Enhancement of cluster yield under gold dimer oblique bombardment of the silicon surface

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Abstract

Recent experiments of silicon bombardment show that kilo electron volt heavy polyatomic projectiles can increase the non-linear enhancement of the total yield of secondary silicon ions as well as the cluster ones. To understand why the heavy polyatomic projectiles increase the yields, molecular dynamics simulations of the bombardment of a Si(100)-(2×1) surface by Al_n and Au_n, $n = 1, 2$ with an initial energy of 1.5 keV/atom at the incident angle of $45°$ are carried out. The microscopic analysis shows that upon penetrating into the substrate the Au_2 constituents disintegrate slowly and the collision cascades overlap with a large probability. The process of sharing and depositing energy near the surface is very effective in the Au_2 case under these bombardment parameters. Thus the probability of high yield events for enhancement of cluster yield is increased.
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Keywords: SIMS; Sputtering; Energy dissipation; Silicon

1. Introduction

The specific question, which is still open in analytical applications, is how to optimize the different primary beam parameters, such as primary particle mass, energy, incident angle, and the number of projectile constituents for a given sample to create the desire improvement of the sensitivity of secondary ion mass spectrometry (SIMS). One approach for improving SIMS performance is using polyatomic projectiles. In experiments [1,2] where polycrystalline silicon target was bombardment by Au_n^- ($n = 1–3$) and Al_n^- ($n = 1, 2$) with the initial energies, E_0, from 6 to 18 keV/atom at the incident angle $\theta = 45°$, have shown that the Au_n^- bombardment produces an anomalously high non-linear enhancement of the large cluster Si_n^+ ($n \geq 4$) yields. In contrast, the Al_n^- bombardment is less effective at producing large cluster ion emission. To understand why the bombardment of the silicon surface by heavy polyatomic projectiles results in the non-linear enhancement of the cluster yields, molecular dynamics (MD) simulations of the bombardment of a Si(100)-(2×1) surface by Al_n and Au_n, $n = 1, 2$ with $E_0 = 1.5$ keV/atom at $\theta = 45°$ have been carried out.

2. Method

The MD simulations are used to model the interaction of Al, Al_2, Au, and Au_2 with the silicon surface. This method is described extensively elsewhere [3,4]. Details of the calculation have been described in Ref. [5]. The dimer reconstructed Si(100)-(2×1)

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microcrystallite consisting of about 9000 atoms with 10 layers is used. The projectiles hit the surface with $E_0 = 1.5$ keV/atom at $\theta = 45°$. The orientation of the dimer projectiles with respect to the surface is selected randomly. The forces among the atoms are described by the best available empirical potential energy functions. The MD-MC/CEM potential developed by DePristo and co-workers [6–8] is used for the Al–Al and Au–Au interactions. The Tersoff potential [9] is used for Si–Si interaction. The Al–Si and Au–Si interactions are described by a purely repulsive Molière potential.

To obtain the average yields, the 1950 trajectories are calculated except for Au_2 projectile where 150 trajectories are simulated. The integration of any run is terminated either when no particle within the target volume has sufficient energy to eject, or on when a cutoff time of 3 ps was reached. Because the action in the substrate is much larger in the case of Au_2 bombardment at $\theta = 45°$ than for the other systems, the average yields in this case are calculated with 150 trajectories with a cutoff time of 8 ps. At the end of each simulation, the atoms and clusters, which have velocities directed towards vacuum and are at the distances of more than 6 Å above the original sample, are regarded as ejected. All the Si atoms linked in the clusters are taken into account as a cluster fraction of the total yield. The short time dissociation of clusters is neglected.

3. Results and discussion

The average total yield, monomer yield, and cluster yield of silicon atoms are given in Table 1. The average yields are calculated as the raw number of corresponding sputtered Si atoms divided by the total number of trajectories. The enhancement factor is calculated by $k_{2,1} = Y_2/2Y_1$, where Y_1, Y_2 are the corresponding yields induced by atomic and dimer bombardment at the same impact velocities for the given type of projectile atom, respectively. The cluster fraction is calculated as a ratio of the number of atom comprising the sputtered clusters to the total number of sputtered Si atoms.

It is seen in Table 1 that $k_{2,1} > 1$ when going from the atomic to dimer bombardment except for the sputtered silicon monomers. The bombardment by Au_2 vs. Al_2 causes more non-linear enhancement of both the total and cluster yields. In case of the Au_2 projectile almost all sputtered atoms are linked in clusters. The monomer yield does not exhibit any non-linear enhancement for either aluminum or gold bombardment.

The yield distributions of ejected Si atoms over the given number of simultaneously sputtered Si atoms are shown in Fig. 1 for atomic and dimer bombardment. It is seen that the onset of high yield events takes place for dimer bombardment. In the case of Au_2 bombardment, the yield increase is caused mainly by the trajectories in which more than 40 atoms are ejected. Trajectories that give rise to a large number of sputtered particles are, of course, the ones that are most probable for large cluster formation. The mechanistic reasons for the large number of Si atoms ejected simultaneously at the event with Au_2 bombardment are given below.

The production of high yield events and, as a consequence, the enhanced sputtering yield, are unambiguously connected with the amount of energy deposited in the target and the degree of the energy localization with respect to the depth and the bulk [10]. The difference between the masses of the Al (27 amu) and Au atom (197 amu) is large. As result, completely different collision events after impact of the heavy gold atom and the light aluminum one on the

Table 1
Sputtered silicon atom yields and corresponding $k_{2,1}$ under bombardment of Al, Al_2, Au, and Au_2 projectiles with the initial energy 1.5 keV/atom at $\theta = 45°$

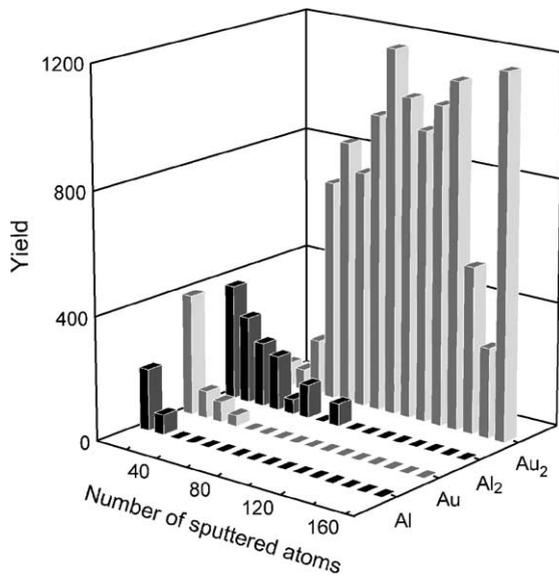| Projectile	Total yield	$k_{2,1}$-total yield	Cluster yield	$k_{2,1}$-cluster yield	Monomer yield	$k_{2,1}$-monomer yield	Cluster fraction
Al	3.0		1.1		1.9		0.37
Al_2	10.0	1.7	6.3	2.9	3.7	1.0	0.63
Au	5.3		3.2		2.1		0.60
Au_2	74.0	7.0	69.3	10.8	4.7	1.1	0.94

Fig. 1. Yield distributions of secondary silicon atoms under atomic and dimer bombardments.

silicon (28 amu) substrate evolve. This difference already becomes apparent in the beginning of the trajectory evolution. Heavy gold atoms have less initial velocity than light aluminum ones at the same E_0 and stay near the surface for a longer time. The

mass ratio of Au and Si atoms is 7 times; consequently only part of the energy can transfer in a single collision. Thus one Au atom can transfer its energy to many Si atoms. The maximum scattering angle for an Au atom hitting a Si atom is only $8°$. The Au atoms do not diverge much from their original paths. Moreover, the two Au atoms remain together as a dimer for a relatively long time. The concept that the Au atoms maintain their identity as a dimer is shown more clearly in Fig. 2. The separation distances expressed in units of the equilibrium internuclear distance between constituents for the each dimer, r_0 vs. the depth of dimer center mass penetration in 28 fs after impact are plotted for the 150 trajectories of Al$_2$ and Au$_2$ bombardments. It is seen that upon penetrating into the substrate the Al$_2$ constituents quickly disintegrate on the initial part of the trajectory, acting independently thereafter. In contrast, the Au$_2$ constituents stay together. The subsequent cascades overlap with a large probability and many Si atoms are displaced in a small region. As a result, at the given initial energy and incident angle, in the case of Au$_2$ the deposited energy is localized within a relatively small narrow region of the target surface, where all atoms are set in motion. More damage is created and it is harder for either Au or Si atoms to penetrate channels
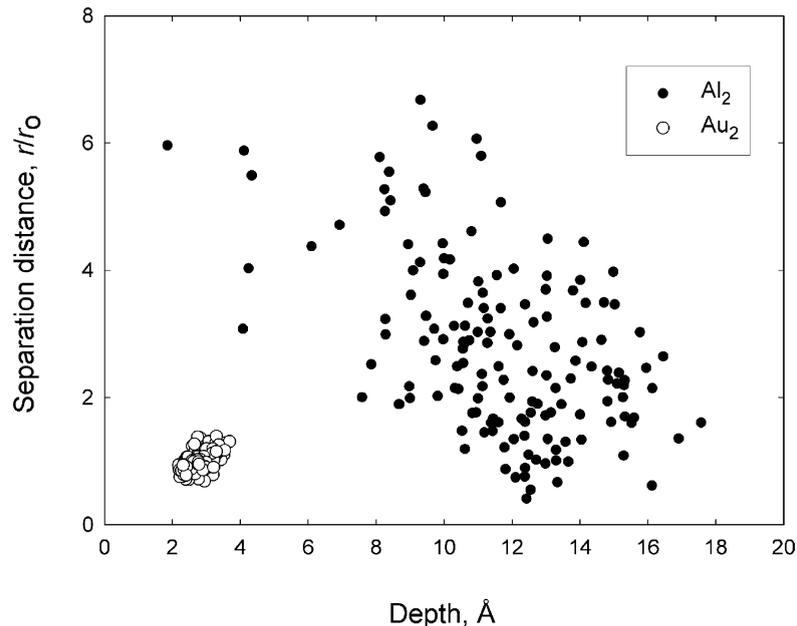

Fig. 2. Separation distance between dimer constituents vs. the depth of the dimer center mass penetration at 28 fs.

and escape from the collision region. The process of sharing and depositing energy is slow and more effective, thus increasing the probability of high yield events for enhancement of both the total and cluster yields.

4. Conclusion

The simulations show that the dimer projectiles with $E_0 = 1.5$ keV/atom at $\theta = 45°$ non-linearly enhance the total and cluster yields of Si. For the Au_2 bombardment, the enhancement factor for the yields is significantly larger than that in the case of Al_2 bombardment. The gold dimer constituents have smaller initial velocity than aluminum ones at the same E_0. Au atoms transfer small amounts of energy to numerous Si atoms, creating many slow recoils near the surface. The atoms of the gold dimer stay in the proximity of each other for a much longer time than the constituents of an aluminum dimer. Cascades created by them overlap with a large probability. All these factors increase the density of deposited energy in the subsurface region. The probability of high yield events becomes large.

Acknowledgements

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