Formation Mechanisms of High-density Growth Twins in Aluminum with High Stacking-Fault Energy

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Nanotwins readily form in numerous face-centered-cubic (fcc) metals with low stacking-fault energy (SFE). However, growth twins rarely form in Al due to its high SFE, \( \sim 120–165 \, \text{mJ/m}^2 \). Here, using thin inter-layers or buffer layers of a low-SFE fcc metal (Ag), we overcome the SFE barrier and successfully grow high-density coherent and incoherent twin boundaries into Al. We identify three mechanisms that induce growth twins in Al and demonstrate enhanced mechanical strength in twinned Al. This study reveals an effective means that may be generalized to control growth twin formation in fcc metals with high SFE.

Keywords: Aluminum, Interfaces, Mechanical Properties, Silver, Twin Boundaries

Novelty Statement: High-density growth twins are introduced in aluminum despite its high stacking-fault energy. Twin formation mechanisms are discussed. Nanotwinned Al shows high mechanical strength.

Introduction Since the initial synthesis of high-density growth twins in Cu [1,2] and 330 stainless steel films [3,4], extensive studies have investigated the mechanical and physical properties of nanotwinned (nt) face-centered-cubic (fcc) metals. The \( \Sigma 3(111) \) coherent twin boundary (CTB) in nt metals provides resistance to dislocation transmission similar to high-angle grain boundaries [5–7], and dense twins lead to high strength, higher strain rate sensitivity [6,8], and enhanced work hardening rate [2,9–11] in comparison with their bulk non-twinned counterparts. CTBs have a symmetrical structure and low-energy configuration [12], so nt metals have better fatigue resistance [13,14], thermal stability, electrical conductivity, and ductility than nanocrystalline (nc) metals with similar strength [8,15–18].

Most previously studied nt metals have low stacking-fault energy (SFE) and hence readily form growth twins during synthesis [1,4,17,19,20]. We recently characterized magnetron-sputtered Ag films on Si (111) substrates via cross-sectional and plan-view transmission electron microscopy (TEM) [19]. Ag has the lowest SFE among monolithic fcc metals, so during deposition, the films form epitaxial nt microstructures with high-density CTBs oriented perpendicular to the growth direction. However, the probability of twin formation during deposition rapidly diminishes with increasing SFE, as twin boundaries become more and more unstable energetically [4]. High SFE thus becomes a fundamental barrier to the formation of high-density growth twins in numerous fcc metals.

Al is one such metal in which twins rarely form due to its high SFE, hence twins in Al are associated only with specific conditions. Molecular dynamic simulations predicted deformation twinning and twin boundary migration in nc Al [21–23]. Deformation twins appeared in highly strained regions at the tip of an indented area and in nc Al powders subjected to high strain rate cryomilling.

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Additionally, the stress concentration near a crack tip can lead to deformation twinning in Al, and these twins reverse as the stress relaxes [26]. Under these extreme conditions (large stress concentration, high strain rate, and low temperature), deformation twins occasionally form in Al to assist dislocation slip and accommodate plastic deformation. Various models have been developed to explain the generation of partial dislocations and twins in nc Al [21–23,27–29], and recent reviews provide detailed information about deformation twinning processes in various metals [30,31]. In spite of the generation of deformation twins in Al, the introduction of high-density growth twins into Al remains a major scientific challenge.

In this article, we demonstrate the growth of high-density twins and stacking faults in Al films for the first time by using a template method. Methods employing template or buffer layers have been known for some time in the study of epitaxial semiconductor thin films [32,33], but have not been used for fabricating nt metals. Here, we incorporate thin inter-layers or a buffer layer of a low-SFE fcc metal (Ag), so that coherent Ag/Al interfaces allow the replication of twins and stacking faults from Ag into Al. We present three mechanisms through which twins may form in Al and show enhanced mechanical strength in these nt Al films. This template approach may be generalized to form high-density twins in numerous other fcc metals with high SFE.

**Experimental** All films were deposited at room temperature using DC magnetron sputtering. Single-crystal silicon substrates with (111) orientation were etched with HF prior to deposition to remove the native oxide layer. The base pressure prior to deposition was typically $5 - 9 \times 10^{-8}$ torr, and Ar gas (99.99%) pressure during sputtering was $\sim 3.6 \times 10^{-3}$ torr. Pure Al (99.99% or better) and Ag (99.99%) targets were used. Single-layer Al films were deposited directly onto the etched Si substrates to a thickness of $2 \mu m$. For all Ag/Al multilayer films, a 100-nm Ag seed layer was deposited first to ensure epitaxial growth. For comparison, single-layer Al films with $1 \mu m$ film thickness were grown on 100 nm thick Ag seed layer. Next, alternating Ag and Al layers of different individual layer thicknesses, $h$, were deposited. Ag 100-nm/Al 100-nm, Ag 5-nm/Al 5-nm, and Ag 5-nm/Al 100-nm multilayer films, and pure 1-µm Al were grown on seed layers to a total film thickness of $1.5 \mu m$. X-ray diffraction (XRD, PANalytical X’Pert PRO Materials Research Diffractometer, Cu Kα radiation) was used to examine film quality. Both out-of-plane θ–2θ and in-plane phi scans were conducted. XRD and TEM experiments were performed immediately after deposition. Cross-sectional TEM specimens were prepared by mechanical grinding and polishing, followed by low-energy ion milling, exercising caution to avoid heating during preparation. The specimens were examined along the Al [011] zone axes with an FEI Tecnai F20 ST electron microscope operated at 200 kV. High resolution high-angle annular dark-field scanning TEM (HAADF-STEM) was performed at the National Center for Electron Microscopy (Lawrence Berkeley National Laboratory) using the aberration-corrected TEAM 0.5 microscope, an FEI Titan electron microscope enhanced by an advanced STEM corrector. Operating at 300 kV, the microscope can form electron probes as small as 0.5 Å, yielding directly interpretable information about the local chemistry of columns of atoms in the sample [34]. Hardness was measured via an instrumented nanoindentation method performed using a Fischerscope HM2000XYp micro-/nano-indenter with a Vickers indenter tip. Hardness vs. depth analysis was performed, and a hardness plateau (i.e. proper depth to avoid erroneous hardening influences of both small indentation size and substrate effects) was typically observed. A minimum of 9–12 good indentations were performed at each indentation depth.

**Results** The single-layer Al films grown directly on Si (111) substrates at a relatively high deposition rate were epitaxial with {111} orientation and developed only a miniscule amount of twinned variant, as evidenced by the XRD phi scan from the film {111} planes (Figure 1(a)). Multilayer Ag/Al films on Si (111) substrates, however, behaved much differently. The phi scan from the Ag 5-nm/Al 100-nm epitaxial multilayer film clearly reveals two sets of peaks, each with three-fold symmetry, corresponding to respective matrix and twin variants. The phi scan from the 1-µm-thick epitaxial Al film on a 100-nm Ag seed layer (referred to as Al 1-µm/Ag seed layer hereafter) also shows two sets of peaks, arising from the matrix and twin variants. As can be seen in the XRD θ–2θ profiles (Figure 1(b)), the single-layer and multilayer samples were all epitaxial {111} textured films on Si (111), hence matrix and twin variants comprise the entirety of the films. There is no evidence of AgAl intermetallic compounds in the XRD profiles.

The HAADF-STEM micrographs collected from the Ag 10-nm/Al 10-nm multilayer film specimens (Supplementary Figure 1(a)) revealed minimal intermixing even at such small $h$. (Note that the Al layer is dark, as the contrast is proportional to $Z^{1/7}$ (where $Z$ is the atomic number)) [35]. The remainder of this article focuses on Al films with much larger $h$, where any contribution of intermixing to twin formation diminishes even further.

TEM examinations of single-layer epitaxial Al films on Si (without Ag seed layer, Supplementary Figure 1(b)) revealed an epitaxial structure with dense dislocation tangles. As suggested by the XRD profiles, essentially all of the Al films consisted of a single-crystal-like epitaxial variant, that is, nearly no twins formed. We observed a few small stacking faults on various {111} planes,
Figure 1. (a) XRD $\{111\}$ phi-scan profiles of epitaxial films. In the pure epitaxial Al film (no Ag seed layer), a single matrix variant (denoted M) with three-fold symmetry dominates. In the Ag 5-nm/Al 100-nm epitaxial multilayer specimen, three twin peaks (denoted T) arise from the greatly increased volume fraction of twin variant and coexist with the three matrix peaks. In the epitaxial Al 1-μm/Ag seed film, the twin variant peaks are smaller than in the multilayer sample, but still indicate a significant twinned fraction. (b) Conventional 2θ XRD profiles plotted with log intensity to enhance any small peaks. The only peaks present correspond to the Si substrate or Ag and Al (111) planes, indicating an epitaxial film structure.

and a scarcity of small variants appeared at the film/substrate interface. These variants were exceedingly rare and extended just a few nanometers into the films.

TEM examination of epitaxial Ag/Al multilayer films revealed high-density twins and stacking faults within Ag layers, as expected. In the epitaxial Ag 100-nm/Al 100-nm films (Figure 2(a)), layers appeared wavy due to island growth mechanisms and residual growth stresses. Strikingly, many ITBs stretched nearly vertically across several Ag and Al layers. The inset selected area diffraction (SAD) pattern taken from a large area confirms an epitaxial multilayer film structure with high-density CTBs and abundant ITBs.

Figure 2(b) shows a typical example wherein groups of ITBs in Al connect CTBs with lengths of over tens of nanometers. Some ITBs also have widths exceeding 100 nm (Figure 2(b)). The high-resolution TEM (HRTEM) micrograph in Figure 2(c) shows a wide ITB (confirmed by the inset fast Fourier transform (FFT)), the structure of which can also be considered as a 9R phase, similar to those observed previously in low-SFE metals, such as Cu, Ag, and Au [36–38]. ITBs in epitaxial nt Cu and Ag typically align vertically (parallel to the growth direction) [15,19]. However, many ITBs in Al form a TB structure in which CTB and ITB units combine to form meandering boundaries, a structure previously observed in Au and termed the chain unit model [39]. Meanwhile, many longer CTB segments were not confined to a single \{111\} plane. A closer examination (Figure 2(d)) revealed that CTBs comprised atomic steps due to numerous adjacent stacking faults.

These observations show that 100-nm-thick Ag layers sufficed to induce abundant twins in Al. To investigate whether thinner Ag layers might produce a similar microstructure, we examined epitaxial Ag 5-nm/Al 100-nm multilayer films. As shown in Figure 3(a), indeed, thinner Ag layers still induced twins in Al, with structures similar to those observed in Ag 100-nm/Al 100-nm films. Again, many ITBs extended through multiple Ag and Al layers. The inset SAD pattern confirms the formation of high-density twins and shows no components other than the matrix and twins. Figure 3(b) shows a typical ITB nucleating at the Ag/Al interface and propagating into the Al layer. The ITB extended through the Al layer by intermingled CTB and ITB segments and eventually terminated at the upper Ag/Al interface. The HRTEM micrograph in Figure 3(c) presents an atomic-resolution view of a CTB decorated by an adjacent SF and a mixed TB consisting of zig-zag steps of CTBs and ITBs. In both multilayer films, most twins extended all the way through the Al layers and propagated horizontally by ~50 nm. Since only one TB usually extended through one layer in a given area, the average twin thickness in Al layers was approximately $h/2$. Twins that did not extend through the entire Al layer typically terminated within 10–20 nm of the interface.

The epitaxial Al 1-μm/Ag seed layer film revealed how far TBs might grow into Al. While high-density
Figure 2. Bright-field cross-sectional TEM micrographs (Al\textlangle011\rangle zone axis) from the Ag 100-nm/Al 100-nm epitaxial multilayer film. (a) At low magnification, ITBs can be seen passing through several layers, and the SAD pattern from the film alone reveals evidence of both ITBs and CTBs. (b) A higher magnification view shows a twin boundary with mixed ITB and CTB components in Al. (c) The HRTEM micrograph shows a magnified view of box (c) in Figure 2b. Here, the ITB has wide stacking faults inside, and the inset FFT confirms the ITB structure. (d) A magnified view of box (d) shows stacking faults leading to a number of atomic steps along a long CTB. The inset FFT confirms the formation of a CTB.

twins still formed, the microstructure differed substantially from the aforementioned multilayer films. Adjacent twin variants extended hundreds of nanometers into Al along the growth direction, resulting in a columnar structure with long ITBs separating adjacent domains (Figure 3(d)). The average columnar width was \(\sim200\) nm. ITBs of various widths were predominant in these films (Figure 3(e)), although some mixed ITB/CTB segments were still present, as shown by the HRTEM micrograph in Figure 3(f).

Discussion

**TB Formation Mechanisms in Al.** Controlled formation of stable growth twins and stacking faults has not been accomplished in Al previously. A thermodynamic model predicts that twin formation probability during vapor deposition depends on the difference in critical radius of formation between a defect-free nucleus and a twinned nucleus [4], which in turn depends on numerous parameters, most notably deposition flux and SFE. Increasing deposition rate effectively ‘freezes’ defects in place by piling up more material before atoms can diffuse to their ideal crystallographic sites and hence introduces more growth defects, such as twins. However, for metals such as Al, SFE is so high that increasing deposition rate alone is insufficient to nucleate twins during growth. The scarce few twins observed at the Al/Si interface in single-layer epitaxial Al films on Si (without the Ag seed layer) suggested that growth twins may form in Al near interfaces. Ag was chosen for inter-layers and buffer layers because it grows coherently with Al due to a small lattice mismatch (\(<1\)%). Furthermore, our previous studies, including plan-view and cross-sectional TEM, show that nt Ag can grow epitaxially on Si while readily forming high-density growth twins [19]. Hence, Ag layers act as templates, whereby twins nucleate in Ag or at the interface and then extend into Al.

We identified three mechanisms by which TBs form in Al. Schematics in Figure 4(a–c) illustrate
these mechanisms, and the corresponding TEM micrographs, Figure 4(a′–c′), provide experimentally observed evidence for each mechanism. In mechanism (1), shown schematically in Figure 4(a), domains in epitaxial Ag layers assume only two possible orientations, twin or matrix, between which ITBs form. Al grows epitaxially on Ag, and thus the orientations of Ag domains and ITBs are replicated by the Al layer, as observed in the HRTEM micrograph in Figure 4(a′). Therefore, mechanism (1) enables the formation of abundant vertical ITBs in Al. The waviness seen in the layers may be related to the formation of adjacent domains. Waviness was most notable in the Ag 100-nm/Al 100-nm films (Figure 2(a)), and a significant number of vertical ITBs emanated from the valley of such waves. In some areas, nearly 50% of the valleys had corresponding ITBs, while other areas had only ∼15%. Additionally, once a vertically propagating ITB formed, it often extended through many Al and Ag layers (Figure 2(a)). However, vertically propagating ITBs appeared elsewhere away from the valleys, and the same propagation mechanism appeared to be active in the Ag 5-nm/Al 100-nm films, where little waviness was observed (Figure 3(a)).

In mechanism (2), CTBs are replicated laterally from Ag into Al, as illustrated in Figure 4(b). Should CTBs terminate at the layer interface, ITBs would have to nucleate inside Al to join the variants, consequently increasing the total system energy. Hence, the propagation of CTBs across the layer interface into Al is more energetically favorable than termination. The HAADF-STEM micrograph in Figure 4(b′) provides an example of this mechanism.

Mechanism (3) shows that twin variants may form in Al even if the underlying Ag consists of a single variant, as shown in Figure 4(c). The HRTEM micrograph in Figure 4(c′) reveals a narrow ITB nucleated from dense stacking faults at the Ag/Al interface. In this mechanism, jumbles of stacking faults in Ag result in similar jumbles in Al near the interface and ITBs may nucleate from these high-density stacking faults. This mechanism requires only a few atomic layers of stacking faults to initiate ITB nucleation in Al. Coherency is probably necessary here as it allows Al to replicate the microstructure of stacking faults in Ag and eventually triggers the nucleation of ITBs that propagate vertically into Al. Each of these mechanisms was observed experimentally; however, there still may be other as-yet unidentified mechanisms through
Figure 4. Illustrations of the observed twin formation mechanisms at the Ag/Al interface. (a) Epitaxial growth forces Al to replicate the microstructure of twinned Ag, causing an ITB to propagate vertically through Al. The corresponding HRTEM micrograph (a’) confirms the formation of twins in Al via this mechanism, with the dotted line delineating the coherent interface. (b) Coherent interfaces allow a CTB in Ag to extend laterally into Al. (b’) The HAADF-STEM micrograph (from the Ag 10-nm/Al 10-nm multilayer film) shows lateral propagation of a CTB from Ag into Al. In this micrograph, Al appears darker as contrast is proportional to atomic number. The same micrograph also shows that Ag contamination is essentially non-existent around the CTB, hence intermixing does not play a role in the twin formation and stabilization processes in Al. The schematic (c) and corresponding HRTEM micrograph (c’) illustrate ITBs nucleating from high-density SFs at the Ag/Al interface. Note that the ITB deviates leftward after propagating ~20 nm, forming a CTB.

which a template (buffer) layer enables twin formation in Al. The preferred propagation mechanism appears to be influenced by the local stress state. For instance, in some areas, the three mechanisms appeared with essentially equal frequency, while in others a single mechanism, typically (1), accounted for nearly 100% of twins in Al. XRD phi scans (Figure 1(a)) indicate a greater prevalence of the matrix variant than the twin variant; approximately 2/3 of the film is matrix in the multilayer films, while 3/4 is matrix in the Al 1-μm/Ag seed films.

We note that the Ag/Al system is ideal for defect propagation from layer to layer. The lattice mismatch of <1% ensures that each Ag/Al interface is a nearly perfect Σ1 boundary; identical crystallographic indices are parallel in both underlayers and overlayers, and there is no need for misfit dislocations, domain matching, or a change in crystallographic orientation to allow epitaxial growth. Hence, Al overlayers nearly exactly replicate Ag underlayers including both twin variant orientation and TBs. This relationship is clearly highlighted in the HRTEM micrographs in Figure 4(a’) and (b’), where atomic planes are continuous across layer interfaces, and the inset SAD patterns indicate bicrystal structures (due to twins) without any tilt, rotation, or misorientation between the layers. Given a [111] growth direction, the matrix and twin variants are related by a 180° rotation about the growth axis, so that the (111) plane is the twinning plane. While theoretically twins may form on other {111} planes, we did not observe such twins, which is similar to our previous observations of twins in pure Ag films [19]. There is no evidence of variants other than the single matrix and twin variants described here.

While mechanisms (1) and (2) are conceptually straightforward, the formation of stacking faults and twins in case (3) requires further explanation. During island growth, an interface traction arises between the island and the substrate. This traction is a shear stress whose magnitude depends on the distance from the island edge. We consider the growth of an island with z parallel to the growth direction and x perpendicular (Supplementary Figure 2), so the shear stress, \( \tau(x) \), can be expressed as [40]

\[
\tau(x) \sim \frac{\sigma_m}{\sqrt{2\pi x}} \quad (1)
\]
asymptotically as \( x/h_f \to 0^+ \). Here, \( \sigma_m \) is the biaxial mismatch stress (defined as \( \sigma_m = \varepsilon_{\text{max}} E_f / (1 - \nu_f) \)) between the film and the substrate, \( \varepsilon_{\text{max}} \) is the mismatch strain between Ag and Al, \( h_f \) is the film thickness, and \( x = 0 \) at the free edge of the island. Also, \( k \) is the plane strain elastic modulus ratio between the substrate and film, computed by

\[
    k = \frac{E_s}{1 - \nu_s^2} \frac{E_f}{1 - \nu_f^2},
\]

where \( E_s, \nu_s, E_f, \) and \( \nu_f \) are the elastic moduli and Poisson’s ratios of the substrate and the film, respectively. The magnitude of \( \tau(x) \) increases with proximity to the free edge or with increasing film thickness. Considering an Al film and an Ag substrate, \( k \approx 1.2 \). Mismatch stress is approximately 1.1 GPa, as mismatch strain, \( \varepsilon_{\text{max}} \), between Al and Ag is \( \sim 1\% \). Therefore, we estimate \( \tau(x) \) of 470–1,140 MPa when \( h_f/x \) is 1–6, that is, near the island edge.

The growing Al layer must somehow accommodate these significant stresses. The shear stresses needed to nucleate full and partial dislocations, as well as those needed to either erase or extend the leading partial dislocations, depend on grain size. By comparing these critical stresses, it has been determined that for Al twinning is favored when grain size, \( d \), is \( \sim 6 \) nm or smaller [41,42], which is consistent with the size of islands nucleated during growth. This analysis gives a critical shear stress, \( \tau_c \), of \( \sim 0.9 \) GPa required for partial nucleation, while \( \leq 0.5 \) GPa is sufficient to enlarge an existing twin [41]. Other models and simulations [43,44] have yielded comparable values for \( \tau_c \). Hence, the traction near the Al island edge provides sufficient stress to nucleate Shockley partial dislocations at the interface during deposition. We also note that mechanisms (1) and (3) only result in the formation of adjacent twin variants separated by ITBs. CTBs may form directly by mechanism (2) or serve as horizontal junctions between ITBs, as seen in Figure 4(c’). Usually, mixed ITB and CTB segments comprised most boundaries, resulting in meandering TB paths throughout Al, as illustrated in Figures 2 and 3.

**Stabilization of TBs.** TBs in high-SFE metals can be stabilized in two ways: by reducing TB energy or by constraining the system such that the twinned state has the lowest possible energy. Numerous previous studies have demonstrated that alloying can decrease SFE, thereby enhancing the energetic stability of TBs and stacking faults [45]. However, the mechanisms at work in our study differ from this well-established method.

An alternative way to stabilize TBs is to use the epitaxial structure to ensure that only twin and matrix variants exist. Epitaxial growth of Ag on Si results in the formation of one of only two possible variants, and epitaxial growth of Al on Ag causes the Al layer to take the same structure. Hence, only ITBs and CTBs separate domains in Al instead of more conventional high-angle or low-angle grain boundaries. Our study of the Cu/Ni system examined both incoherent and coherent multilayer films and revealed that twins penetrated into Ni layers only when layer interfaces were coherent [46].

The relative abundance of ITBs in comparison with CTBs in Al may be related to the total grain boundary energy within the system. ITB energy in Al varies from 223 to 357 mJ/m² [38,47], while the energy of its CTBs is \( \sim 75 \) mJ/m² [48]. In comparison, Cu has an ITB energy of \( \sim 550–714 \) mJ/m² and a CTB energy of \( \sim 24–39 \) mJ/m², while Ag has corresponding energies of \( \sim 126 \) mJ/m² and \( \sim 8 \) mJ/m² [12,48–50]. The ITB/CTB energy ratios in Al, Ag, and Cu are \( \sim 3.5, \sim 16, \) and \( \sim 22 \), respectively. The smaller ratio in Al helps explaining the relative abundance of ITBs to CTBs in Al, whereas in Ag and Cu, CTBs are predominant. ITBs in Al can be described equivalently by dislocation theory [51] or as a volume of 9R phase. Although the presence of ITBs (and hence at least some volume of 9R phase) is certain, as can be seen in Figures 2(b and c) and 3(d and e), the actual volume of 9R phase is unclear. While some ITBs appear to have a very large volume (Figure 2(b)), there is a possibility that this is an artifact caused by a meandering boundary with a component running parallel to the foil plane. When projected onto a two-dimensional surface, such a boundary may appear much wider than it actually is. One can postulate that some distortion of the projected image might occur under such conditions, which may explain the distorted appearance of the ITB in Figure 3(e). However, some sizable boundaries (Figure 2(c)) do not appear to have such distortion and may indeed be thicker slabs of 9R Al. Local stresses may cause an ITB to dissociate into a greater width [52–54]; however, the typical width of a dissociated ITB is much smaller than that of the seemingly huge ITBs observed in this study. Future HRTEM analysis will reveal whether the ITBs observed in this study are the result of projecting typical ITBs onto a flat surface or if the samples actually do contain large volumes of 9R phase.

**Limits and Applicability of the Methodology for the Fabrication of Bulk nt Metals.** In the Al 1-μm/Ag seed layer film, meandering ITB/CTB segments propagated through the entire film thickness, as can be seen in Figure 3(d). We have synthesized other nt Al films, a few micrometers in thickness, where TBs formed via similar mechanisms. It is likely that the ITB/CTB segments could propagate through a film that is tens of micrometers in thickness. For multilayer films, new twins nucleate in each Ag layer, so the structure might be repeated essentially indefinitely. The mechanisms discussed here should
be immediately applicable for the fabrication of a
variety of nt metals with high SFE in the form of coatings.
However, sputtering techniques are limited to relatively
thin (<1 mm) coatings. Adaptation of the mechanisms
discussed in this article to other techniques may help to
fabricate bulk nt metals for structural applications.

High-Strength Al Enabled by TBs. Nanoscale twins
often contribute to improved mechanical strength, and
strengthening indeed occurred in these nt Al films.
Figure 5 shows indentation hardness data compiled
from the literature [55–61] and includes only monolithic
Al in the form of films and bulk specimens. Hardness of
the as-deposited and annealed untwinned single-layer Al
films falls in the expected range given by other studies
(∼0.6–0.8 GPa), whereas hardness of twinned epitaxial
Al films reaches ∼1.2 GPa. The nt Al films are ∼50–
100% harder than untwinned Al films and harder than
crystal Al with an average grain size of 20–50 nm [60,61].
Average spacing between ITBs in epitaxial nt Al films
is ∼200 nm, greater than the columnar grain size of Al
films reported previously by Lim et al. [58], but the hardness
is also greater. Increased strength in nt Al films likely reflects contributions from both ITBs and CTBs,
as well as growth-in dislocations (Figure 3(d)), resulting
in an effective barrier spacing smaller than the ∼200-nm columns defined by the ITBs. A recent mechanistic
model formulated to rationalize feature size-dependent
strengthening caused by both grain boundaries and TBs
shows that contributions from both types of boundaries are important, especially if they are of similar length
scales [62]. Furthermore, a substantial volume fraction of 9R phase (broad ITBs) may also enhance the strength
of the Al films. The hardness of Ag 5-nm/Al 100-nm
transparent films approaches 2 GPa. However, in multi-
layer films, ITBs, CTBs, and layer interfaces influence
strength, and a full discussion of these mechanisms is
beyond the scope of this article [63,64].

Conclusions We have shown that high-density growth
twins can be introduced into metals with high SFE, such as Al, with assistance from coherent layer interfaces.
Twins may extend from Ag into Al via three experimentally observed mechanisms. However, this is not
meant to be an exhaustive collection of mechanisms by
which twins may enter Al with the assistance of a seed
layer. Coherent interfaces force the epitaxial growth of
Al into one of only two possible twin variants defined by
the Ag underlayer. The template methods described here
may provide a critical forward step toward creating nt
structures in other high-SFE metals. Abundant twins and
stacking faults have exciting implications for improving
mechanical properties in Al and many other metals with
high SFE.

Supplementary online material. A more detailed
information on experiments is available at http://dx.doi.
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