Plastic Flow Induced by Single Ion Impacts on Gold

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The formation of holes in thin gold foils as a result of single ion impacts by 200 keV Xe ions has been followed using transmission electron microscopy. Video recording provided details of microstructure evolution with a time resolution of 1/30th sec. Hole formation involves the movement by plastic flow of massive amounts of material, on the order of tens of thousands of Au atoms per ion impact. Plastic flow, as a consequence of individual ion impacts, results in a filling of both holes and craters as well as a thickening of the gold foil. Change in morphology during irradiation is attributed to a localized, thermal-spike induced melting, coupled with plastic flow under the influence of surface forces. [S0031-9007(96)01682-1]

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The interaction of energetic ions with solids results in a wide variety of phenomena including defect production, sputtering, and changes in surface topography. When the energy deposition exceeds a few hundred electron volts, these processes are dominated by atomic displacements localized in defect cascades. Previous investigations of the effects on surfaces of single ion impacts have been made using postirradiation, field ion microscopy (FIM) [1], transmission electron microscopy (TEM) [2], and scanning probe microscopy (SPM) [3]. While such studies reveal adatoms and surface vacancies or surface craters and hillocks, they yield little or no information on the evolution of surface features or the processes responsible. Theoretical studies, using molecular dynamics (MD) modeling [4], of the impact of a 20 keV Au ion on an Au surface indicate a large amount of plastic deformation at the specimen surface associated with the ballistic phase of the displacement cascade. The calculated topography resulting from the deformation and flow may persist and give rise to a permanent crater. However, the calculations do not yet extend long enough in time to fully reveal the way in which the ballistic phase of a displacement cascade, lasting a few picoseconds, gives rise to the surface structures observed in some materials.

In this Letter we present an *in situ* TEM investigation of the microstructural evolution due to the impact of individual 200 keV Xe ions on Au. Ion irradiations were made at room temperature in a Hitachi A-9000 TEM operating at 300 keV at the IVEM Accelerator Facility located at Argonne National Laboratory [5]. TEM specimens were prepared by jet polishing [6] 99.999 at.% pure Au with grain size greater than 10 μm having a (110) texture. After irradiation, specimen thickness was determined using a Philips CM30 TEM operating at 200 keV, equipped with a Gatan parallel electron energy loss spectrometer (PEELS). Grains used for observations were those with (110) surface normal. In the IVEM Accelerator Facility, the ion beam is oriented 30° from the microscope axis; in our experiments the specimen was tilted 15° towards the ion beam so that both ions and electrons were incident on the specimen at 15° to the foil normal. Specimens were irradiated with 200 keV Xe^+ ions at dose rates between 1 and 25×10^{10} (ions/cm^2)/s. Ion range and damage production were estimated from full cascade calculations using TRIM-95 [7]. Images from a Gatan 622 video camera and image-intensification system were viewed with total magnifications of approximately 2×10^6 and recorded on video tape with a time resolution of 1/30th sec (a single video frame). Because the video image is made up of two sequential interlaced images (each resulting from a scan and raster lasting 1/60th sec), when a rapid change occurs, one video frame may show an image that contains the new feature as a "ghost" existing on only one of the interlaced half frames. For clarity such images are not shown. The temporal resolution is clearly insufficient to yield information on the ballistic phase of the collision cascade which persists for only picoseconds. However, the system has clearly identified plastic flow processes associated with individual impacts.

Although we also observe crater formation, our most striking observation is that in the thinnest areas of the foil many single ion impacts resulted in the creation of holes having diameters between 5 and 10 nm as shown in Fig. 1. Approximately $\frac{1}{2}$% of the Xe ions produced holes. Measurements in the area shown in Fig. 1, made with a 50 nm diameter electron beam, indicate that no holes were created in regions with thickness greater than about 50 nm. This thickness is consistent with Monte Carlo calculations, using TRIM-95 [7], of the ability of 200 keV Xe ions to produce damage though the entire depth of an Au foil. On the order of 1% of all ions stop in the last 2 nm of a 50 nm thick specimen. This suggests that it is only when ion damage extends through the entire specimen thickness that a hole may be formed.

Figure 1(3) shows a hole (B) appearing next to a previously created hole (A). (The number under each section of the figure refers to the video frame from which

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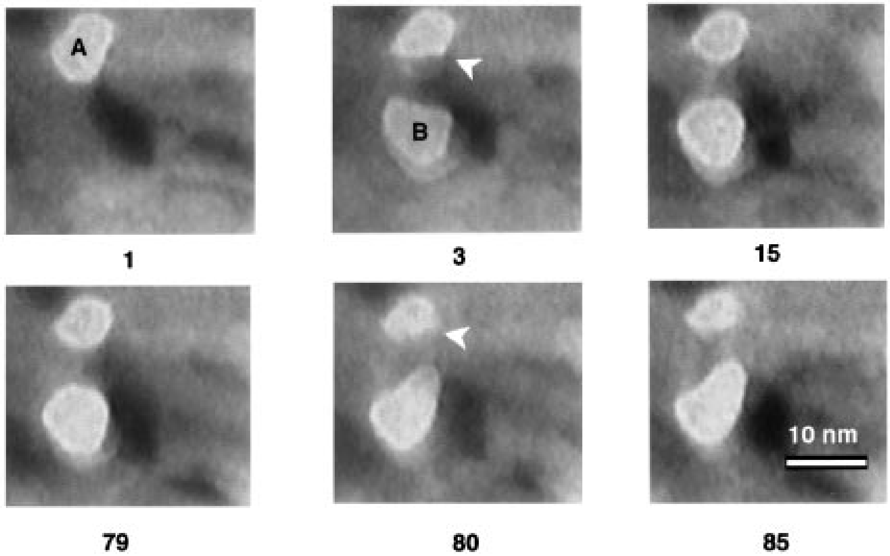

FIG. 1. Microstructural evolution in Au during 200 keV Xe irradiation at room temperature. Parts (1) and (3) illustrate the creation of a hole by the impact of a single 200 keV Xe^+ ion. Parts (3) and (15) illustrate the "rounding" of holes as a result of plastic flow processes caused by single ion impacts. Parts (79), (80), and (85) illustrate a single ion impact modifying both holes. (The numbers under each part of the figure refer to the video frame from which the image was taken, with the first frame of the figure being numbered (1). The frames were recorded at 30 per sec.)

the image was taken, with the first frame of the figure being numbered 1. The contrast within the holes is due to noise in the imaging system.) The shape of the hole is recorded, by the nature of the image recording, long after the cascade that produced it has ended and as such includes any annealing that occurs during the cascade quenching. Note that we also observe formation of craters on one or the other surface, visible only in defocus, but their low contrast renders them more difficult to capture on videotape. An ion strikes the entire area shown in Fig. 1 on average every 10 frames or 0.3 s which is much longer than the cascade lifetime of a few tens of picoseconds. The holes in Fig. 1 appear between successive video frames (within a time period of 1/30th sec) and have been made by single ion impacts. Assuming a foil thickness between 20 to 50 nm, between 20 000 to 50 000 gold atoms were removed to create hole (B). This would imply an enormously high sputtering yield if the atoms were ejected from the gold surface. However, the change in image contrast suggests that these atoms have been moved to the specimen surface. Although expelled material is likely to be closely associated with a hole, occasionally small particles appear far from any hole or crater. This is consistent with STM images that show adatom islands on ion irradiated Pt surfaces [3].

A second equally striking observation is the change in the shape and size of holes during continued irradiation as also shown in Fig. 1. Although holes have been annealed by the quench phase of their own cascade, additional ion impacts may produce further annealing. These changes occur in both large steps, such as when hole B is formed, and small steps as illustrated between Figs. 1(3) and 1(15). The partial filling of hole A resulted from plastic flow of material away from the site of hole (B) as indicated by the arrow on Fig. 1(3). No further major changes are observed until frame 80 when an ion impact causes an enlargement of hole B, and material from the impact partially fills hole A in the region indicated by the arrow on Fig. 1(80). In general, filling of holes does not depend on a second hole being near, and frequently the impact event responsible for a change does not itself produce a visible feature.

Another clear example of plastic flow is displayed in Fig. 2. This figure shows additional details of the filling of a hole during irradiation. An ion strikes the area shown in Fig. 2 on average every 57 frames. The real-time movement of material into the hole, at this dose rate of 2.5×10^{11} Xe/cm^2 sec, appears liquidlike during continuous observation; however, frame-by-frame examination reveals that the process occurs in discrete steps associated with individual ion impacts. Although not shown, when the ion irradiation is stopped, flow of material ceases and there is no further change to the morphology of the gold foil.

Many ion impact events change the shape of existing holes. Rounding of holes in Figs. 1 and 2 is a persistent process that takes place in discrete events. Changes in hole shape are generally due to cascade events that do not produce new holes but generate plastic flow near to existing ones. Although an energetic ion impact may initially give rise to an explosive outflow of material, during the quenching phase of the molten zone to the solid state, surface tension forces will act on any free surfaces

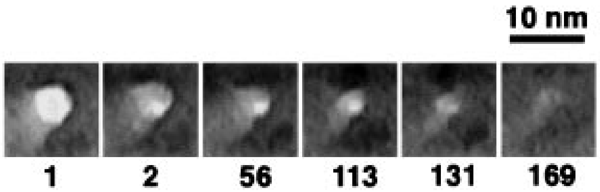

FIG. 2. Filling of a hole in Au during 200 keV Xe irradiation at room temperature. Only frames showing changes are displayed. The image remains unchanged on intermediate frames such as from 3 to 55. Changes to the hole occur in steps as a result of individual ion impacts. (The numbers under each part of the figure refer to the video frame from which the image was taken, with the first frame of the figure being numbered (1). The frames were recorded at 30 per sec.)

involved with the melt zone. This gives rise to changes in the shape of edges and the tendency of holes to become more circular. This process has not been followed with MD simulations.

It has been a long-standing observation in our laboratories that thin areas of irradiated gold specimens disappear. This is a consequence of plastic flow during heavy-ion irradiation even if holes do not form [2]. The dynamics of this process are shown in Fig. 3. An ion strikes the area shown in Fig. 3 on average every four video frames. Our *in situ* observations reveal that the thickening occurs in pulses as a result of ion-beam induced plastic flow. When observed at a high dose rate this process appears to be similar to changes that occur when thin gold foils are heated to close to the bulk melting temperature and surface tension forces cause the material to flow. Frame-by-frame analysis such as that shown in Fig. 3, however, shows this process to be a pulsed localized flow identical to that responsible for the rounding of holes. Between frames 1 and 3 of Fig. 3, the foil morphology changes due to an ion impact probably in the region indicated by the letter "A". The structure remains relatively constant until an impact in region "B" on frame 138 causes another significant modification. Similar discrete events occur on frames 214 and 511 in the regions marked with the letters "C" and "D". As in the case of hole production, only a small fraction of ion impacts result in major changes.

Estimates of the recoil cascade size using TRIM-95 [7] yield, for 200 keV Xe ions incident on 50 nm thick gold, cascades with an approximately cylindrical volume (through the foil thickness) on the order of 5–10 nm in diameter into which almost all of the incident ion energy is deposited. After approximately 10^{-11} sec, atoms within this volume have a mean energy of about 2 eV. Calculations using the thermal spike model of Kelly, in which the integrated thermal sputtering from the surface is calculated over the spike lifetime, indicate that, during the short spike lifetime, only a small number of atoms at the surface could evaporate. This is insufficient to produce a crater or a hole [8]. However, the thermal spike may cause melting and yielding [9]. The sudden melting of this volume of material, with its concomitant volume change of approximately 10%, may give rise to an explosive flow of gold atoms to either (or both) surfaces(s). We believe that events such as that illustrated in Figs. 1(1) and 1(3) are the first observed occurrences of such an ion-induced, pulsed, localized flow process. MD simulations by Averback *et al.* of a 20 keV Au ion incident on Au [4] lend support to the existence of such a plastic flow process. They found a cascade region on the order of 6 nm in diameter by 6 nm in depth in which atoms had a mean energy of approximately 2 eV. At 7.0 ps after ion impact, a crater remained which they described as resulting from plastic flow of the melt zone to the surface. In addition to the rapid, short range processes identified in the MD calculations, longer range annealing effects are experimentally observed. These may include enhanced surface diffusion as well as direct material transfer by a flow process.

Ion-induced, pulsed localized plastic flow is a fundamental process that changes the morphology of an

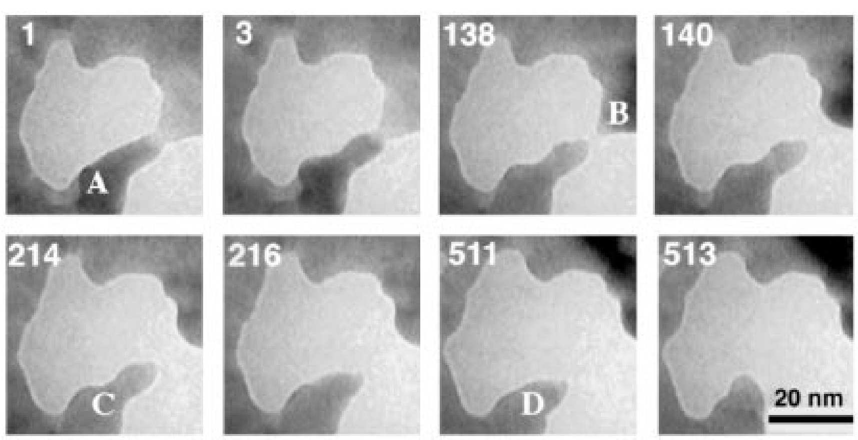

FIG. 3. Discrete structural changes of a Au foil during 200 keV Xe irradiation at room temperature. (The numbers under each part of the figure refer to the video frame from which the image was taken, with the first frame of the figure being numbered (1). The frames were recorded at 30 per sec.)

irradiated gold foil. It is likely to occur in other high-Z materials where displacement cascades are strongly localized such that molten zones form. Holes result from only a small percentage of ion impacts with others causing cratering on one surface (visible under defocus contrast) while the majority of ion impacts result in plastic-flow annealing of structures having small radii of curvature. Any post-irradiation examination of surfaces, such as that carried out by Merkle and Jäger [2], in which craters on a gold surface were assumed to result from single ion impact sputtering, significantly underestimates the actual number of crater-creation events that occurs while misidentifying the origin of the craters. Pulsed localized flow is also important during gas bubble evolution and provides an explanation for ion-induced bubble motion observed during Xe irradiation of Au containing the bubbles [10]. We believe that plastic flow may also be responsible for other processes occurring in gold under ion irradiation such as grain-boundary movement [11]. Since a single ion impact may affect tens of thousands of gold atoms, plastic flow may be the single most important mechanism in determining the behavior of gold under ion irradiation. The degree to which this phenomenon depends on material properties and affects other materials is not yet known but is under investigation in our laboratories.

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