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In situ TEM observations of fast grain-boundary motion in stressed nanocrystalline aluminum films

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Abstract

Free-standing nanocrystalline Al thin films have been strained in situ in a transmission electron microscope at room-temperature. Extensive grain-boundary migration accompanies the in situ loading and has been observed to occur preferentially at crack tips and only in the presence of the applied stress. This grain growth precedes dislocation activity, and measured boundary velocities are greater than can be explained by diffusive processes. The unambiguous observations of stress-assisted grain growth are compatible with recently proposed models for stress-coupled grain-boundary migration. The growth occurs in a faceted manner indicative of preferential boundaries. The fast collapse of small grains with sizes of 30–50 nm demonstrates the unstable nature of a nanocrystalline structure. Clearly observable shape changes testify to the effectiveness of grain-boundary migration as a deformation mechanism, and preferential grain growth at crack tips resulted in efficient crack tip blunting, which is expected to improve the films' fracture toughness.

Keywords: In situ transmission electron microscopy (TEM); Grain-boundary migration; Nanocrystalline film; Aluminum

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1. Introduction

Room-temperature plasticity in polycrystalline metals and alloys is commonly understood and modeled in terms of dislocation nucleation, mobility, and interaction with various obstacles. For metals and alloys, the primary role of grain boundaries is thought to be that of obstacles to dislocation motion, and the widely referenced Hall–Petch relation [1,2] predicts that decreasing the average grain size, effectively increasing the density of grain boundaries, results in increased strength. This relation has been shown to hold for a wide range of materials and grain sizes, and considerable interest has been placed on extrapolating this relationship to the nanocrystalline regime. There is clear evidence to indicate that the strength of nanocrystalline metals falls short of Hall–Petch predictions at the smallest grains sizes (below 100 nm) [3–6]. This realization and the observation that nanocrystalline metals do possess at least moderate levels of ductility have led the community to consider alternative deformation mechanisms. Proposed alternative room-temperature deformation mechanisms of nanocrystalline metals have included: grain-boundary sliding [7], grain rotation [8,9], diffusional creep [10,11], grain-boundary migration, dislocation nucleation or absorption at grain boundaries [12–16] and enhanced partial dislocation activity [17–22]. The focus of this manuscript is on the motion of grain boundaries (GBs).

At variance from dislocation plasticity, GB-mediated plasticity occurs mainly through diffusive processes. In coarse-grained or microcrystalline materials, GB migration and Coble creep are temperature-dependent and related to the diffusion of atoms across or along the grain-boundary. Diffusion-induced grain-boundary migration (DIGM) has been observed in a number of materials and is a mechanism that applies to all types of grain boundaries. DIGM has been widely studied [23–25] and requires a driving force that derives from a difference in free energy on either side

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of a GB. This energy difference can arise from several factors such as solute concentration, GB surface tension (curvature), or non-uniform stress states. Balluffi and Cahn [25] proposed a mechanism for DIGM in which differences in the diffusion coefficients of the diffusing species along the GBs cause a self-sustaining climb of grain-boundary dislocations and motion of the associated GB steps. Other mechanisms and driving forces that have been used to describe diffusion-related GB motion include: Coble creep [10], which is only effective at high temperatures [26] in large-grained polycrystals; reduction in the number and curvature of GBs through homogeneous grain growth [27]; and local shuffling of atoms between the GB and the adjacent grains (short-range diffusion process) [28]. The last process is based on in situ TEM experiments [29] showing defect-free jerky GB migration and is described in detail by Babcock and Balluffi in [30]. The idea is that it is energetically favorable, as shown for a $\Sigma 5$ boundary, to displace atoms by shuffling over very short distances in each unit cell of the GB.

GB migration can also be carried out by athermal shear, without diffusion. This is predictable when the misorientation between two grains is small, as in the case of low-angle tilt boundaries that can be described as discrete networks of dislocations [31]. The low-temperature displacement of a low-angle GB thus results from the glide of this dislocation array. This has been demonstrated experimentally in Al polycrystals under creep conditions at 200 °C [32] and for Al bicrystals [33]. Traditionally this mechanism is envisaged to operate for a low-angle GB, but recent theoretical descriptions by Cahn and colleagues [34,35], molecular dynamics (MD) [36,37] simulations and experimental observations [38–40] have shown that stress-coupled grain-boundary migration operates for high-angle grain tilt boundaries as well. The perpendicular motion of both low- and high-angle GBs can be framed in terms of their dislocation structure, as determined by the Frank–Bilby equation. The coupling between the applied shear stress and the GB motion, either positive or negative, can result in potential back-and-forth motion of the GB. As shown by MD, the change in shear stress/motion coupling can be attributed to a change in the dislocation structure of the GB during the movement. This entirely geometric model is very effective at predicting the results obtained from MD, but both approaches are employed on relatively simple, planar, GBs.

Despite obvious fundamental differences between diffusion-based and stress-coupled GB motion, experimental attempts to separate both mechanisms has proven elusive. For example, high-temperature measurement of GB motion in Al bicrystals leads to displacements predicted by the shear coupling GB model [33], but migration energies (E_m) are very similar to those found for DIGM [40]. In fact, E_m, determined from the GB mobility [38,41,42], seems to depend more on its structure [43–45] than on its mode of motion. Attempts to quantify GB mobility are also complicated by the fact that GB structures can change and evolve as they migrate, as evidenced by observations of dislocation emission and absorption during GB migration experiments [46–49]. There is also no clear relationship between the GB energy (E_{GB}) and the grain-boundary migration energy. Plotting E_m as a function of GB characters shows a sharp transition between low- and high-angle GB motion. Typically, E_m for low-angle GBs has been reported to be close to the activation energy for bulk diffusion, while that for high-angle GBs is closer to the activation energy for diffusion along grain boundaries. Although well documented, this low-to-high angle transition is not fully understood [39]. The situation becomes even more complex in polycrystalline materials, where E_m differs significantly from bicrystals [46,50] because of multiple junctions [51]. In real polycrystals, GBs are also not predominantly pure twist or tilt character, and GB migration cannot be conservative and requires diffusion and climb of the various GB dislocations.

In nanocrystalline materials, the influence of GB migration is expected to be critical for several reasons: the small grain size and associated GB curvature increase the stored energy, the high density of GBs and vacancies coupled with reduced diffusion pathways accelerates diffusive processes, and dislocation-mediated plasticity demands much higher stresses. Recent MD simulations suggest that dislocation-mediated plasticity never completely disappears [52] but that the role of stress-assisted GB deformation is increased at nanocrystalline grain sizes. MD simulations suggest that GB deformation can occur by GB sliding [53,54] or by a Coble creep-like diffusive process involving grain growth and coalescence [55,56]. A few authors also consider atoms in high-energy GBs to be very close to their melted state, making the migration of these GBs easier, for instance by atomic shuffling [57]. The possibility of stress-coupled GB motion [35] offers yet another possibility for GB-related plasticity but is yet to be established for a general population of grain boundaries and in the specific case of nanocrystalline metals.

Clean experimental observations of GB sliding and grain rotation in nanocrystalline metals have proven tenuous and evidence in support of these mechanisms remains controversial [9,58–60], mainly because it is extremely challenging to image individual and adjacent nanocrystalline grains with conventional in situ TEM. By contrast, the observed growth of nanocrystalline grains by post mortem TEM observations of specimens that have been subjected to an applied stress, through indentation [61–63] or uniaxial loading [64,65], has been clearly documented. Tensile testing of nanocrystalline specimens has proved to be challenging, but testing very small specimens also provides a way to bypass the inherent residual porosity of many nanocrystalline samples [66–68]. Recent advances in micro-tensile testing [3,69,70] allows one to determine the stress–strain response of tiny samples and free-standing films. By coupling micro-tensile thin film testing, in situ synchrotron diffraction experiments and post mortem TEM, Gianola and co-workers recently established grain growth as an active deformation mode in abnormally ductile nc-Al thin films at room-temperature [71]. Moreover, by varying the applied strain-rate during testing, it was