon ion beam used in the experiment was produced with a 1.7MV Cockcroft–Walton accelerator. The TOF ion detector is in the normal direction of the sample surface. The system vacuum was kept better than 4×10^{-6} Pa during the measurement. A 400nm α-Al_2O_3 layer on a pure aluminium substrate was used in the measurement as a target. Sputter cleaning was applied to the sample surface for 30 min before measurement. Other details of the experiment can be found elsewhere.[24,26]

4. Result and discussion

4.1. Sputtering yield of Al_2O_3 sample bombarded with MeV silicon ions

The simulated sputtering yields for Al_2O_3 bombarded with MeV Si ions are shown in Fig.1 as a function of ion energies; curves 1, 2 and 3 are obtained with R_{coupling}=0, 0.016 and 0.05, respectively. The measured yields for these ions bombarding Al_2O_3 are also shown in Fig.1. All the curves have been normalized to 1 at 1MeV for comparison, and the lines in the figure are only to guide the eyes.

It is well known that, with the ion energy increasing, S_n becomes smaller but S_e becomes larger. As a result, the contribution of the nuclear sputtering process should decrease and that of the electronic sputtering should increase.

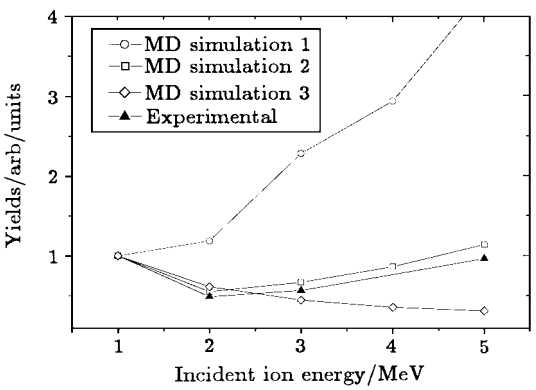


Fig.1. Simulated and experimental sputtering yields for Al_2O_3 bombarded with MeV Si ions as a function of incident ion energies with R_{coupling}=0.05, 0.016 and 0 in simulations 1, 2 and 3, respectively.

It can be seen that the yields obtained with R_{coupling}=0 decrease monotonically while increasing the ion energy. This is only consistent with the measured yield when the ion energy is lower than 2MeV. Beyond this energy, the simulated Y is much lower than the experimental value. R_{coupling}=0 means that the sputtering process is pure nuclear sputtering and the electronic effect is neglected, so this result indicates that the electronic sputtering cannot be neglected, especially when the ion energy is higher than 2MeV.

The yields obtained with R_{coupling}=0.05 increase quickly while increasing the ion energy, and they are much higher than the measured yields. This means that the electronic sputtering process in the simulation is stronger than the real process, and R_{coupling}=0.05 is too high for the simulation.

For R_{coupling}=0.016, the simulated results fit well with the measured values. When the ion energy changes from 1 to 2MeV, the yield decreases, which indicates that the nuclear sputtering process is dominant. While increasing the ion energy further, the

contribution of electronic sputtering becomes significant, so the yield increases again. Therefore, 0.016 is a suitable parameter to describe the coefficient for the electronic energy transferred to the atom's kinetic energy.

The above results show that the sputtering of Al_2O_3 induced by MeV Si ions is the cooperation of nuclear and electronic sputtering mechanisms, none of which can be neglected.

4.2. Secondary ion kinetic energy distribution

The simulated KED of Al particles from Al_2O_3 bombarded with 2 MeV Si ions is shown in Fig.2, and the measured KED of Al^+ is also shown in this figure. The maximum counts in these KED spectra have been normalized to 1 for easy comparison.

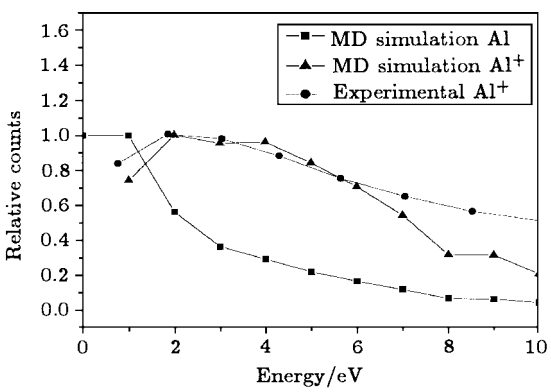


Fig.2. Simulated and experimental kinetic energy distribution of Al particles from Al_2O_3 bombarded with MeV Si ions.

The KED spectrum obtained with the MD simulation contains all the sputtered Al particles, both neutral and charged. It cannot be compared directly with the measured KED of Al^+, because the ionization probability for an Al particle from a solid surface is not a constant but depends on its velocity/energy.[25,26]

To date, no experimental data on the ionization probability for Al particles from Al_2O_3 have been found. The electron tunnel mode assumes that the ionization probability ($\boldsymbol{P}^+$) of particles depends on the resonance charge transfer process that occurs between the departing atoms and the solid surface.[27] This model predicts that

$$\boldsymbol{P}^+ \propto \exp(-(I-\phi)/v) = \exp(-\beta/v)), \qquad (7)$$

where I is the ionization potential, ϕ is the surface workfunction, β is a parameter related to the sample surface property and v is the particle velocity in the surface normal direction. It is possible to obtain the KED of Al^+ from that of the simulated Al particles using Eq.(7).

By carefully adjusting the value of β, the simulated Al^+ KED can fit well with the measured KED in the low-energy region as shown in Fig.2, the counts are higher than those measured only when $E > 8$eV. The value of β used in the above fitting is 1.1×10^4cm/s, which is similar to that for Al^+ from pure Al samples.[25,26] This might indicate that the ionization mechanisms for Al^+ from pure Al and Al_2O_3 are also similar. However, more experimental data on the ionization probability for Al^+ from Al_2O_3 are needed for a further comparison.

5. Summary

Both MD simulation and experimental measurements on the sputtering behaviour of Al_2O_3 bombarded with MeV Si ions have been performed. Both the nuclear and electronic energy loss processes were taken into account in the MD simulation.

By carefully adjusting the simulation parameters, the simulated sputtering yields fit well with the experimental results, and the simulated KED of Al^+ fits roughly with the experimental KED after being modified theoretically. These results indicate that both the nuclear and electronic sputtering are important for the sputtering of Al_2O_3 bombarded with MeV Si ions. When the ion energy is lower than 2MeV, the nuclear sputtering is dominant. In the higher-energy region, the electronic sputtering becomes significant.

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References

[1] Sigmund P 1981 *Sputtering by Particle Bombardment I* (Berlin: Springer)

[2] Toulemonde M, Dufour C and Paumier E 1992 *Phys. Rev.* B **46** 14362

[3] Meftah A, Brisard F, Costantini J M, Dooryhee D, Hage-Ali M, Hervieu M, Stoquert P, Studer F and Toulemonde M 1994 *Phys. Rev.* B **49** 12 457

[4] Dunlop A, Lesueur D and Dural J 1989 *Nucl. Instrum.*

Methods B **42** 182
[5] Dunlop A, Lesueur D, Morillo J, Dural J, Sphor R and Vetter J 1990 *Nucl. Instrum. Methods* B **148** 419
[6] Johnson R E 1989 *Phys. Rev.* B **140** 49
[7] Fenyo D 1993 *Phys. Rev.* B **147** 8263
[8] Skuratov V A, Efimov A E and Zagorskii D L 2002 *Phys. Solid State* **44** 171
[9] Kyoh S, Takakuwa K, Umezawa M, Itoh A and Imanishi N 1995 *Phys. Rev.* A **51** 544
[10] Pereira J A M, deCastro C S C, Jeronymo J M F, Ponciano C R, daSilveira E F and Wien K 1997 *Nucl. Instrum. Methods* B **129** 21
[11] Eckstein W 1991 *Coumpter Simulation of Ion-Solid Interaction* (Berlin: Springer-Verlag)
[12] Becker C 2001 *Chin. Phys.* **10** 174
[13] Sundaqvist B U R and Karlsson B R 1990 *Phys. Rev.* B **42** 1895
[14] Bringa E M and Johnson R E 1998 *Nucl. Instrum. Methods* B **143** 513
[15] Bringa E M, Johnson R E and Dutkiewicz L 1999 *Nucl. Instrum. Methods* B **152** 267
[16] Harrison D E, Gay Jr. W L and Effron H M 1969 *J. Math Phys.* **10** 1179
[17] Feschwind S and Remerika J P 1961 *Phys. Rev.* **122** 757
[18] Belonoshko A, Ahuja R and Johansson B 2000 *Phys. Rev.* B **61** 3131
[19] Gutierrez G 2000 A *Phys. Rev.* E **61** 2723
[20] Lewis G and Catlow C 1985 *J. Phys. C: Solid State Phys.* **18** 1149
[21] Rambaut C, Jobic H, Jaffrezic H, Kohanoff J and Fayeulle S 1998 *J. Phys.: Condens. Matter* **10** 4221
[22] Wilson M, Exner M, Huang Y and Finnis M 1996 *Phy. Rev.* B **54** 15 683
[23] Ziegier J F, Biersack J P and Littmark U 1985 *The stopping and Range of Ions in Solids* (New York: Pergamon)
[24] Imanishi N, Shimizu A, Ohta H and Itoh A 1999 *Applications of Accelerators in Research and Industry, edited by J.L. Duggan and I.L. Morgen* p 396
[25] Garrett R F, Macdonald R J and O'Connor D J 1983 *Nucl. Instrum. Methods* **218** 333
[26] Xue J M and Imanishi N 2002 *Chin. Phys.* **11** 245
[27] Yu M L 1978 *Phys. Rev. Lett.* **40** 574