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Surface Trapping of Hyperthermal Particles: A Molecular Dynamics Simulation

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We have studied the trapping probability of hyperthermal particles impacted on the surface by molecular dynamics simulations. The angle dependence of the trapping probability, the structural factor of the impacted surface and the sputtering yields have been calculated. We find that, for the normal incidence, the trapping probability decreases with the incident energy, while for 45° incidence, the trapping probability shows a non-monotonic behavior in agreement with a recent experiment.

1. Introduction

The interaction of energetic particles with a surface is fundamental to a number of thin film deposition techniques and applications of the ion beam processing. Due to the technical importance, the implantation of high energy particles was intensively studied. However, relatively fewer studies have been done for the process of energetic particles impacting on a surface in the hyperthermal energy (energy between a few eV and a few hundred eV) region [1 to 7]. The particles with the hyperthermal energy impacted on the surface could lead to very complicated processes such as surface trapping, near-surface implanting, and even deep implanting. It is well known that the trapping of the particles can greatly affect the properties of a deposited film, so it is important to study the interaction process of energetic particles with the surface, especially the trapping probability for energetic particles.

Generally the trapping probability decreases with increasing incident energy, which has been confirmed by many experiments. This behavior of the trapping probability of energetic particles is understandable. The particles with low incident energy easily transfer a large fraction of their kinetic energies to the substrate so that they do not have enough energy to scatter back. Hence a large trapping probability for low energy incidence is expected. For high energy incidence, the particles have enough energy to recoil back, so the trapping probability decreases. However, a very recent work by Goodstein et al. [8] indicated that the surface trapping probability shows a non-monotonic behavior at a large incident angle $\theta = 45°$, while at a small angle $\theta = 8°$ it still decreases monotonically with the increasing energy. Although their classical trajectory calculations obtained the qualitative trend similar to the experimental observation at $\theta = 45°$, the angle dependence of the trapping probability was not studied. Moreover, the ignorance of the force between the surface atoms makes it impossible to understand thoroughly how the surface atoms respond to the energetic particles, which could be important for the trapping process.

In this paper, based on molecular dynamics (MD) simulations, we have studied the trapping probability of hyperthermal particles at the surface. For simplicity, the interaction between surface atoms and the interaction between energetic particle and surface atoms are described by a model potential, which will not prohibit us to get a general trend. We find that for a large angle incidence, the surface trapping probability decreases non-monotonically, while it shows a monotonic behavior for normal incidence. The non-monotonic behavior of surface trapping probability could be attributed to the onset of surface atoms being kicked off.

2. Simulation Details

The evolution of the system with time is determined by the numerical integration of the Newton equation, i.e., the MD simulation. The f.c.c. (100) surface is modeled by eight atomic layers with each atomic layer containing 100 (10×10) atoms. The bottom four atomic layers of the simulation cell are kept at their equilibrium positions to mimic the underlying solid. The periodic boundary conditions along the lateral directions are imposed to simulate the infinite surface.

To get a quantitative agreement with experimental results on the trapping probability, the proper description of the interaction between surface atoms, and the interaction between surface atoms and the energetic particle, is crucial. However, it is extremely difficult to get an accurate classical potential to describe such a complex process in which the charge transfer could take place. So in our present simulations, for simplicity, we just take a very simple potential, the Lennard-Jones potential, to model the interaction between energetic particle and the surface atoms. The interaction between surface atoms is also modeled by the Lennard-Jones potential, where the parameters are fitted to copper. The potential has the form

$$V(r_{ij}) = 4\epsilon_{\alpha\beta}\left[\left(\frac{\sigma_{\alpha\beta}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{\alpha\beta}}{r_{ij}}\right)^{6}\right], \tag{1}$$

where ϵ is the bond energy and σ the atomic size. α and β denote the incident particle A and the surface atom B, respectively. In all calculations, we assume $\sigma_{\mathrm{BB}} = 1.0$, $\sigma_{\mathrm{AB}} = 0.76$, and $\epsilon_{\mathrm{AA}} = \epsilon_{\mathrm{BB}} = 1.0$, $\epsilon_{\mathrm{AB}} = 0.5$. In reduced units, the time $t = (m\sigma^2/\epsilon)^{1/2}$, and the temperature $T^* = kT/\epsilon$, where m is the atomic mass ($m_{\mathrm{A}} = m_{\mathrm{B}} = 1$), k the Boltzmann constant. From this simple model, we do not expect that the obtained results can quantitatively agree with experimental data, however, they could shed some light on understanding the microprocess for the energetic particles impacted on the surface.

In our simulations, the surface atoms are coupled to an external heat bath by using the Langevin MD [9]. In this scheme, the force on the atoms consists of three terms, the gradient of the potential energy, a friction term proportional to the velocity of the particles and a random force exhibiting a Gaussian distribution. The latter two terms are not independent of each other, which keep the temperature of the system to be constant. The friction coefficient is chosen to be 0.1 [9]. However, the force on the energetic particle stems only from the gradient of the potential surface and no friction is applied to it. The simulation temperature is kept at $T^* = 0.02$, the equation of motion is integrated by a modified Beeman procedure [10] with a time step $\Delta t_{\mathrm{MD}} = 0.005$.

The energy dependence of the trapping probability at two incident angles, $\theta = 45°$ and $\theta = 0°$, has been studied. Since most kinetic energy of the energetic particle is transferred to the surface in a very short time (200 to 500 MD time steps), we consider the energetic particle to be trapped at the surface if it remains at the surface after 2000 MD time steps. The trapping probability, the possibility of an energetic particle to be trapped at the surface with an incident energy, is obtained from the average of 5000 impact events. The structural factor of the surface after the energetic particles have impacted has also been calculated.

3. Results and Discussions

The trapping probability, P_{tr}, for the normal incident particles decreases with the energy, as shown in Fig. 1. We have calculated the surface trapping (above the first layer of the surface) and subsurface trapping (below the first layer of the surface), respectively. At very low incident energy, P_{tr} is close to unity, but it decreases quickly as the incident energy increases, while the subsurface trapping increases with the incident energy.

We observed a non-monotonic behavior of P_{tr} for the incident angle $\theta = 45°$ in our simulations, as shown in Fig. 2. Similarly, nearly all the particles are trapped at very low incident energy, while P_{tr} decreases quickly as the incident energy increases up to about 70. P_{tr} curve shows a peak at energy around 80. Another small peak at energy around 160 can also be observed, while in the experiment (see Fig. 4a of Ref. [8]) a pronounced peak of the surface trapping is at about 140 eV. In fact, if one looks into the experimental results carefully, there is a small bump around its double energy 280 eV. In this sense, our present simulations have qualitatively reproduced the basic behaviors of the surface trapping, although it is hard to quantitatively compare the present results with experimental data.

It is interesting to note that P_{tr} for the normal incidence is always larger than that for $\theta = 45°$ case (see Figs. 1 and 2), in agreement with experimental observation [8]. A comparison of the subsurface trapping for the different incident angles shows that the subsurface trapping starts at a lower energy for normal incidence, which could be attributed to the easier surface penetration or implantation.

Figure 3 presents the sputtering yields (SY) changing with the incident energy. We define SY as the average number of surface

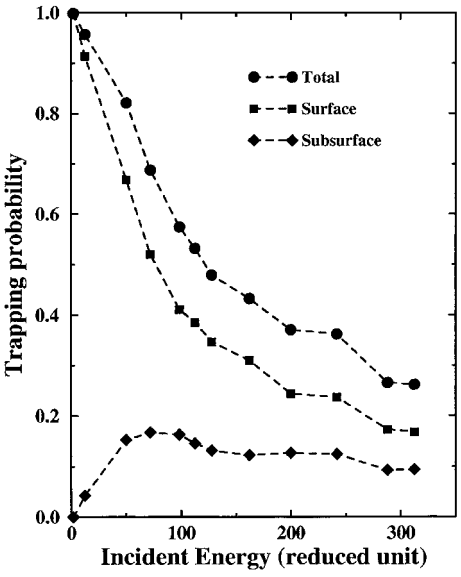

Fig. 1. Trapping probability of the energetic particles with normal incident. The total and surface trapping probabilities monotonically decrease with the incident energy, the substrate trapping begins at low incident energy

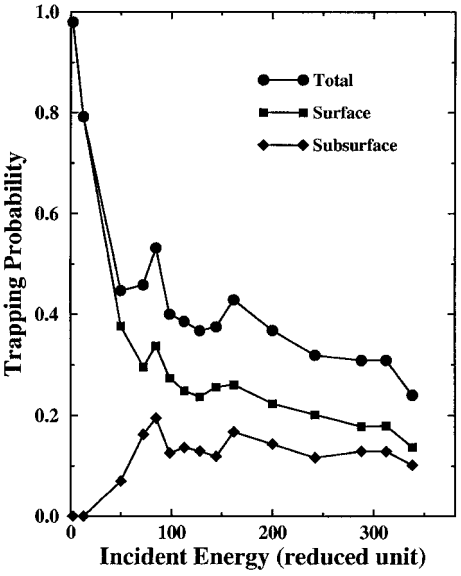

Fig. 2. Trapping probability of the energetic particles with the incident angle $\theta = 45°$. The total and surface trapping probabilities show non-monotonically decreasing behaviors

atoms being kicked off. For the normal incidence, the onset for surface atoms to be kicked off starts at around energy 100, SY increases linearly with the increasing energy. In this figure, we have also shown the probability of kicking off two or even more atoms, which starts at the energy 160 and also increases almost linearly with energy. However, for the case with incident angle $\theta = 45°$, the change of SY with incident energy shows an interesting behavior. SY becomes non-zero at around energy 70, a lower energy than that for the normal incidence, and its linear increasing stops at around 120. From 120 to 160, SY increases with energy slowly, while after that, it increases almost linearly. This means that, energetic particles impacted at θ = 45° have more chance to kick surface atoms off at the low incident energy, while they have less chance in the high energy region.

The non-monotonic decrease of P_{tr} for the incident angle $\theta = 45°$ could be attributed to the sudden increase of SY. The sudden increase of SY at energies 80 and 160 indicates more energy transfer from the incident particle to surface atoms, which leads to an increase of P_{tr}. For the normal incidence, however, the onset of the increasing SY

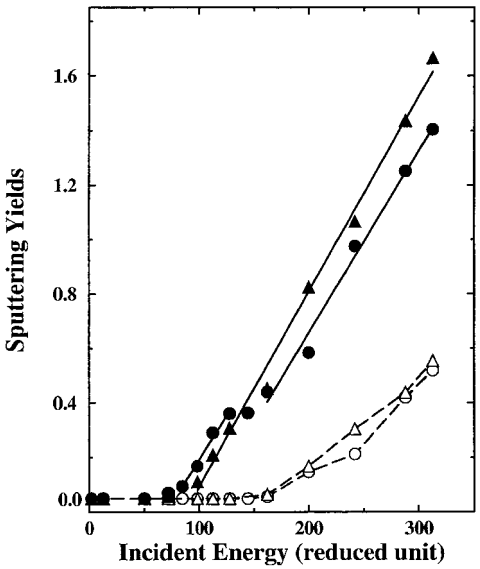

Fig. 3. Sputtering yields vs. the incident energy. Circles: incident angle θ = 45°, triangles: normal incidence. The solid lines are guides for eyes. The filled symbols show the total sputtering yields and the empty symbols indicate the probabilities of kicking out two or more surface atoms. Different behaviors of the sputtering yields for the two angles can been observed from the figure

appears at a higher energy, the incident particles still have enough energy to recoil back, so P_{tr} does not necessarily show a peak.

The incident particles at $\theta = 45^\circ$ could corrugate the surface, the onset of non-zero SY at a low energy, which makes the particles unable to recoil back and leads to the appearance of the first peak of P_{tr}. When the energy increases up to about 120, the corrugation of the surface does not help to kick more surface atoms off and thus SY does not change significantly. When the energy increases further, SY increases again, and a sudden increase of energy transferred to the surface leads to another small peak for the surface trapping.

Different sputtering yields have different effects on the surface structure. We use the structural factor $S_l(K_\alpha)$ as a parameter to measure the structural order of the top two atomic layers,

$$S_l(K_\alpha) = \left\langle \frac{1}{n_l} \sum_{i \in l} \mathrm{e}^{iK_\alpha \cdot r_i} \right\rangle ; \qquad \alpha = x, y , \tag{2}$$

where, $K_x = (2\pi/a_0)(2,0,0)$ and $K_y = (2\pi/a_0)(0,2,0)$, the angular brackets indicate an average over the simulated trajectories. The smaller $S_l(K_\alpha)$ is the more disorder of the surface structure exists. $S_l(K_\alpha)$ decreases to zero when the surface structure becomes completely disordered. The obtained results versus the incident energy for θ = 45° are shown in Fig. 4. A general trend, the higher the incident energy the smaller the structural factor, can be clearly observed. It is interesting to note the different slopes at different energy regions. $S_l(K_\alpha)$ decreases with a relatively small slope from the very low energy to an energy about 70 and from 120 to 160. While there is a larger slope indicating a more disorder of the structure, from 70 to 120 and from 160 to higher energy. In fact, these changes of $S_l(K_\alpha)$ can be easily correlated to SY. At low incident energy, the transfer of the kinetic energy to surface atoms makes the surface structure disorder thermally, the disorder is enhanced with the increase of the energy transferred from the particles. While SY becomes non-zero at energy 70, the surface structure changes with the occurrence of some defects. With the energy increasing, SY increases linearly and the number of defects increases, which leads to a disorder different from the thermal disorder at low energy, so $S_l(K_\alpha)$ decreases with a large slope. As the energy increases up to 120, SY increases slowly again, thus $S_l(K_\alpha)$ decreases slowly. When the energy increases

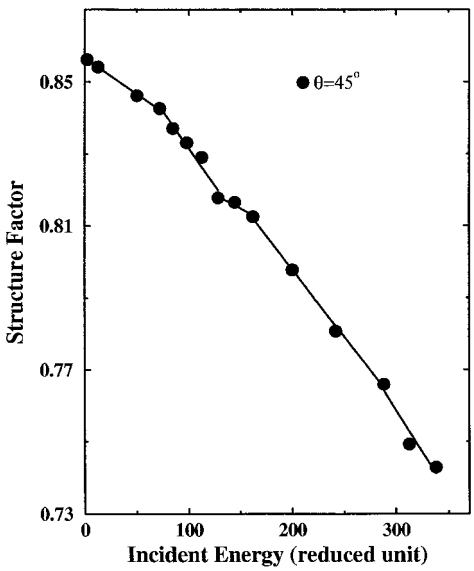

Fig. 4. Surface structural factors vs. the incident energy. Filled circles: calculated results, solid line: guide for eyes. Different slopes of structural factor at various energy regions imply different mechanisms of structure changes

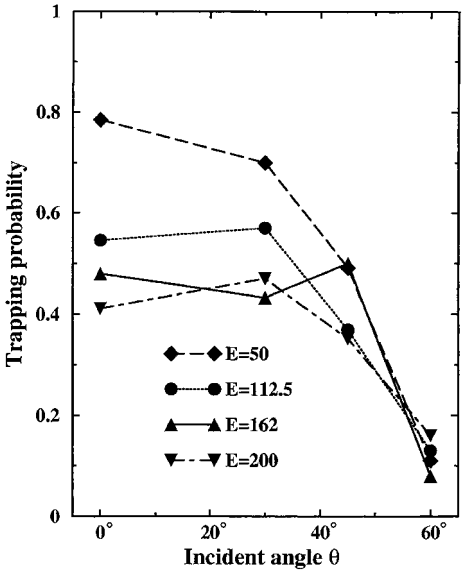

Fig. 5. Trapping probability vs. incident angle for different incident energies. The filled symbols are the present calculated results, the lines are guides for eyes. Except the normal incidence, the trapping probabilities show non-monotonic behaviors with the incident energy

to 160, SY increases linearly with energy, therefore $S_l(K_\alpha)$ decreases faster again.

The trapping probability shows a non-monotonic behavior with the incident energy. In Fig. 5 $P_{\rm tr}$ is shown for different incident energies at different incident angles. We can see, for non-zero incident angles, there are line crossings, which indicates a strong dependence of $P_{\rm tr}$ on the incident energy. Furthermore, it is confirmed that, except for normal incidence, $P_{\rm tr}$ is a non-monotonic function of the energy.

In summary, we have studied the trapping properties of energetic particles on a f.c.c.(100) surface by MD simulations. We have correctly produced the basic features of surface trapping probability as observed in the experiment. We do find a non-monotonically decreasing behavior of surface trapping for the incident angle $\theta = 45^\circ$ and a monotonic behavior for the normal incidence, both of which are in agreement with experimental results.

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