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QUANTITATIVE *IN SITU* NANOINDENTATION OF ALUMINUM FILMS

Andrew M. Minor*, Eric A. Stach†, and J. W. Morris, Jr.*

*Department of Materials Science and Engineering, University of California, Berkeley, and Materials Science Division, Lawrence Berkeley National Laboratory, One Cyclotron Road, Berkeley, CA 94720
†National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, One Cyclotron Road, Berkeley, CA 94720

We report the development of a method for quantitative, *in situ* nanoindentation in an electron microscope and its application to study the onset of deformation during the nanoindentation of aluminum films. The load-displacement curve developed during *in situ* nanoindentation shows the characteristic "staircase" instability at the onset of plastic deformation. The instability corresponds to the first appearance of dislocations in previously defect-free grains, and occurs at a force near that measured in conventional nanoindentation experiments on similarly oriented Al grains. Plastic deformation proceeds through the formation and propagation of prismatic loops punched into the material, and half-loops that emanate from the sample surface. This new experimental technique permits the direct observation of the microstructural mechanisms that operate at the onset of deformation.

In nanoindentation a material surface is indented with a small diamond pyramid that has a tip radius in the range 50-400 nm. This method of hardness testing has become an important tool for both scientific research and materials characterization.[1,2] Since nanoindentation measures the mechanical properties of volumes so small that they can be made defect-free, it probes the fundamental processes that initiate deformation. More practically, it measures the mechanical behavior of small systems, including thin films and microelectromechanical devices (MEMS).

The interpretation of nanoindentation data is not always clear. For example, since most metals form native oxides, yielding under the nanoindenter may be governed by fracture of the oxide film rather than the onset of plastic deformation in the material itself.[3] It is difficult to resolve such issues when the microstructure of the material can only be studied after the fact. However, the recent development of a unique *in situ* stage for transmission electron microscopy (TEM) has made it possible to image nanoindentation in real time.[4] The force-displacement relation can be measured simultaneously with a calibrated piezo-ceramic control element. Initial applications of these new tools are described below.

During *in situ* nanoindentation, as we practice it, a 3-sided boron-doped diamond indenter approaches the sample in a direction normal to the electron beam (Fig. 1). The sample is a thin film deposited onto a silicon substrate that includes a narrow wedge. The indentation is made in the cap of film on the flat top of the wedge. In order to be transparent to a 200 kV electron beam, the cap width must be less than about 500 nm.

The indenter is mounted on a piezo-ceramic actuator, which both controls its position and forces it into the edge of the sample. The piezo-ceramic actuator is also used to measure the force developed as a function of displacement during the test. It must be calibrated in order to do this. The force is determined by the combination of voltage and displacement, and the relation between them must be measured. In the present work the voltage-force-displacement characteristic of the actuator was found by bending micro-scale silicon cantilevers *in situ* in the TEM. (Conveniently, for a given voltage the force on the actuator is linearly related to the displacement.) During a nanoindentation test the voltage on the actuator is measured electronically, the displacement is measured by direct observation in bright-field TEM, and the force is computed from the actuator characteristic. (The appropriate conversion from force-displacement to the stress and strain distribution is under investigation.)

Fig. 2 shows an example force-displacement curve from an *in situ* nanoindentation test. The film was vapor-deposited Al, with a grain size of 250-400 nm. An Al grain at the cap was indented in a <113> direction, and monitored continuously in bright field TEM. The test was filmed, and example TEM micrographs were copied from the film to illustrate characteristic stages of the indentation process.
Fig. 2a shows the indenter and the defect-free Al grain just before contact. In Fig. 2b (point B on the curve) the indenter imposes an elastic strain, indicated by the dark strain contour in the micrograph. The indentation remains elastic until the film yields, at point C. At yield (Fig. 2c) a dislocation forms at the surface and propagates into the film. After yielding the indenter penetrates at nearly constant load (region D), while dislocations form and flow into the grain to preserve an essentially constant dislocation density (Fig. 2d). The test was stopped after about 30 nm indentation since the diameter of the contact area (> 100 nm) had become significant with respect to the radius of curvature of the sample tip. While the native oxide at the film surface is not clearly imaged, oxide rupture does not appear to play an important role; there is no yield point, and dislocations appear at the onset of plastic deformation.

This particular example was chosen because the force-displacement relation can be directly compared to one published by Gouldstone, et al.,[5] who indented an epitaxial Al film in the same direction with an indenter of similar tip geometry (radius ~50 nm). The initial portion of the Gouldstone data is superimposed on the *in situ* nanoindentation curve in Fig. 2. The data are in good agreement through yield and initial plastic flow. However, the Gouldstone data has the "staircase" shape that is typical of nanoindentation of ductile metals.[6] The steps in the staircase are, presumably, due to the fact that hardening or defect exhaustion requires successively higher stresses to continue plastic deformation. In the *in*

situ nanoindentation tests done to date on Al, plastic flow is not exhausted at displacements up to 30 nm, and the second step of the staircase is not seen. We believe this is due to the geometry of the *in situ* specimen, which provides a much larger free surface to absorb dislocations and minimize hardening.

In order to study the crystallographic mechanisms of nanoindentation in detail it is more useful to use dark-field TEM. In a dark field condition, the image is formed by using a strongly diffracted beam of electrons from one grain only. Unfortunately, it is not possible to image the indenter simultaneously with a dark-field image of the grain, so the relevant force-displacement relations must be acquired separately.

Figs. 3 and 4 show examples of the information that can be obtained from dark-field TEM images. Fig. 3 includes a series of micrographs from the video record of an *in situ* nanoindentation of an Al grain that was oriented in approximately the [-1-11] direction. Figs. 3(a) and 3(b) are micrographs taken prior to indentation in bright field, and dark field respectively. Figs. 3(c) and 3(d) show the evolution of nanoindentation damage. As expected, dislocations glide along close-packed {111} planes, and multiply and tangle as the deformation proceeds.

Fig. 4 is a more detailed examination of the development of dislocations near the yield point of the sample shown in Fig. 3. The top picture shows the entire Al grain at +0.6 seconds, followed by a series of pictures taken 0.1 seconds apart and cropped from the region outlined by the white box in the top picture. The most striking feature is the sequential appearance of prismatic dislocation loops on an axis approximately 45° from the direction of indentation, a mechanism of nanoindentation-induced deformation that has been suggested by several researchers.[5,7,8] The loops have <110> Burger's vectors and lie in {111} planes. While they presumably form at the surface, their nucleation and migration to the site at which they appear is too rapid to capture, as is their motion to new equilibrium positions as additional prismatic loops are punched into the grain.

Finally, we note that the dislocations introduced during nanoindentation anneal out of the foil very quickly after the load is removed. This observation suggests that TEM studies of indentation damage taken after nanoindentation experiments should be viewed with some caution.

The nanomechanical behavior of small solid volumes exhibits fundamental phenomena that are obscured on coarser scales. We have presented a novel experimental technique that can image in real time the discrete deformation mechanisms of nano-scale volumes, while simultaneously correlating this behavior with quantitative information.

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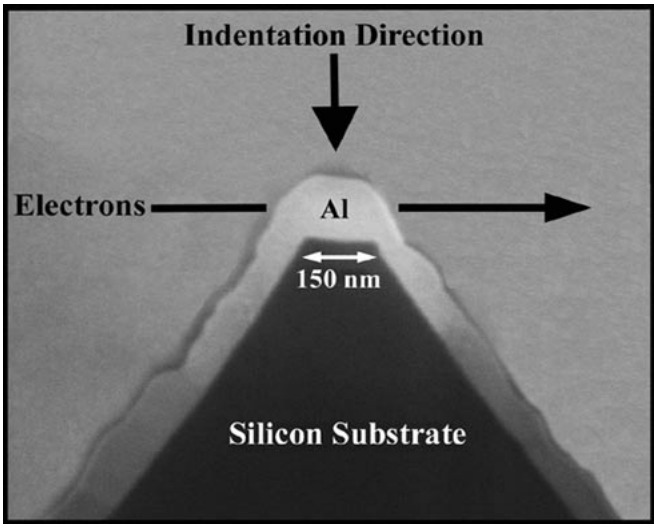

Figure 1. Cross-section of an *in situ* nanoindentation sample. An Al film ~250 nm thick is deposited onto a wedge-shaped perturbation on a Si substrate.

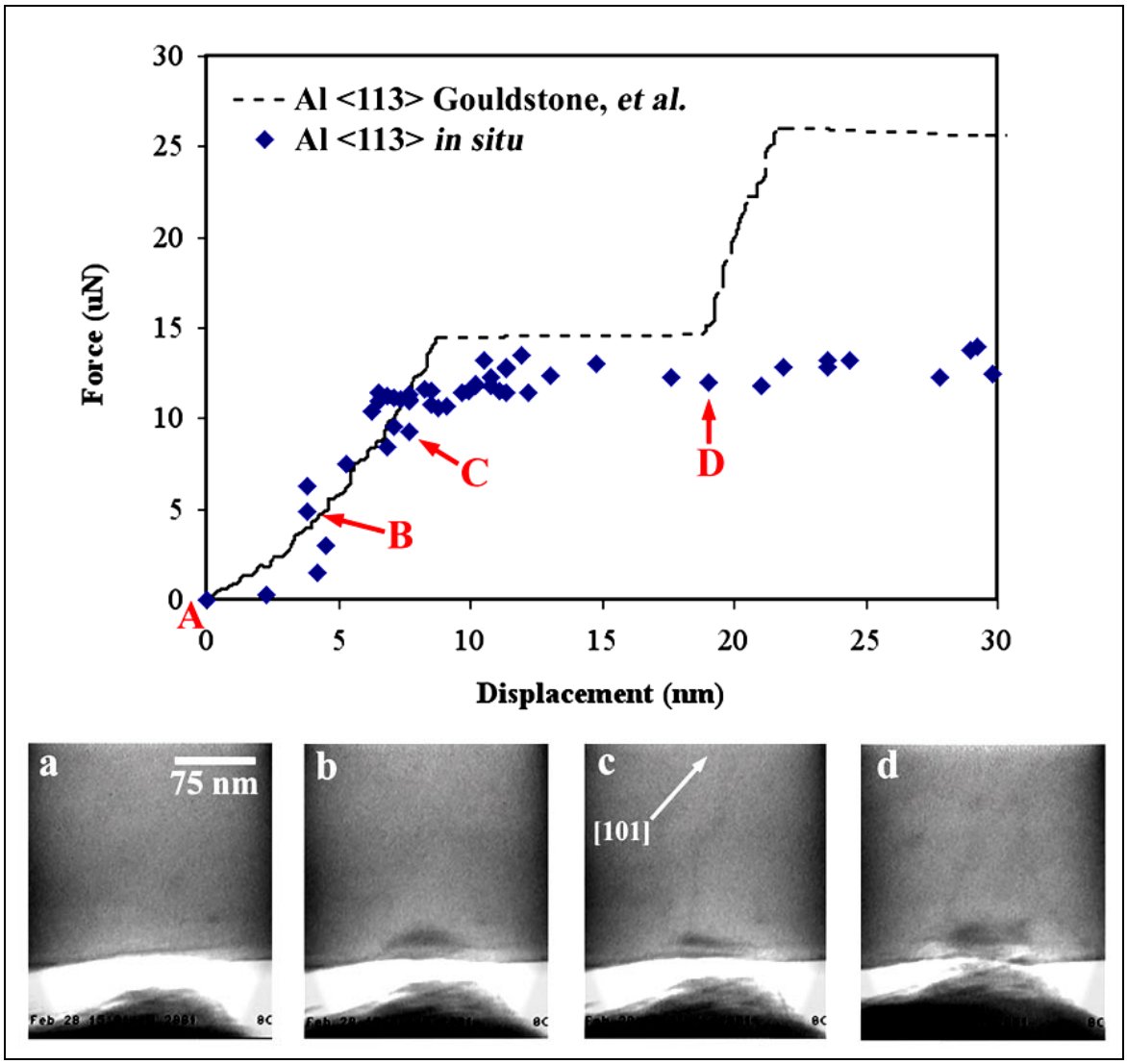

Figure 2. Force-displacement curve and representative micrographs from an Al grain indented *in situ* in the <113> direction. The nanoindentation data of Gouldstone, et al.[5] is superimposed for comparison. Pictures a, b, c and d correspond to points A, B, C, and D on the graph, respectively. Due to contrast effects, the very tip of the diamond indenter is not visible in the micrographs.

Figure 3. Time series of an Al grain showing the evolution of plastic deformation during an *in situ* nanoindentation: (a) Bright field and (b) dark field image before indentation. The dark field condition used was [1-11]. The stripes are thickness contours due to the wedge shape of the specimen. (c) Dark-field micrograph at t =1.4 seconds showing dislocations on {111} planes. (d) Dark field micrograph at t = 2.8 seconds showing multiple dislocations.

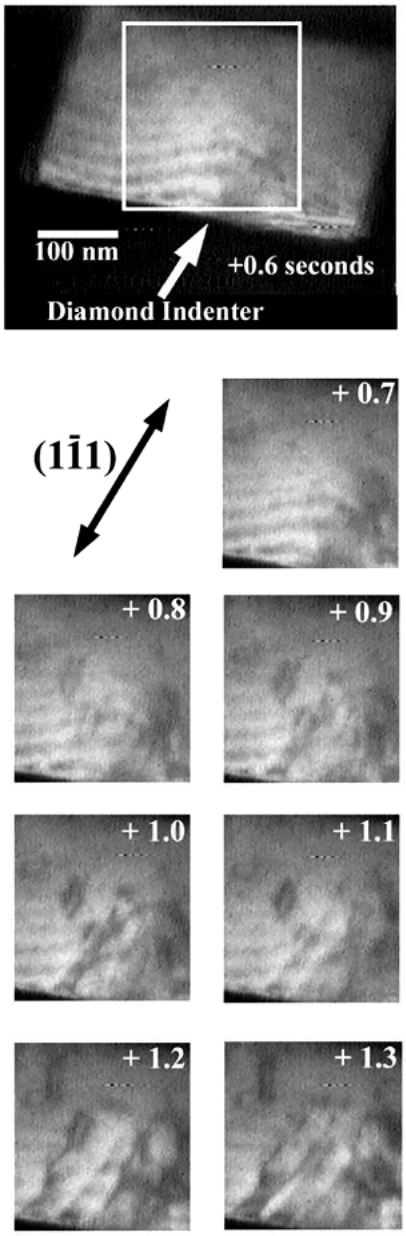

Figure 4. Series of dark field transmission electron micrographs showing the appearance and evolution of prismatic dislocation loops in a previously undeformed part of the Al grain during an *in situ* nanoindentation. The numbers in the micrographs are the time in seconds from the start of the indentation.