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The formation mechanisms of growth twins in polycrystalline Al with high stacking fault energy

S. Xue [a], Z. Fan [a], Y. Chen [b], J. Li [c], H. Wang [d], X. Zhang [a,c,*]

[a] Department of Mechanical Engineering, Texas A&M University, College Station, TX 77843, United States
[b] Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, United States
[c] Department of Materials Science and Engineering, Texas A&M University, College Station, TX 77843, United States
[d] Department of Electrical Engineering, Texas A&M University, College Station, TX 77843, United States

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ABSTRACT

Growth twins are scarcely observed in metals with high stacking fault energy, such as pure Al. In this study, however, we report the observation of growth twins in sputtered polycrystalline Al films on amorphous substrates and a majority of these growth twins are inclined to the growth direction (inclined twins). Although the fraction of twinned grains is low in general, it increases monotonically with increasing film thickness, reaches a maximum at the film thickness of 80 nm, and decreases gradually thereafter in the thicker films. The nucleation mechanism for the inclined twins is compared with that of the parallel growth twins in Al. Different twin formation mechanisms are discussed. This study provides an alternative perspective to evaluate the formation of growth twins in metals with high stacking fault energy.

1. Introduction

Nanotwinned (nt) metals have been intensely studied and show various unique properties. Cu with nanotwins have excellent electrical conductivity [1,2] while maintaining high mechanical strength. Nt metals, such as Cu and Ag [3–6], also show better thermal stability than monolithic nanocrystalline (nc) metals [7–9]. The mechanical properties of nt metals have also been extensively studied [2,3,9–19]. Nanotwins enhance the mechanical properties of metals via various mechanisms, some of which are briefly summarized as follows. First, molecular dynamics (MD) simulations have shown that glide dislocations can be blocked by the $\sum 3(1\,1\,1)$ coherent twin boundary (CTB) [12,14,20,21] and $\Sigma 3$ $(1\,1\,2)$ incoherent twin boundary (ITB) [22–24]. A very high stress is necessary to transmit dislocations across TBs [25,26]. *In situ* nanoindentation studies of nt Cu confirm significant interactions between dislocations and CTBs/ITBs [27,28]. Second, high density dislocations can accumulate at the CTBs and thus enhance work hardening capability and ductility comparing with nc metals [10,29,30]. Third, TBs are mobile manifested as detwinning in nt Cu as have been observed experimentally by *ex situ* shear deformation [16,31–33] or *in situ* nanoindentation and validated by MD simulations [15,27]. The stress for detwinning of fine nanotwins can be exceptionally low, ~100 MPa, much lower than the yield strength of nt Cu [27].

Prior studies on twins focus primarily on metals with low stacking fault energy (SFE), such as Cu, Ag, and 330 stainless steels [9–11,16,34–43]. Because of the appealing mechanical properties induced by TBs, there are increasing interest in synthesis of twinned light-weight metals, such as Al. Although twins can be introduced into fcc metals by annealing (annealing twins), deformation (deformation twins) and growth (growth twins), the twinnability of fcc metals remains largely controlled by their SFE [34,35,44–48]. Consequently it is much more difficult to introduce twins in high SFE metals than in metals with low SFE. The prediction of deformation twin in nc Al by MD simulations [46,49] leads to a series of successful discovery that shows twins can indeed be introduced via nanoindentation [50], tensile test [10] or cryomilling [51] in nc Al. These evidence give us a hint that although Al has an inherently high SFE barrier to form twins, other factors, such as grain size [52,53], strain rate [54,55] and high stress concentration [56], may trigger the formation of twins.

Recently high-density growth twins and stacking faults (SFs) have been fabricated in Al by introducing nt Ag buffer layers [57–59] by the magnetron sputtering technique. The Al film grown epitaxially on Ag replicates the microstructures including twins from the Ag seed layer because Ag and Al have identical lattice parameter and crystal structure. A systematic study on various

* Corresponding author at: Department of Mechanical Engineering, Texas A&M University, College Station, TX 77843, United States.
E-mail address: zhangx@tamu.edu (X. Zhang).

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metallic multilayers leads to two criteria for the introduction of growth twins into high SFE metals [60]. The first criterion emphasize the need for a low SFE buffer layer that can readily form high-density twinned seeds; and the second criterion highlights the significance of global coherency between coherent similar interface (between constituents with identical planar indices) or local coherency between coherent dissimilar interfaces (between constituents with different planar indices) that allow twins to propagate across layer interfaces. Although high-density growth twins have been introduced into Al with the assistance of the buffer layers, whether growth twins can be synthesized in Al without low SFE metal buffer layers remains an unanswered question.

In this article, we present a simple method to introduce twins into polycrystalline Al films by DC magnetron sputtering without low SFE metallic seed layer. By controlling the film thickness, the fraction of twinned grains in Al can increase to nearly 10%. We also investigated the inverse film thickness effect on the formation of twins in thicker Al films. The twin formation mechanisms, including nucleation and growth of twins, are discussed.

2. Experimental

Polycrystalline Al films were deposited at room temperature by DC magnetron sputtering using 99.99% purity Al target onto Si substrates and sample grids supported with carbon film for transmission electron microscopy (TEM) studies. The aluminum films of different thickness (20–180 nm) were deposited by controlling deposition time under the same deposition rate, 0.7 nm/s. The base pressure of the vacuum system prior to deposition was $\sim 8 \times 10^{-8}$ torr or better and Ar gas pressure was $\sim 2.5 \times 10^{-3}$ torr during magnetron sputtering. TEM studies were performed on an FEI Tecnai F20 ST electron microscope operated at 200 kV and equipped with a Fischione ultra-high resolution high-angle annular dark field (HAADF) detector. For the statistic studies on the distributions of grain size and twin thickness, ~500 grains were measured for each specimen with different film thicknesses, and over 1200 grains were examined for each film to calculate the fraction of twinned grains at different locations in order to establish statistical significance.

3. Results

Plan-view TEM micrographs are shown in Fig. 1 for polycrystalline Al films with different film thickness (h) deposited on TEM grids. In the 20 nm thick Al films in Fig. 1a, growth twins were observed as labeled selectively by arrows. The average grain size (D_{ave}) is 30 nm. The inserted selected area diffraction (SAD) pattern (collected with a large aperture to include numerous grains) shows numerous continuous rings, such as (111), (220) and (200) diffractions, arising from nc grains. When the film thickness (h) increases to 100 nm, the SAD pattern in Fig. 1e shows film texture barely changes. Meanwhile the grain size of the Al film increases to over 100 nm when h = 140 nm. Fig. 2a shows a CTB multi-junction containing two 2-fold twins. The white dash box b in Fig. 2a is magnified in the high resolution TEM (HRTEM) micrograph in Fig. 2b. Each of the 2-fold twins contains 2 CTBs (CTB1 and 2). The two nodes are connected by a highly distorted boundary that is nearly parallel to the {111} plane. The fast Fourier transform (FFT) of Fig. 2b shows the relationship between the two sets of TBs in Fig. 2c. The two CTBs in node 1 are identified to be $(1\bar{1}1)$ and $(\bar{1}\bar{1}1)$ plane, respectively. The HRTEM micrograph of the box d (in Fig. 2a) shows SFs adjacent to the TB (Fig. 2d).

Statistical studies in Fig. 3a show that the average grain size (D_{ave}) increases monotonically from 30 to ~110 nm with film thickness when $h \leqslant 80$ nm; and it then approaches a plateau, ~140 nm, in thicker films. In parallel with the increase of film thickness, the average twin spacing (T_{ave}) also increases, from 12 to 44 nm, when $h \leqslant 80$ nm; and approaches 57 nm when h = 180 nm. The ratio of T_{ave}/D_{ave} is ~40% with little dependence on film thickness. Fig. 4 summarizes the evolution of T_{ave} and D_{ave} with film thickness. Meanwhile the fraction of twinned grains rises monotonically and reaches a maximum of 9.5% when h = 80 nm, which is twice as much as the fraction of twinned grains in the 20 nm-thick films. The fraction of twinned grains then decreases thereafter in the thicker films.

4. Discussion

4.1. The formation mechanisms of growth twins in Al

Our previous studies showed that the epitaxial growth of Al on highly twinned Ag seed layer prompts the extension of nanotwins (nucleated in Ag) into Al films [58]. In this study, however, the twinned polycrystalline Al thin films were synthesized by DC magnetron sputtering without the assistance of any Ag seed layers. From the thermodynamics point of view, it has been shown that an increase in deposition rate will prompt the formation of growth twins in metals [35]. Such a prediction has been validated in Cu experimentally [61]. Bufford et al. [4] used the same twin nucleation model [35] to illustrate that twins are rarely observed in Al films even at a very high deposition rate because of its high SFE. However, the previous thermodynamic model, which was constructed based on the assumption that the TB is parallel to the substrate (referred to as a parallel twin hereafter), cannot predicate the emergence of growth twins (most of which are inclined twins) in the current polycrystalline Al films. Parallel twins have been frequently observed in sputtered nt metals, such as Cu, Ag and 330 stainless steels [1,4,26]. These sputtered twinned films typically have a strong {111} texture. In comparison, when the nt Ag films have [110] texture, inclined twins have been observed [4]. In the current study, nanotwins in polycrystalline Al were observed in plan-view TEM specimens, implying that a majority of these are inclined twins and TBs intersect the film surfaces.

Cross-section TEM (XTEM) micrographs in Fig. 5 (for the Al films grown on Si substrate) show several examples where inclined twins formed. In the first case as shown in Fig. 5a, the inclined twins (manifested by CTB) nucleated from Si-film interface and extended into the film (to a height of ~220 nm) until it is terminated by the columnar grain boundary. Note that this film is thicker than most of the films that were grown on TEM washer. The SAD pattern of the same film in Fig. 5b shows the formation of a {111} CTB. Based on the observation of these inclined twins, we will discuss the revised thermodynamics model to describe the formation of the inclined twins in Al.

4.2. A revised thermodynamics model for the formation of inclined twins in metals with high SFE

We will compare the nucleation mechanisms of a perfect nucleus, a nucleus with a parallel twin and a nucleus with an inclined CTB as shown schematically in Fig. 6. The perfect nucleus in Fig. 6a has the {111} texture. The parallel twin nucleus in Fig. 6b has the same texture and the CTB is normal to the growth direction. For the inclined twin nucleus with {100} texture (the mechanism is similar for {110} texture), the {111} CTB formed a finite angle (≠90°) with respect to the growth direction. As shown in Fig. 6c, the CTB separates the twin nucleus into a right and left section. The right portion of the nucleus has essentially a coherent interface with the matrix, whereas the left portion forms an ITB with the matrix. The angle θ between the CTB and the