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Size effects on the mechanical properties of thin polycrystalline metal films on substrates

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Abstract

An analysis of the effects of the thickness and grain size of polycrystalline thin films on substrates is presented with the objective of linking the film mismatch stress to the underlying characteristic size scales. The model is predicated on the notion that the relaxation of mismatch strain in the film is accommodated by the introduction of dislocation loops whose population, dimensions and interaction energies are controlled by the film thickness and microstructural dimensions. The model is capable of capturing the combined effects of these size scales by accounting for the interaction energies of the constrained dislocation structure, and provides quantitative predictions of the evolution of film stress during thermal excursions. The predictions of the analysis are compared with available experimental results for polycrystalline films of face-centered cubic materials on Si substrates. It is shown that the model correctly predicts the observed influence of film thickness and grain size on stress evolution during thermal excursions. Aspects of strain hardening in thin polycrystalline films with high dislocation densities are also discussed. © 2002 Acta Materialia Inc. Published by Elsevier Science Ltd. All rights reserved.

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1. Introduction

The physical dimensions of thin film materials are generally comparable to the characteristic microstructural length scales that strongly influence their mechanical properties [1]. The mechanical behavior of thin films has also become an important technological issue in such applications as microelectronics, integrated optoelectronics,

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data storage, and micro-electro-mechanical systems (MEMS). In many cases, the reliability and performance of such devices are strongly influenced by their mechanical properties. It is now well known that the mechanical properties of thin metal films can be substantially different from those of their bulk counterparts [2–5]. Specifically, the flow stress of thin films can exceed that of bulk materials of the same composition and grain size by as much as a factor of three to ten. For example, a 1-µm thick Al film on a relatively thick Si substrate can withstand up to 200 MPa in tension prior to plastic yielding at room temperature [6], whereas the yield stress of pure bulk Al is about

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70 MPa. Prior studies, to be reviewed later in this section, have sought to provide a rationale for the observed high strength of thin films, vis-à-vis bulk materials, by invoking mechanisms of constrained dislocation motion within the film or the grain. However, accurate quantitative predictions of film stress evolution during large temperature changes, in terms of the film geometry and microstructure, are presently unavailable.

There exists a large body of experimental evidence which suggests that the coupling of various length scales influences the plastic response of film-substrate systems in a complex manner. Blakely [7] reported that the tensile stress-strain behavior of monocrystalline and polycrystalline gold films was very similar, suggesting that the microstructural size effect was not an important factor in thin film strength. However, more recent and systematic experiments [8-11] indicate that both the grain size and the film thickness have a marked influence on the strength of thin films. Murakami [8] studied the effect of film thickness and average grain size on strain relaxation due to the thermal mismatch using X-ray diffraction of thin Pb films on Si substrates. The residual strains after strain relaxation, directly related to stress states of thin films, were inversely proportional to both thickness and grain size. Murakami also assumed that dislocation glide was the dominant strain relaxation mechanism in the thin film system. Venkatraman and Bravman [10] measured, by monitoring changes in substrate curvature, stress variations with temperature as a function of film thickness, for a fixed grain size, in pure Al and Al-0.5% Cu films on Si substrates. They found that the film strength varied inversely with film thickness, and examined their results in the context of the classical Hall-Petch relationship for the effect of grain size on strength:

$$\sigma_{\mathbf{v}} = \sigma_0 + K d^{-1/2},\tag{1}$$

where d is grain size and σ_0 and K are empirical constants. Venkatraman and Bravman found the value of K to be slightly greater for the Al thin films as compared with bulk Al, and that the data for thin films followed a d^{-1} variation instead of the $d^{-1/2}$ functional form for bulk materials. More recently, Keller et al. [11] showed that the grain

size dependence of strength in Cu thin films on Si substrates followed a Hall–Petch type relation. They also sought an alternative explanation for thin film strengthening in the Cu–Si system with a superposition of the strain hardening effect by incorporating the increase in dislocation density with grain size reduction.

The effect of film thickness on the flow stress has been modeled [5] by considering the movement of a single threading dislocation that sweeps across a slip plane of the film. Such models invoke the assumption that when a single crystalline thin film, attached to a rigid and comparatively thick substrate, is subjected to a stress in the film plane, dislocations are generated which subsequently glide to relax the film mismatch stress. The minimum shear stress (τ_y) on the slip plane along the slip direction that is necessary to initiate yielding by dislocation glide is expressed as:

$$\tau_{\rm y} \approx \frac{\mu_{\rm f} b {\rm cos} \phi}{h_{\rm f}},$$
(2)

where μ_f is the shear modulus of the film, b is the magnitude of the Burgers vector, h_f is the film thickness, and ϕ is the angle made by the inclined slip plane with the plane of the film/substrate interface. In this expression, the obstacles that pin the dislocation are the film/substrate interface and the native oxide layer formed on the film. However, this simple equation significantly underestimates the yield stress of thin films. (For a thin pure Al film of 1 μ m thickness, it predicts that τ_y is less than 10 MPa.)

Following the analysis of threading dislocations by Freund [12,13], Nix [6] developed a model that incorporates the effect of thickness on the strength of the thin film. This model is based on the balance between the work done by external stress in the gliding dislocation along the slip plane of the film and the stored energy in the interfacial misfit dislocation. The critical biaxial stress required to generate the unit length of misfit dislocation is given by:

$$\sigma_{\text{flow}} = \frac{\sin\phi}{m} \left\{ \frac{\mu_{\text{eff}} b}{4\pi (1 - \nu_f) h_f} \right\},\tag{3}$$

where m is the Schmid factor, v_f is Poisson's ratio of the film, and μ_{eff} is an effective shear modulus

that takes into account the effect of the substrate and native oxide (or passivation layer) on the line tension of the interfacial dislocations, which is defined as:

$$\mu_{\text{eff}} = 2 \left[\frac{\mu_{\text{f}} \mu_{\text{s}}}{\mu_{\text{f}} + \mu_{\text{s}}} \ln \left(\frac{\beta_{\text{s}} h_{\text{f}}}{b} \right) + \frac{\mu_{\text{0}} \mu_{\text{s}}}{\mu_{\text{0}} + \mu_{\text{s}}} \ln \left(\frac{\beta_{\text{0}} h_{\text{0}}}{b} \right) \right]. \tag{4}$$

Here, h_0 is the thickness of oxide on the film, $\mu_{\rm f}$, $\mu_{\rm s}$ and μ_0 are the shear modulus of the film, substrate and oxide, respectively, and $\beta_{\rm s}$ and β_0 are constants of the order of unity. If an oxide or passivation layer is not present, then the second term on the right-hand side of Eq. (4) is omitted.

Eq. (3) predicts that the flow stress of the film is approximately proportional to the inverse of film thickness. This thickness dependence of flow stress is supported by experimental results for largegrained and relatively thick (≥1 µm) films. For example, the calculated stress values from Eq. (3) agreed well with experimental results of coarsegrained Al films on Si substrate, in which grains were enlarged by the laser reflow method [14]. However, the experimental results from polycrystalline Al and Cu films of typical grain size were significantly higher than the predictions from Eq. (3): for a film of 1 µm thickness, the predicted stresses are 23 and 45 MPa for Al and Cu films, respectively, while the corresponding experimental results are 200 and 280 MPa, respectively. Nix [15] incorporated strain hardening into the above model, considering the elastic interaction of a moving threading dislocation with an array obstacle dislocation already present at the film/substrate interface. As the number of obstacle dislocations increases with strain, the elastic strain field in front of the moving dislocation, which inhibits the advance of the moving dislocation, increases substantially. This results in the decrease of effective thickness of the film as the strain increases. Although this mechanistic model is reasonably suitable to predict the experimental results for thin films with large lattice resistance and low dislocation densities, such as hetero-epitaxial films of semiconducting covalent bond materials, the predicted stresses do not agree well with experimental results in the case of metallic thin films, where lattice resistance is negligible and high dislocation densities are commonly observed. In addition, the film stress is only a function of the elastic properties of the film—substrate system in Eq. (3); as a result, explicit variations in film mismatch strain and microstructural length scales are not easily analyzed by this approach.

Grain size is a particularly important microstructural constraint and relevant length scale in polycrystalline thin films. During normal grain growth, the film grain size is usually limited by the film thickness [16]. The grains often extend through the thickness of the film with a preferred orientation, thereby developing a columnar grain structure. The discrepancy between experiments and the predicted strength values has been reduced by linearly superposing a grain size effect, such as the classic Hall–Petch relation, with Eq. (3). Thompson [17] extended Eq. (3) to include the deposition of dislocations at grain boundaries, creating an additional term to account for the grain size effect on film stress:

$$\sigma_{\rm gb} = \left(\frac{2}{m}\right) \frac{\mu_{\rm f} b}{4\pi (1 - \nu_{\rm f}) d} \ln\left(\frac{d}{b}\right),\tag{5}$$

where d is the grain size. This suggests a d^{-1} dependence of strength rather than the classical $d^{-1/2}$ dependence, which was noted by Keller et al. [11] in their study of Cu films. However, such grain size dependence of film stress has not been validated by systematic experiments.

A comparison of available experimental results with the predictions of the foregoing models for thin film strengthening reveals that while the models qualitatively capture the general experimental trends of the effects of film thickness and grain size on the flow stress, they substantially underestimate the magnitude of the flow stress in most cases. This discrepancy is not surprising because these models consider only the movement of a single dislocation, ignoring the interaction between dislocations. In addition, although stresses are limited by the thermal mismatch strain during thermal excursion, these models overestimate the stresses due to the simple linear relation of $1/h_f$ in Eq. (3) for very thin films. Finally, since the only temperature dependence on mechanical response in these models arises from the weak temperature dependence of elastic modulus, they are ill-suited to account for the large variation of stress and plastic properties with temperature throughout the thermal cycle.

In this paper, we present a simple model for the strengthening of thin polycrystalline films subject to a thermal mismatch strain. Instead of a single dislocation or dislocation loop, we consider arrays of circular dislocation loops that are equally spaced and confined within the slip planes of a grain. The generation of each dislocation loop in the set consecutively relaxes the elastic thermal strain, which arises from thermal mismatch between the thin film and the substrate during thermal cycling. With this simplified model, we are able to evaluate the total energy of the system as the sum of the energies stored in dislocation loops, the interaction energies among the loops, and the elastic energy of the grain. By minimization of the total energy of system, time-independent equilibrium strains that are sustained by the metal film are calculated and converted to stresses. This approach allows us to describe stress evolution in films as a function of their thickness, grain size and thermally induced strain (or equivalently, temperature). The predictions of the model are found to agree well with available experimental data for Al and passivated Cu films on Si substrates. The interaction energies among the dislocation loops are shown to play an important role in stress evolution of the thin film.

2. Analysis

For a single-crystal film deposited on a substrate with a different lattice spacing, misfit dislocations form at the film/substrate interface when the film thickness exceeds a critical thickness. Matthews [18] developed a model to predict the critical thickness of the thin epitaxial layer on the substrate by minimizing the total energy in the layer with respect to the elastic strain due to the lattice mismatch between the film and the substrate. The total energy of the system, which is minimized at equilibrium, is the sum of the elastic strain energy and the misfit dislocation energy.

Based on similar energetic considerations, we can develop a model to evaluate the equilibrium

elastic strain that can be sustained in a polycrystalline thin film on a substrate. The model assumes that dislocations or dislocation loops are generated in response to a uniform elastic strain. The total energy of the system can be described as the sum of two parts: the energy associated with the uniform elastic strain in the film ($E_{\rm elastic}$) and the energy associated with the elastic strain fields of dislocations ($E_{\rm disl}$):

$$E_{\text{total}} = E_{\text{elastic}} + E_{\text{disl}}.$$
 (6)

Note that if the substrate is much thicker than the film, then it experiences a much lower in-plane stress than the film. Therefore, we make the usual approximation that all of the strain energy is accommodated by strain in the film, rather than in the substrate. We further assume that the polycrystalline thin film has a preferred texture normal to the film surface, but is of random in-plane orientation; the in-plane properties can then be considered isotropic. If we denote the in-plane quantities by the symbol ' \parallel ' and out-of-plane quantities by the symbol ' \perp ', the elastic energy, $E_{\rm elastic}$, in a single grain of thin film under the equibiaxial strain state is given by:

$$E_{\text{elastic}} = \frac{1}{2} (2\sigma_{\parallel} \varepsilon_{\parallel} + \sigma_{\perp} \varepsilon_{\perp}) V_{\text{g}}$$

$$= 2 \left[\frac{1 + v_{\text{f}}}{1 - v_{\text{f}}} \right] \mu_{\text{f}} \varepsilon^{2} d^{2} h_{\text{f}},$$
(7)

where $V_{\rm g}$ is the volume of a grain, and ε is the equibiaxial mismatch strain to which the film is subjected. Since the film is free to expand out of the plane, σ_{\perp} vanishes. The shape of the grains is assumed to be a rectangular prism.

Transmission electron microscope (TEM) observations by Kuan and Murakami [19] of Pb films on Si substrates cooled to -173 °C from room temperature indicate that most dislocations glide across the grains along {111} planes that are inclined at an angle of $\sim 70^{\circ}$ to the film surface or along {111} planes that are nearly parallel to the film surface. These observations also show that, although the as-deposited Pb film contains very few dislocations at room temperature, a high density of dislocations is introduced in the grains upon cooling, suggesting that dislocations originate at

grain boundaries and subsequently glide across the grain. All dislocation motion is confined within each grain by the native surface oxide, the substrate, the grain boundaries or twin boundaries. It is also found that most of the dislocation glide events are reversible for a small number of thermal cycles [19]. Dislocations induced upon cooling disappear during heating, and the new dislocations appear and glide on another set of slip planes during subsequent cooling. No tangling of dislocations gliding in the parallel glide planes occurs, and no appreciable increase in dislocation density is observed. Similar dislocation characteristics were also reported by Venkatraman et al. [20] for Al-0.5%Cu films on Si substrates that were heated from room temperature to 450 °C.

Drawing on the above observations, we may reasonably represent the dislocation structure in most face-centered cubic (FCC) metal film grains on a substrate with the idealized schematic shown in Fig. 1. The substrate and the native oxide (or passivation) layer on the top of the grain are not shown in the schematic for simplicity. The grain with an arbitrary shape (solid line) is reduced to a parallel prism (dashed line) with a square base. The size of the base and the height of the grain are defined by the effective in-plane grain size and the film thickness. A set of dislocation loops is gener-

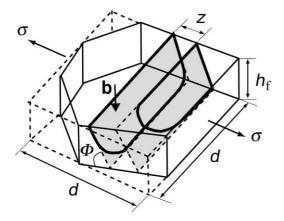


Fig. 1. A schematic of a dislocation loop structure along an inclined slip plane of a grain, where d is the grain size, h_t is the film thickness, z is the distance between loops, Φ is the angle between slip planes and the film surface normal, \mathbf{b} is the Burgers vector, and σ is the applied stress or the film mismatch stress.

ated at grain boundaries and they spread out across the grain on one of the inclined slip planes. However, in order to evaluate $E_{\rm disl}$ for the system, the details of the dislocation configuration should be considered, including the self-energy of different parts of the dislocation loop that result from the different shear moduli of the oxide and the substrate, the image force on those dislocation segments, and the interaction energy of dislocations within a loop and between loops. The number of loops and the size of each loop should also be considered as a function of the applied strain.

Assume that N dislocation loops have glided across the (111) textured grain and relax a given applied strain ε_{app} (which in the context of thermal cycling represents the thermal mismatch strain, $arepsilon_{ ext{thermal}}$, during temperature excursions) to the equilibrium residual elastic strain $\varepsilon_{\rm eq}$. If the film is not bonded to the substrate (e.g., a free-standing film without a surface oxide), and dislocation loops can sweep the entire grain and make slip steps at the surface of the grain, the amount of strain that each generated dislocation loop can relax is $\varepsilon_0 = b'/d$, where b' is the projection of the edge component of the Burgers vector on the film surface (for $\{111\}\langle 110\rangle$ mixed dislocations in an FCC film, $b' = \sqrt{3b/6}$). If the film, however, is bonded to a rigid substrate and/or surrounded by its own oxide layer such that the generated loop is constrained from expanding throughout the entire grain, the resulting strain relaxation due to dislocation glide should be less than that of the free film. The extent of actual strain relaxation would be reduced to $\varepsilon_0 = \eta b'/d$, where η ranges from 0 to 1. The correction factor η is expected be a function of the film thickness, the relative orientation of grains in the film plane, boundary conditions at the film/substrate or film/oxide interface, the elastic constants of the film and the substrate, and the dislocation density [19]. The number of dislocation loops generated in the grain is directly related to the extent of strain relaxation for an imposed thermal strain such that

$$N = \frac{\varepsilon_{\text{thermal}} - \varepsilon}{\varepsilon_0} = \frac{d(\varepsilon_{\text{thermal}} - \varepsilon)}{\eta b'}.$$
 (8)

The biaxial stress state of the film would cause the strain to be relaxed by generating two sets of dislocation loops in two distinct directions. Thus, the dislocation energy can be expressed by:

$$E_{\text{disl}} = 2\sum_{i}^{N} W_{\text{self}} + \sum_{i}^{2N} W_{ij}, \tag{9}$$

where W_{self} is the self-energy of the single loop and W_{ij} is the interaction energy between each dislocation loop in the set.

In order to evaluate the exact total energy of dislocation loops including the interaction energy, several approximations are made so that a tractable model can be formulated.

- 1. Both the film and the substrate are elastically isotropic.
- Image stresses of the edge component of dislocation loops that arise due to the film/substrate and film/oxide interfaces are not considered.
- 3. Dislocation loops are coaxial, circular loops of radius R and uniform separation z (z = d/N).
- 4. The radius of the dislocation loops is an average of the grain size and thickness of the film [for example, $R = (d + h'_f)/4$ for the arithmetic average or $R = \sqrt{dh'_f/\pi}$ for the geometric average, where $h'_f = h_f/\sin\phi$].
- 5. The interaction energy between parallel dislocation loops is limited to the nearest neighbor.

The self-energy and interaction energy of coaxial loops depend not only on loop radius and separation, but also on the direction of their Burgers vector. For two coaxial dislocation loops of the same radius R, with the same Burgers vector that exists in the plane of the loop, and the same sense of dislocation line direction ξ , the interaction energy is given by [21]:

$$W_{\text{inter}} = 2\pi R \frac{2 - v_f}{2(1 - v_f)} \frac{\mu_f b^2}{2\pi} \left(\ln \frac{4R}{z} - 2 \right), \tag{10}$$

where z is the distance between dislocation loops. This equation is valid only when $z \ll R$. The self-energy of a circular dislocation loop with a radius R is obtained by setting $z \approx b$ and dividing Eq. (10) by a factor of two:

$$W_{\text{self}} = 2\pi R \frac{2 - v_f \ \mu_f b^2}{2(1 - v_f) 4\pi} \left(\ln \frac{4R}{b} - 2 \right). \tag{11}$$

Combining these energy terms in Eqs. (10) and (11) with Eqs. (8) and (9), in conjunction with the above assumptions, the total energy of the system is found to be:

$$E_{\text{total}} = E_{\text{elastic}} + 2NW_{\text{self}} + 2(N-1)W_{\text{inter}}$$

$$= 2\mu \frac{1 + v_{\text{f}}}{1 - v_{\text{f}}} \varepsilon^{2}b^{2}h_{\text{f}} \qquad (12)$$

$$+ \frac{d(\varepsilon_{\text{thermal}} - \varepsilon)}{\eta b'} \frac{2 - v_{\text{f}}}{2(1 - v_{\text{f}})} \mu_{\text{f}}b^{2}R \left(\ln \frac{4R}{b} - 2\right)$$

$$+ 2\left(\frac{d(\varepsilon_{\text{thermal}} - \varepsilon)}{\eta b'} - 1\right) \frac{2 - v_{\text{f}}}{2(1 - v_{\text{c}})} \mu_{\text{f}}b^{2}R \left(\ln \frac{4R}{z} - 2\right).$$

For a given thermal strain $\varepsilon_{\rm thermal}$, the equilibrium strain $\varepsilon_{\rm eq}$ is the value of strain ε which minimizes $E_{\rm total}$. The elastic film stress can then be calculated as the product of the film biaxial modulus and the equilibrium strain $\varepsilon_{\rm eq}$.

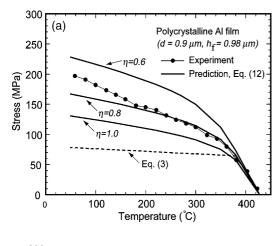
3. Comparison with experimental data

In this section, we compare the predictions of the above model with available experimental data for polycrystalline Al and passivated Cu thin films on Si substrates.

3.1. Polycrystalline Al thin films on Si substrates

Venkatraman and Bravman [10] performed a systematic experimental study of the variations of mismatch stress with temperature as a function of film thickness and grain size for Al films on Si substrates. The variation in film thickness for a given grain size was achieved by the repeated, controlled growth and dissolution of an anodic oxide layer on the surface of the Al film. Stress measurements were conducted as a function of temperature using the substrate curvature method.

Fig. 2(a) and (b) shows the effect of film thickness on the flow stress of polycrystalline Al films, with a fixed grain size, as a function of temperature. The effective radius of dislocations loops is evaluated by setting the arithmatic average of the film thickness and grain size $[R = (d + h'_1)/4]$. While Eq. (3) poorly predicts the stress-tempera-



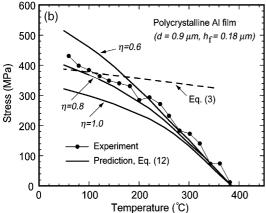


Fig. 2. Stress evolution in thin polycrystalline Al films on Si substrates during cooling: (a) for $d=0.9~\mu m$ and $h_f=0.98~\mu m$, and (b) for $d=0.9~\mu m$ and $h_f=0.18~\mu m$. Symbols are experimental data from Venkatraman and Bravman [10], solid lines are calculated from Eq. (12) and dashed lines are calculated from Eq. (3).

ture relation of these thin films, the stresses calculated on the basis of the present model, Eq. (12), match the experimental trends well for $\eta = 0.6$ to 0.8. In the case of the 0.98 μ m thick Al film, Eq. (3) underestimates the experimental flow stress at room temperature by 40%. This discrepancy is due to the fact that Eq. (3) does not take account the effect of grain size. In the case of the 0.18 μ m thick film, the stresses predicted using Eq. (3), especially in the lower temperature regime, are similar to those predicted by Eq. (12) and the experimental data. This agreement may be attributed to the fact that the thickness of the film is

relatively small compared with its grain size, so that the assumption of a single crystalline film is better suited for this geometry. However, the temperature dependence of film stress in Eq. (3) is not sufficient to describe the stress evolution during the cooling process.

For both film thicknesses, the correction factor η in Eq. (12) that gives the best fit for values is about 0.6–0.8, with the calculated stress at the same temperature increasing with decreasing η . It is noted that these values for η lie within the experimentally measured range. Kuan and Murakami [19] reported that an empirical factor \bar{f} , which is similar to the parameter η in the present model, ranged between 0.6 and 1 for 0.2 μ m thick polycrystalline Pb films of various grain size upon cooling from room temperature to 100 K. They also estimated a theoretical value for \bar{f} in their system to be ~0.6.

The magnitude of stresses evaluated from Eq. (12) also depends on the method used to calculate the radius of the dislocation loops. In Fig. 3, the estimated stress evolution is presented for two different calculations of R, using the same experimental data as shown in Fig. 2. The difference between these two methods becomes significant when the thickness of the films decreases, forcing

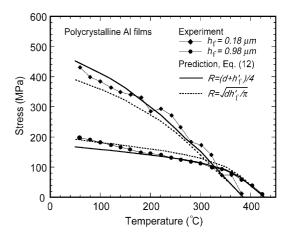


Fig. 3. Stress evolution in thin polycrystalline Al films on Si substrates during cooling, for $h_{\rm f}=0.98~\mu{\rm m}$ and $h_{\rm f}=0.18~\mu{\rm m}$ for a fixed grain size of 0.9 $\mu{\rm m}$. Symbols are experimental data from Venkatraman and Bravman [10], solid lines are calculated from Eq. (12) when $R=(d+h_{\rm f}')/4$ and dashed lines are calculated from Eq. (12) when $R=\sqrt{dh_{\rm f}'/\pi}$.

the actual shape of loops to become more elliptical. This leads to errors in the choice of proper grain shape and, consequently, the assumed radius of the loops R fails to represent the length scale effects well in Eq. (12) for cases in which the ratio of film thickeness to grain size (h_f/d) deviates substantially from unity.

Fig. 4 shows the experimentally measured and calculated film stress as a function of film thickness at 60 °C, after cooling from about 450 °C, for three different grain sizes of Al films. The stresses calculated using Eq. (12) are based on the same temperature difference from a stress-free reference temperature ($\Delta T = 370$ °C). The effects of thickness and grain size on the strength of thin films are reasonably well predicted by Eq. (12) for the choice of η described above; by comparison, the predictions of Eq. (3) match experiments only for the large-grained films. It should be noted that the value of η for a given thickness decreases when grain size decreases. This trend is similar to that shown Fig. 2, which suggests that when either the grain size or the thickness of the Al film decreases, the strain relaxation afforded by dislocation glide is reduced. Another important point to note here is that the slopes of the stress vs. inverse film thickness plots in Fig. 4 are not the same for three dif-

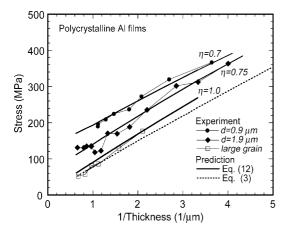


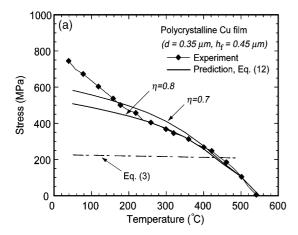
Fig. 4. Stresses in thin polycrystalline Al films on Si substrates with $d=0.9~\mu m$, $1.9~\mu m$ and laser reflowed large grains as a function of film thickness at 60 °C. Symbols are experimental data from Venaktraman and Bravman [10], solid lines are calculated from Eq. (12) and dashed line is calculated from Eq. (3).

ferent grain sizes; this indicates that the effects of grain size and film thickness on film stress are not independent.

3.2. Passivated polycrystalline Cu thin films on Si substrates

Thermal stresses of thin Cu films on Si substrates were examined by Keller et al. [11] as a function of film thickness and the presence of a silicon nitride passivation layer. Three different thicknesses of passivated Cu films were examined and the grain sizes of films were measured by TEM. Dislocation densities were also determined at room temperature via X-ray diffraction. Fig. 5 shows the stress evolution in passivated Cu films of 1.0 µm and 0.45 µm thickness on Si substrates during cooling from 600 °C. The average grain size for the two film thicknesses was 1.3 µm and 0.83 um, respectively. The films were passivated by 50 nm of Si₃N₄. This nitride passivation layer limits diffusional creep through the surface and grain boundaries at high temperatures. Thus the passivated Cu films are more suitable for assessing the validity of the present model than unpassivated films that could develop different stress-temperature responses due to the thermally activated processes.

The predicted stress values of Eq. (3) clearly underestimate the experimental data for both cases because the film thickness and grain size are comparable. The stresses calculated from Eq. (12) correctly predict stress evolution at high temperature in both Fig. 5(a) and Fig. 5(b). However, experimental data begin to deviate from predictions of Eq. (12) around 200 °C, and stresses increase rapidly below that temperature. Similar experimental results were reported by Kobrinsky and Thompson [22] in Ag thin films on oxidized Si wafers. These researchers used in situ TEM observations to show that the rapid increase of stress in the low-temperature regime is due to the thermally activated motion of dislocations through forest dislocation obstacles. Since the present model uses the equilibrium energy criterion as a necessary condition for dislocation generation, and hence does not consider the energy required to nucleate and propagate dis-



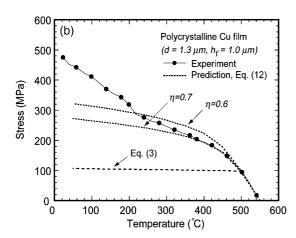


Fig. 5. Stress evolution in passivated thin polycrystalline Cu films on Si substrates during cooling: (a) for $d=0.35~\mu m$ and $h_{\rm f}=0.45~\mu m$, and (b) for $d=1.3~\mu m$ and $h_{\rm f}=1.0~\mu m$. Symbols are experimental data from Keller et al. [11], solid and dotted lines are calculated from Eq. (12) and dashed lines are calculated from Eq. (3).

locations, it cannot capture such mechanisms operating at low temperatures.

In Fig. 6, the experimentally measured and predicted stresses at room temperature and at 250 °C are shown for passivated Cu films as functions of inverse film thickness. At room temperature, the difference between experiments and predictions increases as the film thickness and grain size decrease. This discrepancy is related to the mechanism transition described above. A smaller grain size and/or film thickness would be expected to inhibit the generation and motion of dislocations

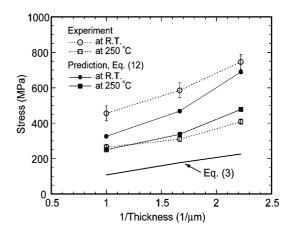


Fig. 6. Stress at room temperature and at 250 °C for passivated thin polycrystalline Cu films on Si substrates as a function of film thickness. Open symbols are experimental data from Keller et al. [11], filled symbols are calculated from Eq. (12) and solid line is calculated from Eq. (3).

more effectively. Above 200 °C, the calculated stresses agree well experiments.

4. Discussion

The dislocation interaction model presented in this paper describes the stress evolution of a film–substrate system during thermal excursion, as shown through the comparison with experimental data. It also indicates the relative importance of the coupled effects of two length parameters (e.g., film thickness and grain size) on the strength of the thin films.

Venkatraman and Bravman [10] and Keller et al. [11] analyzed their experimental assessments of film stress with a linear combination of the effects of film thickness and grain size. In order for the linear superposition of these effects to be valid, the relationship between film stress and inverse thickness must be independent of grain size. From Fig. 4, it is clear that the variation of stress with thickness is indeed a function of grain size: the variation decreases with decreasing grain size: The reported ambiguity concerning the grain size dependence of film strength $(d^{-1} \text{ or } d^{-1/2} \text{ variation})$ could arise from a lack of consideration of this coupling.

In Fig. 7, the residual strains at room temperature after stress relaxation caused by dislocation loop generation are shown for a polycrystalline Al film as a function of inverse film thickness for two values of n, where the film grain size is taken to be equal to film thickness. The system was cooled down from 375 °C to 25 °C. The residual strain increases with decreasing film thickness and the increase in residual strains seems to be mitigated. For the range of film thicknesses shown here, the residual strain does not reach the value of the imposed thermal strain. This trend was experimentally observed in Pb films on Si substrates by measuring strain relaxation via X-ray diffraction [8,23]. They show that the residual strain becomes almost constant when the thickness or grain size is reduced below a critical value, and the plateau in the residual strain occurs at a smaller value than the imposed thermal strain; this difference has been attributed to strain relief by grain boundary relaxation. In situ TEM experiments of Pb and Al films during thermal cycling [5,20] also show that no extensive slip or plastic deformation occurred within extremely small grains, even among different sizes of grains in the same film to which the same magnitude of thermal strain was imposed. The agreement of the present model predictions

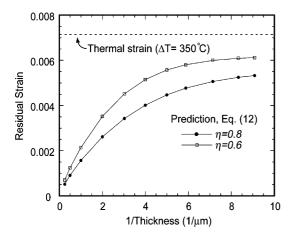


Fig. 7. Residual strain at room temperature as a function of film thickness in thin polycrystalline Al films on Si substrates for an imposed thermal strain. Grain size is assumed to be the same as the film thickness. Solid lines with symbols are calculated from Eq. (12) for $\eta=0.6$ and 0.8, and dashed line represents the imposed thermal strain ($\varepsilon_{\text{thermal}}=0.00714$).

with these experimental observations thus suggests that the limited formation and motion of dislocations inside very small grains is responsible for the high flow stress of thin films and high strain hardening, as described in Section 3.2.

In bulk materials, the increase of yield stress with decreasing grain size is well known as the Hall–Petch relation [24,25]. The basic idea of this microstructural length scale effect is predicated upon the assumption that a dislocation pile-up within a grain can initiate slip across a grain boundary due to the stress concentration at the leading edge of a pile-up. The number of dislocations required to initiate this slip is quite large, and such large dislocation pile-ups have not been observed in thin film systems [20]. One may expect a transition from this pile-up mechanism to another mechanism as the grain size becomes extremely small, as in the case of nanocrystalline bulk materials [26].

As noted by Keller et al. [11] and Kobrinsky and Thompson [22], strain hardening due to the high dislocation density is one of the important sources of thin film strengthening. The model developed in this study accounts for strain hardening as a function of film thickness and grain size by invoking rate-independent flow processes. The calculated stress—strain relation of Al films with different thicknesses, for a fixed grain size, is shown in Fig. 8. It is assumed here that the developed thermal

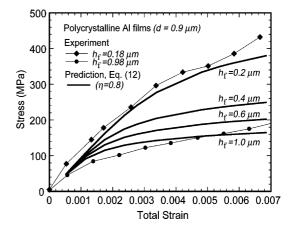


Fig. 8. Stresses in thin polycrystalline Al films on Si substrates as a function of total strain with different film thicknesses. Grain size, d, is fixed at 0.9 μ m. Symbols are replotted from Fig. 2 and solid lines are calculated from Eq. (12).

strain induced during cooling equals the total strain. It is evident that thinner films show greater strain hardening. If power-law strain hardening is assumed to evaluate the apparent hardening exponents n (from $\sigma = H\varepsilon^n$, $\sigma > \sigma_v$), the hardening exponent becomes 0.23 for a 1.0 µm thick film and its value increases to 0.55 for a 0.2 µm film. We obtain a similar variation in n for different grain sizes with a fixed film thickness. (For a 1μm thick Al thin film, n become 0.26 and 0.52 with grain size of 1 µm and 0.2 µm, respectively.) These values of n are significantly higher than that of bulk Al alloy [27]. We believe this high strain hardening in thin metal films is due to the same mechanism previously described, in which the dislocation structure and interaction in thin film grains are completely different from those in the bulk. For example, an Al thin film of 1 µm thickness and 1 um grain size, the interaction energy terms become 35%, 69% and 85% of the self-energy terms in Eq. (12) as the total strain increases from 0.0021 to 0.0041 and 0.0062 due to the increase of temperature difference (ΔT) .

A model for strain hardening in thin films as an extension of Eq. (3), has been proposed [15]. The reduction of effective thickness, resulting from the strain field of a predeposited misfit dislocation array at the film/substrate interface, generates high strain hardening behavior. However, this requires a large array of misfit dislocations in front of an advancing dislocation, which has thus far not been observed in in situ TEM experiments. Furthermore, such a process cannot account for the effect of grain size. The exceptionally high strain hardening exponent $(n\sim0.5)$ has also been observed in a 0.4 um thick passivated Cu film bonded to a glass substrate [28]. The proposed mechanism is that the high strain hardening is accounted for by strong barriers to inelastic shear, provided by the inhibition of lateral spreading. This alternative mechanism arises from the limited volume of the confined thin film.

The dislocation density of thin film can be calculated using Eq. (8) and the volume of grain after we evaluate the equilibrium strain $\varepsilon_{\rm eq}$ from Eq. (12). The calculated dislocation densities of three different passivated Cu films are compared with published experimental results by Keller et al. [11]

in Table 1. The variation of calculated values is caused by the selection of η in Eq. (12), which varies from 0.6 to 0.8. X-ray diffraction was used to determine dislocation densities of the films, and grain size of films were measured by TEM. The calculated dislocation densities are quite high, but smaller than experimental values. Keller et al. determined dislocation densities by an X-ray peak broadening model which was developed for bulk materials and assumed that the peak broadening originated only from the strain fields around dislocations. Thus the cited experimental dislocation densities should be considered an upper bound of dislocation densities. Since the actual dislocation densities can be smaller than the reported values, the difference between calculated and measured dislocation densities by a factor of two may not be significant. The dislocation densities increase as the film thickness and grain size decrease. The typically reported values of dislocation densities for polycrystalline thin films of FCC metals range from 10^{14} to 10^{15} m⁻² [22], which is in good agreement with our estimation shown in Table 1.

5. Conclusions

This work has shown a simple way by which the interaction energies among dislocations can be used to rationalize a variety of experimental observations in thin polycrystalline metal films subjected to thermal excursions. The model successfully predicts the overall trends pertaining to the effect of length scale, such as film thickness and grain size, on stress evolution in film–substrate systems. Through comparison of experimental data for Al and Cu films (with different thickness and grain size) on Si substrates, the stress–temperature curves of those films upon cooling from a high temperature could be estimated without considering the possibility of rate-dependent processes such as diffusional creep.

However, thermally activated processes leading to stress relaxation must be taken into account in order to broaden the scope of this model to those metal films that are not covered by a native oxide or effective passivation layer. Nevertheless, the present approach demonstrates the strong coupling

Table 1 Experimentally measured and calculated dislocation densities of passivated thin polycrystalline Cu films on Si substrates at room temperature after cooling from 600 °C. All data in the table, except the predictions from Eq. (8), are taken from Keller et al. [11]

Film thickness (µm)	Grain size (µm)	Dislocation density (10 ¹⁴ m ⁻²)	
		Experiment	Prediction, Eq. (8)
1.0	1.3	8.6	5.0–6.1
0.6	0.83	13.8	7.28.3
0.45	0.35	22.3	10.3

between the dependence of film stress on two key length scales: the film thickness and the grain size of the film.

These size effects are closely related to the increased strain hardening behavior of thin film systems, which is caused by the confinement of dislocation generation and movement within a small volume of grain. Since the model predicts that dislocation densities of thin films increase as the film thickness and grain size decrease, strain hardening due to high dislocation densities can be considered as an important source of thin film strenghening.

The correspondence between experiments and predictions of the model indicates that the assumed dislocation structure is a plausible model of thin polycrystalline films (with thickness and grain size of the order of µm). This dislocation structure may be chiefly responsible for the unique mechanical behavior of these system. Although the model developed in this study can give us an insight into the relations between size parameters and macroscopic mechanical properties, further work, including more detailed microscopic observations of dislocation evolution and glide during thermal cycling, is required to more accurately calculate the energy of dislocation structures and to understand in detail the evolution of the dislocation structure in a thin film system.

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