Vol 11 No 3, March 2002
1009-1963/2002/11(03)/0245-04
Chinese Physics
© 2002 Chin. Phys. Soc.
and IOP Publishing Ltd

Molecular dynamic simulation of secondary ion emission from an Al sample bombarded with MeV heavy ions*

Xue Jian-Ming(薛建明)[a] and N. Imanishi(今西信嗣)[b]

[a] *Institute of Heavy Ion Physics, Peking University, Beijing* 100871, *China*

[b] *Department of Nuclear Engineering, Kyoto University, Kyoto* 606-8501, *Japan*

(Received 14 November 2001)

Sputtering yields and kinetic energy distributions (KED) of Al atomic ions ejected from a pure aluminium sample under MeV silicon ion bombardment were simulated with the molecular dynamic method. Since the electronic energy loss S_e is much higher than the nuclear energy loss S_n when the incident ion energy is as high as several MeV, the S_e effect was also taken into consideration in the simulation. It was found that the simulated sputtering yield fits well with the experimental data and the electronic energy loss has a slight effect at incident ion energies higher than 4 MeV. The simulated secondary ion KED spectrum is a little lower in the peak energy and narrower in the peak width than that in the experiment.

Keywords: sputtering, molecular dynamic simulation, kinetic energy distribution, energy spectrum
PACC: 3450D, 7115Q

1. Introduction

It is well known that an energetic ion loses its energy via two nearly independent processes in a solid: nuclear energy loss (S_n) and electronic energy loss (S_e). When the incident ion energy is low (keV), the nuclear energy loss is dominant. The incident ion loses its energy mainly through elastic collisions with target atoms. Some target atoms obtain enough energy so that they can be ejected from the sample, and the contribution of S_e to the sputtering process can be neglected. This sputtering phenomenon (nuclear sputtering) can be well explained by the elastic collision theory.

When the incident ion energy is high, S_e becomes much higher than S_n, its influence on sample sputtering behaviour can no longer be neglected. However, the mechanism of the electronic-energy-loss-induced sputtering (electronic sputtering) is still not clear, despite the fact that some models have been set up to explain it. Among these models, the thermal spike model[1,2] and the ionic model[3,4] are the most popular. The thermal spike model assumes that the electronic energy loss can be transferred to lattice atoms through an electron–phonon coupling process. The ionic model assumes that the lattice atoms can obtain energy from the repulsive Coulomb force between the high-density excitations created by the electronic energy loss. No matter what the energy transfer mechanism is, it is sure that a part of the electronic energy loss can be transferred to the lattice atom, and this will influence the target sputtering behaviour. Since the energy that target atoms can receive depends on both the magnitude of S_e and the S_e-lattice transfer property, the electronic sputtering behaviour also depends on both of these. It was found[5−11] that some materials, such as low-temperature condensed-gas solid, organic materials,[13] oxides,[6,7] have strong S_e-lattice energy transfer efficiencies, while some other materials, such as Al and Ni,[12] have weak S_e-lattice energy transfer efficiencies. Therefore, the electronic sputtering effect in different materials should be different even if S_e is the same.

For most MeV heavy ions, the values of S_e are higher than those of S_n when they travel in a solid, and both S_n and S_e possibly influence the sputtering behaviour of materials in this energy region. It was found that the sputtering yield of Si^{4+} from a SiO_2 sample depends on both S_e and S_n.[14] In order to study the mechanism of the electronic sputtering process, it is necessary to investigate the S_e and S_n effect on the sputtering behaviour of materials with different S_e-lattice transfer properties. For this purpose, Al, Al_2O_3 and SiO_2 samples which have different S_e-

*Project supported by the National Natural Science Foundation of China.

lattice transfer properties,[6,7,12] have been or will be used in our sputtering measurements.

The molecular dynamics (MD) simulation method is a very powerful tool to investigate the ion sputtering process. Much MD simulation work has been carried out to simulate the sputtering process induced by low-energy ions,[15] and the simulated results fit well with the experimental data, including the secondary ion kinetic energy distribution (KED), angular distribution and sputtering yield.[16,17] For electronic sputtering, fewer MD simulations have been done on weakly bonded materials, such as solid gas samples and biological samples. Though the S_e-lattice energy transfer mechanism is not clear, some simulated results are in agreement with the experimental results.[18−20] These results have shown that the MD simulation is suitable to study the electronic sputtering process. However, up to now, very few works have been done on the sputtering process of tight-binding-chemical materials irradiated with MeV heavy ions.

In this paper, the MD method is used to simulate the sputtering process of pure aluminium bombarded with MeV silicon ions. Both S_e and S_n effects are taken into consideration in the simulation. The simulated sputtering yields and secondary ion KED spectra are compared with our experimental results.

2.Simulation method

The code used in the simulation was developed in our group on a PC computer. The average force method[21] was used to calculate the target atom movement. A Hookean-like force was applied to the boundary atoms to keep the sample stable,[15] except those in the sample free surface, which the incident ions hit.

The simulated Al sample contains 14×14×10=1960 atoms organized in a fcc structure. The pair potential is used to describe the interaction between Al atoms in a combined form as given by

$$V(r)=\begin{cases} A\mathrm{e}^{-Br}, \quad r<R_1 & (1)\\ \text{cubic polynomical}, \quad R_1<r<R_2, & (2)\\ V_0(\mathrm{e}^{-2\alpha(r-R_0)}-2\mathrm{e}^{-\alpha(r-R_0)}), & \\ \qquad R_2<r<R_3, & (3)\\ 0, \quad r>R_3, & (4)\end{cases}$$

where r is the distance between the pair of Al atoms. The values of A, B and α are 4285.1eV, 2.718nm^{-1} and 12.1nm^{-1} respectively, which are adopted from Ref.[15]. V_0 is the depth of the attractive well, R_1 and R_2 are chosen carefully to ensure the smooth connection between the potentials (1) and (3), and R_3 is the cut-off distance for the potential.

The screened Coulomb potential with ZBL parameters is used to describe the interaction between the incident silicon ion and the target atoms.[22] Nuclear energy loss could be taken into account in the simulation directly by using the Coulomb force between the incident ion and the target atoms. As to the electronic energy loss, it is impossible to simulate the S_e-lattice energy transfer process since its mechanism is still not clear. In this simulation, we assume that the electronic energy transferred to the lattice atoms within a very short time, and the transferred energy is distributed only to the atoms within a cylindrical region around the ion track as follows

$$E_{\mathrm{atom}} \propto S_e \times R_{\mathrm{se}} \times \frac{1}{r}, \quad r<R_0, \tag{5}$$

$$E_{\mathrm{atom}}=0, \quad r\geq R_0, \tag{6}$$

where E_{atom} is the energy transferred to the lattice atom, R_{se} is the S_e-lattice energy transfer coefficient, two arbitrary values of 0.025 and 0.1 are used in the simulation, r is the distance of the atom from the ion track, and R_0 is the assumed cylinder radius. Atomic momentum is distributed in random directions.

In the simulation, silicon ions with energies ranging from 100 keV to 6 MeV are incident on the Al (111) surface at an angle of 60° to the surface normal, and the incident ion number is 100 for every energy. The total simulation time is 5 ps, and those atoms that go beyond 0.8nm from the sample surface are picked out as sputtered atoms.

3. Experimental details

The energy spectra of sputtered secondary ions were measured with a conventional time of flight (TOF) set-up at Kyoto University. A silicon ion beam used in the experiment was produced with a 1.7MV Cockcroft–Walton accelerator. The TOF ion detector was in the normal direction of the sample surface. The system vacuum was kept at better than 4×10^{-6} Pa during the measurement. A surface barrier detector was placed at an angle of 60° to the incident beam to monitor the beam current intensity, and a single-crystalline Al sample was used in the measurement. An ion-beam-sputtering cleaning was applied to the sample surface for 30min before every measurement. Other details of the experiment can be found elsewhere.[23]

4. Result and discussion

4.1. Secondary ion sputtering yield

The simulated sputtering yields for R_{se}=0, 0.025 and 0.1 are shown in Fig.1 as functions of incident ion energy (E_{in}). The measured sputtering yields of Al bombarded with MeV silicon ions are also shown in Fig.1. All the simulation data are normalized with the experimental data at E_{in}=1 MeV.

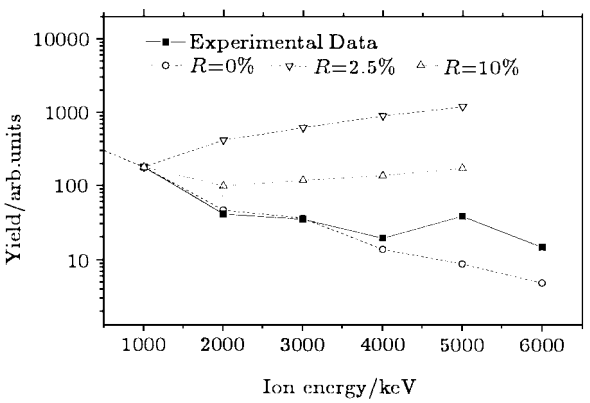


Fig.1. Comparison between the simulated and measured sputtering yields for the Al sample bombarded with silicon ions as functions of the incident ion energy.

It can be seen that, if R_{se}=0, which means only the nuclear sputtering is taken into account, the simulated yields fit well with the experimental data when E_{in} is lower than 4 MeV. However, when E_{in} is higher than 4 MeV, the simulated data become a little lower than the experimental data.

Though the statistical error of the simulation is high at high energies, it can still be seen that there is an obvious deviation between the tendencies of the experimental and the simulated data. This deviation may come from the S_e effect. When E_{in} is low, S_e is also low, its contribution to the sputtering yield can be neglected, and the simulated yield of R_{se}=0 fits the experimental result well. But when the incident energy becomes high, the effect of S_e becomes comparable with that of S_n, and then it can no longer be ignored. The measured yield is a little higher than the simulated data for R_{se}=0 in the high-energy region.

The simulated sputtering yields for R_{se}=0.025 and 0.1 increase monotonically with the increase of E_{in}. This behaviour disagrees with the experimental behaviour. Even for R_{se}=0.025, the simulated sputtering yield increases much faster than the measured one. The above results show that the S_e-lattice energy transfer process is very weak in Al. This is consistent with the theoretical analysis.[12]

4.2. Secondary ion KED

The KED spectrum obtained from the MD simulation contains all sputtered atoms, both the neutral and the charged. In order to compare with the measured Al^+ KED, some corrections must be made. According to the tunnelling model, the single-charged secondary ion yield from metal can be written as[24–27]

$$Y^+ \propto N_0(E)\exp(-\beta/v\cos(\phi)), \qquad (7)$$

where $N_0(E)$ is the KED for all the sputtered atoms, β is a special parameter, and usually its value depends on the energy of the secondary ions.[26] However, when the secondary ion energy is lower than 50 eV, its value for Al^+ from the Al material is nearly a constant and can be estimated to be 1.1×10^4 m/s.[26] v is the secondary ion velocity and ϕ is the angle between v and the sample surface normal direction, with ϕ=0 in our experimental arrangement.

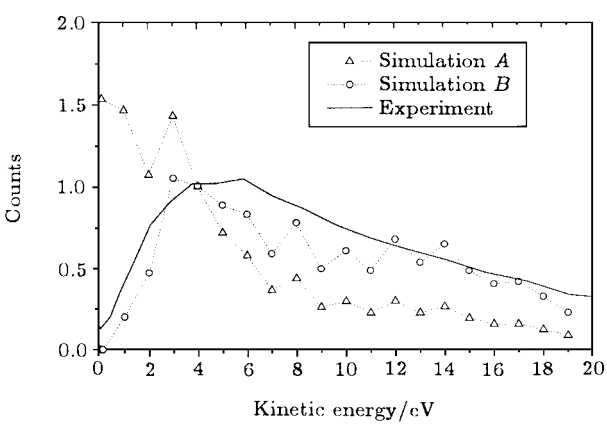


Fig.2. The simulated and measured secondary ion kinetic distribution spectra for the Al sample bombarded with 1MeV Si^+ ions: curve A, simulated KED spectrum of all the sputtered atoms; curve B, simulated Al^+ KED spectrum obtained with Eq.(7).

Using Eq.(7), the Al^+ KED spectrum can be obtained from the simulated KED spectrum of the secondary ions. The simulated secondary ion KED spectra for the Al sample bombarded with 1 MeV Si^+ ions, both for the total atoms and for the Al^+ atoms, are shown in Fig.2. The experimental Al^+ KED spectrum is also shown in Fig.2 for comparison. All the spectra are normalized to the experimental data at E_k=4 eV.

It can be seen that the Al^+ KED spectrum of the MD simulation fits roughly with the experimental spectrum. Compared with the experimental result, the peak energy of the simulated spectrum is a little lower, and the simulated peak width is narrower.

Both the simulated and measured results show that the shape of the secondary ion KED spectra does not depend significantly on the incident ion energy in the range from 1 to 6 MeV.

In our simulation, the pairwise potential was used, since it is simple and allows a faster simulation speed. If the embedded-atom type potential is used, it can be expected that the peak energy of the KED spectrum of the sputtered atoms increases, and the energy peak can become broader.[16,27] The potential used may be the cause for the above discrepancy between the simulated spectrum and the experimental result.

The above results indicate that nuclear sputtering is still the dominated process for the Al sample irradiated with MeV silicon ions. For ion energies higher than 4 MeV, S_e shows a slight influence on the sputtering yield, though S_e is tens times higher than S_n. Since SiO_2 or Al_2O_3 has a stronger S_e-lattice transfer efficiency compared with Al, it could be expected that the electronic sputtering effect in them could be more significant than that in Al.

5. Summary

We have carried out both the MD simulation and the measurement for the Al sample sputtered with MeV silicon ions. It was found that, if only S_n is concerned, the simulated sputtering yields fit well with the experimental ones at incident ion energies lower than 4 MeV. By using the tunnel mode, the Al^+ KED spectrum was derived from the simulated KED spectrum of all the secondary ions. The simulated Al^+ KED spectrum fits roughly with the measured one, though its peak energy is a little lower compared with the measured spectrum.

These results show that, when Al is irradiated with silicon ions in the energy range from 1 to 6 MeV, nuclear sputtering is still the dominant process, and the electronic sputtering effect is very slight. This means that the S_e-lattice energy transfer process in Al is very weak.

Acknowledgments

This work was done with the Experimental System for Ion Beam Analysis at Kyoto University. We thank A. Itoh, K. Yoshida and K. Norizawa for their useful advice and technical support during the experiment.

References

[1] Toulemonde M, Dufour C and Paumier E 1992 *Phys. Rev.* B **46** 14362
[2] 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** 12457
[3] Dunlop A, Lesueur D and Dural J 1989 *Nucl. Instrum. Methods* B **42** 182
[4] Dunlop A, Lesueur D, Morillo J, Dural J, Sphor R and Vetter J 1990 *Nucl. Instrum. Methods* B **48** 419
[5] Toulemonde M, Bouffard S and Studer E 1994 *Nucl. Instrum. Methods* B **91** 108
[6] Meftah A, Brisard F, Costantini J M, Dooryhee E, Hage-Ali M, Hervieu M, Stoquert J P, Studer F and Toulemonde F 1993 *Phys. Rev.* B **48** 920
[7] Canut A, Benyagoub A, Marest G, Meftah A, Moncoffre N, Ramos S S M, Studer F, Thevenard P and Toulemonde P 1995 *Phys. Rev.* B **51** 12194
[8] Itoh N and Stoneham A M 1998 *Nucl. Instr. Meth.* B **146** 362
[9] Toulemonde M, Costantini J M, Dufour C H, Meftah A, Paumier E and Studer F 1996 *Nucl. Instrum. Methods* B **116** 37
[10] Xu Y, Pan Z Y and Wang Y X 2001 *Acta Phys. Sin.* **50** 88 (in Chinese)
[11] Meftah A, Djebara M, Khalfaoui N, Stoquert J P, Studer P and Toulemonde M 1997 *Nucl. Instrum. Methods* B **122** 470
[12] Wang Z G, Dufour C, Paumier E and Toulemonde M 1994 *J. Phys. Matter.* **6** 6733
[13] Runne M, Becker J, Lassch W, Varding D, Zimmer G, Liu M and Johnson R E 1993 *Nucl. Instrum. Methods* B **82** 301
[14] Kyoh S, Takakuwa K, Umezawa M, Itoh A and Imanishi N 1995 *Phys. Rev.* A **51** 544
[15] Eckstein W 1991 *Computer Simulation of Ion-Solid Interaction* (Berlin: Springer-Verlag)
[16] Garrison B J *et al* 1987 *Phys. Rev.* B **36** 3516
[17] Garrison B J, Nicholas Winograd, Deaven D M and Reimann C T 1988 *Phys. Rev.* B **37** 7197
[18] Sundaqvist B U R and Karlsson B R 1990 *Phys. Rev.* B **42** 1895
[19] Bringa E M and Johnson R E 1998 *Nucl. Instrum. Methods* B **143** 513
[20] Bringa E M, Johnson R E and Dutkiewicz L 1999 *Nucl. Instrum. Methods* B **152** 267
[21] Harrison D E, Gay Jr W L and Effron H M 1969 *J. Math Phys.* **10** 1179
[22] Ziegier J F, Biersack J P and Littmark U 1985 *The Stopping and Range of Ions in Solids* (New York: Pergamon)
[23] Imanishi N, Shimizu A, Ohta H and Itoh A 1999 *Applications of Accelerators in Research and Industry* ed J L Duggan and I L Morgen, p 396
[24] Yu M L and Lang N D 1986 *Nucl. Instrum. Methods* B **14** 403
[25] Garrison B J 1986 *Surf. Sci.* **167** 225
[26] Garrett R F, Macdonald R J and O'Connor D J 1983 *Nucl. Instrum. Methods* **218** 333
[27] Vasile M J 1982 *Surf. Sci.* **115** 1141