Available online at www.sciencedirect.com

Acta Materialia 58 (2010) 4772–4782

www.elsevier.com/locate/actamat

In situ TEM study of microplasticity and Bauschinger effect in nanocrystalline metals

Jagannathan Rajagopalan [a], Christian Rentenberger [b], H. Peter Karnthaler [b], Gerhard Dehm [c], M. Taher A. Saif [a,*]

[a] Mechanical Science and Engineering Department, University of Illinois at Urbana-Champaign, 1206 W Green Street, Urbana, IL 61801, USA
[b] Physics of Nanostructured Materials, Faculty of Physics, University of Vienna, Boltzmanngasse 5, A-1090 Wien, Austria
[c] Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, and Dept. Materials Physics, University of Leoben, Jahnstr.12, 8700 Leoben, Austria

Received 11 March 2010; received in revised form 3 May 2010; accepted 4 May 2010
Available online 9 June 2010

Abstract

In situ transmission electron microscopy straining experiments with concurrent macroscopic stress–strain measurements were performed to study the effect of microstructural heterogeneity on the deformation behavior of nanocrystalline metal films. In microstructurally heterogeneous gold films (mean grain size $d_m = 70$ nm) comprising randomly oriented grains, dislocation activity is confined to relatively larger grains, with smaller grains deforming elastically, even at applied strains approaching 1.2%. This extended microplasticity leads to build-up of internal stresses, inducing a large Bauschinger effect during unloading. Microstructurally heterogeneous aluminum films ($d_m = 140$ nm) also show similar behavior. In contrast, microstructurally homogeneous aluminum films comprising mainly two grain families, both favorably oriented for dislocation glide, show limited microplastic deformation and minimal Bauschinger effect despite having a comparable mean grain size ($d_m = 120$ nm). A simple model is proposed to describe these observations. Overall, our results emphasize the need to consider both microstructural size and heterogeneity in modeling the mechanical behavior of nanocrystalline metals.

Keywords: Nanocrystalline materials; Plastic deformation; Yield phenomena; Microstructural heterogeneity; Thin films

1. Introduction

The deformation behavior of nanocrystalline metals, which typically have mean grain sizes around 100 nm or less, has attracted extensive interest in recent years. Unlike coarse-grained polycrystalline solids, where plasticity is dominated by dislocations generated by intragranular dislocation sources [1], alternative deformation mechanisms can be activated in nanocrystalline metals due to the paucity of intragranular dislocation sources and the difficulty in activating them [2,3]. The alternative deformation mechanisms include grain boundary (GB) diffusion and sliding [4], GB migration [5], twinning [6] and grain rotation [7]. As a result, nanocrystalline metals exhibit several unusual characteristics, such as high strength [8], high strain rate sensitivity [9,10] and stress-induced room temperature grain growth [11,12].

Molecular dynamics (MD) simulation has been one of the most widely used approaches to model deformation processes in nanocrystalline metals [13,14]. MD simulations suggest that plasticity in nanocrystalline metals is carried by partial dislocations nucleated at GBs; in materials such as Cu and Ni only one partial is emitted, leading to the formation of stacking faults, whereas in Al a trailing partial follows, resulting in a twin or a full dislocation [15]. Recent studies [16], though, have emphasized the need to consider the constraints in MD simulations, such as the small time scales, in interpreting these results. Qualitative in situ transmission electron microscopy (TEM) [17–19] has been

---

[*] Corresponding author. Tel.: +1 217 333 8552; fax: +1 217 244 6534.
E-mail address: saif@illinois.edu (M.T.A. Saif).

1359-6454/$36.00 © 2010 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.actamat.2010.05.013

another major technique used to probe deformation mechanisms in nanocrystalline metals. These studies have shown that dislocation-mediated plasticity continues to be a dominant mechanism in nanocrystalline metals with mean grain sizes above 30 nm. In single crystal gold films, transition from perfect to partial dislocation plasticity has been observed at a thickness of 80 nm [20]. However, despite the tremendous advances in the modeling and experimental characterization [21], some major aspects of the deformation behavior of nanocrystalline metals still remain unclear.

One such aspect is the elastic–plastic transition and the macroscopic yielding of nanocrystalline metals. In coarse-grained metals the macroscopic yield stress is usually defined by the stress at 0.2% offset strain, with the assumption that the majority of the grains are deforming plastically at this point. So far, this criterion has largely been applied to determining and comparing the yield stresses of nanocrystalline metals. However, recent reports have revealed a significantly extended microplastic regime in nanocrystalline metals and questioned the validity of this yield criterion [22,23]. To account for the extended microplasticity, a tangent modulus-based approach [24,25] to determine the elastic–plastic transition has been suggested. Combining such an approach with in situ X-ray diffraction experiments, a new criterion for yielding in nanocrystalline metals has recently been proposed [26].

Another aspect that has received relatively little attention is the effect of microstructural heterogeneity on the deformation behavior of metals in the nanocrystalline transition regime (mean grain size between 50 and 150 nm). This is surprising, since nanocrystalline metals synthesized by severe plastic deformation often have grain sizes in this regime and are known to have a highly heterogeneous microstructure [27]. Understanding the relationship between microstructural heterogeneity and the deformation behavior is especially relevant since recent investigations [28,29] have revealed that the unusual plastic strain recovery in nanocrystalline metals [30,31] is a direct consequence of microstructural heterogeneity. However, a detailed mechanistic understanding of how microstructural heterogeneity influences the mechanical behavior is still lacking. TEM experiments allow such mechanistic investigations, provided the macroscopic material response is measured while the microstructure is visualized. Until now, such studies have been limited due to the lack of appropriate instrumentation.

In this work, we studied how microstructural heterogeneity affects the deformation behavior of nanocrystalline metal films (mean grain size 70–140 nm) in which dislocation-mediated plasticity dominates. Towards this end, we synthesized free-standing nanocrystalline gold and aluminum films with highly dissimilar microstructures and performed in situ TEM straining experiments with concurrent macroscopic stress–strain measurements. Our experiments show that the extent of microplasticity as well as the overall deformation response of nanocrystalline metals is strongly influenced by heterogeneity. The gold films, which have a heterogeneous microstructure, show large microplastic deformation and do not yield macroscopically even at 1.2% applied strain. In contrast, the microstructurally homogeneous aluminum films show limited microplastic deformation and a markedly sharper elastic–plastic transition. Furthermore, our experiments reveal the mechanism for the unusual Bauschinger effect observed in free-standing metal films [32].

2. Experimental details

A 160 nm thick gold film and a 225 nm thick aluminum film were sputter deposited on Si (0 0 1) wafers. Before deposition, the native silicon dioxide layer on the Si wafers was removed by hydrofluoric acid etching and the wafers were immediately transferred to the sputtering chamber to avoid regrowth of the oxide layer. From these wafers, free-standing gold and aluminum tensile specimens were co-fabricated with microtensile devices using the process described in Ref. [33].

The thickness of the thin film specimens fabricated using this process is highly uniform, with a variation of less than 5 nm, unlike conventional TEM samples. In addition, these tensile testing devices ensure uniform uniaxial loading and unloading of the specimen and eliminate bending and torsion to a large extent [34]. They are also compatible with the standard TEM straining specimen holders, and have built-in force and displacement sensors that allow the measurement of the macroscopic stress and strain during in situ straining. This allows us to directly relate the microstructural changes during deformation to the macroscopic behavior and thus establish the structure–property relationship. The maximum strain that can be imposed on the thin film specimens with these devices is around 6–7%, which precludes the observation of very large ductility in our specimens. However, this is not usually a limitation because the failure strain does not exceed 2% in majority of the specimens. The relatively low failure strain is due partly to the large aspect ratio of the specimens (length 350 μm, thickness 150–250 nm) and partly to flaws introduced during the fabrication.

Uniaxial tensile load–unload experiments were performed using a displacement controlled, single tilt, straining specimen holder in a Philips CM200 transmission electron microscope at an accelerating voltage of 200 kV. The loading and unloading on both the gold and aluminum specimens were performed in a series of steps; a set of two to three displacement pulses (strain of ≈0.05–0.1%) were imposed and the evolution of the microstructure was observed for a period of a few minutes at constant displacement. During all cycles, the microstructural observations were made under bright field conditions and recorded using a real-time TV-rate camera (30 frames per second) with an image intensifier.

In the case of the gold specimen, grains with favorable orientation for dislocation glide were identified before straining using the following procedure: dark-field images were taken using diffraction spots that were aligned parallel