Statistical characterization of surface defects created by Ar ion bombardment of crystalline silicon

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Ion bombardment of crystalline silicon targets induces pattern formation by the creation of mobile surface species that participate in forming nanometer-scale structures. The formation of these mobile species on a Si(001) surface, caused by sub-keV argon ion bombardment, is investigated through molecular dynamics simulation of Stillinger-Weber [Phys. Rev. B **31**, 5262 (1985)] silicon. Specific criteria for identifying and classifying these mobile atoms based on their energy and coordination number are developed. The mobile species are categorized based on these criteria and their average concentrations are calculated. © *2008 American Institute of Physics*.
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I. INTRODUCTION

Ion bombardment of silicon targets at moderate temperatures drives pattern formation by the creation of mobile surface species that can diffuse and assemble into nanometer-scale structures. When the energy of the ion is transferred to the target, some of the substrate atoms are sufficiently energized to break all of their bonds and leave the surface (sputtered atoms), while some other substrate atoms break their bonds partially, but remain attached to the surface. These atoms, also known as adatoms, are relatively mobile. It has also been shown that, on Si(001), adatom pairs, or "ad-dimers" may form more stable configurations than those of single adatoms.[1]

Surface evolution equations, like that developed by Bradley and Harper,[2] are the basis for most of the theoretical studies in ripple formation due to ion bombardment and they depend upon local concentrations of mobile species. According to the Bradley-Harper model, the time rate of change of the average height of the surface can be obtained by

$$\frac{\partial h}{\partial t} = S\kappa + B\frac{\partial^2}{\partial s^2}\kappa, \tag{1}$$

where s is the surface arc length measured perpendicular to the ion beam direction. The roughening prefactor is given by $S=-(fa/n)Y_0(\theta)\Gamma_2(\theta)$ in which f is the ion flux, a is the average depth of ion energy deposition, n is the atomic volumetric density, $Y_0(\theta)$ is the sputter yield as a function of the angle of incidence and $\Gamma_2(\theta)$ is a coefficient governing the erosion rate dependence on the local surface curvature κ. The relaxation prefactor is $B=D_sC\gamma/n^2k_BT$, where γ is the surface energy density, k_B is the Boltzmann constant, T is the temperature, D_s is the surface diffusivity, and C is the concentration of mobile species which take part in the surface diffusion.[2,3] It can be seen from Eq. (1) that among all the parameters involved in surface evolution, only the product D_sC is unknown. Elrebacher *et al.* showed that D_sC does not always obey an Arrhenius form, but under high flux and high temperature, C (number of mobile species/area) is not thermally activated. They suggested that instead it follows the rate equation:

$$\frac{\partial C}{\partial t} = fY_1 - fC\sigma_c - \frac{C}{\tau_1} - \frac{2C^2}{\tau_2}, \tag{2}$$

where the mobile species are considered to be ad-dimers, f is the ion flux, Y_1 is the number of ad-dimers created per incident ion, σ_c is the area of the surface over which the ion's damage is deposited, τ_1 is the characteristic time for an ad-dimer to diffuse to a surface trap and τ_2 is the characteristic time for two ad-dimers to merge which can be expressed in terms of C, D_s, the surface lattice parameter and the number of surface sites.[3]

As Eq. (2) indicates, Y_1 is a key parameter that remains to be determined, which raises the question of how to identify mobile atoms on an ion bombarded surface. Textbook definitions of adatoms (e.g., Ref. 4) are applicable to simple cubic crystals. For more complicated atomic structures such as the diamond structure, equilibrium configurations of adatoms and ad-dimers have been predicted on a perfectly crystalline surface.[5,6] However, on surfaces damaged by ion bombardment, identifying and distinguishing the various defects created is not straightforward. Some adatom definitions are based on their distance from the ion bombarded surface. For instance, in a molecular dynamics study of Al ion bombardment, all of the atoms that fall between half of the NN distance and twice the cut off radius of the interatomic potential from the surface are categorized as adatoms.[7] This method is not applicable for studying damaged Si(001) because it is unclear how to define a reference coordinate for a highly damaged surface. Moreover, as depicted in Fig. 1, the position of an adatom on a Si(001) surface can be equivalent to the level of the initial surface height. Finally, these basic definitions fail to distinguish between various types of mobile surface species.

The goal of this paper is to develop a definition for mobile species on a damaged surface and apply it to the case of

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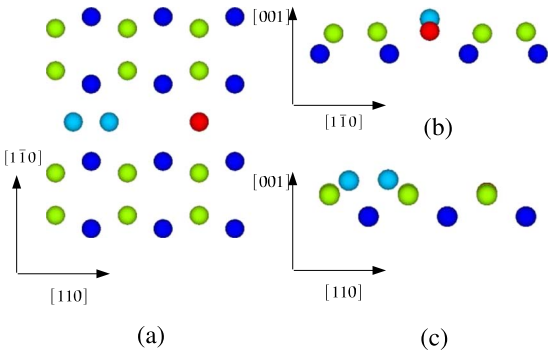

FIG. 1. (Color online) Typical configurations of mobile species observed from (a) [001], (b) [1$\bar{1}$0] and (c) [110] directions. Green atoms are the dimers on a reconstructed Si(001) surface. Light blue atoms show a typical ad-dimer and the red atom is a typical adatom.

ion bombarded Si(001) to predict the types and concentrations of these species. We investigate the mechanism of adatom creation via statistical analysis of molecular dynamics simulation results of many single ion impacts on a (001) surface of Stillinger-Weber[8] silicon. We develop specific criteria for identifying the mobile atoms based on their energy and coordination numbers. The formation of adatoms and ad-dimers as well as other types of surface defects is observed. The mobile species are categorized based on our criteria and their average concentrations are calculated for the cases of 500, 700, and 900 eV incidence energy.

II. SIMULATION METHODOLOGY

The bombardment target in the simulation has 2400 silicon atoms in a $3.84\times3.84\times3.08$ nm^3 box with a Si(001) $\times(2\times1)$ reconstructed surface and lateral periodic boundary conditions, as shown in Fig. 2. The Stillinger-Weber potential (SW) (Ref. 8) is used to model silicon-silicon interactions. Although SW has proved an effective tool in modeling bulk silicon, it has some shortcomings in reproducing a number of surface phenomena. However, it has been shown to model the (001) surface reasonably well.[9] The widely used Moliere[10] potential is used to model the argon-silicon interactions. The energy of the incident argon ion is taken to be 500, 700, or 900 eV. A thermostat based on velocity rescaling[11] is applied to a few bottom layers of the target to guide the temperature to 550 K, although the results are ultimately shown to be fairly insensitive to the substrate temperature.

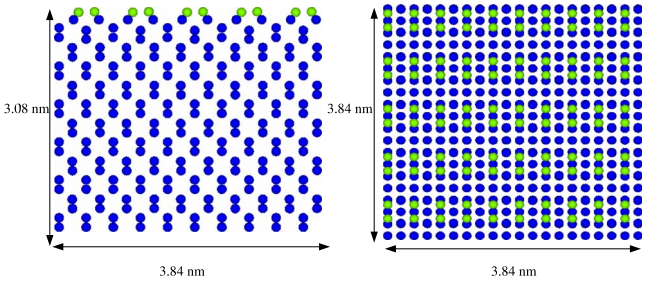

FIG. 2. (Color online) The [1$\bar{1}$0] and [001] views of the simulation box. The surface dimers are shown in green.

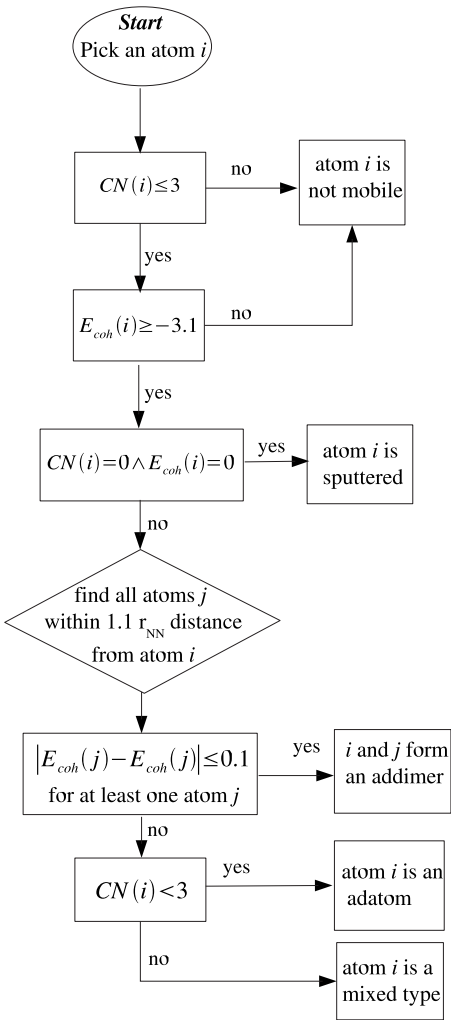

FIG. 3. Summary of the defect diagnostic algorithm.

Figure 1 depicts the basic types of mobile surface defects on a perfect Si(001) surface, which is otherwise clean and has well defined energies and coordination. To automatically count the surface species created during bombardment, we construct diagnostics based on these basic forms, considering both their cohesive energies and coordination numbers. The cohesive energy of a silicon atom in the bulk structure is known to be 4.3 eV and it has four NNs (i.e., its coordination number, *CN*, is equal to 4). On the dimerized Si(001) surface, a surface atom has three NNs (and, thus, one dangling bond) and its cohesive energy must be less than that of a bulk atom because it is not in the perfect diamond structure. The Stillinger-Weber[8] potential gives cohesive energy of 4.3 eV for a bulk silicon atom and 3.2 eV for a surface atom. We calculate the number of NNs of an atom by counting the atoms that are within the $1.1r_{NN}$ of the atom of interest where $r_{NN}\approx2.35$ Å represents the NN distance in silicon. By this method the calculated coordination numbers of a bulk atom and a surface atom are 4 and 3, respectively. Surface mobile species should have lower cohesive energies and coordination numbers. Our simulations show that either one of these criteria alone cannot identify the adatoms and ad-dimers. For instance, after an impact many examples can be found in which two atoms have a *CN* equal to 3 (or approximately the

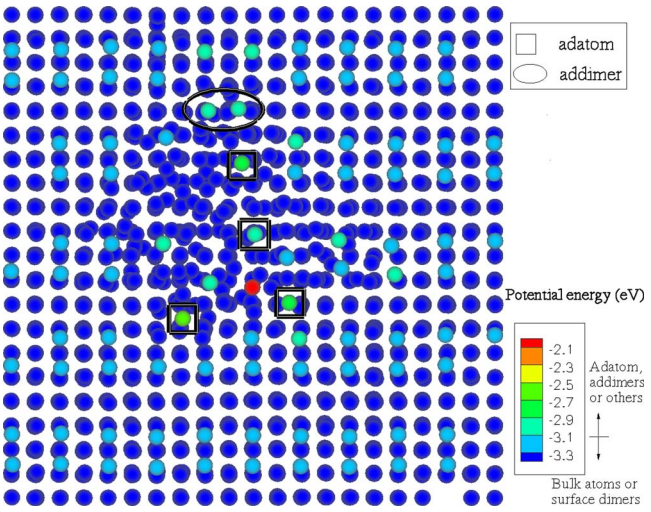

FIG. 4. (Color online) The [001] view of the simulation box after a single 500 eV impact. The color scheme indicates the cohesive energy of the atoms which is part of the defect diagnostic. An ad-dimer pair and four adatoms are observed. Also, there are eight mixed types that have high potential energy but are not classified as either adatom or ad-dimer participants. It can be seen that most of the unclassified atoms are still part of surface dimers and some are low energy/high *CN* atoms in the damaged area. The red atom is the implanted ion.

same cohesive energy) but one is in an ad-dimer while the other is a regular surface atom. Thus, we combine the two factors of *CN* and cohesive energy to develop a unified defect diagnostic.

Figure 3 shows the summary of the defect diagnostic. To apply this diagnostic method, all atoms with E_{coh} and *CN* less than a regular surface atom (in a surface dimer) are considered to be potentially mobile and are tagged first. Among these atoms those with zero cohesive energy and *CN* are regarded as sputtered atoms and are not considered in the future stages. In the next step, ad-dimers are identified using the fact that both atoms in an ad-dimer pair should have approximately the same cohesive energy. Thus, among all of the atoms of interest obtained from the first step, NNs are examined and if only one atom among the NNs can be found with nearly the same energy the two atoms qualify as an

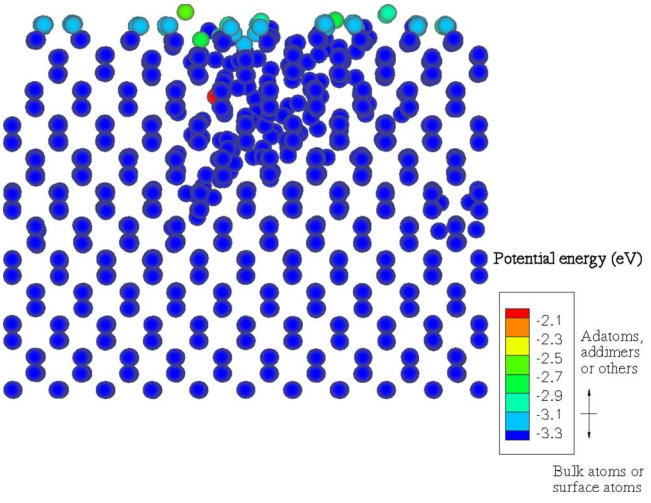

FIG. 5. (Color online) The $[1\bar{1}0]$ view of the domain shown in Fig. 4.

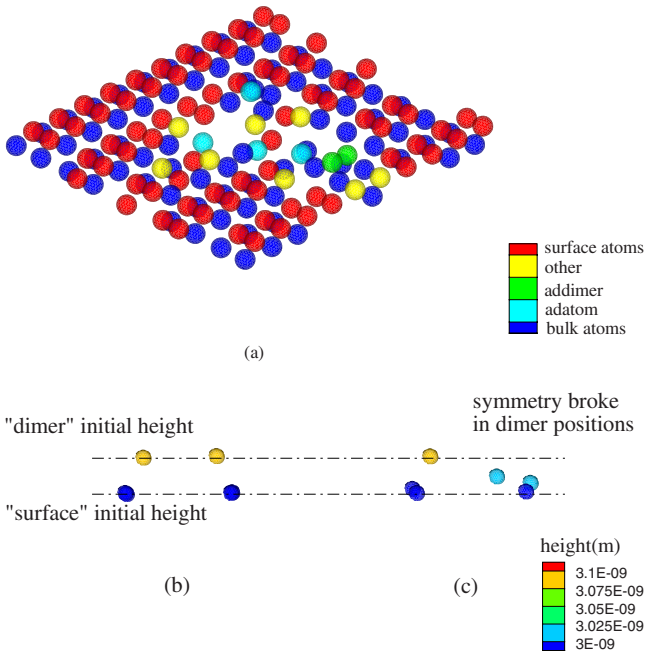

FIG. 6. (Color online) (a) Typical adatoms, ad-dimer, and other surface defects created after a single impact. Atoms below the surface are omitted for clarity. A closer look at a typical defect of other type (b) initially as a dimer and (c) after a single ion impact. These species are excited surface atoms that do not gain enough energy to break their original bonds.

ad-dimer pair. In the next stage the remaining atoms are considered and only atoms with *CN* less than 3 are characterized as adatoms. At this point all sputtered atoms, ad-dimer pairs, and single adatoms are identified. However, there may be some atoms among the total interesting species that remain unclassified. These atoms might be atoms of a broken surface dimer or excited surface atoms that may eventually become adatoms or return to their lattice sites.

III. RESULTS AND DISCUSSION

We statistically study the average single impact response of the Si surface, shown in Fig. 2, by performing molecular dynamics over 95 ensembles for each of the three incidence energies. In each individual impact case, the position of the ion is chosen randomly within the range of a surface unit cell. Figures 4 and 5 show the results of a typical single impact. The existence of adatoms, ad-dimers, and other high

TABLE I. Average values of mobile species concentrations. The total number of surface atoms is equal to 100 in each of the 95 ensembles of 500 eV bombardment over which the statistics are collected. The sputter yield agrees with experimental data (Ref. 12) and the sum of ad-dimer atoms and adatoms agrees well with the estimate of Erlebacher *et al.* (Ref. 13).

	Total high potential energy atoms	Sputtered atoms	Ad-dimer pairs	Adatoms	Other
Average value	11.73	0.51	0.88	2.65	6.71
Standard deviation	6.26	0.67	1.06	2.30	4.4
Standard error	0.64	0.07	0.11	0.24	0.45

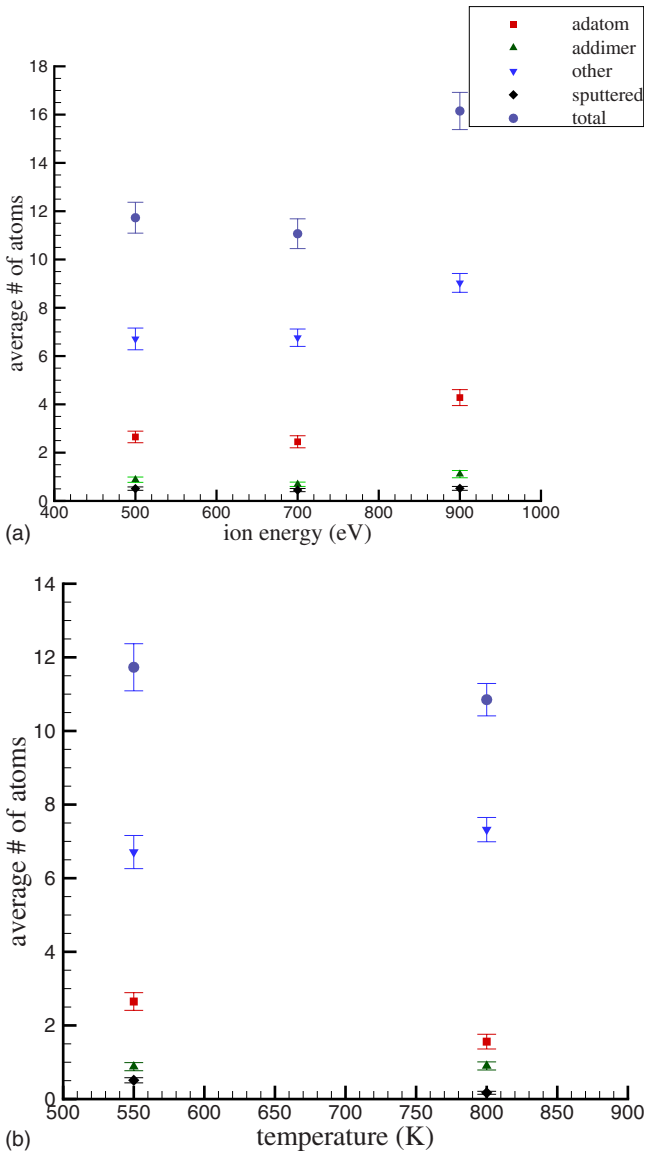

FIG. 7. (Color online) Statistics of a single ion impact for different values of ion energy at 550 K (left) and substrate temperature for 500 eV bombardment (right). Error bars indicate the standard error.

potential energy species is observed. An "other" type atom has higher potential energy and the same *CN* as a surface atom. Therefore, based on our algorithm, the relatively high potential energy criterion qualifies the "other" atoms as interesting species but the high *CN* distinguishes them from adatoms. Figure 6 provides a closer look at typical "other" atoms. We note that this category includes excited surface atoms that do not gain sufficient energy to break their bonds but instead, displace slightly, breaking the symmetry of their original dimerized positions.

Table I contains the values of sputter, adatom, and ad-dimer yield that are calculated by averaging the results of 95 simulations of 500 eV bombardment. The sputter yield value obtained here agrees well with previous results.[12] The total average number of atoms in adatom and ad-dimer forms is 4.41 from Table I. Since there are 100 total surface atoms, the concentration obtained by our method is in good agreement with the estimate of 4% suggested by Erlebacher *et al.*[13]

The above calculations are repeated to compute the statistics of a single impact for different values of ion energy and substrate temperature and the results are shown in Fig. 7. The results show there is a mild variation in the numbers of different surface species identified by our metrics but there is not strong sensitivity to either impact energy or temperature.

IV. CONCLUDING REMARKS

The goal of this paper is to identify and calculate the concentration of different mobile species created by 500, 700, and 900 eV argon ion bombardment on the (001) surface of SW silicon. We developed a defect diagnostic to identify and classify different mobile types based on their cohesive energies and coordination numbers. We then counted different types of mobile species created by statistical analysis of molecular dynamics simulation results. It is shown that upon bombardment, mobile species are created in the form of adatoms, ad-dimers, and some mixed configurations. The number of such species produced is insensitive to temperature for the range considered. The average concentrations agree well with experimental sputter yields. The average concentrations of mobile species could be used in solving rate equations such as Eq. (2). Also, the statistical effects of single ion impacts could be incorporated into Monte Carlo type models to predict long time behavior of the surfaces during ion bombardment.

ACKNOWLEDGMENTS

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