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Mechanisms of Defect Production and Atomic Mixing in High Energy Displacement Cascades: A Molecular Dynamics Study.

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ABSTRACT

We have performed molecular dynamics computer simulation studies of displacement cascades in Cu at low temperature. For 25 keV recoils we observe the splitting of a cascade into subcascades and show that cascades in Cu may lead to the formation of vacancy and interstitial dislocation loops. We discuss a new mechanism of defect production based on the observation of interstitial prismatic dislocation loop punching from cascades at 10 K. We also show that below the subcascade threshold, atomic mixing in the cascade is recoil-energy dependent and obtain a mixing efficiency that scales as the square root of the primary recoil energy.

INTRODUCTION

The nature of the damage that ensues in a metal upon its irradiation with energetic particles has been a topic of interest for over 40 years [1,2]. Initially, this interest resulted from the need to understand radiation effects in materials in a nuclear reactor environment; today it is driven by these same concerns and by the emergence of energetic ion beams as a common tool in non-equilibrium processing and modification of materials [3].

Essential to gathering a knowledge of radiation damage in

solids is understanding the cascade of atomic displacements that results when an energetic particle (e.g., a heavy ion or a fast neutron) penetrates a solid. Displacement cascades present a difficult problem to study by either theoretical or experimental means. Their many-body and highly non-linear nature precludes use of treatments based on the linearized Boltzmann transport equation and the binary collision approximation; such collisional models consider only the interaction between moving and stationary atoms [4,5]. Experimentally, the small size ($\sim 3 \times 10^5 \text{ \AA}^3$) and very short lifetime ($\sim 10^{-11} \text{ s}$) of cascades has hampered attempts to dynamically resolve their evolution. Models based on applications of the heat transport equation [6] are limited by the inhomogeneous nature and extreme thermal gradients imposed in the core of the cascade. Caro et al. [7] have recently proposed a model that employs results from molecular dynamics simulation to establish the time scale of the heat transport, thereby avoiding explicit definition of the thermal diffusivity in the cascade. This approach is nevertheless limited in its scope since it provides no information on defect production and clustering.

Molecular dynamics computer simulation (MD) has been used as a tool to study the structure and dynamics of energetic displacement cascades for the last 30 years. The early pioneering work of Gibson et al. [8] demonstrated that Frenkel defect production in electron irradiated metals proceeds via replacement collision sequences (RCSs) along close-packed directions. These seminal studies, performed at Brookhaven around 1960 with recoil atom energies up to 400 eV, set the stage for higher energy MD simulations. Until recently, however, progress in the field has been slow mainly because of the large number of atoms required to perform these simulations, but also because of the lack of reliable interatomic potentials to describe the energetics of metals. The recent surge in the availability and power of supercomputers has made the use of MD plausible for radiation damage studies at higher energy. MD studies of 5 keV cascades in Cu and Ni with pair potentials led to a model of formation of the primary state of radiation damage in metals based on the concept of local melting during the thermal spike [9-11]. Within this simple "liquid droplet" model, defect production, point defect clustering and the amount of atomic rearrangement produced in a displacement cascade (i.e., ion beam mixing) are all governed by the extent and lifetime of the melt zone and therefore by the melting behavior of the material under study. Due to the limited recoil atom energy (maximum of 5 keV) employed in those studies, ideas put forth in the model are based on extrapolation of results to higher energies where subcascade formation becomes important. Furthermore, the nature of the pair potentials employed sheds some questions into the reliability of some quantitative aspects of the results.

In order to increase the PKA energy in MD simulations we have developed a new code, termed MOLDYCASKE, that takes advantage of the vector architecture of Cray supercomputers [12]. This code has enabled us to perform MD simulations of energetic displacement cascades in Cu at energies as high as 25 keV ($E_R = 0.11$ in reduced energy units [13]) with more realistic isotropic many-body potentials of the EAM type [14,15]. It makes use of the model of

Caro and Victoria [16] to describe lattice-electron interactions.

We present results of 1, 2.5, 5, 10, and 25 keV PKA cascades in copper at 10 K. Also included are results of dimer-recoil ($E_{\text{Tot}}=10$ keV) events. At the highest reduced energy treated in these simulations ($E_R = 0.11$), the break-up of a cascade into individual isolated subcascades is seen to take place. The collapse of a displacement cascade to a vacancy dislocation loop is observed and more significantly, prismatic dislocation loops are seen to appear near the edge of the molten zone of 25 keV and dimer-recoil cascades. We have recently proposed [17] that a new, cooperative mechanism, not related to RCSs but rather akin to dislocation loop punching [18] is responsible for this interstitial prismatic dislocation loop formation. We analyze evidence for this new mechanism of defect production and discuss its consequences on the microstructural evolution of metals in high-radiation environments.

The results show that at the energies treated, atomic mixing in the cascade is energy dependent. The degree of clustering of vacancies and SIAs in the cascades is also observed to depend on energy (and energy density). For the highest energies treated, it is observed that the size and fraction of interstitials in clusters exceeds that of vacancies, in good agreement with recent experimental results [19,20]. The results reproduce the correct magnitude of ξ in cascades and shed new light into defect production mechanisms.

MOLECULAR DYNAMICS MODEL

The MOLLYCASK code employs the vectorized link cell method (VLC) of Heyes and Smith [21] as modified by M.W. Finnis [22] to find the neighbors of a given atom in the computational crystal. With this technique, the calculation of the forces on all atoms in an MD step can be fully vectorized on a Cray-2. In the MD studies described below we have employed crystals containing up to 500,000 atoms for the case of 25 keV events. With this crystal size the code performs a complete MD step (including I/O operations) in 111 s. of CPU.

The cascades were initiated by imparting kinetic energy to one atom in the crystal. The direction of the primary knock-on atom (PKA) was chosen at random although it was insured that it was away from a channeling or a close-packed direction. Simulations were also performed for dimer recoils with a total energy of 10 keV. In this case, a pair of nearest neighbors endowed with 5 keV/atom is employed as the primary knock-on. For simplicity, and to maximize overlap effects at this energy, the initial direction of travel of the 2 recoiling atoms in the dimer was chosen to differ by only 5° . Henceforth, these events will be referred to as di-5 keV cascades. All events were performed in computational crystals coupled, via Langevin dynamics applied to the boundary atoms [23], to a thermal reservoir at a constant ambient temperature of 10 K. Energy that arrived at the boundary layers was prevented from reentering the crystal via the periodic boundary conditions by virtue of a damping coefficient applied to the atoms in the boundary link cells.

Details of this procedure have been given elsewhere [12].

The embedded atom method (EAM) potentials of Foiles, Baskes and Daw [15] have been shown to accurately reproduce many equilibrium properties of fcc metals, and are used in a modified version throughout this work. In order to treat the highly non-equilibrium atomic configurations and distances encountered in the course of these simulations, we modified these potentials to improve the description of the pressure derivative of the bulk modulus [12] and to correctly characterize high energy scattering [24]. It has been shown that the EAM potential for Cu reproduces the threshold displacement energy correctly [25] and therefore our modification does not alter the characteristics of the potential for energies below ~ 40 eV. At very high energy the pair potential contribution to the total energy is chosen to reproduce the scattering from a Moliere potential and the embedding function is forced to become a constant. In this manner, the EAM potential is effectively transformed into a pair potential at high energy.

The electronic energy loss has been included in our simulations by means of the model of Caro and Victoria [16]. In this model, the high energy electronic stopping enters the motion of the atoms in the MD crystal via the Langevin equation of motion. Furthermore, the model reproduces the correct limit of the electron-phonon coupling at low energy for weakly coupled metals such as Cu. We have previously employed this model to investigate the effect of electron-phonon coupling on the dynamics of thermal spikes in Cu in reference [24] where details of its implementation are given.

RESULTS

Previous MD studies of 5 keV displacement cascades in Cu and Ni [9-11] with pair potentials showed that local melting takes place at the core of the cascade. It was concluded that the melting point is the physical quantity that determines the behavior of a metal under cascade-producing irradiation. The present study confirms the melting behavior of the cascade and provides the first dynamical simulation evidence for subcascade formation and interaction. Moreover, the large damage energy density deposited in the local melt region of the 25 keV and dimer-recoil cascades is shown to give rise to new defect production phenomena not observed before.

Evidence for the break-up of a high energy cascade into subcascades and their subsequent interaction is presented in figure 1 where a snapshot of the atom positions in a {010} plane of width $a_0/2$ are shown (a_0 is the lattice parameter of the perfect crystal). The plane cuts across the centroid of a 25 keV cascade defined as the "center of gravity" of the kinetic energy distribution in the crystal. Figure 1a shows such a cut 0.2 ps after the initiation of the primary recoil event. A region in which a large degree of crystalline order is preserved is seen surrounded by two "lobes" of highly disordered material. At a later time, 1.02 ps, figure 1b shows that the two original subcascades have

overlapped to form a single melt zone. This represents the first time that the dynamical interaction between subcascades has been observed in an MD simulation. From these results it may be expected that for irradiations of low melting temperature materials where a large amount of damage energy is deposited, a molten tube connecting subcascades may be formed.

Figure 1. Evidence for the formation of subcascades in a 25 keV PKA cascade in Cu. The filled circles represent atomic positions in a (010) plane of width $a_0/2$ at (a) $t=0.19$ ps and (b) $t=1.2$ ps.

value of the melting point of Cu), for a 25 keV cascade. The temperature remains above the melting point for over 3 ps. At early times (0.2-0.4 ps) the cooling rate achieved in the core of the 25 keV displacement cascades is $\sim 1 \times 10^{15}$ K/s and the temperature gradient is ~ 90 K/Å in the central region.

In order to study the local atomic structure of the cascade region, we calculate the square of the magnitude of the planar structure factor, $|S_p(k)|^2$, for regions of (010) planes that contain ~ 250 atoms in the core area of the cascade. This quantity, where k is a primitive lattice vector in the plane, is highly sensitive to local order and takes a value of 1 for a perfect crystal at 0 K and fluctuates near a value of 0 for a crystal with no long range order. Figure 2 shows the average value of $|S_p(k)|^2$ for 12 planes centered on the centroid of the cascade at several times. No long range order is appreciable at times longer than about 0.25 ps until late in the event. It is interesting to note however, that at 0.2 ps $|S_p(k)|^2$ fluctuates near a value of 0.1 indicating, as expected, that the cascade core has not yet reached a true molten state.

Further evidence for the molten state of the cascade core is gained when the temperature (here defined from the atom kinetic energies, E_k , by $E_k = 3/2 K_B T$, where K_B is Boltzmann's constant) is plotted as a function of distance from the centroid of the cascade region. Figure 3 shows the temperature (normalized to the experimental

Figure 2. Average value of the square of the magnitude of the planar structure factor at several times in the region near the centroid of the cascade.

The large cooling rates and temperature gradients described above result, as expected, in a large degree of undercooling in the melt region. In figure 3, it can be seen that by 4.6 ps the crystal temperature is everywhere below the melting point. However, a large disordered region is still present near the center of the cascade as confirmed by the very low value of $|S_p(k)|^2$ at 4.6 ps seen in figure 2.

For sufficiently high interfacial undercooling during the rapid quenching of a pure metal melt, the formation of structures departing widely from those at equilibrium is expected [26]. Following the analysis of ref. [26], we estimate that for the 25 keV events discussed above the scaled interfacial undercooling at the crystallization front at ~ 4.6 ps is $\Delta T_l/T_l = 0.26$ where $\Delta T_l = T_l - T_i$ and T_l and T_i are the liquidus and interfacial temperatures, respectively. This value is over a factor of 2 larger than the estimates of Lin and Spaepen [27] following laser melting of pure Fe.

Atomic mixing

The amount of atomic rearrangement that takes place during the lifetime of a displacement cascade is obtained from the total mean square displacement for the atoms in an MD crystal, which we define as

$$\langle R^2(t) \rangle = \sum_{i=1}^N [x_i(t) - x_i(0)]^2 \quad (1)$$

Figure 3. Radial temperature profile for a 25 keV PKA cascade in Cu.

where $r_i(0)$ denotes the perfect lattice position of atom i , and $r_i(t)$ indicates its displaced position at time t .

Because of experimentally observed break-up of high energy cascades into subcascades, the magnitude of the mixing efficiency obtained in heavy ion irradiations may be compared to the results of the simulations. In experiments of ion beam mixing in pure metals, the broadening of a marker of tracer atoms embedded in the matrix is measured [28]. The experimentally observed mixing efficiency is then related to the mean square displacement obtained in the simulations by

$$\frac{Dt}{\Phi F_D} = \frac{1}{6N} \frac{\langle R^2 \rangle}{E_D'} \quad (2)$$

where Φ is the ion dose employed in the experiment, F_D is the damage energy deposited per unit length perpendicular to the surface, E_D' is the damage energy per atom in the simulation and N is the atomic density of the target. Figure 4 shows the results of our simulations normalized according to equation 2. The actual magnitude of mixing is about a factor of 2 smaller for our 25 keV events than that observed experimentally [28], but this is not unreasonable since isolated subcascades are not expected in Cu until higher recoil energies are reached.

Figure 4. Normalized value of the mixing parameter for cascades in Cu. The fit through the data results in a square root dependence of the atomic mixing in the cascade.

More important is the increase in atomic mixing with PKA energy. This reflects the long lifetime and high energy density of the melt zone found in the 25 and di-5 keV PKA events. Energy density is defined by considering the radius of gyration of the region of the crystal in which total atom energies exceed 0.48 eV (i.e., the molten zone). The time at which this definition is applied is arbitrarily chosen as ~ 0.25 ps (i.e., near the end of the collisional phase) for all events. The energy density in the 5, 10, di-5 and 25 keV cascades is 0.54, 0.68, 0.97 and 0.96 eV/atom, respectively. We note that atomic mixing in the di-5 keV cascades is almost a factor of two larger than in the 10 keV monomer events and very similar in magnitude to the 25 keV events. A fit through the data, excluding the di-5 keV cascade, results in a magnitude of atomic mixing that scales $\sim$ as the $1/2$ power of the cascade energy.

This observation that the mixing is energy dependent in this regime is consistent with the model that assigns individual liquid droplets to different subcascades. It is not until the energy range where subcascades are formed is reached that mixing becomes energy independent. The quantitative results of experiments are in excellent agreement with calculations based on diffusion in the molten region of the cascade [28].

Defect production

In the absence of nuclear transmutation effects, defect production and clustering in energetic displacement cascades determine, to a large extent, the response of a material to a high-radiation environment.

Figures 5a-d show the primary state of damage resulting from 5, 10, di-5 and 25 keV cascades in Cu. The usual pattern, originally described by Seeger [29], of a vacancy-rich core region surrounded by a cloud of SIAs can be seen. While small clusters that contain 2 or 3 SIAs are seen in all cases, figures 5c,d reveal the presence of several large clusters of SIAs. As we will see below, these large SIA clusters are formed as the result of a new, cooperative mechanism of defect production [17].

The absolute number of defects observed in the monomer PKA events agrees well with the experimental observation of a defect production efficiency, ξ , $\sim 1/3$ for cascade-producing irradiation in most metals [30]. The average number of Frenkel pairs observed at the end of the events is 14, 27, and 59, for 5, 10 and 25 keV events, respectively. These correspond to $\xi \sim 0.26, 0.23, 0.2$ respectively [31]. Under experimental conditions where highly non-linear spike effects are important, e.g., in 40 keV di-atomic molecule irradiation of W [32], ξ has been found to approach unity. In the dimer-recoil events discussed here, the ratio of defects produced (per recoil) to that produced in the monomer 5 keV events is 1.2. This number is lower than the value of 1.55 found by Seidman et al. [32]; this is not unexpected since the recoil energy in our dimer cascades is well below the subcascade threshold. The trend towards an increased ξ in highly non-linear cascades is reproduced. The new defect production mechanism revealed by these studies helps explain some of these experimental results. Its consequences will be discussed in more detail below.

Recent MD studies by Foreman et al. [33-35] of low energy cascades (up to 2 keV) in Cu with a many-body potential have largely confirmed previous 5 keV results with pair potentials regarding the ejection of SIAs by RCSs from the molten core of the cascade. Also, the previously observed clustering of small numbers of SIAs as the result of the elastic interaction among SIAs ejected from the cascade core via RCSs [9] has been confirmed. These studies have also provided statistically significant information on the absolute number of defects produced in low energy cascades in Cu.

Cascade collapse

Although pair potential studies of 5 keV cascades in Cu and Ni [9-11] provided extensive evidence for vacancy clustering, the results of this work are the first theoretical confirmation of experimental results that show cascade collapse to vacancy dislocation loops at ambient temperature below annealing stage I.

Analysis of the spatial configuration of the vacancies at the end of a 25 keV cascade reveals a dislocation loop that contains 14 vacancies in a $\{111\}$ plane. Figure 6a shows the empty lattice sites where the vacancies are located. Figure 6b shows the positions of the atoms in the $\{110\}$ plane normal to section AA' in figure 6a. As can be seen from this figure, the vacancies form a Frank loop with the Burgess vector $\mathbf{b} = a_0/3[111]$.

Figure 5. Primary state of damage resulting from (a) 5 keV, (b) 10 keV, (c) di-5 keV, and (d) 25 keV cascades in Cu. Filled circles () indicate self interstitial atoms and open circles vacancies.

Figure 6. (a) Vacant lattice sites for a vacancy dislocation loop following a 25 keV cascade in Cu. (b) Location of the atoms in a (110) plane normal to the (111) plane of figure 5(a) (section AA') in (a)).

Experimental evidence shows that in many metals energetic displacement cascades collapse into vacancy dislocation loops even at irradiation temperatures below stage I [36]. The present results, as well as our previous MD studies with pair potentials, have suggested a mechanism of vacancy clustering driven by the rapid resolidification of the molten core of the cascade [37]. This resolidification kinetics-based interpretation of cascade collapse is borne out of the difference in atomic density between the molten zone of the cascade and the surrounding crystal. For materials with low melting point, cascades with high energy density, or events in a lattice at high ambient temperature, the resolidification process should proceed slowly and therefore the cascade collapse probability should be

large.

Although detailed calculations based on these premises are currently under way, the present results support these ideas. The fraction of 'effects' (both vacancies and SIAs) found in clusters larger than λ in all these events is shown in figure 7. For each energy, an average is taken over the two events simulated. As the energy density in the cascade increases, there appears to be a trend towards both a larger fraction of defects in clusters, as well as larger defect clusters. Figure 8 shows the size of the largest vacancy and SIA clusters for all these events. The large reduction in atomic density in the core of the cascade and long lifetime of the 25 keV events are consistent with the mechanisms of cascade collapse described above. For 25 keV PKA cascades, the number of defects produced is sufficiently large that a vacancy dislocation loop is actually observed. As we shall see below, the production of large SIA clusters, and in some cases interstitial prismatic dislocation loops, results from a new, cooperative mechanism of defect production.

Figure 7. Fraction of vacancies and SIAs in clusters for 5, 10, di-5, and 25 keV cascades in Cu.

Figure 8. Size of the largest defect clusters for 5, 10, di-5, and 25 keV cascades in Cu.

Interstitial loop punching - A new mechanism of defect production

The spatial distribution of SIAs that surrounds the cascade core is not well understood. It is commonly assumed that Frenkel pairs are produced as isolated point defects free to migrate above stage I annealing. Whether SIAs can be found in clusters following low temperature (below stage I) irradiation of metals has been a topic of controversy for many years. Previous MD studies, as well as the cascades described here, have shown that the large elastic

interaction of SIAs may lead to clustering of RCSs ejected from the cascade core. Evidence for this process has been presented before [9] and is also depicted in figure 9 where the replacement "cloud" (i.e., all the lattice sites in which an atomic replacement has taken place during the lifetime of the cascade) resulting from a 5 keV PKA event is shown. Clearly visible are RCS trails leading to SIAs that are well separated from the edge of the disorder region. Also evident from this figure are small clusters of SIAs that result from the interaction of RCSs. We note here that small clusters of SIAs (4) that did not appear to be the result of RCSs were also reported in reference 9.

Figure 9. Replacement "cloud" and RCS trails leading to final position of SIAs for a 5 keV cascade in Cu.

Diffuse X-ray scattering experiments provide evidence for the formation of small SIA clusters in cascades produced by low dose neutron irradiation of Cu at 77 K [19]. At this temperature SIAs in Cu are mobile and their clustering could be a result of long range diffusion. Earlier experiments at liquid He temperature in which SIA clusters were identified by diffuse X-ray scattering employed very high neutron doses; therefore questions remained as to the effect of cascade overlap on the observed distribution of defects [38,39]. Transmission electron microscopy studies at low temperature provide evidence for SIA clustering in 14 MeV neutron irradiated Au and Cu but experimental uncertainties remain [20].

The largest SIA cluster found in figure 5d (indicated by an arrow in the figure) reveals the presence of 17 nearest neighbor SIAs. The individual SIAs, oriented along the $[110]$ direction, are all observed to lie on a single (111) plane forming a prismatic dislocation loop. Figure 10a shows the lattice sites, defined as those that contain two atoms per Wigner-Seitz cell, containing these SIAs. Figure 10b shows the actual positions of atoms in the (110) plane normal to section AA' in figure 10a. The SIAs in the cluster form a prismatic dislocation loop with Burgess vector $b = a_0/2[110]$. The lines in figure 10b indicate the original positions of the (111) planes. By looking at a grazing angle at this figure, the interstitial loop (i-loop) is clearly visible. Such an i-loop is expected to glide easily under the influence of image forces and could migrate towards the surface of a thin film.

Figure 11 displays the time evolution of the location of the cluster of SIAs relative to the boundary of the molten zone. The cluster moves away from the surface of the cascade, ahead of the molten-zone solid interface, along $[110]$ with a speed just under than that of a C_{44} shear wave. Since RCSS travel at a speed exceeding the longitudinal sound velocity, the SIAs in the loop were not produced as the result of RCSS. Later the transient thermal stresses produced by the outgoing shock-wave [40] relax as the cascade cools and the SIAs in the cluster glide back along the $[110]$ direction towards the center of the cascade. Due to the high speed of the resolidification front the SIA cluster remains separated from the liquid-solid interface and is not absorbed by the molten region. During the ejection and relaxation, all the SIAs remain confined within a few adjacent (111) planes and at longer times coalesce into a single (111) plane.

Figure 10. (a) Lattice sites on a (111) plane on which the SIAs in the large cluster indicated by an arrow in figure 4d are centered. (b) Location of the atoms in a (110) plane normal to the (111) plane of figure 9(a) (section AA' in (a)).

Further evidence for the formation of i-loops in high energy

displacement cascades in Cu is presented in figure 12 where atomic positions in the neighborhood of a 7-SIA cluster found at the end of a di-5 keV event are shown. By glancing at the figure along the [110] direction, the 7 SIAs in the loop and its associated stress field are visible.

DISCUSSION

Based upon the results of these studies, we have recently proposed that two distinct mechanisms of defect production operate in cascades [17]. One is based on RCS transport of SIAs and the other on interstitial prismatic dislocation loop punching from the molten zone.

RCSs are important for events of all energies, i.e., from near threshold recoils (~ 20 eV) to PKA energies in the keV range. The length of RCSs is not well known but MD studies in Cu show their average length in 5 keV cascades to be ~ 23 Å [25]. In W however, RCSs were found to have lengths of the order of 160 Å [41]. As the size of the cascade increases with primary recoil energy a larger fraction of RCSs produced during the collisional phase is absorbed in the molten core of the cascade. These point defects become delocalized in the melt and lose their identity. The Frenkel pair production efficiency therefore decreases with increasing PKA energy as observed experimentally. Above 5-10 keV, as the subcascade threshold is approached, ξ approaches an asymptotic value of $\sim 1/3$. The present simulations have shown that as the cascade energy (or energy density) increases, the fraction of SIAs produced as isolated defects decreases, i.e., fewer RCSs transport SIAs beyond the liquid-solid interface. In the 25 keV events, only 35% of the SIAs are seen to be the result of RCS transport. If RCSs were the only defect production mechanism in these cascades, this would have resulted in $\xi \sim 0.07$. However, as seen in figure 7 (see also figures 5a-d), an increase in the PKA energy (or energy density) results in an increase in the fraction of SIAs produced in clusters. As we have shown above, these SIAs are in the form of prismatic dislocation loops that have been punched from the periphery of the molten core of the cascade. The i-loop punching mechanism acts to maintain the asymptotic behavior of ξ at high

Figure 11. Evolution of location of SIAs in the loop shown in figure 9. C_L is the longitudinal, and C_{44} and C' are the shear sound velocities along [110].

recoil energies and may help explain its large value for very high energy density cascades [32].

Figure 12. Atomic positions in the neighborhood of a 7-SIA cluster found at the end of a di-5keV cascade in Cu.

The RCS mechanism of defect production in cascades establishes 2 relevant length scales [9]: the radius of the molten zone and the length of the RCSs. It is therefore governed by the value of the melting point of the material and by parameters (such as atomic radius and mass) that determine RCS focusing [42]. On the other hand, the threshold energy density for loop ejection depends on thermo-mechanical properties of the material. In particular, the i-loop punching probability should depend on the Grüneisen constant of the material which determines the pressure in the cascade zone produced by a given energy density [40], and on the shear moduli which determine the pressure required to punch a dislocation loop [18]. As the energy density (and thus the pressure in the highly compressed region of the cascade periphery) increases, the probability of punching a dislocation loop increases and a larger fraction of SIAs are produced in clusters relative to those produced as isolated SIAs via RCSs.

The process of i-loop punching in nascent cascades has important consequences regarding the microstructural evolution of

irradiated metals. The fraction of point defects free to migrate over long distances will be strongly dependent on this i-loop formation. Since SIA clusters are expected to be stable up to very high temperatures (above operating temperatures in fission or fusion reactor environments), i-loop production in nascent cascades may lead to large excess vacancy fluxes at elevated temperatures where vacancy dislocation loops are unstable but i-loops are not. Woo and Singh have recently shown that interstitial clustering in cascades can lead to an anomalously high rate of void swelling by adding a large bias term in the chemical rate theory equations that govern microstructural evolution [43].

CONCLUDING REMARKS

We have presented recent results of energetic displacement cascade studies in Cu with realistic interatomic potentials. These studies are the first ever to simulate cascades at PKA energies in the subcascade threshold regime where direct comparison to fast neutron and heavy ion irradiation experiments is possible. Our results have shown, for the first time in a dynamical simulation, that cascades may collapse to vacancy dislocation loops. Moreover, based on the results of our 25 keV cascade studies, we have postulated a new, cooperative mechanism of defect production not related to RCSs but rather based on punching of interstitial prismatic dislocation loops from the core of the molten zone of the cascade. We have proposed that this new mechanism of defect production may have profound implications regarding the primary state of damage and the microstructural evolution of metals in high-radiation environments.

Our defect clustering results are consistent with recent experimental evidence that at low temperature the number of SIA clusters produced in Cu and Au cascades is greater than that of vacancy clusters [19,20]. At room temperature, the experiments show roughly equal numbers of both. The observation of an increase in the number of vacancy clusters with temperature is compatible with the resolidification kinetics model of cascade collapse, since at elevated temperatures cascades are expected to cool at a slower rate [23].

We have also discussed the energy dependence of the atomic mixing parameter in cascades and have shown that the mixing efficiency increases towards a constant, asymptotic value at energies beyond the subcascade formation threshold. Our results confirm the "liquid droplet" model of atomic mixing in cascades and show that for diatomic molecule irradiation mixing increases substantially over that expected in monomer recoil cascades.

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