Dynamic Monte Carlo Simulation of Compositional and Topography Changes Induced by Ion Beam Irradiation

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A computer simulation model is presented for ion induced deposition and sputtering in two-dimensional topographic surfaces. The model combines dynamic Monte Carlo simulation of the collision cascade with surface topography simulation, which treats compositional changes self-consistently with the evolution of the surface. An application is presented: erosion and deposition of a ripple surface of carbon irradiated by tungsten ions. Due to the deposition of implanted particles and the redeposition of sputtered particles, the surface topography drastically changes. Sharp wedges are formed on the ripple surface. With increasing ion fluence and energy, the surface is dominated by an oscillatory feature, where the initial ripple structure disappears. Since the redeposition is more for a steep slope than for a gentle slope, the redeposition produces both a peaked region and a shadowed region on the surface against the next redeposition. The surface topography changes strongly influence the W concentration in a W-C mixed layer formed on the C bulk. [DOI: 10.1380/ejssnt.2006.32]

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I. INTRODUCTION

In recent research on dynamic ion-solid interactions, two different computer simulations, i.e., molecular dynamics (MD) simulation and dynamic Monte Carlo (MC) simulation, are performed to obtain useful information from comparing their results with experimental observations [1, 2]. The MC simulation has been conventionally used to describe the slowing down of energetic ions and particle emission such as ion reflection and sputtering. The MD simulation describes dynamic interactions involved in ion implantation more realistically, but require much larger amounts of computer capacity than the MC simulation.

High fluence ion irradiation modifies surface composition and topography. The former is due to deposition of the projectile ions and sputter erosion and mixing of the target and deposited atoms [3]. The latter partly occurs in terms of differences in the deposition rate and the sputtering yield as a function of the angle of incidence relative to the local surface [4].

In this article, we present a MC simulation code that treats compositional changes self-consistently with the evolution of the surface topography. To test the simulation model, it is applied for the topographic evolution of a ripple surface of carbon (C) bombarded by high-fluence tungsten (W) ions. With no reflection of projectile ions, the heavy non-volatile W ion incidence on the light C material strongly changes the surface composition with increasing ion fluence. At the fluence of \(10^{17}\) cm\(^{-2}\), where a steady state composition is reached, a W-C mixed material is produced at the energies of >1 keV whereas at low energy (<1 keV), there appears a thick deposition layer [5]. Therefore, another aim of this article is to reveal the influence of the surface compositional change and the redeposition of sputtered W and C on the evolution of the surface topography.

II. COMPUTER SIMULATION MODEL FOR COMPOSITIONAL CHANGE AND SURFACE TOPOGRAPHY DEVELOPMENT

The binary collision approximation method [5, 6] simulates the slowing down of projectile ions penetrating into a solid material and the associated formation of recoil atom cascades. A dynamic composition change arises from the deposition of implanted ions and the collisional transport of implanted and material atoms, assuming that each N projectile ions represents a differential ion fluence, \(\Delta \Phi\). The surface layer is divided into k slabs of constant thickness \(\Delta d\); in this article, \(\Delta \Phi = 10^{14}\) cm\(^{-2}\), \(N = 1000\), \(k = 10\) and \(\Delta d = 10\) nm.

The physical processes associated with the collisional transport of the projectile and recoils atoms, such as ion implantation, sputtering and atomic relocation, cause removal or deposition of particles in different layers. Each particle removed from (or deposited in) a layer causes a change in the partial atomic density of the component atoms. The fractional composition and the total atomic density in each layer are calculated by the reciprocal addition of atomic densities of pure elements according to local composition. For bombardment of carbon (C) with tungsten (W) ions, \((n_{W-C})^{-1} = f_W(n_W)^{-1} + f_C(n_C)^{-1}\), where \(n_{W-C}\), \(n_W\) and \(n_C\) are the atomic densities of the mixed, pure W and pure C solids, respectively, and \(f_W\) and \(f_C\) are the compositions of W and C in each layer, respectively; \(f_W + f_C = 1\). As a consequence the atomic volume ratio for the component atoms is the same as that of pure elements. This relaxation results in an altered thickness of the layer due to the lack of or excess densities of the component atoms. Before following the collisional transport of a new ion, the depth profile of the projectile ions and the change in total thickness of the surface layer, i.e., the surface erosion (or deposition), are calculated.
The recombination processes of the vacancies and interstitial atoms generated in the collisional mixing [3] are not taken into account. The surface binding energy of the mixed material is not known as a function of the composition. The simplest assumption is to use a binding energy [3], which varies linearly between the cohesive energies of pure elements, e.g., 7.37 eV for C and 8.90 eV for W, according to surface composition.

At elevated temperatures, the surface composition changes will be influenced by their diffusion in the bulk material and phase formation such as WC and W2C. The simple procedure applied for an inclusion of the diffusion effect, is to combine the dynamic MC model with an analysis of the Fickian equation [7]. After simulating the projectile implantation and collisional mixing with ∆Φ, the implantation profile is used as an initial profile in the Fickian equation with the diffusion time ∆t = ∆Φ/Γ, and the profile after diffusion serves as input for the calculation with the next ∆Φ, where Γ is the flux of projectile ions.

The dynamic MC simulation for compositional change, surface erosion and deposition combines with topography simulation. The structured surface is divided into segments by a two-dimensional rectangular grid with a segment size of 10 nm×10 nm. In this combined program, it is assumed that the topography changes of the surface occur only in terms of the differences in the deposition rate and sputtering yield as a function of the local angle of incidence relative to neighboring segment surfaces. The topography simulation starts with surface erosion (or deposition) and composition change with ∆Φ at each segment. The local angle of incidence with a next ∆Φ is the slope calculated with the difference of eroded depth (or deposited thickness) between neighboring segments on both side, except for the bottom and the top of the structured surface where the angle against the surface normal is zero. The next ∆Φ with the local angle causes erosion (or deposition) at each segment and changes composition. This sequence is performed M times to obtain the surface topography evolution with projectile ion fluence, which is proportional to time, where total ion fluence is used instead of ∆Φ.

To avoid non-correctable computer errors on eroded depth (or deposited thickness) Δy due to finite ∆Φ and limited number of N simulating ∆Φ, an averaging technique [8] in the calculation of Δy, i.e., (Δyn−1 + 2Δyn + Δyn+1)/4, is used instead of Δyn at the n-th segment. This correction are based upon the practical situation that the erosion and deposition processes are dictated by a collision cascade of finite dimensions and an impurity diffusion [3], so that it may be unrealistic to consider that each segment is unaffected by neighboring sputtering and diffusion.

The ion bombardment of the surface causes particle emission such as projectile reflection and sputtering from each segment. Since the angle of the reflected projectiles and sputtered atoms distributes, some of them redeposit (or re-enter) on another segment after moving above the surface. Such redeposition process from the mixed material surface is also calculated using the trajectory simulation of the particles emitted from each segment. Therefore, the redeposition changes the surface topography and composition.

In this article, to test the simulation model, it was applied for the topographic evolution of a ripple surface of C with a sinusoidal profile, a = cos(2πx/b), as shown in Fig. 1 (thick line): a = 0.25 μm and b = 0.5 μm. The whole surface was bombarded in the negative y-direction by W ions with the energies from 0.5 keV to 10 keV and the total fluence of 2 × 10^{17} cm^{-2}, in difference conditions which are the local deposition of ions, local sputtering of implanted and target atoms and local redeposition of sputtered atoms.

III. APPLICATION TO W ION IRRADIATION TO A C RIPPLE SURFACE

As long as both no deposition of implanted projectiles and no redeposition of sputtered particles are assumed, the topographic evolution of the surface is determined only by the angle dependence of the sputtering yield within our simulation. With increasing angle mea-
FIG. 2: Development of ripple C surface with W deposition and of W atomic fraction \( (f_W) \) distribution in C at shallow region during (a) 0.5 keV and (b) 5 keV W ion irradiation. The thick line represents the initial ripple surface \( (\Phi = 0) \). Upper figure: the surface topography, lower figure: the W atomic fraction, \( f_W \), in the depths of <10 nm.

sured from the surface normal, the sputtering yield increases. However, at an oblique angle of \( 75^\circ \sim 85^\circ \) for W ion impacts on C, the yield reaches maximum values, 3.6 and 16.0 atoms/ion at 0.5 keV and 5 keV, respectively; whereas for normal incidence the yields are approximately zero and 0.6 atoms/ion, respectively. With further increasing angle, it falls zero due to increasing projectiles reflected from the surface just after incidence. Therefore, as shown in Fig. 1(a), the surface topography shrinks with ion irradiation at most area of the surface but the apical point is a wedge-shape with glancing angle due to strong erosion at inclined neighboring points.

At high energies where the sputtering yield is not ignored at normal incidence, the apical point also shrinks (Fig. 1(b)). The apical point is sharpened to form a wedge with increasing fluence, and with further increasing fluence, the wedge is divided into many wedges. This is due to the steep change in the sputtering yield at grazing angles, which makes additional wedges neighboring the original wedge. Furthermore, a small roughness due to statistical fluctuation in the MC calculation grows during further irradiation and as a result, it strongly influences the surface topography.

In addition to the sputter erosion, Fig. 2 shows the development of the ripple C surface with the deposition of projectile W ions and the W atomic fraction \( (f_W) \) distribution in C at shallow (<10 nm) region. For normal incidence, the surface is dominated by the W deposition due to the small sputtering yields, so that a thick W-rich layer is formed on the C bulk. For oblique incidence, however, the strong sputtering erodes W deposited on the inclined surface, so that a W-C mixed layer with small \( f_W \) is formed. For low energy (Fig. 2(a)), the W deposition flattens more, although the apical point is not influenced whether the W deposition is taken into account or not. It should be noted that the W concentration, \( f_W \), at each position of the surface is strongly affected by the surface topography.

As shown in Ref. [5], even for high energy bombardment (Fig. 2(b)) the flat surface is deposited by W at the early stage of irradiation (\( < 2 \times 10^{17} \text{cm}^{-2} \)). The W deposition localizes the collision cascades near the surface, resulting in the enhanced C sputtering. With increasing ion fluence, therefore, a transition from the W deposition to the surface erosion occurs where a steady state is reached for the W concentration, \( f_W \), in the C bulk. The erosion rate is more for the W-C mixed layer than for the C bulk, due to the enhanced C sputtering and W
self-sputtering. As a result, the $f_W$-distribution changes due to the development of the surface topography.

Figure 3 shows the topography changes of the ripple C surface after ion irradiation ($2 \times 10^{17} \text{cm}^{-2}$) due to the redeposition of sputtered particles, in addition to the projectile W deposition. The redeposition of W sputtered from the surface drastically changes the development of the surface topography. Sharp wedges are formed all over the ripple surface at low energy (0.5 keV, Fig. 3(a)), whereas at high energy (10 keV, Fig. 3(d)) the surface is dominated by an oscillatory feature, where the initial ripple surface disappears and the surface is only featured by oscillations. The oscillatory feature is strong for high sputtering yield and for high oblique angle where the maximum yield occurs. The formation mechanism of the surface oscillation is that the redeposition produces both a peaked region and a region shadowed against the next redeposition on the surface. This is because the redeposition is more for a steep slope than for a gentle slope. After the formation of the oscillatory structure, the surface erosion is suppressed all over the surface whereas the structure continues to change with ion irradiation. In our case, the calculation results are less influenced by reflection of projectile ions at the surface due to the heavy non-volatile ion (W) incidence on the light material (C). Nevertheless, the reflection should be taken into account at a steep oblique angle.

Figure 4 demonstrate the relationship between the surface topography and the redeposition profile at the early stage of irradiation of 5 keV W ions. At the first stage of irradiation (Fig. 4(a)), only bulk C atoms redeposits at the inclined surface. With increasing ion fluence (Figs. 4(b), 4(c) and 4(d)), the W redeposition is obtained due to additional sputtering of W deposits. The oscillatory surface enhances the redeposition of the next sputtered particles, and then enhances the amplitude in the oscillation. The oscillations in the number of W and C redeposits, which are strongly correlated with the surface topography, occur at the same phase as each other.

When the surface is inclined, the sputtering yield of redeposited W is increased. Therefore, $f_W$ is small at the inclined surface, whereas at the tops and bottoms of the oscillatory surface it is large. As a result, $f_W$ oscillates in the same phase of the oscillatory surface structure at
both shallow (< 10 nm) and deep (40 nm ~ 50 nm) regions in the bulk, as shown in Fig. 5.

IV. CONCLUSIONS AND DISCUSSION

We developed a MC simulation model that treats both the dynamic composition change and the surface topography development during ion irradiation on a solid surface. The model was applied to the W ion induced erosion and deposition on a ripple surface of C where the redeposition of the sputtered projectile (W) and target (C) atoms on the surface are taken into account.

For no deposition of implanted W projectiles and no redeposition of sputtered C and W particles, the ripple structure shrinks at inclined area of surface but the apical point is a wedge-shape with glancing angles. When the W deposition occurs, the surface topography change is concerned with the W concentration, $f_W$, in C. The redeposition of the sputtered particles drastically changes the development of the surface topography again. The wedges are formed all over the ripple surface and the surface is dominated by an oscillatory feature, where the initial ripple structure disappears. Since the redeposition is more for a steep slope than for a gentle slope, the redeposition produces both a peaked region and a shadowed region on the surface against the next redeposition.

Although the reflection of projectile ions less influences due to the heavy non-volatile ion incidence on the light material, it should be taken into account at a steep oblique angle. At elevated temperatures, thermal effects, such as impurity diffusion, induce the W composition change in the C bulk, resulting in the changes of the surface topography, which is not shown here. In recent long-pulse operation of magnetic fusion devices, prompt or plasma-transported redeposition of impurities changed the net erosion rate of plasma facing materials, but also modified the surface morphology. The surface morphology of redeposited materials strongly depended on the plasma bombardment condition [9]. Furthermore, the erosion rate of
a thin boron film (a-B:H) under carbon deposition was not constant with time at a fixed position and the non-uniform deposition and topographic change were observed [10]. In the designed International Thermonuclear Experimental Reactor (ITER), C and W are used as the diverter components, which results in the bombardment of C (or W) by sputtered and subsequently ionized W (or C) [5]. Therefore, the compositional and topographic changes of W and C during the operations are a critical issue in the ITER project. These plasma-surface interactions, which include similar problems to the ion processing for device applications, are a subject of the evaluation of the present models and assumptions in our simulation code, so that the existing and planned observations in surface measurements will be compared with our calculation in the next step of our code development.

Finally, if the deposited and redeposited particles are sufficiently mobile, they tend to agglomerate [4]. The surface mobility and W and C agglomeration is perhaps one process to form growing islands on the surface. This may be an important process for the surface development but it is a future problem that our simulation model should take.
into account. Nevertheless, the deposition of projectile ions and the redeposition of sputtered particles strongly influence not only the surface topographic development but also the dynamic surface concentration change with high fluence ion irradiation.

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