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Cryogenic indentation-induced grain growth in nanotwinned copper

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Nanocrystalline copper thin films with as-deposited Σ3 twin boundaries were indented while immersed in liquid nitrogen. Quantification using precession-enhanced electron diffraction determined the crystallographic texture and grain-to-grain misorientation of the undeformed, pile-up and compressed regions. Grains in the undeformed region retained a high density of Σ3 recrystallization twins, whereas the pile-up showed significant coarsening, prevalent Σ7 subgrain formation and a decrease in twin boundaries. The abnormal grain growth is attributed to a detwinning mechanism. The compressed region showed significant grain refinement.

Keywords: Grain growth; Microindentation; Twinning; Coincidence lattice; Precession enhanced electron diffraction

Classically, grain growth is considered to be the product of a driving force and intrinsic boundary mobility. Driving forces originate from stored deformation energy, grain boundary energy, surface energy, elastic energy, or from a chemical and/or thermal gradient [1]. Boundary mobility refers to the kinetics necessary to allow the atoms to move in response to that driving force. Lack of sufficient mobility and driving force prevents systems from achieving their lowest-energy microstructural configuration. Since mobility is thought to be related to atomic diffusion, temperatures of approximately one-half or higher of the homologous temperature (T_m) are usually required to observe substantial grain growth [2]. When the grain growth is self-similar, it is referred to as normal growth. In some cases, a single grain or an aggregate of grains coarsen at a faster rate than others in the microstructure, a phenomenom referred to as abnormal grain growth (AGG).

As grain sizes approach the nanometer regime, they can exhibit peculiar grain growth behavior not observed in larger grains. For example, nanocrystalline metals have been shown to undergo both moderate, widespread coarsening as well as dramatic AGG as a result of monotonic tensile deformation [3], high-cycle fatigue [4], low-cycle fatigue [5] and indentation [6] at moderate ($<0.5T_m$) to low ($<<0.5T_m$) temperatures. In the case of fine-grained Cu, samples that are simply left at room temperature have been shown to display abnormal coarsening [7]. Zhang et al. [6] have reported transmission electron microscopy (TEM) studies in which AGG at room temperature occurred under in situ indentation. In this report, the starting grain size was 40 nm. After indentation, certain grains achieved sizes of nearly 300 nm. Even more surprising was a series of ex situ cryogenic indentations at −190 °C where the grains grew up to 700 nm. At temperatures of a few hundred °C, typical boundary velocities for Cu are on the order of 10^{-9} m s^{-1} [1]; at −190 °C one might expect grain boundary migration to occur only on geological timescales. To date, the ability for grains to grow under cryogenic indentation, where low temperatures are insufficient for reasonable mobility, is not well understood.

In this paper, a series of nanocrystalline copper films with a high density of twins was subjected to indentation at liquid nitrogen temperatures. The films' grain boundary misorientations were quantified to understand which grain boundary types are present after cryogenic mechanically induced grain growth. These remnant grain boundaries could be either pre-existing from the parent grain structure or induced by indentation, such as subgrains or cell walls; they could be either active boundaries which are responsible for grain growth or static boundaries which impede grain growth.

Copper films ~500 nm thick were grown by a pulsed laser deposition (PLD) technique onto a polished 0.5 mm thick silicon substrate. The deposition was accomplished using a previously described setup [8] which has been upgraded with a 248 nm KrFeximer

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laser operating at 35 Hz and 500 mJ. The base pressure during deposition was 4×10^{-7} torr. The deposition process led to columnar-type grains normal to the substrate with a high density of twin boundaries, oriented either horizontally or inclined to the substrate, as determined through TEM micrographs shown and discussed below. This type of microstructure is described as "nanotwinned". The twin-to-twin spacing was typically of the order of tens of nanometers. X-ray diffraction (XRD) measurements confirmed that the film had strong {1 1 1} and weak {2 0 0} out-of-plane texture intensities.

Cryogenic indentation of the Cu film was carried out using a custom-built immersion indenter, shown schematically in Figure 1a. A sample is held in place by a spring-loaded platen, visible in Figure 1a. The hollow tube allows a long rod to raise and lower the carriage containing the Vickers indenter tip, attached to a lightweight shuttle (Fig. 1b). The shuttle holds the indenter tip normal to the sample, as well as serving to support small cylindrical tungsten weights, which can be added to increase the normal force applied during indentation (Fig. 1c). For this experiment, additional weights were used to achieve 0.5 N normal force. The assembly was designed to operate either during immersion in a liquid cryogen or in air. For this experiment, the sample was submerged in liquid nitrogen (LN_2). A 1 cm × 1 cm film-on-substrate sample was held in place by a spring-loaded platen (Fig. 1b), after which the assembly head was submerged until rapid boiling of the LN_2 ceased, signifying that the indenter temperature had equilibrated with its environment. The shuttle carriage was then lowered at a rate of approximately 0.08 mm s^{-1} until the indenter tip contacted the sample surface (Fig. 1c) and held for 60 s. After 60 s of indentation, the tip was retracted and the sample was removed from the LN_2 and allowed to warm by sitting at room temperature.

Samples from the pre- and post-indentation film were prepared for TEM analysis with a focused ion beam (FIB) milling and lift-out procedure [9] using a FEI Quanta 3D dual-beam FIB with an in situ Omniprobe lift-out micromanipulator. For the post-indentation sample, the site-specific region was extracted such that the undeformed, pile-up and compressed regions of the indent were captured in one foil. During the final milling steps, the acceleration voltage was reduced from 30 to 5 keV to reduce surface milling damage. After the foil had been extracted and attached to a TEM grid, the TEM specimen was polished further using a Gatan Precision Ion Polishing System (PIPS) at 2.5 keV, 1.0 mA and an inclination angle of 8°. This last step was found to significantly improve foil quality by removing any residual FIB damage, yielding crisp bright-field and precession-enhanced diffraction patterns. Particular care in preparation minimized any possibility of ion-beam-induced effects on grain growth [9]. The localized nature of the indentation facilitated side-by-side comparison of the as-deposited and deformed grain structures.

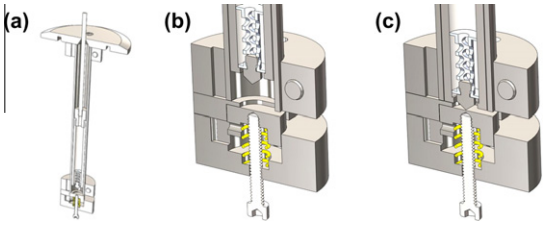

Figure 1. Cross-section schematic showing (a) components of the indenter apparatus as well as the (b) unloaded and (c) loaded positions. Cylindrical weights and sample are not shown.

The TEM foils were analyzed in a FEI Tecnai F20 Supertwin microscope operating at an accelerating voltage of 200 kV and fitted with a NanoMEGAS ASTAR precession-enhanced diffraction orientation identification platform [10]. The NanoMEGAS platform generates pseudo-kinematic electron diffraction conditions, which allow grain orientation to be measured at a spatial resolution of ~10 nm. This facilitates the collection and analysis of not only grain size but also misorientation distributions. The precession angle used was 1° at step sizes of 10 nm.

The cross-sectional inverse pole figure orientation map of the undeformed film is shown in Figure 2b. A prevalence of Σ5 boundaries and low-angle boundaries at the substrate surface, where growth was initiated, is evident in the color trace map of boundary type. The columnar grains from the initial growth surface are separated by these series of low-angle grain boundaries (Fig. 2c). Within these columnar grains, Σ3 twin boundaries spanned the grain diameters. Brandon's criterion [11] was used to distinguish the misorientation range of these coincident site lattice (CSL) values. In Figure 2d, the twins shown in red are the primary recrystallization twins with a plane normal of {1 1 1} and a direction of ⟨111⟩.

After cryogenic indentation, a complimentary series of images was captured in the indented material. Figure 3a shows SEM plan and tilted views of an indented region, with indication of the orientation of the cross-sectional FIB milling. The grain orientation map of the indented sample is shown in Figure 3c. Within this figure, clear evidence of grain coarsening is present in the pile-up region near the edge of the indent. Many of these coarsened

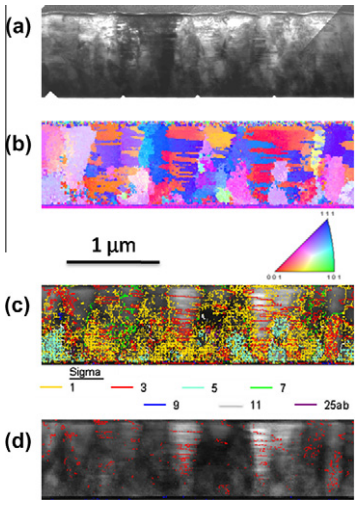

Figure 2. As-deposited nanotwinned Cu film in cross-section: (a) TEM bright-field image; (b) inverse pole figure orientation map; (c) image quality map with CSL boundaries highlighted; (d) image quality map with twin boundaries highlighted in red.