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PLASTIC FLOW INDUCED BY SINGLE ION IMPACTS ON GOLD

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ABSTRACT

In situ transmission electron microscopy has been used to follow room temperature irradiation of thinned bulk and 62 nm thick gold films with Xe ions in the energy range 50 to 400 keV. Energy spikes resulting from single ion impacts give rise to surface craters and holes which exist until annihilation by subsequent ion impacts. Video recording provided details with a time-resolution of 33 milliseconds. Craters were produced on the irradiated surface at all ion energies and on the opposite surface when the ions had sufficient energy to traverse the specimen. Crater sizes were as large as 12 nm for the higher energy irradiations. On average, approximately 6% of impinging ions result in craters.

A single 200 keV Xe ion may produce a hole in thin gold foils. Hole formation involves the movement by plastic flow of massive amounts of material; on the order of tens of thousand Au atoms per ion impact. Individual ion impacts also result 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.

EXPERIMENTAL

In situ ion irradiations were carried out in a Hitachi A-9000 TEM operating at 300 keV at the IVEM/Accelerator Facility located at Argonne National Laboratory [1]. The ion beam is oriented 30° from the microscope axis, and 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 Xe ions at energies of 50 keV, 200 keV or 400 keV at dose rates between 1 and 25 10^{10} ions/cm²/s.

Thin Au films, 62 ± 5 nm with a $\langle 100 \rangle$ surface normal, were made by evaporation of 99.999 at % pure starting material onto NaCl at a temperature of 350°C. Small pieces of Au were floated off on a water/alcohol mixture and picked up on either Au or Cu TEM grids. Bulk TEM specimens were prepared by jet-polishing [2], 99.999 at. % pure Au with grain size greater than 10 μ m having a (110) texture.

Surface features were made visible in the TEM by defocusing the objective lens of the microscope (typically ≈ 1000 nm of underfocus). Photographic recording were made during interruptions of the irradiations, and images from a Gatan 622 video camera and image-

intensification system were continuously recorded during irradiation on video tape with a time resolution of 1/30th second (a single video frame). The temporal resolution is clearly insufficient to yield information on the ballistic phase of the collision cascade which persists for picoseconds or less. However, the system clearly identifies the plastic flow processes associated with individual impacts.

RESULTS

Figure 1 shows images digitized from video recordings made during the continuous Xe ion irradiation. Figure 1A, recorded during a 50 keV Xe irradiation, shows a small crater observed for irradiations at all energies studied. Figure 1B was recorded during a 200 keV Xe irradiation and shows a larger faceted crater and pre-existing small craters. The craters in figures 1C through 1E were produced by 400 keV Xe ions. In each case, material is deposited on the surface concomitantly with crater formation. In figure 1C, a faceted crater is surrounded by a faceted island. In figure 1D, a two-part "lid" appears to have been ejected from the crater. Such lids were not observed during 50 keV irradiations. The contrast in figure 1E is consistent with a faceted crater and a quenched droplet of ejected molten material. (The light contrast to the right of this crater is the remnant of a pre-existing crater). The expelled material

Figure 1: Images digitized from video recordings resulting from single Xe ion impacts at A) 50 keV, B) 200 keV and C), D) and E) 400 keV Xe ions. f) an isolated particle which appeared on the surface (between video frames) during irradiation with 400 keV Xe ions.

contains approximately 20,000 gold atoms. The largest craters involved as many as 40,000 gold atoms. Figure 1F shows a particle on the surface that has appeared between successive video frames with no associated crater within a field of view approximately six times the area shown. Craters were also found by Merkle and Jäger after Bi ion irradiation of Au at room temperature [3].

Analysis of more than 200 craters recorded on videotape reveals that the mean crater area increases from 14 nm^2 at 50 keV to 32 nm^2 at 200 keV and 34 nm^2 at 400 keV. For the small number of craters studied using stereoscopic techniques, both small and large craters appeared on both surfaces during 200 keV and 400 keV irradiations of 62 nm thick films. The number of craters imaged in this manner, however, was insufficient to draw conclusions concerning differences in size distribution for the two surfaces.

The images recorded on videotape during continuous irradiation reveals a rapidly changing crater population with crater creation and annihilation [4]. If the irradiation is interrupted, both creation and annihilation cease. At any time approximately 7 craters were in the field of view ($110 \text{ nm} \times 85 \text{ nm}$). At the dose rate used in the experiment, $2.4 \cdot 10^{11} \text{ ions/cm}^2/\text{s}$, statistically, there is one ion impact per second on the area of each segment of figure 1. The efficiency of crater creation is on the order of 6% (ranging from 2% to 10%) implying that the majority of ion impacts annihilate craters without creating new ones.

Under steady-state conditions of continual irradiation, at a dose rate of $2.4 \cdot 10^{11} \text{ ions/cm}^2/\text{s}$, craters have a lifetime in the range 1 - 12 s at all ion energies studied. Small craters are generally annihilated in a single discrete event whereas larger craters disappear in two or more steps. Each event occurred within the time of 1 video frame capture or 1/30 sec and was produced by a single ion. Lifetime measurements on 14 craters with a mean diameter of 4.4 nm yield a mean lifetime of 4.9 s for craters annihilated in a single step. With the assumption that all ion impacts are capable of annihilating existing craters, the Xe ion has an annihilation cross section for small craters of approximately 85 nm^2 , i.e. an ion impact within a radius of approximately 5 nm of the center of a small crater will annihilate the crater.

The use of carbon collector foils shows that crater formation is not, in general, accompanied by the emission of crater-sized particles although occasionally such a particle is emitted from the surface. A crude estimate of the probability of cluster emission by a 400 keV ion is $\approx 3 \cdot 10^{-5}$ particles/ion corresponding to approximately one particles per 2,500 craters [4].

In the thinnest areas of bulk foil many single ion impacts resulted in the creation of holes [5] having diameters between 5 and 10 nm as shown in figure 2. Approximately 1/2 % of the Xe ions produced holes.

Measurements in the area shown in fig. 2, made with a 50 nm diameter electron beam, indicate that no holes were created in regions with thickness greater than about 50 nm. 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 2-(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 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 [5]. An ion strikes the entire area shown in figure 2 on average every 10

FRAME 1

FRAME 3

FRAME 79

FRAME 80

Figure 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 second)

frames or 0.3 s which is much longer than the cascade lifetime of a few tens of picoseconds. The holes in figure 2 appear between successive video frames (within a time period of 1/30th second) 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 in an enormously high sputtering yield if the atoms were ejected from the gold surface. However, the change in image contrast suggest 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 [6].

A second equally striking observation is the change in the shape and size of holes during continued irradiation as also shown in figure 2. 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 figures 2-(3) and 2-(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 2-(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 figure 2-(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. 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 figure 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 involved with the melt zone. This gives rise to changes in the shape of edges and the tendency of holes to become more circular.

It has been a long-standing observation in our laboratories that thin areas of irradiated gold specimens disappear [3]. This is a consequence of plastic flow during heavy-ion irradiation even if holes do not form. Our *in-situ* observations [5] 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, however, shows this process to be a pulsed localized flow identical to that responsible for the rounding of holes. As in the case of hole production, only a small fraction of ion impacts result in major changes.

SUMMARY

In situ observations demonstrate that impacts of individual xenon ion in the energy range of 50 to 400 keV continuously create and annihilate craters on the surface of gold during room temperature irradiation. These structures are stable at room temperature when irradiation ceases. The efficiency of crater production is ≈ 0.06 craters per ion and the cross section for crater annihilation is approximately 85 nm^2 . Crater formation is not, in general, accompanied by the emission of crater-sized particles although occasionally (≈ 1 per 10000 craters) such a particle is emitted from the surface. Crater creation and annihilation result from radiation-induced flow and surface diffusion driven by cascade energy spikes.

In the thinnest areas of the bulk foils single ion impacts resulted in the creation of holes with diameters between 5 and 10 nm. Approximately 1/2 % of the Xe ions produced holes. Holes result from only a small percentage of ion impacts with other impacts cause cratering while the majority of ion impacts result in plastic-flow annealing of features, holes, craters and specimen edge, having small radii of curvature.

Ion-induced, pulsed localized plastic flow is a fundamental process that changes the morphology of an irradiated gold foil. It is likely to occur in other high-Z materials where displacement cascades are strongly localized such that molten zones form. We believe that plastic flow may also be responsible for other processes occurring in gold under ion irradiation such as ion-induced bubble motion observed during Xe irradiation of Au containing He bubbles [7] and grain-boundary movement [8].

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