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

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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.

Key words: nanotwinned Al; growth twin; defect nucleation kinetics

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 $\Sigma 3$ (111) coherent twin boundary (CTB) [12, 14, 20, 21] and $\Sigma 3$ (112) incoherent twin boundary [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, GaP and 304 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 those with low SFE. The prediction of deformation twin in nc Al by molecular dynamics simulation [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 the 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 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 has identical lattice parameter and crystal structure. A systematic study on various 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.

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 1,200 grains were counted for each film to calculate the fraction of twinned grains at different locations in order to establish statistical significance.

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)

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4 diffractions, arising from nc grains. When the film thickness (h) increases to 100 nm, the SAD
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6 pattern in Fig. 1e shows film texture barely changes. Meanwhile the grain size of the Al film
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8 increases to over 100 nm when $h = 140$ nm. Fig.2a shows a CTB multi-junction containing two
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10 2-fold twins. The white dash box b in Fig. 2a is magnified in the high resolution TEM (HRTEM)
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12 micrograph in Fig. 2b. Each of the 2-fold twins contains 2 CTBs (CTB1 and 2). The two nodes
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14 are connected by a highly distorted boundary that is nearly parallel to the $\{111\}$ plane. The fast
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16 Fourier transform (FFT) of Fig. 2b shows the relationship between the two sets of TBs in Fig. 2c.
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18 The two CTBs in node 1 are identified to be $(1\bar{1}1)$ and $(\bar{1}\bar{1}1)$ plane, respectively. The HRTEM
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20 micrograph of the box d (in Fig. 2a) shows stacking faults (SF) adjacent to the TB (Fig. 2d).
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27 Statistical studies in Fig. 3a show that the average grain size (D_{ave}) increases
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29 monotonically from 30 to ~110 nm with film thickness when $h \leq 80$ nm; and it then approaches a
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31 plateau, ~140 nm, in thicker films. In parallel with the increase of film thickness, the average
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33 twin spacing (T_{ave}) also increases, from 12 to 44 nm, when $h \leq 80$ nm; and approaches 57 nm
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35 when $h = 180$ nm. The ratio of T_{ave}/D_{ave} is ~ 40% with little dependence on film thickness. Fig.
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37 4 summarizes the evolution of T_{ave} and D_{ave} with film thickness. Meanwhile the fraction of
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39 twinned grains rises monotonically and reaches a maximum of 9.5% when $h = 80$ nm, which is
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41 twice as much as the fraction of twinned grains in the 20 nm-thick films. The fraction of twinned
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43 grains then decreases thereafter in the thicker films.
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53 Discussion

54 *The formation mechanisms of growth twins in Al*

Our previous studies show 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.

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 ($\neq 90^\circ$) 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 matrix/nucleus interface plane is the angle between (111) and (100) plane in this scenario. Based on the twin nucleation model developed by Zhang et al [35], during physical vapor deposition, the total Gibbs free energy of a disc-shaped ‘perfect’ nucleus (ΔG_1), the parallel twin nucleus (ΔG_2) and the inclined twin nucleus (ΔG_3) are given as:

$$\Delta G_1 = 2\pi r h \gamma - \pi r^2 h \Delta G_V \quad (1),$$

$$\Delta G_2 = 2\pi r h \gamma - \pi r^2 h \Delta G_V + \pi r^2 \gamma_{CTB} \quad (2), \text{ and}$$

$$\Delta G_3 = 2\pi r h \gamma - \pi r^2 h \Delta G_V + l(h / \sin \theta) \gamma_{CTB} + A_{ITB} \gamma_{ITB} \quad (3);$$

where the r and h are the respective nucleus radius and height. γ is the surface energy and ΔG_V is the bulk free energy difference between solid and vapor per unit volume driving the nucleation. γ_{CTB} and γ_{ITB} are the respective energy of CTB and ITB. A_{ITB} is the area of ITB at the film-substrate interface. The CTB in the inclined twin nucleus has a truncated ellipse shape, which can be approximately to be a rectangle. The area of CTB can thus be calculate as lh , where l , the length of the rectangle, is the intersection between the CTB and matrix as shown in Fig. 6c, and the other edge of the rectangle can be estimated as $h/\sin\theta$. If the area of the ITB $A_{ITB} = f\pi r^2$, where f is the fraction of the ITB area with respect to the area of the nucleus/matrix interface, then $l = 2\lambda r$, where λ is determined by f .

Then following the prior practice [35], the critical nucleation radius for different type of nucleus can be calculated as:

$$r_{perfect}^* = \frac{\gamma}{\Delta G_V} = \frac{\gamma}{\left(\frac{kT}{\Omega} \ln \left[\frac{J\sqrt{2\pi mkT}}{P_s} \right] \right)} \quad (4)$$

$$r_{parallel}^* = \frac{\gamma}{\Delta G_V - \frac{\gamma_t}{h}} = \frac{\gamma}{\left(\frac{kT}{\Omega} \ln \left[\frac{J\sqrt{2\pi mkT}}{P_s} \right] - \frac{\gamma_t}{h} \right)} \quad (5)$$

$$r_{inclined}^* = \frac{\gamma + \frac{\lambda\gamma_t}{\pi \sin \theta}}{\Delta G_V - \frac{f\gamma_{ITB}}{h}} = \frac{\gamma + \frac{\lambda\gamma_t}{\pi \sin \theta}}{\left(\frac{kT}{\Omega} \ln \left[\frac{J\sqrt{2\pi mkT}}{P_s} \right] - \frac{f\gamma_{ITB}}{h} \right)} \quad (6)$$

Where k is the Boltzmann constant, Ω is the atomic volume and T is the substrate temperature, J is the deposition flux, m is the atomic mass of the deposited metal, P_s is the vapor pressure above solid.

The critical radius difference between the twin nucleus and the perfect nucleus can be used to gauge the possibility of forming different types of twins. The critical radius difference between the parallel (or inclined) twin nucleus and the ‘perfect’ nucleus is expressed as $\Delta\gamma_{pp}^* = \gamma_{para}^* - \gamma_{perfect}^*$ or $\Delta\gamma_{ip}^* = \gamma_{inclined}^* - \gamma_{perfect}^*$. The area fraction of the ITB in the inclined twin is an important variable that directly impacts the probability of nucleation of an inclined twin. By using $f = 25\%$, 12% , 6% , we can compare the probability of nucleation of an inclined twin with that of a parallel twin. The following values were used in the calculation: $\gamma = 1.3 \text{ J/m}^2$, $K = 1.38 \times 10^{-23} \text{ J/K}$, $T = 300 \text{ K}$, $\gamma_t = 0.075 \text{ J/m}^2$ and $\gamma_{ITB} = 0.3 \text{ J/m}^2$, $h = 2.33 \times 10^{-10} \text{ m}$, $P_s = 1.07 \times 10^{-5} \text{ Pa}$, and $\Omega = 1.66 \times 10^{-29} \text{ m}^3/\text{atom}$. J is deposition flux in units of monolayers deposited per second. A monolayer of Al has an atomic area density of $1.41 \times 10^{19} \text{ atom/m}^2$ and the deposition rate in our experiments is 0.705 nm/s , thus J is estimated as $\sim 4.2 \times 10^{19} \text{ atom/m}^2\text{s}$.

As shown in Fig 7, for the Al film with parallel twins, the value of $\Delta\gamma_{pp}^*/rp$ is more than 22% even if the deposition rate is 10 nm/s , implying that it is difficult to form parallel twins. When $f = 25\%$, $\Delta\gamma_{ip}^*/rp$ is nearly the same as that of $\Delta\gamma_{pp}^*/rp$. However, a continuous decrease in f value can significantly reduce $\Delta\gamma_{ip}^*/rp$. When $f = 6\%$, $\Delta\gamma_{ip}^*/rp$ drops to below 10% (the horizontal dash line in Fig. 7) at a deposition rate of 0.6 nm/s , implying the possibility to form inclined twins may have increased significantly at the current deposition conditions, 0.7 nm/s . This revised thermodynamic model suggests that the probability for the formation of an inclined growth twin could be much greater than that of a parallel twin, consistent with the observation of predominantly inclined growth twins in polycrystalline Al with high SFE.

Promoting the formation of growth twins during the growth of films

Beside the twin embryo formed during the nucleation stage, the twin formation process in Al may also be closely related to the continuous growth of the films. Fig. 5b shows that the formation of an inclined CTB originated from the columnar grain boundary in Al film. The SAD pattern in Fig.5d indicates the formation of the inclined growth twin.

As shown in Fig. 8a, nuclei with different orientations formed randomly on substrate. Some of the nuclei may contain twins (Fig. 8a) and occasionally certain adjacent nuclei may happen to have the twinned orientation as evidenced in XTEM studies in Fig. 5e. During island coalescence, larger islands with low surface and interface energy may consume smaller or unfavorably orientated islands driven by the decrease in interface and surface energy (Fig. 8b). Meanwhile as shown in Fig. 8c, the high angle grain boundaries between the grain 1 and 2, and 1 and 4, could be replaced by a CTB between the grain 1 and 4 as the CTB has much lower energy than a high angle grain boundary, leading to the formation of CTBs during the subsequent growth of the film.

The increase of the fraction of the twinned grains with film thickness (when $h \leq 80$ nm)

As shown in Fig. 4, the fraction of the twinned grains rises monotonically to ~ 9.5% when the film thickness increases to 80 nm and decreases afterwards. The fraction of the twinned grains is defined as the number of twinned grains per unit area (P_{twin}) divided by the total number of grains per unit area (P_{grain}). So mathematically it appears that either an increase in P_{twin} or a decrease in P_{grain} will result in a greater fraction of the twinned grains.

However our studies in Fig. 9 shows that both P_{twin} and P_{grain} decrease continuously with increasing film thickness. For instance, the P_{twin} decreases from 46 to 18, and then to 7 twinned grains/ μm^2 when $h = 20, 40$ and 80 nm, respectively. In parallel, the value of P_{grain} decreases at a greater rate, from 861 to 261 and then to 84 grains/ μm^2 when $h = 20, 40$ and 80 nm, respectively.

The reduction in the number of grains in thicker films arises from the island coalescence process [62]. When the island coalesced, the twins were terminated at the same time. However, as CTBs typically have lower energy than high angle grain boundaries, they are more stable and may have greater probability to survive during the island coalescence process. Furthermore new TBs may form during impingement of islands as evidenced in Fig. 5c and illustrated in Fig. 8c. Consequently the fraction of twinned grains increases with increasing film thickness when h is 80 nm or less.

The decrease of twinned grain fraction during continuous growth of the films (when $h > 80$ nm)

When $h > 80$ nm, the fraction of the twinned grains decreases as shown in Fig. 4. A notable phenomenon in this regime is that the average grain size approaches a plateau ~ 140 nm (or P_{grain} becomes a constant), because the films have entered the regime of a continuous growth of the columnar grains instead of islands coalescence. Correspondingly, the decrease of the fraction of the twinned grains results from the reduction of P_{twin} .

To explain the termination of twins during continuous growth of films when grain size saturates, we need to consider the energetics for the growth of CTB and ITB for the inclined twins. As shown schematically in Fig.10, the length of CTBs for inclined twins is proportional to the film thickness and hence the continuous growth of the CTBs with increasing film thickness will increase the total energy of the system. As the inclined CTBs extends to the film surface, there is no ITBs (or there is no extra energy necessary to create ITBs). It is important to realize that the termination of CTBs inside the films imply the necessity to form ITBs within the films. In Fig. 10a, there are two inclined twins (thin vs. thick) inside the film ($h > 80$ nm) during the early growth stage. As shown in Fig. 10b, if the total energy of the CTBs (W_{CTB}) of the thin twin

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4 exceeds the formation energy of the ITB (W_{ITB}), the possibility to form an ITB will increase and
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6 the formation of the ITB terminates the continuous growth of the CTBs. Consequently the
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8 number of twinned grains (examined from plan-view TEM) will decrease. But the thick twin
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10 may continue its growth with the increase of film thickness because the energy $W_{ITB} \geq W_{CTB}$, or
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12 it is much more energetically costly to terminate a thick twin inside the film. The thick twin is
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14 eventually terminated when $W_{CTB} \geq W_{ITB}$ (Fig. 10c).
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20 Foregoing discussions suggest that the growth of twins in thicker films could be affected by
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22 the energy minimization considering the energy difference between W_{ITB} and W_{CTB} . W_{ITB} can be
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24 approximated as $\alpha T_{\text{twin}} E_{ITB}$, where α is a constant related to the inclined twin angle and E_{ITB} is
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26 the formation energy of an ITB per unit area. Similarly, $W_{CTB} = b h E_{CTB}$, where b is a constant
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28 which related to the inclined twin angle and E_{CTB} is the energy of the CTB per unit area. α , b ,
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30 E_{ITB} and E_{CTB} are constants independent of film thickness. When $W_{CTB} = W_{ITB}$, an inclined dash-
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32 dot line is obtained as shown in Fig. 11. As discussed previously, the energy discrepancy
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34 between the ITB and CTB suggests the possibility to form an ITB. When $W_{ITB} > W_{CTB}$ (to the
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36 left side of the straight line), it is energetically feasible to extend the inclined growth twins
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38 during the growth of films. Whereas when $W_{ITB} < W_{CTB}$ (to the right side of the straight line), the
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40 continuous growth of the CTBs becomes energetically unfavorable. Consequently, the growth of
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42 a CTB will be terminated, leading to a reduction of the fraction of the twinned grains.
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50 The reduction of the fraction of the twinned grains may also be related to the lack of
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52 continuous grain growth (or a lack of island coalescence). As mentioned earlier twins may
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54 nucleate during island coalescence, and thus the lack of island coalescence may reduce the
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56 probability to form growth twins. The current study on the formation of growth twins in
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1 polycrystalline Al is an important forward step towards the eventual goal - the synthesis of high
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6 density nanotwins in Al without buffer layers. There are numerous questions that remain to be
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8 examined in future studies, including the peculiar constant ratio between the average twin
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10 thickness and average grain size observed in this study.
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18 **Conclusions**

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21 Sputter-deposited polycrystalline Al films contain growth twins and the fraction of
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23 twinned grain increased with increasing film thickness to a maximum value, $\sim 9.5\%$, when $h =$
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25 80 nm, and decreased thereafter in the thicker films. An inclined twin nucleation model has been
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27 provided from the thermodynamics perspective to explain the formation mechanism of the
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29 growth twins in Al films. The termination mechanism for the growth of the inclined twins in
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31 thicker films may be related to the competition between the energy of CTB and ITB, the
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33 elimination of fine nanotwins, and the lack of grain growth via island coalescence. Although the
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35 fraction of twinned grains remains low, the twin formation mechanisms presented in this study
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37 suggest new perspectives to tailor the formation of grow twins in high SFE metals, such as Al.
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Figure captions

Figure 1. Transmission electron microscopy (TEM) micrographs of Al films with different film thickness showing the formation of growth twins. (a) $h = 20$ nm, (b) $h = 40$ nm, (c) $h = 60$ nm, (d) $h = 80$ nm, (e) $h = 100$ nm, (f) $h = 140$ nm. Twins were frequently observed as labeled selectively by arrows in the micrographs. The inserted selected area diffraction (SAD) patterns indicate the formation of polycrystalline Al films.

Figure 2. Microstructure of long coherent twin boundaries (CTBs) in twinned Al films. (a) Low magnification TEM micrograph of the CTB multi-junction in Al film, $h = 40$ nm. (b) HRTEM micrograph showing the intersection of CTBs at the junctions. (c) The fast Fourier transform

(FFT) of the CTB junctions confirmation the formation of two sets of twins. (d) HRTEM micrograph showing the CTB1 decorated with stacking faults.

Figure 3. Statistic distributions of grain size and twin thickness for Al films with various film thickness. (a) Statistical variation of grain size with different film thickness. The average grain size increases monotonically up to ~138 nm with increasing film thickness to 100 nm, and then reaches a plateau. (b) In parallel the average twin thickness increases to ~ 52-56 nm when $h = 100$ nm, and reaches a saturation.

Figure 4. The evolution of fraction of twinned grains, average twin thickness and average grain size with Al film thickness. The fraction of twinned grains increases continuously with increasing film thickness, reaches a maximum of ~ 9% when $h = 80$ nm, and decreases thereafter in thicker films. The average twin thickness and grain sizes increase monotonically with film thickness and approach plateaus when $h = 100$ nm.

Figure 5. Microstructure of inclined twins in Al films on Si substrate. (a) Low magnification cross-section TEM micrograph of the twinned Al films, the inclined twin nucleated at the Si surface. (b) The SAD pattern of R1 in Fig. 5a. (c) Low magnification cross-section TEM

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4 micrograph of the twinned Al films, the inclined twin nucleated inside the Al films. (d) The SAD
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7 pattern of R2 in Fig. 5c. (f) the SAD pattern of R3 in Fig. 5e.
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13 Figure 6. Schematic diagrams comparing the nucleation of (a) a perfect nucleus, (b) a parallel
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15 twinned nucleus and (c) a nucleus with inclined growth twin on Al matrix. (a) The perfect
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17 nucleus and the matrix has the same $\langle 111 \rangle$ crystal orientation. (b) The parallel twin nucleus
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19 forms (111) CTB on the (111) matrix. (c) A nucleus contains both inclined CTB and incoherent
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21 twin boundary (ITB) with the matrix. Note an example of (100) matrix is used to illustrate the
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23 concept without losing the generality of the model. A fraction of the matrix-nucleation interface
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25 contains ITB with an area of A_{ITB} , whereas the remaining of the nucleus has the same crystal
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27 orientation as that of the matrix.
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40 Figure 7. Plots of the percentage of critical radius difference between perfect nuclei and parallel
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42 twin nuclei $\Delta r_{para}/r_p$ (and between perfect and inclined twin nuclei $\Delta r_{incl}/r_p$, where $\Delta r_{para} = r_{para} - r_p$,
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44 $\Delta r_{incl} = r_{incl} - r_p$, and r_p , r_{para} , r_{incl} represent the radius of perfect, parallel and inclined twin nuclei
45
46 respectively. For parallel twins, r_{para} is at least 25% greater than that of r_p even at a very high
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48 deposition rate, implying the nucleation of parallel growth twins in Al is very difficult in
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50 comparison for inclined twins, by reducing the area fraction of A_{ITB} to 12% (or less) of the
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52 contact area (between matrix and twin nuclei), r_{incl} is merely $\sim 10\%$ greater than that of r_p ,
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54 implying the nucleation of inclined twins in Al is easier than the nucleation of parallel twins.
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Figure 8. Schematics illustrating different twin formation mechanisms. In the first case, inclined twins may nucleate directly on the matrix as shown in (b). In the second case, two grains, 1 and 4, with twin orientation may impinge upon each other as shown in (c) to form a twin boundary during the growth of films.

Figure 9. Plots of twinned grain density and grain density versus film thickness. Twinned grain density follows the tendency of grain density with the increase of film thickness. The density drops quickly before film thickness reach 60 nm and goes to plateaus gradually.

Figure 10. Schematics illustrating the termination of thin inclined twins and continuous growth of thick inclined twins in Al by using the energy balance concept. When the energy of CTBs is less than or equal to the formation energy of ITB, the inclined twins may grow continuously (a). In contrast, the growth of inclined twins become difficult when the opposite holds true. Thus, the growth of thinner twins terminates earlier than thicker twins (b-c).

Figure one

Figure two

Figure three

Figure four

Figure five

Figure six

Figure seven

Figure eight

Nucleation

Growth + Nucleation

Impingement of twin

Figure nine

Figure ten
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Thicker twin $W_{CTB} > W_{ITB}$
Stable

$W_{CTB} > W_{ITB}$
Stable

$W_{CTB} < W_{ITB}$
Unstable

Thinner twin $W_{CTB} > W_{ITB}$
stable

$W_{CTB} < W_{ITB}$
unstable

Figure eleven