SECTION 1

PHYSICS OF RADIATION DAMAGES AND EFFECTS IN SOLIDS

SURFACE DIFFUSION INDUCED BY LOW-ENERGY BOMBARDMENT WITH He IONS: AN EXCHANGE MECHANISM

O.V. Dudka[1], A.A. Mazilov[1,2], J.A. Gordienko[1]
[1]National Science Center "Kharkov Institute of Physics and Technology", Kharkov, Ukraine;
[2]Belgorod State University, Belgorod, Russia
E-mail: alexey.mazilov@gmail.com

The radiation-induced surface diffusion of molybdenum adatoms was studied using molecular dynamics simulations based on a many-body tight-binding potential interpolated to the short-range repulsive screened Coulombic interactions. It was shown that the He ion impact is accompanied by an extensive surface mobility of Mo atoms. The long radiation-induced atomic jumps, spanning more than a nearest-neighbor distance, were revealed on the {110} terrace. The radiation induced exchange of Mo atoms colliding with Mo {110} surface was found in our mathematical simulations: there were observed exchange processes in which the radiation excited atom entered the surface and another surface atom emerged nearby. These results of MD simulations appear to be the first observation of exchange events in radiation-induced surface diffusion.

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INTRODUCTION

The key materials science problem in designing fusion reactors is associated to radiation stability of the first wall and divertor materials. In particular, the surface of these materials is bombarded with high doses of low-energy He ions and neutral atoms which cause significant erosion [1, 2]. Basic problems foreseen at the operation of fusion power reactors are connected with the ability of structural materials to retain their properties under heavy radiation environments. The bombarded surface can be very beneficial for healing radiation damage by trapping and recombining point defects. A firm understanding of the effect of surface diffusion on materials is necessary to serve as a guide for creating novel advanced materials. Radiation tolerant materials would exploit the presence of surfaces to act as the recombination area and suppress damage accumulation.

Recent understanding of the atomic level response of solid materials to radiation yields insights into surface-defect interactions enable for atomic scale design of radiation tolerant structural materials for advanced fission and future fusion reactors. However, advanced fission and future fusion reactors call for significant progress materials able to sustain radiation doses up to the order of magnitude higher than in existing nuclear reactors. Motivated by the progress in understanding surface morphological events, and by the technological need of having a detailed information on the ion-bombarded surfaces, recently a number of experimental and theoretical studies have been conducted to evaluate the structure and microtopography of surfaces eroded by low-energy ion bombardment [3, 4].

The impact-induced surface diffusion has been first revealed in early field ion microscopy researches on the low-energy ion bombardment [5–8]. This method gives the direct information with an atomic resolution on various elemental surface processes [9–12], but it is impossible to image short lived configurations and diffusion pathways. For this purpose, MD simulations have been used. At low temperatures the surface diffusion is dominated by its radiation-induced component, which is independent of temperature. Despite over 40 years' theoretical and technological interest in this phenomenon, there are still very little direct atomic-scale observations of elementary events of ion-induced surface diffusion. The MD simulations of corresponding cascade events provide a basis for understanding the special effects of surfaces including phenomenon of radiation-induced surface diffusion. In spite of the known wide possibilities of MD simulations studies of the ion-surface impacts, the most of them have primarily been concerned with the sputtering events, and not with surface damage and impact-induced diffusion.

Statistically significant information on an atomic scale concerning the surface damage production by impacts of ions of inert gases was obtained in conjunction with STM experiments [13]. In this paper, the impact-induced surface diffusion of molybdenum adatoms has been studied using MD simulation. Molybdenum was chosen as the construction material for the divertors in present prototype nuclear fusion reactors and the plasma-facing structure material for future reactors due to its high melting point and physical sputtering yields [1]. As a result, a new mechanism of radiation-induced surface diffusion based on the exchange of surface atoms was revealed.

SIMULATION MODEL

In this paper we used a software package "Kalypso" for MD simulation of projectile collisions with metallic targets, which is described in [14]. This package is well-suited for MD simulation of bombardment phenomena in surface atomic layers that involves short time and length scales ($< 10^{-8}$ s, $< 10^{-7}$ m). Kalypso comprises user-friendly integrated suite of software instruments based on many-body potentials for design and analysis

of simulation experiments that relate to ion impacts or atomic recoil events. Noble gas atoms interact with the target metal atoms via repulsive screened Coulombic potentials. Interactions between the target atoms are defined by composite potentials, consisting of an attractive a many-body tight-binding potential interpolated to a short-range repulsive screened Coulombic potential [15].

The equations of motion of the many-body dynamical system are integrated over certain number of time-steps using the Verlet method. In our MD experiments in most cases the free boundaries conditions were used. An adaptive time-step is used to improve the speed of the MD simulation. The size of simulated crystallites was changed by varying their vertical and lateral dimensions over a wide range. The simulated Mo nanocrystal contained $2 \cdot 10^4$ atoms on the average. The crystallite size was chosen to be large enough to include most collision cascades. The direction normal to the Mo (110) surface was set as the z-axis, and the tangential directions ([-110] and [001] directions) were set as the x-axis and y-axis, respectively. The excellent agreement between the experimental data and Kalypso MD simulations is replicated across the wide ion energy range. Minor differences persist between results obtained by these methods, but the overall agreement is encouraging. The projectile impact coordinates for the simulations were randomly distributed across the entire crystallite surface.

The initial time step for integration of the Newton equations was chosen in the range of 0.01…0.1 fs. The duration of the surface collision cascades was in the range of 500…1000 fs. After this period, there is not sufficient kinetic energy for further atoms be sputtered or displaced in the lateral direction.

RESULTS AND DISCUSSION

The MD simulation experiment showed that the projectile impacts on the upper surface of the target were followed for about 0.5 ps by collision cascades. However, the morphological evolution of the bombarded target caused by atomic recoils was followed until about 10 ps have elapsed. The surface morphology after irradiation at normal projectile incidence is dominated by random adatom coverage and not by surface vacancies. The obtained data demonstrated that the low-energy He atomic bombardment is able to induce a substantial mobility of adatoms. The atomic displacements in the surface layers responsible for mass-transport along the surface are induced by linear atomic displacements along the <111> close-packed directions (shown by arrow in Fig. 1,b). Such linear displacements can be considered as dynamical crowdions or focusons.

In these MD experiments, the surface atom starts from a given site, and then due to radiation activation overcomes the surrounding migration barriers. So, it may make a flight and is thermalized in a surface potential well which most probably is one of the neighbors, thus making a single short jump (atom A in Fig. 1,b and Fig. 2) or a long-range jump (atom B in Fig. 1,b–d).

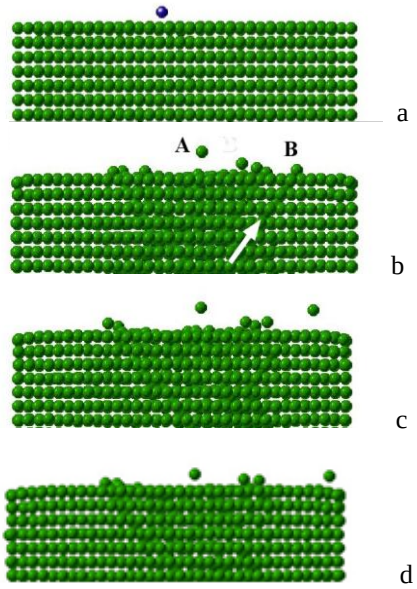

Fig. 1. Ion-impact MD patterns of Mo (110) surface bombarded by He (5 keV) at a normal angle after 2.4 fs (a), 233 fs (b), 360 fs (c), 495 fs (d), and 501 fs (e) from the beginning of the MD experiment.

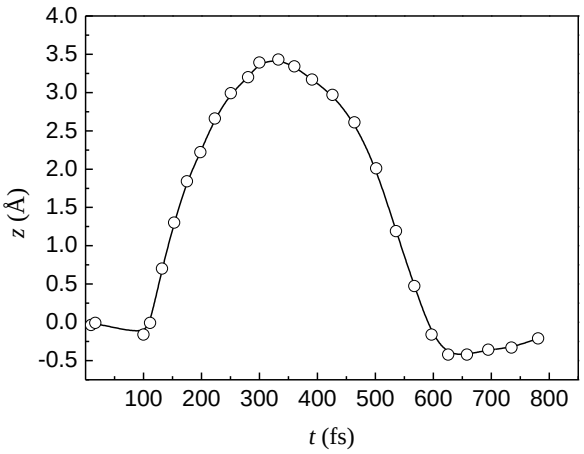

Fig. 2. Time dependence of the jump height for impact-induced diffusion of atom (pointed by A) to the nearest-neighbor position

The kinetic energy of displaced atoms (Fig. 3) may be high enough to induce the surface diffusion processes that may modify the atomic topography. The jumping atom can be also retrapped in a surface site which is far away from the starting point (the long-jump lateral replacement in this case, see the atom B in Fig. 1,b). As it is shown in Fig. 4, large surface jumps of atoms more than a nearest distance contribute meaningfully to the impact-induced diffusion. The nearest-neighbor diffusion jumps of molybdenum atoms are the utmost probable but the double or more complicated atomic jumps are significant. The MD data are satisfactory fitted by the exponential equation $N(L) = B\exp(-(L-L_0)/L^*)$, where N is the number of atomic hops; L_0 is the adjusted parameter; L^* is the reduced amplitude of the atomic jumps equal to 2.942; C is the dimensionless constant 61.39.

Long jumps of surface atoms during a thermally activated diffusion process, with length of several

surface lattice parameters, were first discovered by MD simulations [16], and later were revealed in FIM studies. The other evidences of long jumps during surface diffusion of adatoms and the mechanisms of this phenomenon were discussed in two comprehensive reviews [17, 18].

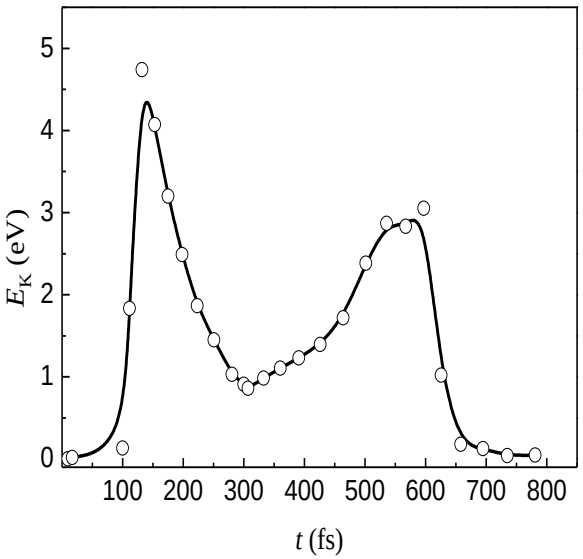

Fig. 3. Dependence of the kinetic energy for impact-induced diffusion of atom A to the nearest-neighbor position

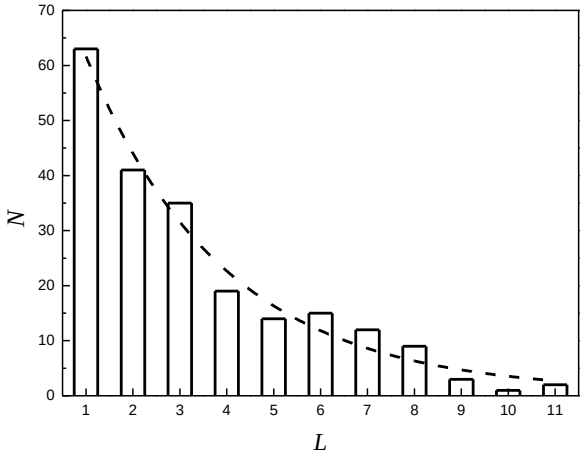

Fig. 4. The probability distributions of radiation-induced jump length of molybdenum atoms. The dash line is an exponential fit to the diffusion data

Our MD simulations of the impact induced surface diffusion reveal a variety of atomic jumps along bombarded surface, including the nearest-neighbor hops. This observation is agreed with results of FIM studies reported in [5, 6]. In this FIM study simulation of the low-energy ion bombardment processes, it was shown that the most atoms sputtered from an atomically rough surface can originate from adatom sites, and the rest from interior terrace regions. The exponential jump length probability distribution theoretically revealed and observed in MD experiments [19] for the thermal surface diffusion of adatoms is characteristic of friction due to discrete events rather than as a continuous process. So, the strictly exponential distribution of radiation-induced jumps obtained in our experiments indicates the instantaneous mechanism of interatomic interactions that drain the excess energy of adatoms by discrete events rather than as a continuous friction process.

At described above impact condition, we also found exchange processes in which the radiation excited atom entered the surface and another surface atom emerged nearby. Such an exchange event is illustrated in Fig. 5, which shows the same MD run as in Fig. 1 for the atom B. The excited migrating atom (shown by the arrow in Fig. 5,a) with kinetic energy of 6.5 eV enters the surface layer near the step (pointed by the arrow head in Fig. 5,b) and pushes aside two of atoms, the last of which emerges and transforms into an adatom on the {100} terrace (see Fig. 5,b,c). The arrows in Fig. 5,b and c show the direction of velocity of the emerged atom. An initial kinetic energy of this atom was 2.4 eV. The arrow head in Fig. 5,d points the position of this atom after thermolization and its termination. The hopping atom embedded in the {110} surface near the terrace edge (see Fig. 5.a,b) travels two surface sites down the {100} surface plane. Similar movement was also found in several other MD simulations.

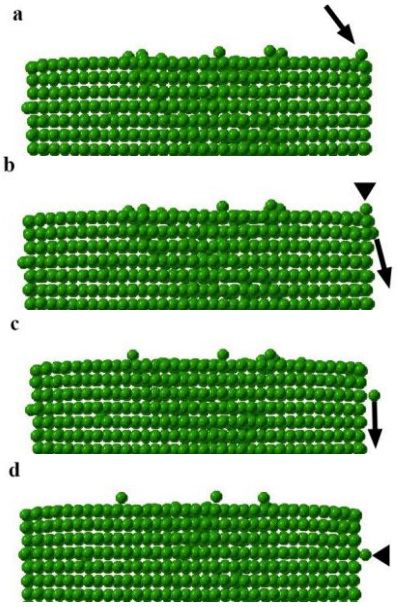

Fig. 5. The MD run patterns of bombarded Mo (110) in the continuation of the same experiment as shown in Fig. 1 after 536 fs (a), 595 fs (b), 710 fs (c), and 830 fs (d) from the He ion impact

An analogical exchange phenomenon was revealed and analyzed earlier by Evangelakis and Papanicolaou [20] and considered in details in several MD studies of thermally activated self-diffusion processes on copper surfaces [21–24]. They observed a complicated mechanism with the surface atom exchange processes with comparatively low energy barriers: a surface adatom entered the first monolayer, and another atom adjacent to the entering one popped out of the surface layer. This emerging adatom can substantially contribute to the surface diffusion. The total reduced length of diffusion hops for this atom L equals to the length L_h before the exchange event and after it L_{ex}: $L = L_h + L_{ex}$. The described above impact-induced long-range diffusion hopping is characterized by $L_h = 6$ and

$L_{ex} = 3$. So the total number of jumps for this single impact event equals to 9.

MD patterns shown in Fig. 5 appear to be the first observation of exchange events in impact-induced surface diffusion. From these MD simulations, it is clear that radiation-induced exchange events on metal surfaces may contribute meaningfully to surface diffusion. It must be stressed, however, that this conclusion is just the result of MD simulations without direct experimental confirmations at this time.

CONCLUSIONS

The elementary events of impact-induced surface diffusion on molybdenum surface at the atomic level were studied using MD simulations. It was shown that:

1. The He ion impact at normal incidence is accompanied by an extensive mobility of Mo surface atoms. The kinetic energy-induced hyperthermal mass-transport has a relevant influence on the development of the Mo atomic topography.

2. The long radiation-induced atomic jumps, spanning more than a nearest-neighbor distance, were revealed on the {110} plane at 0 K.

3. An exponential distribution of impact-induced jumps obtained in our experiments indicates the instantaneous mechanism of interatomic interactions that drain the excess energy of adatoms by discrete events rather than as a continuous friction process.

4. At described above impact condition, we also found exchange processes in which the radiation excited atom entered the surface and another surface atom emerged nearby. These results of MD simulations appear to be the first observation of exchange events in impact-induced surface diffusion.

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ПОВЕРХНОСТНАЯ ДИФФУЗИЯ, ИНДУЦИРОВАННАЯ НИЗКОЭНЕРГЕТИЧЕСКОЙ БОМБАРДИРОВКОЙ ИОНАМИ He: МЕХАНИЗМ ОБМЕНА

О.В. Дудка, А.А. Мазилов, Ю.А. Гордиенко

Радиационно-индуцированная поверхностная диффузия адатомов молибдена была изучена с помощью моделирования методом молекулярной динамики, основанном на многочастичном потенциале жесткой связи с интерполяцией короткодействующего отталкивания экранированным кулоновским взаимодействием. Было показано, что бомбардировка ионами He сопровождается обширной поверхностной подвижностью атомов Mo. Длинные радиационно-индуцированные атомные скачки, охватывающие расстояния более чем ближайшие межатомные, были выявлены на {110} террасах. Радиационно-индуцированный обмен атомов Mo, сталкивающихся с поверхностью Mo {110}, был обнаружен в нашем математическом моделировании: наблюдались обменные процессы, в которых радиационно-возбужденный атом проникал в поверхностный слой, а поблизости выходил на поверхность другой атом. Эти результаты моделирования представляют собой первое наблюдение обменных эффектов в радиационно-индуцированной поверхностной диффузии.

ПОВЕРХНЕВА ДИФУЗІЯ, ІНДУКОВАНА НИЗЬКОЕНЕРГЕТИЧНИМ БОМБАРДУВАННЯМ ІОНАМИ He: МЕХАНІЗМ ОБМІНУ

О.В. Дудка, О.О. Мазілов, Ю.О. Гордієнко

Радіаційно-індукована поверхнева дифузія адатомів молібдену була вивчена за допомогою моделювання методом молекулярної динаміки, що базується на багаточастковому потенціалі жорсткого зв'язку з інтерполяцією короткодіючого відштовхування екранованою кулонівською взаємодією. Було показано, що бомбардування іонами He супроводжується великою поверхневою рухливістю атомів Mo. Довгі радіаційно-індуковані атомні стрибки, що охоплюють відстані більш, ніж найближчі міжатомні, були виявлені на {110} терасах. Радіаційно-індукований обмін атомів Mo, що стикаються з поверхнею Mo {110}, був виявлений в нашому математичному моделюванні: спостерігалися обмінні процеси, в яких радіаційно-збуджений атом проникав у поверхневий шар, а поблизу виходив на поверхню інший атом. Ці результати моделювання являють собою перше спостереження обмінних ефектів у радіаційно-індукованій поверхневій дифузії.