Bauschinger effect in thin metal films

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Abstract

The Bauschinger effect in thin sputter-deposited Al and Cu films is studied by isothermally deforming the films alternately in tension and compression. Passivated films exhibit an unusual Bauschinger effect with reverse flow already occurring on unloading, while unpassivated films show little or no reverse flows when the film is fully unloaded.

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1. Introduction

It is well known that the mechanical response of a metallic material depends not only on its current stress state but also on its deformation history. One of the most important examples is the observation that after a metal is deformed plastically in one direction, the yield stress in the reverse direction is often lower. Fig. 1 schematically shows a typical stress-strain curve for metallic materials. The stress $\sigma_f$ is the forward flow stress, and $\sigma_r$ at the start of reverse plastic flow is the reverse flow stress. If $\sigma_r$ is equal to $\sigma_f$, the material hardens isotropically. For many metals, however, the reverse flow stress is found to be lower than the forward flow stress. This anisotropic flow behavior was first reported by Bauschinger [1] and is referred to as the Bauschinger effect. The loss of strength due to the Bauschinger effect is of practical importance since the strength of a metal part may be impaired if the working stress acts in the reverse direction compared to the manufacturing stress. Furthermore, a good understanding of the physical origin of the Bauschinger effect may lead to more refined plasticity theories and may ultimately result in materials with superior mechanical behavior. Many experimental and theoretical efforts have been devoted to studying the Bauschinger effect in bulk metals since the phenomenon was first reported [2–15]. The physical origins are generally ascribed to either long-range effects, such as internal stresses due to dislocation interactions [6,7], dislocation pile-ups at grain boundaries [8,9] or Orowan loops around strong precipitates [10–14], or to short-range effects, such as the directionality of mobile dislocations in their resistance to motion or annihilation of the dislocations during reverse straining [10]. Satisfactory agreement has been achieved between models and experimental results obtained in various bulk metals and alloys [3–5,7–11].

Thin metal films are widely used in many advanced devices across a wide range of industries. Reliability problems encountered in these applications have motivated a strong interest in the mechanical behavior of thin films [16,17]. Besides this technological driving force, thin films also provide a unique opportunity to investigate fundamental problems in materials science. For example, at least one dimension of a thin film is comparable to the characteristic length scales associated with material defects such as dislocations; thus, free surfaces and interfaces are expected to play an important role in the mechanical behavior of thin films [18].

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Furthermore, as a result of the special manufacturing techniques and the small materials dimensions, films often have unique microstructures. For example, thin films often have a columnar grain structure with an average grain size that is much smaller than in bulk materials; they are frequently highly textured [19]. As a result of these dimensional and microstructural constraints, many materials behave mechanically very differently in thin film form than in the bulk, especially in the plastic regime [18]. Various theoretical models and numerical simulations have been proposed recently to describe thin-film plasticity including strain-gradient plasticity theories [20,21], crystal plasticity theories [22], and discrete dislocation simulations [23,24]. The plasticity theories and the discrete dislocation models explain the strengthening effects associated with the film thickness and microstructure reasonably well; they predict, however, very different behavior in reverse loading. In the discrete dislocation simulations [22,25] and some crystal plasticity theories [25], passivated films show a distinct Bauschinger effect upon unloading after plastic pre-straining in tension. Reverse plastic flow starts early even though the overall stress in the film is still in tension. This type of Bauschinger behavior is not predicted in other models [20–22] and is also very different from that typically found in bulk materials. Up to date, however, there has been no direct experimental evidence of such a Bauschinger effect in thin metal films.

2. Experiment and results

The new experimental method is based on the plane-strain bulge test technique [26,27]. In this technique, the film of interest is deposited on a Si wafer and long rectangular membranes are fabricated using standard micromachining technology. Fig. 2(a) shows a perspective view of a typical bulge test sample. The as-prepared membrane is initially flat and under tension. It is then deflected by applying a uniform pressure to one side causing a state of plane strain in the film. The applied pressure, \( p \), and corresponding membrane deflection, \( h \), are measured and converted to a stress–strain curve using the following two simple formulae [26,27]:

\[
\sigma = \frac{pa^2}{2ht} \quad \text{and} \quad \varepsilon = e_0 + \frac{2h^2}{3a^2},
\]

where \( t \) is the film thickness, \( 2a \) the membrane window width, as shown in Fig. 2(a), and \( e_0 \) the residual strain.
in the film, which is equal to the residual stress divided by the biaxial modulus of the film. Fig. 2(b) illustrates how this technique can be modified to test films in compression. A bilayer membrane is microfabricated consisting of the metal film of interest on a thin ceramic layer such as Si$_3$N$_4$ that has a much higher elastic modulus than the metal film. During the test, the metal film first flows in tension, while the Si$_3$N$_4$ layer only deforms elastically. Upon unloading, the tensile stress in the Si$_3$N$_4$ layer drives the metal film into compression, while the overall stress in the bilayer is kept in tension to prevent buckling of the membrane. The Si$_3$N$_4$ film thus serves a dual purpose: It provides the driving force to deform the metal film into compression and it passivates one of the surfaces of the metal film. By adjusting the thickness ratio of the Si$_3$N$_4$ layer and the metal film, different levels of compressive stress can be obtained in the metal film.

If the Si$_3$N$_4$ film and the metal film are denoted as layers 1 and 2 with thickness $t_1$ and $t_2$, respectively, the average stress in the bilayer, $\sigma_b$, is given by

$$\sigma_b = \frac{t_1}{t_1 + t_2} \sigma_1 + \frac{t_2}{t_1 + t_2} \sigma_2,$$

where $\sigma_1$ and $\sigma_2$ are the stresses in the respective layers. These stresses are related to the corresponding strains in each layer through the following constitutive equations:

$$\sigma_1 = f_1 \left( \varepsilon_{01} + \frac{2h_1^2}{3a^2} \right)$$

and

$$\sigma_2 = f_2 \left( \varepsilon_{02} + \frac{2h_2^2}{3a^2} \right),$$

where $\varepsilon_{01}$ and $\varepsilon_{02}$ are the residual strains in layers 1 and 2, respectively. Equilibrium requires that the average stress in the bilayer, $\sigma_b$, is equal to the total pressure supported by the bilayer and the Si$_3$N$_4$ membrane gives the deflection $h$. According to Eq. (5), $p_2(h)$ can be obtained by subtracting the pressure–deflection curve of a freestanding Si$_3$N$_4$ film from that of the bilayer. The stress–strain curve of the metal film, $f_2$, can be calculated from $p_2(h)$ using Eq. (1). This method is generally applicable as long as the constitutive equation of layer 1, $f_1$, does not change when layer 2 is removed.

The detailed sample preparation method is discussed elsewhere [27]. Briefly, 1 $\mu$m thick Al films were sputter deposited onto a Si wafer, coated on both sides with 75 nm of Si$_3$N$_4$ by means of low-pressure chemical vapor deposition (LPCVD). Immediately prior to the Al deposition, a thin TiN sticking layer was grown using reactive sputtering. Freestanding Al/TiN/Si$_3$N$_4$ composite membranes were microfabricated by opening rectangular windows in the Si substrate using standard silicon micromachining techniques. Freestanding Cu/Si$_3$N$_4$ bilayers were prepared by first microfabricating freestanding Si$_3$N$_4$ membranes followed by sputter depositing 600 nm Cu films directly on top of the Si$_3$N$_4$ membranes. Both sets of samples were vacuum annealed at 300 °C to stabilize the microstructure. Freestanding Al and Cu membranes were prepared by etching the Si$_3$N$_4$ or Si$_3$N$_4$/TiN layer beneath the metal films using reactive ion etching (RIE). Transmission electron microscope (TEM) micrographs in Fig. 3 show that grains in both films are roughly equiaxed and that the average grain size is 2.1 $\mu$m for Al and 0.9 $\mu$m for Cu. Both films have a columnar grain structure with grain boundaries traversing the thickness of the film. Annealing twins are only found in Cu grains.

Both composite and freestanding films were tested in multiple loading/unloading cycles with a bulge test apparatus [27]. For the Cu/Si$_3$N$_4$ samples, the elastic contribution of the Si$_3$N$_4$ films was measured independently by dissolving the Cu in dilute nitric acid and testing the Si$_3$N$_4$ membranes separately. The pressure–deflection of the Cu/Si$_3$N$_4$ bilayer and the freestanding Si$_3$N$_4$ membrane are plotted in Fig. 4. The curve of the freestanding Si$_3$N$_4$ membrane consists of both loading (solid line) and unloading (solid squares) sections. It can be seen that the deformation of the Si$_3$N$_4$ membrane is elastic. For a given deflection, the difference between the pressures supported by the bilayer and the Si$_3$N$_4$ membrane gives the contribution of the Cu film, as illustrated by the insert in Fig. 4. The stress–strain curve of the Cu film is then calculated using Eqs. (1) and (6). The same method could not be applied to the Al samples because the TiN had a large compressive stress that caused the freestanding Si$_3$N$_4$/TiN bilayer to buckle if the Al was dissolved. Instead, the properties of the Si$_3$N$_4$/TiN were obtained from the elastic unloading curves of both Si$_3$N$_4$/TiN/Al and freestanding Al films [26]. The elastic contribution of Si$_3$N$_4$/TiN bilayer can then be subtracted from the data of the Si$_3$N$_4$/TiN/Al composite films.
The resulting stress–strain curves are presented in Fig. 5(a) and (b) for passivated Cu and Al films, respectively. Also plotted in Fig. 5 are typical stress–strain curves for unpassivated Cu and Al films. For both materials, the passivated films show a very strong Bauschinger effect: during unloading, reverse plastic flow starts when the applied stress is still in tension and continues when the films are loaded in compression. Each loading cycle shows significant hysteresis, which increases with increasing plastic strain. By contrast, the stress–strain curves of unpassivated films have unloading cycles with little or no hysteresis.

3. Discussion

A dislocation-based mechanism is proposed to explain the difference between passivated and unpassivated films. Fig. 6 depicts a thin metal film with a columnar grain structure. The lower surface of the film is passivated by a material that forms a strong interface, while the upper surface is free. When dislocations reach an obstacle such as a grain boundary or a strong interface during forward plastic deformation, they form pileups; when they reach a free surface, they simply exit the material to form surface steps. In the presence of pileups, dislocations in the film are subject to two types of stress: the externally applied stress and the internal stress imposed by the pileups; the former provides the driving force for further plastic flow, while the latter acts as a...
resistance. During tensile loading of a passivated film, dislocation pile-ups form at the film/passivation interface resulting in significant back stresses in the film. Upon unloading, these back stresses cause dislocations to glide in the opposite direction resulting in a nonlinear unloading curve. The different behavior of passivated and freestanding films arises as follows. In freestanding films, many dislocations can exit the film due to the proximity of the free surface and no significant Bauschinger effect is observed. If one of the film surfaces is passivated (Fig. 6) and the interface is strong, a boundary layer with high dislocation density is formed near the interface [23]. This boundary layer, which may consist of simple pileups or may have a more complex structure, has an associated back stress that is not present in freestanding films. As a result of this high back stress, reverse plastic flow starts much earlier during the reverse loading cycle. If the back stress is large enough, reverse flow can start even when the externally applied stress is still in tension, as observed in the stress–strain curves of the passivated Cu and Al film in Fig. 5. It should be noted that the Bauschinger effect will occur only when the obstacle to dislocation motion is strong enough to cause significant back stresses. The data in Fig. 5(b) suggest that this is not the case for the thin native oxide that forms on freestanding Al films.

In addition to the composite-film technique proposed here, the substrate curvature technique can also be used to load thin films deposited on a substrate alternating in tension and compression. In this technique, a metal film on a silicon substrate is loaded into compression by heating the sample until the residual stress in the film becomes compressive due to the thermal mismatch with the substrate. Upon cooling, the film goes into tension. From the change in substrate curvature, one can determine the stress in the film and thus obtain an estimate for the forward and reverse flow stress. Using this technique, Baker et al. [28] observed “anomalous” thermomechanical behavior for Cu films encapsulated between Si₃N₄ barrier and passivation layers, when a small amount of oxygen was introduced in the deposition system during film growth: the films exhibited a Bauschinger-like behavior that is large compared to that observed for bulk Cu. The result is difficult to interpret, however, because the applied strain cannot be decoupled from the temperature change and because at elevated temperatures other deformation mechanisms such as diffusional creep may come into play [29]. Compared to the substrate curvature technique, the composite film technique proposed here has the unique advantage of measuring the isothermal stress–strain behavior and is thus ideal for studying Bauschinger effect in thin metal films.

4. Summary

In conclusion, we have developed a new experimental technique to deform thin metal films alternately in tension and compression and to measure the corresponding isothermal stress–strain curve. This method was used to investigate the Bauschinger effect in thin sputter deposited Al and Cu films. A large Bauschinger effect is found in both materials when one of the film surfaces is passivated with a thin layer of LPCVD Si₃N₄. We propose that the strong interface between the film and passivating layer prevents dislocations from exiting the film and allows the build-up of significant back stresses. On unloading, these back stresses cause reverse plastic flow. By contrast, stress–strain curves of unpassivated films show little or no reverse flows when the film is fully unloaded because the free surface allows dislocations to exit the film and no back stresses are generated. We anticipate that these experimental results will be useful for validating and refining existing models for thin-film plasticity.

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References