

Defect production in tungsten: A comparison between field-ion microscopy and molecular-dynamics simulations

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Molecular dynamics (MD) computer simulations of 20 – 30 keV self-ion bombardment of W were performed and compared to past field-ion microscopy (FIM) studies [M. I. Current *et al.*, *Philos. Mag. A* **47**, 407 (1983)]. The simulations show that the unusually high defect production efficiencies obtained by FIM are a consequence of a surface effect, which greatly enhances defect production compared to that in the crystal interior. Comparison of clustering of vacancies and the formation of interstitial atoms found in the FIM experiments and MD simulations shows overall good agreement. [S0163-1829(98)00530-X]

Field-ion microscopy (FIM) studies of defect production in W and Pt have remained an anomaly in the field of radiation effects for several years. Unlike the findings on other metals, that the number of defects produced in energetic displacement cascades is less by a factor of ~ 3 than the number predicted by the modified Kinchin-Pease (KP) formula N_{KP} ,¹ the FIM results indicate that defect production in W and Pt actually exceeds the KP prediction.^{2,3} In addition, these experiments show that the average lengths of replacement collision sequences (RCS's) in tungsten are very long, ~ 180 Å, which is far in excess of what is now commonly accepted. Since the FIM method offers the most direct and unambiguous view of the defect structure in cascades, the disparity between the findings of FIM and what is currently believed about defect production is mystifying. The present work employs molecular dynamics (MD) computer simulations to clarify this situation.

MD simulations of defect production have now been performed on a large number of metals, and the results support the smaller defect production numbers, $N_{KP} \sim 0.25$.⁴ Although these results appear in contradiction to the FIM results, no simulations have been performed on W using realistic potentials. Moreover, nearly all previous MD studies of defect production examined damage created by a primary recoil atom in the crystal interior, with no nearby surfaces, whereas the FIM experiments were conducted using a 20–30 keV ion beam impinging on the surface of the tip. In previous work using MD, it had been shown that for projectiles with energies less than ~ 10 keV, the proximity of the surface causes a significant enhancement of damage in Au (Ref. 5) and in Ni₃Al and Cu₃Au (Ref. 6) owing to surface melting. The question at hand, therefore, is whether the same effects are important in a metal with a melting temperature 3–4 times higher than those previously investigated and whether surface damage still dominates when the bombardment energy is 2–5 times higher than previously employed.

The present simulations are also of value for judging the accuracy of MD simulations of cascade processes. Although MD simulations have been very successful in elucidating the nature of damage production in irradiated materials, they are based on classical interatomic potentials and thus do not account for any possible influence of the conduction electrons

on the cascade dynamics. It has, in fact, been suggested that electron-phonon coupling may play a strong role in dissipating cascade energy, and various empirical schemes have been devised to include such coupling in MD.^{7,8} Unfortunately, the parameters required for these methods are not known, and present theoretical methods are not capable of providing them. The possibility of comparing MD simulations with FIM experiments, therefore, is uniquely exciting, since the FIM experiments provide an atomic description of the structure of cascades that is then laid open for direct comparison with simulations. Any shortcomings in the simulations become immediately apparent. An important advance represented in the present work is the capability to simulate cascades with energies of 30 keV and to obtain statistically significant results. Our work will show that, in fact, good agreement is found between FIM and MD studies and that the apparent disagreement between FIM and other experimental work derives from the shallow depth of penetration of 20–30 keV heavy ions in tungsten. In the bulk, we find that W behaves quite similarly to the metals investigated by others.^{9,10}

Classical molecular dynamics methods were used to simulate full collision cascades.¹¹ The atoms were arranged in a cubic simulation cell, using periodic boundary conditions in two dimensions, and open boundaries in the third dimension to model a surface. A collision cascade was initiated by placing a W atom ~ 4 Å outside one of the open boundaries, and directing it toward the surface. The cell size was chosen to be sufficiently large to contain the resulting cascade within the cell boundaries and far from the other open surface. The cell temperature was initialized to 15 K, and scaled towards this value at the two atomic layers closest to the nonsurface faces of the cell.

The interactions between W atoms were modeled with the Finnis-Sinclair three-body interatomic potential¹² that was smoothly joined to the Ziegler-Biersack-Littmark universal interatomic potential¹³ at short interaction distances to realistically describe strong collisions. The fit was adjusted to reproduce the experimental value of the threshold displacement energy, 38 eV. The potential also reproduces the experimental melting point of 3680 K to within 200 K. The SRIM96 electronic stopping power¹⁴ was included in the simulations as a nonlocal frictional force affecting the motion of all atoms with a kinetic energy higher than 10 eV.

TABLE I. Average defect production results in W obtained by simulation and experiment. ‘‘S’’ in the event type denotes surface and ‘‘B’’ bulk events. θ denotes the incident angle in degrees off the normal of the surface. N_{vac} is the total number of vacancies, $N_{vac,surf}$ is the number of vacancies produced close to the surface, and N_{int} is the number of interstitials. The statistical uncertainty of the simulations is about 25%, and of the 30 keV experiments, 10% (Ref. 2). The 20 keV experiments consisted of only two events.

Event type	θ	N_{vac}	$N_{vac,surf}$	N_{int}
B 20 keV (sim.)		16		16
S 20 keV (sim.)	61°	123	65	2.3
S 20 keV (sim.)	22–50°	145	111	2.0
20 keV (expt.) ^a	61°	81	23	
S 30 keV (sim.)	70–80°	63	34	0.8
S 30 keV (sim.)	22°	136	28	6.7
30 keV (expt.) ^a	70–85°	125	46	

^aReference 2.

Quantities similar to those used in the FIM experiments were used to characterize the primary state of damage: the average numbers of vacancies, interstitials, sputtered atoms, and adatoms per cascade event; the size distribution of vacancy clusters, and the pair-correlation function of vacancies. Interstitials and vacancies were located using space-filling Wigner-Seitz cells, and vacancy clusters were recognized by finding a continuous network of vacancies connected by nearest-neighbor distances.¹⁵ Atoms less than two lattice constants outside the initial surface of the simulation cell were interpreted to be adatoms, and atoms farther out were considered sputtered.

The primary finding of these simulations is that the number of vacancies produced by an external atom impinging on a surface with an energy of 20–30 keV is 4–8 times larger than the number produced by an interior lattice atom recoiling with the same energy. As illustrated in Table I, the average number of vacancies produced in a near surface cascade is ~ 123 for 20 keV events, and 90 for 30 keV events, whereas it is only ~ 16 for bulk 20 keV events. This enhanced defect production at the surface results directly from local melting in the core of the cascade coupled with the convective flow of the liquid W onto the surface. The liquid-like structure and transport of atoms above the surface are seen in Fig. 1. When the melt resolidifies, many adatoms are trapped on the surface and vacancies are left below. It is interesting to note that even if all of the vacancies in the first two atomic planes are ignored, over 65 vacancies are produced in the 30 keV events, which is ~ 3 times the number produced in the bulk. (The bulk value for 30 keV cascades is obtained by scaling the number at 20 keV by the difference in damage energy, ~ 1.5 .) The added defect production found in the FIM experiments can thus be attributed to the surface effect that was first observed in Au,⁵ even though the melting temperature of W is nearly four times higher than that of Au.

Quantitative comparison of the FIM and MD results for vacancy production shows agreement to within $\sim 30\%$. This falls within the statistical uncertainty of 25% and 10% of the simulations and experiments, respectively. Some disagreement is not unexpected, however, since a wide range of im-

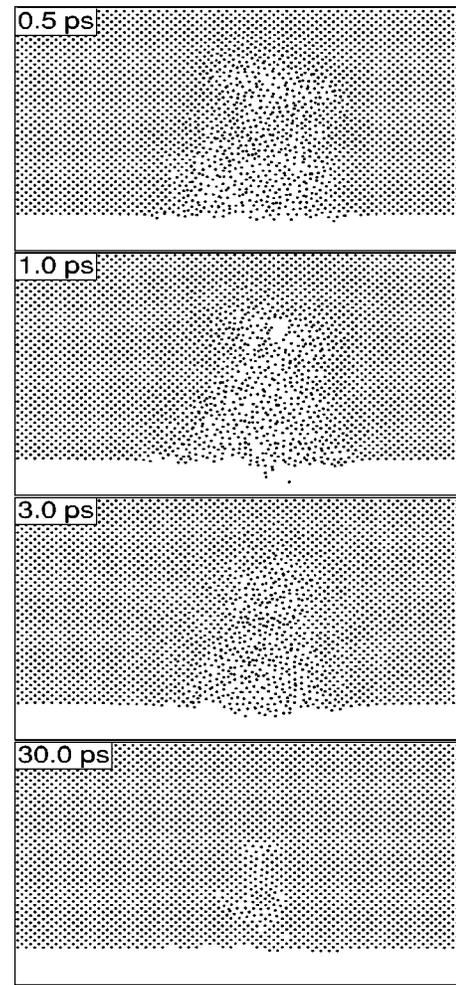


FIG. 1. Illustration of the development of a 30 keV surface cascade in W. The slice shows two layers of atoms through the center of the cascade. The liquid formed is close to the surface, and parts of it flow on the surface, producing adatoms and leaving a vacant region in the center of the cascade.

pact angles and surface planes are encountered in the FIM experiments, whereas only (100) surfaces and a few angles of incidence were examined in the simulations. Previous experience with cascade results in metals,¹⁵ moreover, indicate that MD simulations have an inherent uncertainty of the order of 20% in calculations of defect numbers. Thus the agreement between the surface simulations and experiment can be considered reasonably good, and most importantly, clearly much better than the agreement between the bulk simulations and experiment.

The structure of the vacancies in the W cascades was examined in the FIM by forming various correlations functions. Shown in Fig. 2 are pair-correlation functions of vacancies in three specific experimental cascades obtained from Ref. 16. Superposed are correlation functions obtained by simulation. The latter were selected to provide good fits to the experimental ones. The good agreement thus illustrates that differences between the vacancy structures from one cascade to another are larger than the differences between the simulations and experiments. The pair-correlation functions from all events were also combined in the FIM study to form a normalized plot of the vacancy concentration as a

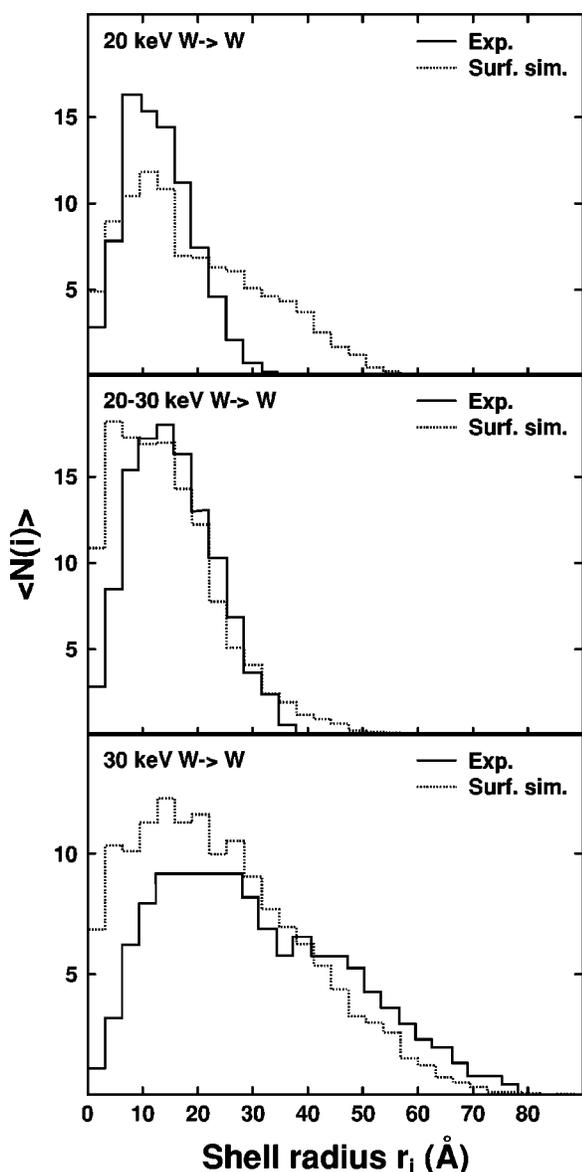


FIG. 2. Comparison of simulated and experimental radial distribution functions $\langle N(i) \rangle$ of three individual 20 – 30 keV surface cascades in W. The definition of the quantity $\langle N(i) \rangle$ and the experimental curves are given in Ref. 16.

function of neighboring site separation. This type of plot is shown in Fig. 3 for the experiments and the simulations. For a homogeneous vacancy distribution, the slope in this plot should be zero. For the bulk cascades obtained from the simulations, the slope is indeed very small. The slope obtained by experiment, on the other hand, is large, illustrating strong correlations for vacancy clustering. Noteworthy is that the slopes were nearly the same in the FIM study for bombardments with ions of different masses even though the absolute concentrations of vacancies were vastly different. This same slope is obtained in the simulations for vacancies produced in the surface cascades. These results provide additional evidence that many of the vacancies in the FIM experiment were indeed formed by the surface effect described above.

The number of interstitial atoms produced in cascades was not accurately obtained in the FIM study, but it was

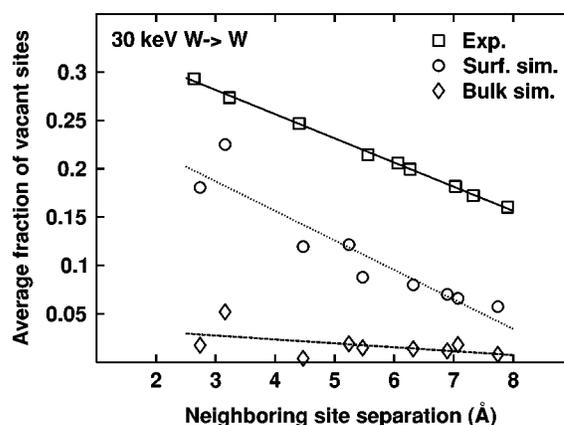


FIG. 3. Comparison of experimental and simulated surface and bulk radial distribution functions $\langle R(i)/Z(i) \rangle$ of three individual 20 – 30 keV surface cascades in W. The definition of the quantity $\langle R(i)/Z(i) \rangle$ and the experimental curves are given in Ref. 2.

clear that their number was far less than that of the vacancies. For example, in experiments where the specimen was warmed following ion bombardment, typically only a few interstitials from each cascade were observed to migrate to the surface.¹⁷ A similarly small number of interstitials was found in the simulations. Table I shows that while ~ 100 vacancies are produced in the surface events, an average of only ~ 3 interstitial atoms are created. The reason for the large imbalance is simply that only vacancies are produced by the flow mechanism near a surface.

It is somewhat surprising that far fewer interstitials are produced in the near surface events than in the bulk events. Some interstitials, of course, could be lost to the surface in the defect production process, for example by RCS's intersecting the surface, but seemingly only about half should be lost in this way. We did observe in the simulations, however, that interstitials migrated during the thermal spike and some of these then recombined in the cascade core or annihilated at the surface. Such defect annealing is likely to be more prominent in surface events than in the bulk owing to the surface sink, the higher vacancy concentration in the cascade, and the slower heat dissipation.

The controversy concerning the long lengths of RCS's found by FIM cannot be answered with certainty by the simulations. We did observe, however, that several interstitials migrated long distances as dynamic crowdions while the cascade cooled (the cold boundary walls limited this diffusion in the simulations). Similar migration has been observed previously in simulations using pair potentials.¹⁸ Although the classical potentials employed here are not sufficiently reliable to ensure that this result can be fully trusted, it nevertheless suggests that the experimentally observed large separation of interstitials from the cascade core may be due to interstitial migration rather than RCS's. Analysis of the simulations for the temperature around the cascade showed that a region extending 5 nm beyond the core of the cascade remained above 200 K for 10 ps, which means that significant interstitial migration can take place.

Our simulations have answered two important questions regarding damage production in collision cascades in W. The unexpectedly large vacancy production observed in FIM experiments is caused by the convective flow of liquid atoms

onto the surface during the development of the cascade, resulting in a much larger production of adatoms and vacancies than in the bulk. The smaller number of interstitials found by FIM is due to interstitials not being produced by the flow mechanism. Quantitative comparison between simulation and experiments shows agreement to $\sim 30\%$, which is within statistical uncertainties. Although the agreement does not rule out some influence of electron-phonon coupling or other errors in the simulation model, it does show that these possible errors cannot be overly significant. Finally, the simulations offer a plausible alternative mechanism to RCS's for

the large separation of interstitials from their nascent cascades observed in the FIM experiments, namely, dynamic crowdion migration.

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