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Molecular dynamics study of the fluence dependence of Si sputtering by 1 keV Ar^+ ions

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Abstract

The fluence dependence of the Si sputtering by 1 keV Ar^+ has been studied by molecular dynamics simulations. To this purpose, previously amorphized samples with different initial argon concentration have been ion bombarded and the sputtered atoms have been analyzed. The calculated sputtering yield increases with the argon content according to the experimental results. The mechanisms involved in this sputtering enhancement are discussed.

1. Introduction

The sputtering of atoms from a solid due to ion bombardment is a common process related to some technological steps in the fabrication of integrated circuits. Also plays a key role in surface analysis techniques like Secondary Ion Mass Spectroscopy (SIMS). In particular, the sputtering of silicon by noble-gas ion bombardment has been studied both experimental and theoretically [1-7]. It is well known [1] that when silicon is bombarded by noble-gas ions, such as argon or xenon, the sputtering yield initially increases with the dose, and finally, steady state conditions are reached. This behavior could be associated to the incorporation of the ions near to the target surface and to its amorphization under high dose bombardment [1,2,4]. In this work, we have investigated on the fluence dependence of the sputtering enhancement in Si under Ar^+ bombardment using the molecular dynamics technique.

Although computer simulations have been carried out to study the mechanisms involved in the sputtering process [6,7], direct comparison with experimental results is not straightforward. In those simulations, each ion is assumed to enter a perfect lattice, with no argon atoms inside. This only could account for a very low dose bombardment, which is different from the experiment. In fact, to our knowledge, there are no experimental sputtering measurements of Ar^+ bombarded Si under these low dose conditions. Simulating the accumulation of ions near the surface and the amorphization of the lattice due to the succesive ions impacting on the target would spend a very large CPU time. In this work, as an approach to this process, we have simulated the 1 keV Ar^+ bombardment of previously amorphized silicon samples that contained different initial concentrations of Ar up to 22 at.%. In this way, we have studied the sputtering process and its dependence on the fluence through the amount of argon atoms contained in the sample.

2. Sample preparation

The samples used in this work were prepared mixing a previously amorphized Si sample with an argon gas, and letting the system relax and stabilize. As for the amorphous Si sample, it was obtained by simulating the melting and subsequently quenching of an 8000 atom (100) Si crystal. The details were described elsewhere [8] (Sample A). The Si-Si interaction has been modeled only using the Stillinger-Weber [9] potential. For the Ar-Ar interaction we used a Lennard-Jones potential with the parameters currently adopted [10]. The Ar-Si interaction is probably repulsive in nature [11] and has been assumed to behave as the Universal Potential [12]. The boundary conditions were free for the two surfaces normal to the subsequent incidence direction of the argon ions and periodic for the four surfaces parallel to this direction.

Once the amorphous sample was prepared, we put inside a spatial uniform distribution of argon atoms. On this process, some argon atoms lay very close to silicon atoms giving rise to very strong repulsions which do not represent in fact the real ones inside the sample. To relax the sample we scaled the velocities of all the particles at

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zero every 3 fs for 10 ps. In the next step we scaled the velocities every 200 fs for 40 ps to reach a temperature of 300 K. During this step, the volume of the sample was expanded in the directions normal to the free boundary conditions surfaces (the total expansion depends on the amount of argon atoms used in each sample). Finally, a final stabilization step was performed for 10 ps. In this way several samples with different argon concentration were prepared: 10 at.% (Sample B1), and 22 at.% (Sample B3). During the step of scaling at 300 K, argon atoms near to the free surfaces released from the sample so that there are no argon atoms left in these two zones (aproximately 5 Å).

In the sample B1 argon atoms are observed to be uniformly distributed, except in the mentioned surface zones. On the contrary, in the sample B3, which would represent a higher argon dose implantation, argon bubble formation takes place, in good agreement with some experimental results [5,13]. It has been observed experimentally that for low ion doses, the argon atoms become randomly implanted throughout the whole sample, and when the dose is increased, argon atoms precipitate and form bubbles due to the low solubility of argon in silicon. This bubble formation has been observed experimentally even at room temperature and at fluences of 10^{14} ions/cm^2 for keV Ar^+ ions on Si [5].

3. Simulation results

The samples previously prepared were bombarded on one of the free surfaces with 1 keV Ar^+ ions at normal incidence. The total sputtering simulation time was set to 0.5 ps. As the two-body term of the Stillinger–Weber potential describing the Si–Si interaction does not represent accurately enough that interaction at energies above a few eV, we have used the Universal Potential of Ziegler and Biersack at short distances. The connection between them was performed using a spline. The size of the sample was made large enough for the results to be insensitive to the boundary conditions in the surfaces parallel to the initial direction of the ions. To check this, we prepared a test sample with the same argon content as the sample B1, but using free boundary conditions in all surfaces. This test sample was bombarded on a selected zone in the center of the surface to avoid boundary effects. The statistical results were the same for the two samples. On the other hand, samples with the same boundary conditions and argon content than B1, but different sizes, were prepared and argon bombarded. For the samples equal to and larger than B1 the results were size independent.

For each sample, we have simulated about 2000 ions, which entered the sample through impact points chosen at random. Accounting for the silicon or argon atoms ejected, we obtain the sputtering yield as the ratio between the number of atoms ejected and that of the simulated ions. The sputtering yields for each of the samples previously prepared are shown in the Table 1. The total sputtering yield increases with the argon content as it was expected, and this enhancement is due both to the ejection of argon and silicon atoms.

In Fig. 1 the distributions of atoms ejected per single ion (ASI) are shown. For the sake of comparison, the distributions for the samples A and B1 are represented together (Fig. 1a), and the same stands for the samples A and B3 (Fig. 1b). As it can be seen, the presence of argon atoms inside the silicon sample reduces the number of ion trajectories that produce no atom ejection and clearly

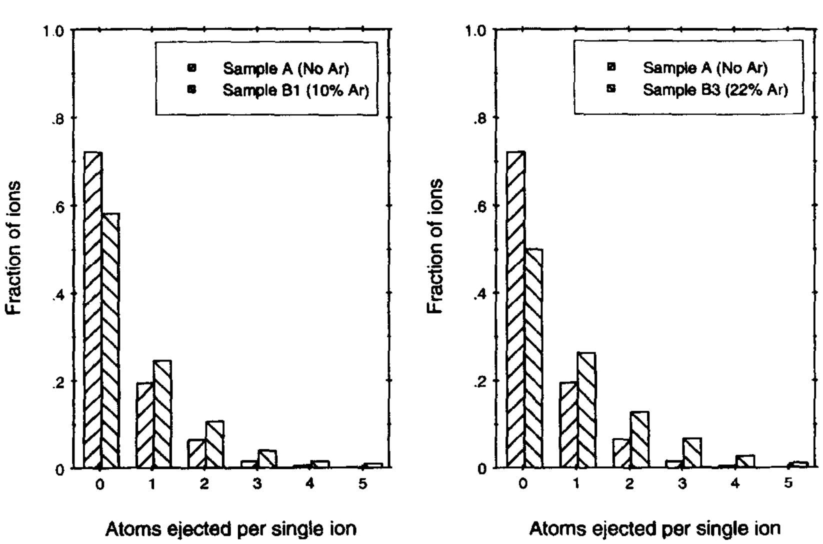

Fig. 1. Distributions of atoms ejected per single ion (ASI). (a) Distributions for the samples A and B1, (b) distributions for the samples A and B3.

Table 1
Total sputtering yield and relative Si and Ar sputtering yields for the samples with different argon content

Sample	Total sputtering yield	Relative Si yield (ejected Si atoms/ions)	Relative Ar yield (ejected Ar atoms/ions)
A (no Ar)	0.39	0.39	–
B1 (10% Ar, matrix)	0.71	0.54	0.17
B3 (22% Ar, bubbles)	0.93	0.68	0.25

increases the number of ions ejecting more than 1 atom. This effect is more noticeable in the case of the sample B3, containing the highest percentage of argon atoms. In fact, the fraction of ions that produce the ejection of more than one atom for the samples A, B1 and B3 are 0.08, 0.17 and 0.24 respectively. So, we can say that the sputtering enhancement observed in Table 1 is probably due to the increase of the number of high yield trajectories.

Furthermore, looking at the surface positions of the incident Ar^+ ions, we have observed that more than 85% of the ions ejecting several atoms impact precisely on zones with a high argon concentration just below the surface. These ions interact with the argon atoms and they can be backscattered or can produce the release of some Ar atoms. The stress exerted at the surface in those cases enhances the sputtering yield, as it has been suggested by Blank and Wittmaack [1], and Wittmaack [4].

Finally, in Fig. 2 the contribution of the different yield events to the total sputtering yield (Fig. 2a) as well as that separated of silicon (Fig. 2b), argon (Fig. 2c) are shown. Fig. 2a represents the ratio of the number of atoms ejected in events that produce the sputtering of one or several atoms relative to the total number of ions. For the amorphous sample without argon (A) the total sputtering yield was 0.39 (Table 1) and the main contribution corresponds to atoms being ejected alone (0.19). Each of the events producing the sputtering of more than one atom has a lower contribution, as can be seen in the figure, and the sum of them is 0.20. On the contrary, for the sample B3, that have the highest Ar concentration, the contribution to the total sputtering yield (0.93) of events ejecting 1 atom is only 0.26 while the fraction of the yield due to higher yield events is 0.67. As a conclusion, the sputtering yield enhancement in silicon samples containing argon seems to be mainly related to the increasing number of high yield ion trajectories when the argon content is raised, as it has been mentioned above.

This sputtering enhancement is due both to the ejection of silicon and argon atoms and in Fig. 2b and 2c, respectively, the contribution of them to the sputtering yield in the different yield events are shown. As for the silicon (Fig. 2b), the contribution of impacts ejecting one atom to the sputtering yield is similar for the three samples shown in the figure. This similarity suggests that silicon atoms

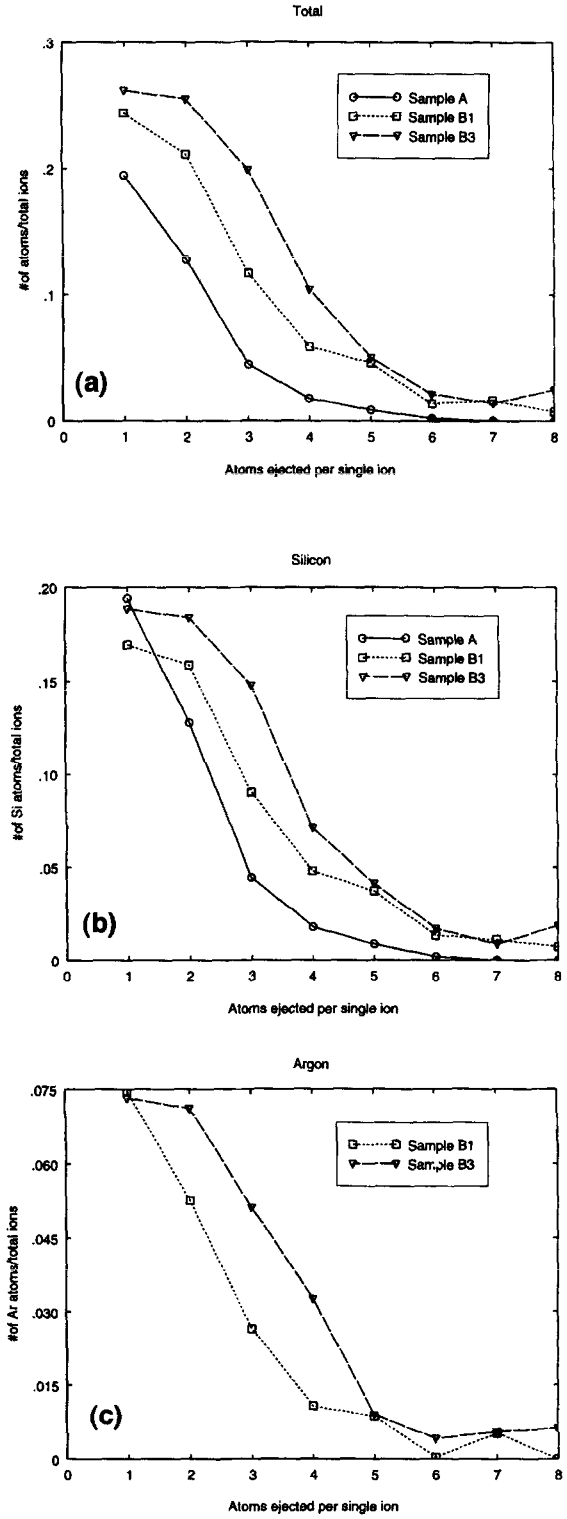

Fig. 2. Contribution of the different yield events (a) to the sputtering yield. Contributions of the (b) silicon and (c) argon atoms to the sputtering yield.

ejected alone are not directly related with the presence of argon near the surface. The differences appearing in the higher yield impacts, however, are presumably due to the argon atoms that during the collision cascade exert stress at the surface, enhancing the ejection of silicon atoms.

The contribution of the ejected argon atoms to the sputtering yield for the samples B1 and B3 is shown in Fig. 2c. Most of the argon atoms from the sample B1 have been ejected alone, which can be explained because in this sample the argon is uniformly distributed through the whole sample and there are no bubbles inside. On the contrary, in the sample B3 most of the argon atoms in the sample B3 are grouped in large bubbles. Hence, when a particle collides with one of these bubbles it is likely to produce the release of more than one argon atom from the bubble. These atoms may produce the ejection of some silicon atoms closer to the surface and may be finally ejected from the sample. Therefore, the role of argon atoms in the sputtering enhancement is double: the argon atoms ejected from the sample increase the total sputtering yield and also push out some silicon atoms thus raising the silicon relative sputtering yield. For the samples used, it has not been observed the experimental effect of the saturation of the sputtering yield, which probably occurs for higher doses.

4. Conclusions

Simulations of 1 keV Ar^+ bombardment on silicon samples with different argon contents have been performed to study the fluence dependence of the sputtering yield experimentally observed. These samples have been prepared starting on an 8000 atom amorphous silicon sample, putting inside a gas of argon and letting the whole sample relax and stabilize. The argon concentration of the resulting samples ranged from 10 to 22 at.% and argon bubbles have been formed for the higher concentration samples.

The sputtering yield calculated under 1 keV Ar^+ bombardment increases with the argon content. This enhancement is related mainly to the raising number of ions producing high yield events. Most of these ions impact on argon-rich zones just below the target surface and can be backscattered or cause some argon atoms to move towards the surface. These atoms will push out some silicon atoms closer to the surface and can eventually be sputtered from the sample, in the two cases producing an increase of the sputtering yield, according to the experiments.

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