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Direct observation of tribological recrystallization

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We present the direct evidence of tribological recrystallization and grain growth in a polycrystalline gold thin film induced by sliding a tungsten tip at ambient temperature using *in situ* transmission electron microscopy.

Keywords: nanotribology; recrystallization; grain growth; *in situ* TEM

1. Introduction

The fundamental processes of tribology related to friction, wear, and adhesion [1–3] are still not well understood [4,5]. When a single asperity touches and slides on a surface, complex phenomena such as local rearrangements of atoms, permanent change of the surface structure [6], and more macroscopic phenomena such as dislocation motion or microstructural changes can occur. As the interface is buried, very few studies that directly observe microstructural changes during sliding have been performed, and much of our existing knowledge is based upon the forensic analysis of probable processes taking place. Related studies have been done using *in situ* transmission electron microscopy (TEM) by controlling the temperature and/or stress on polycrystalline materials, enabling the study of dynamic phenomena in greater detail [7,8]. These have used a strain holder (with or without heating) mounted inside the TEM system [9–11]. While they provide invaluable information about global processes, it is not completely clear how one should translate this to a single asperity problem. More relevant, *in situ* nanoindentation studies have been performed, for instance induced grain growth in polycrystalline aluminum [7]; again, nanoindentation is not the same as tribological sliding. For a recent review of relevant *in situ* studies, see [12].

In this study, an external tungsten tip was gently slid on a polycrystalline gold film and mechanically induced site-specific recrystallization and grain growth were observed. This allows site-specific systematic studies of local changes in the film microstructure related to wear, scratching, and sliding.

2. Experimental details

A thin film of gold, about 27 nm thick, was sputtered at room temperature in a vacuum of 10^{-5} Torr onto lacey carbon films supported on 200 mesh TEM grids

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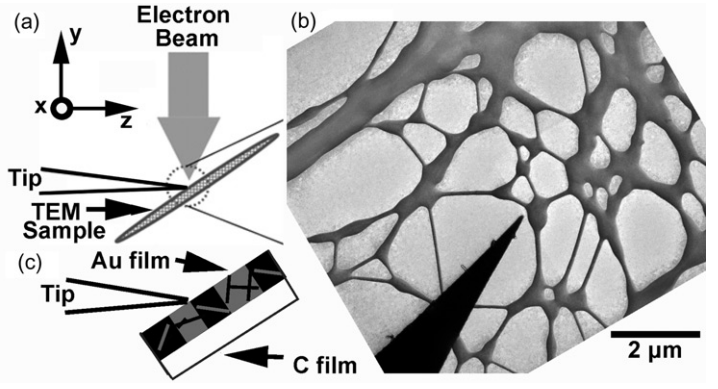

Figure 1. The structure of the tribological system and the sample configuration. (a) Schematic representation of the the positions of the *in situ* TEM holder and tip, (b) BF image showing a W tip above Au film deposited on the network of a C film, and (c) cross-sectional schematic representation of the tip sliding on polycrystalline Au film, sitting on C film. The contrasts in the Au film indicate different orientations and the lines inside the film indicate grain boundaries.

(Ted Pella, Inc., USA). For reference, the nanoscale structure of such a film will be a dense, polyparticle structure [13,14] of partially coalesced multiple-twinned particles (MTPs) with a high density of both twin and grain boundaries as described previously [15], the structure being determined by kinetics, not minimum-energy thermodynamics [16]. The width of the free-standing network typically ranged from ~200 nm to 1.8 μm (Figure 1b). The samples were analyzed using a Tecnai F20 G2 operated at 200 keV, with videos recorded by a conventional in-column TV camera with an acquisition speed of 33 frames per second. Static bright-field (BF) and dark-field (DF) images (using the gold (111)/(200) ring) were acquired using a Gatan CCD camera.

The *in situ* TEM holder (a HS100 STM-TEM-Holder^{TM}, Nanofactory Instruments, Sweden) has been described elsewhere [17]. The TEM grid was mounted onto the holder at an angle of 45° to the horizontal, as shown on the schematics in Figure 1a. The Au polycrystalline film was set facing the tip, as shown in Figure 1c. The TEM sample can be accurately driven by a piezo device with spatial resolutions of 0.2 Å in the X and Y directions and 0.025 Å in the Z direction. The tungsten probe used as a tip was prepared by standard electrochemical polishing methods using a buffered NaOH solution.

3. Results and discussion

Figure 2 shows the frames extracted from a DF video sequence (Supplementary online material) during the sliding of the tungsten tip (asterisks) on the gold film over a distance of ~100 nm, with the time elapsed indicated. The contrast in the figures shows classic diffraction contrast associated with grain orientation and perhaps thickness, not porosity. In Figure 2b and c, the applied force of the tip on the film slightly decreases due to the increase of the relative height when sliding in

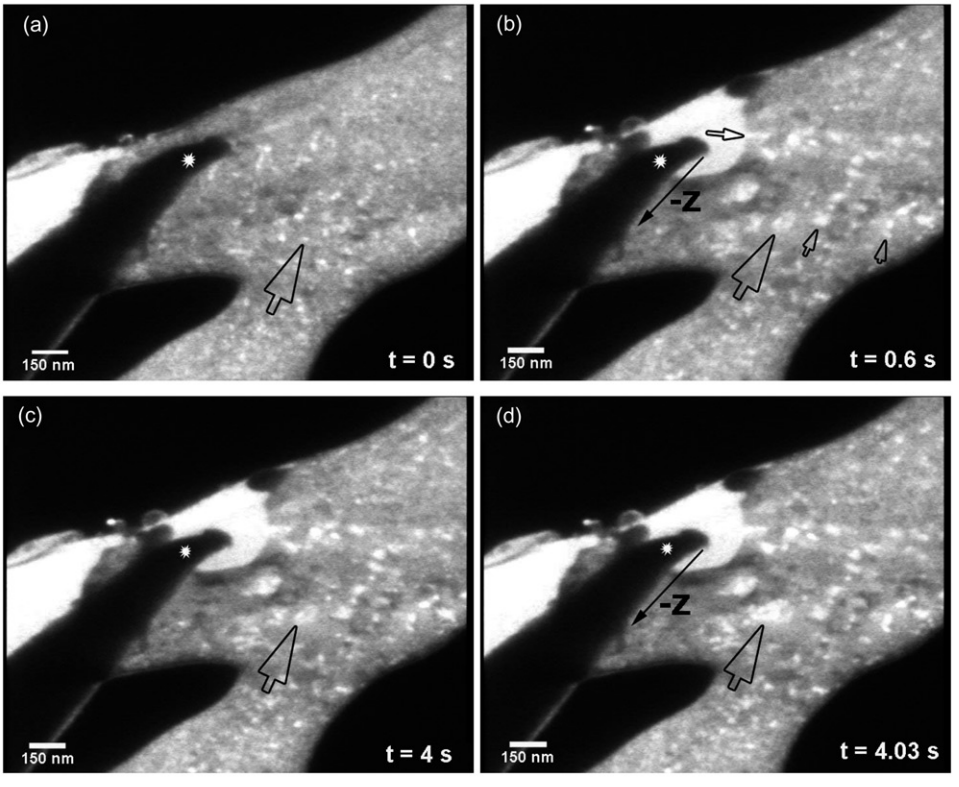

Figure 2. Time-elapsed DF images captured from video recorded by an in-column TV camera. The asterisks indicate the tungsten tip. The long black arrows in (b) and (d) indicate the tip sliding in Z direction. The arrows point to grains undergoing mechanically induced grain-growth. The two big white areas are amorphous lacey carbon and the darker background other than the tip region corresponds to vacuum.

the Z direction. The large white region on the left-hand side in Figure 2a shows an area where the Au film was removed during a previous sliding sequence. The initial size of the grains in the Au film on the right-hand side of the probe in Figure 2a is less than 20 nm and about 9 nm on average. Based on the videos captured by CCD camera, the lateral sliding speed was about 200 nm s^{-1} during the measurement. The radius of the tip was initially about 40 nm and was 50 nm at the end of the measurements.

Comparing Figure 2a before sliding, the video (Supplementary online material) during sliding with Figure 3 after the tip was retracted clearly indicates a remarkable change in contrast near the contact area at distances much larger than the region of contact. In Figure 3a, the edge, indicated by the white arrow, shows the nearly uniform contrast of a single crystal instead of the mottled diffraction contrast of nanograins, indicated by the dark arrow. The dark arrow in Figure 3a also shows a nanocrystalline region as a reference, demonstrating that the structural change is quite local. This is reminiscent of recrystallization observed in some regions of similar thicker films [15]. Clearly, the sliding process is initiating recrystallization

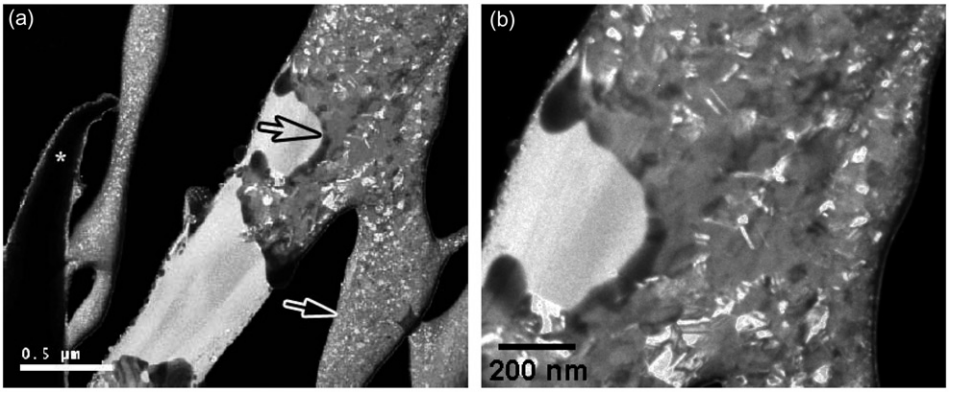

Figure 3. A DF image acquired from the investigated region after mechanical probing. (a) Lower magnification and (b) enlargement around the recrystallized region. The region around where the probe slid on the film shows remarkably uniform contrast indicating a large recrystallized single crystal, which can be contrasted with the nanocrystalline material elsewhere. The large arrow indicates the immediate vicinity of the region where sliding occurred, and the small arrows indicate the virgin Au film. As in Figure 2, the asterisk indicates the position of the tungsten tip.

which is itself driven by reductions in the stored elasticity (from the disclinations in the multiply-twinned particles [18]) and grain-boundary energy.

In addition to recrystallization, rapid grain growths in nearby crystals were also observed. Specifically, after 4 s when the tip was retracted, the grain indicated by an arrow in Figure 2 underwent rapid grain growth faster than the time resolution of the TV-rate camera, as shown in Figures 2c and d. The velocity of the recrystallization front from Figures 2c to d, indicated by the larger arrow, was determined to be faster than $3\,\mu m\,s^{-1}$.

The results we have described are, in some respects, not unexpected; the perturbations induced by the tip initiate processes which lead to an overall relaxation of the microstructure to a lower free-energy configuration in the local region near the tip, where "near" here means within about 0.8 μm. We will argue that such processes almost certainly take place in a very general fashion near a sliding asperity, and as such are almost certainly important processes to consider during tribological microstructure evolution. How fast they occur is somewhat an open question, probably some respectable fraction of the local speed of sound in the material, making this a very good candidate for dynamic TEM experiments (e.g. [19]).

4. Conclusion

We have presented microstructural evolution in thin Au films due to tip sliding on the surface. Mechanically induced rapid recrystallization and grain growth at ambient temperature was observed directly. The driving force for mechanically stimulated recrystallization and grain growth originates from the stored energy in the Au films.

Supplementary material

Video caption: real-time video of the tip sliding on the nanocrystalline gold film. The bright region in the lower left is from an earlier experiment. Single frames from this video are shown in Figure 2 of this article.

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