New Mechanism of Defect Production in Metals: A Molecular-Dynamics Study of Interstitial-Dislocation-Loop Formation in High-Energy Displacement Cascades

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We present molecular-dynamics computer-simulation studies of 25-keV displacement cascades in Cu at low temperature. We observe the splitting of a cascade into subcascades and for the first time show by molecular dynamics that cascades in metals may lead to the formation of both vacancy and interstitial dislocation loops. We propose a new mechanism of defect production based on interstitial prismatic dislocation-loop formation and discuss its consequences regarding the primary state of damage in irradiated metals.

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The role played by energetic displacement cascades in radiation-damage research has received renewed interest in recent years as ion beams have become an important tool for performing nonequilibrium processing and modification of materials.[1] Germane to understanding the response of a given material to radiation is a knowledge of the number and spatial configuration of the elemental point defects produced by a given type of particle irradiation.

Resistivity measurements of metals irradiated under a wide variety of experimental conditions show[2] that the number of defects generated by cascade-producing irradiation corresponds to about $\frac{1}{4}$ of the value calculated with linear cascade models such as the modified Kinchin-Pease equation; see Norgett, Robinson, and Torrens (NRT).[3] Recent molecular-dynamics (MD) studies show that point defects produced by 5-keV cascades in Cu and Ni are the result of the ejection of self-interstitial atoms (SIA) from the core of the cascade via replacement collision sequences (RCS).[4] RCS's in a crystal transport mass and energy at speeds exceeding the longitudinal sound velocity and serve as a mechanism to separate a SIA from its accompanying vacancy.[5] Diaz de la Rubia *et al.*[4] also showed that after the initial collisional phase, melting takes place at the core of displacement cascades. The reduced defect-production efficiency, i.e., defect production relative to the NRT equation, then results from the absorption of RCS's in the volume of the molten zone. Under experimental conditions where highly nonlinear spike effects are important, e.g., in diatomic molecule irradiations, the defect-production efficiency approaches unity.[6]

Experimental evidence shows that in many metals energetic displacement cascades collapse into vacancy dislocation loops even at irradiation temperatures below stage I.[7] Previous MD studies[4,8] suggest that vacancies cluster near the center of a displacement cascade as a result of the rapid resolidification of the molten core of the cascade. Less understood is the distribution of SIA's that surrounds the cascade core. It is commonly assumed that Frenkel pairs are produced as isolated point defects free to migrate above stage-I annealing.[9] Whether SIA's can be found in clusters following low-temperature (below stage I) irradiation of metals has been a topic of controversy for many years.[10] Previous MD studies show that the large elastic interaction of SIA's may lead to clustering of RCS's ejected from the cascade core.[4,11] Diffuse x-ray-scattering experiments provide evidence for the formation of SIA clusters in cascades produced by low-dose neutron irradiation of Cu at 77 K.[12] At this temperature SIA's in Cu are mobile and their clustering could be a result of long-range diffusion. Previous experiments at liquid-He temperature in which SIA clusters were identified by diffuse x-ray scattering employed very high neutron doses;[13] therefore, questions remained as to the effect of cascade overlap on the observed distribution of defects. Transmission-electron-microscopy studies at low temperature provide evidence for SIA clustering in 14-MeV neutron irradiated Au and Cu (Ref. 14), but experimental uncertainties remain.

In this Letter we describe MD simulations of energetic displacement cascades in Cu generated by 25-keV ($\epsilon = 0.11$ in reduced energy units[15]) primary recoil atoms. At this high reduced energy the breakup of a cascade into individual isolated subcascades is observed as expected from binary collision calculations.[16] The collapse of a displacement cascade to a vacancy dislocation loop is observed and, more significantly, at this high reduced-energy prismatic dislocation loops appear near the edge of the molten zone of the cascade. We propose that a new, cooperative mechanism, not related to RCS's but akin to dislocation-loop punching,[17] is responsible for this interstitial-dislocation-loop formation.

The simulations were performed with crystals containing 500000 atoms at an ambient temperature of 10 K. We employed the Cray2 computers at the National Energy Research Supercomputer Center in Livermore. The average CPU time per step was 2×10^{-4} s/atom. The two events were initiated by imparting the primary knock-on atom energy (25 keV) to one atom in an equilibrated crystal. The direction of the recoil atom was chosen at random; it was ensured that all recoil atoms were away from a channeling or close-packed direction. The calculations were performed for a maximum time of 12 ps at which time all atomic migrations were observed to have ceased.

The atomic interactions were described by the embedded-atom-method potentials of Foiles, Baskes, and Daw.[18] At short range these potentials were modified by effectively transforming the interaction function at high energy into a pair potential. Details of the procedure and of the MOLDYCASK program have been given elsewhere.[19] Electronic energy loss was included in our simulations by means of the model of Caro and Victoria.[20]

Evidence for the breakup of a high-energy cascade into subcascades and their subsequent interaction is presented in Fig. 1 where the positions of atoms in (010) planes of width $a_0/2$ (a_0 is the lattice parameter) are shown. The planes cut through the center[21] of a 25-keV cascade. Figure 1(a) shows such a cut 0.2 ps after the initiation of the event. A region in which a large degree of crystalline order is preserved is seen surrounded by two "lobes" of highly disordered material. Later, at 1.02 ps, Fig. 1(b) shows the overlap of the two original subcascades to form a single melt zone. This represents the first observed dynamical interaction between subcascades in a MD simulation.

Figure 2(a) shows the primary state of damage at the end of the 25-keV event corresponding to Fig. 1(a). In Fig. 2(b) a second 25-keV event is shown. The usual

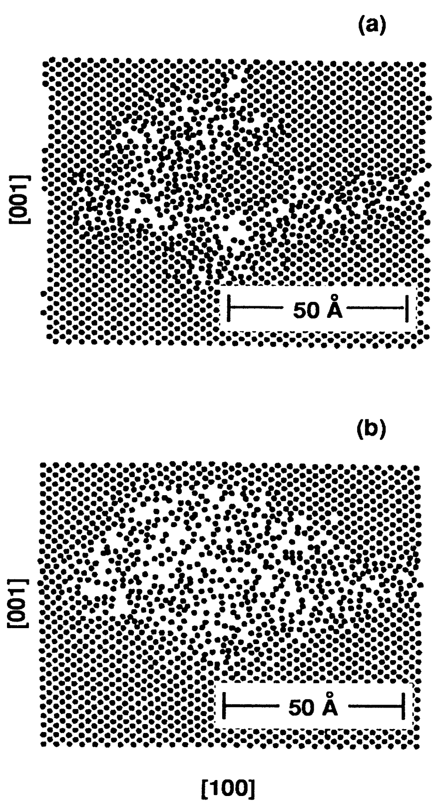

FIG. 1. Evidence for the formation of subcascades and their subsequent interaction following a 25-keV recoil event in Cu. The solid circles represent atomic positions in the (010) plane of width $a_0/2$ at (a) $t = 0.2$ ps and (b) $t = 1.02$ ps.

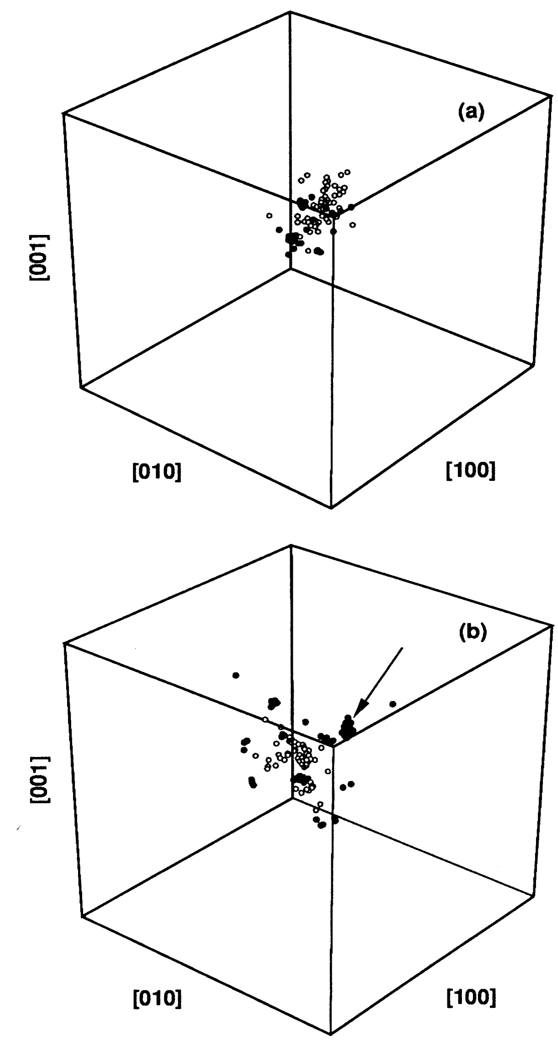

FIG. 2. Primary state of damage at $t = 12$ ps resulting from 25-keV displacement cascades in Cu. Open circles represent vacancies and solid circles indicate self-interstitial atoms. The cascade in (a) is the same as that depicted in Fig. 1. (b) A second independent 25-keV cascade is shown. The arrow in (b) points to the 17-atom SIA cluster shown in Fig. 3(a) (see text). The cubes mark the boundaries of the computational cells which were 216 Å on a side.

pattern of a vacancy-rich cascade core surrounded by a cloud of SIA's is evident. In Fig. 2(a) only one vacancy-rich core is apparent; however, the distribution of SIA's was found to correlate with the spatial distribution of the two original subcascades. Remarkable in these defect distributions is the large number of SIA clusters observed; 67% and 60% of the SIA's produced by the cascades represented in Figs. 2(a) and 2(b) are found in clusters containing at least four SIA's. All the single and di-interstitials seen in these figures are the result of long (up to 33 Å) RCS's and have the expected [100] dumbbell configuration, but larger clusters form as the result of a new cooperative mechanism of defect production.

The absolute number of point defects produced in these 25-keV events is 60 and 58. This corresponds to ≈ 0.20 of the value obtained with the NRT equation and is in good agreement with experiment as well as with previous MD studies.[2,4]

Figure 2(a) contains a dislocation loop containing fourteen vacancies in the (111) plane; these form a Frank loop with Burgers vector $\mathbf{b} = a_0/3[111]$. The distribution resulting from the second cascade [cf. Fig. 2(b)] contains two clusters with ten vacancies each; however, no collapse to a loop was observed in this case. The observed clustering of vacancies near the center of the cascade region is in very good agreement with previous MD studies using pair potentials.[4,8]

The largest SIA cluster found in Fig. 2(b) (indicated by an arrow in the figure) reveals the presence of seventeen nearest-neighbor SIA's. The individual SIA's are oriented along the [110] direction and at the end of the event are all observed to lie on a single (111) plane forming a prismatic dislocation loop. Figure 3(a) shows the lattice sites, defined as those that contain two atoms per Wigner-Seitz cell, containing these SIA's. Figure 3(b) shows the actual positions of atoms in the $(\bar{1}10)$ plane normal to section AA' in Fig. 3(a). The SIA's in the cluster form a prismatic dislocation loop with Burgers vector $\mathbf{b} = a_0/2[110]$. The lines in Fig. 3(b) indicate the original positions of the (111) planes. By looking at a grazing angle in Fig. 3(b), 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 toward the surface of a thin film.

Figure 4 displays the time evolution of the location of the cluster of SIA's 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 that of a C_{44} shear wave. Since RCS's travel at a speed exceeding the longitudinal sound velocity, the SIA's in the loop were not produced as the result of RCS's. Later the transient thermal stresses produced by the outgoing shock wave[22] relax as the cascade cools and the SIA's in the cluster glide back along the [110] direction towards the center of the cascade. Because of the high speed of the resolidification front[23] 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 SIA's remain confined within a few adjacent (111) planes and at longer times coalesce into a single (111) plane.

Based upon the results of the present study, we pro-

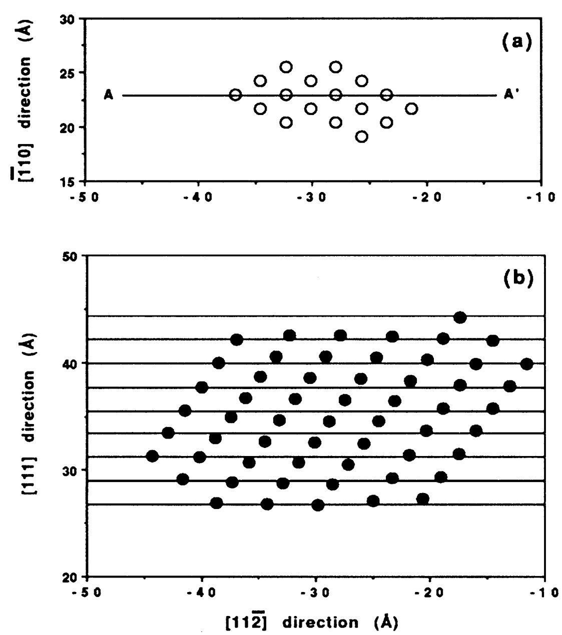

FIG. 3. (a) Lattice sites on the (111) plane on which the SIA's in the large cluster indicated by the arrow in Fig. 2(b) are centered. (b) Location of the atoms in the $(\bar{1}10)$ plane normal to the (111) plane of (a) [section AA' in (a)].

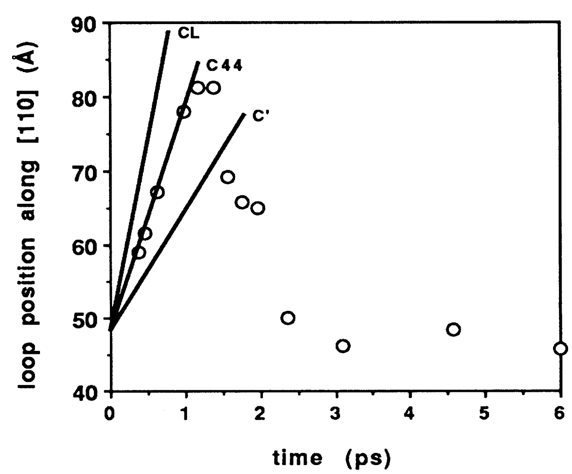

FIG. 4. Time evolution of the location of the SIA's in the cluster depicted in Figs. 2(b) and 3. Positions corresponding to a loop traveling at the longitudinal sound velocity C_L and the two shear velocities C_{44} and C' along the [110] direction obtained for our potential are shown for comparison.

pose that two distinct mechanisms of defect production operate in cascades. RCS's are important for cascades of all energies, i.e., from near-threshold events[5] (25 eV) to events with recoil energies in the keV range.[4,11] The length of RCS's is not well known but MD studies in Cu show that their average length in 5-keV cascades is of the order of 23 Å.[4] In W, however, RCS's were found to have lengths of the order of 160 Å.[24] As the size of the cascade increases with primary recoil energy, a larger fraction of RCS's produced during the collisional phase is absorbed in the molten core of the cascade. In the present simulations, for events in the subcascade threshold energy range, i.e., $\epsilon \geq 0.08$ for fcc metals,[16] the fraction of SIA's produced as isolated defects is very small. We propose that by punching an *i* loop from the surface of the cascade a large number of SIA's are protected from absorption in the molten core of the cascade. As the energy density in the cascade (and thus the pressure in the highly compressed region of the cascade periphery) becomes larger, the probability of punching a dislocation loop increases and a larger fraction of SIA's are produced in clusters relative to those produced as isolated SIA's via RCS's. We expect a threshold energy density for loop ejection dependent on the thermomechanical properties of the material. Above this threshold, the defect-production efficiency would increase with energy density.

This process of *i*-loop formation 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 such *i*-loop formation and the interpretation of such processes as radiation-induced precipitation and void swelling will be affected by this new mechanism. Woo and Singh[25] show that interstitial clustering in cascades could lead to an anomalously high rate of void swelling.

To investigate further this new mechanism of defect production, we are performing simulation studies of diatomic molecule irradiation and lower-energy cascades.

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