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Flow and Fracture of Free-Standing Thin Films and Multilayers

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ABSTRACT

Free-standing polycrystalline films of Ag, Cu and Ag/Cu multilayers were tested in tension using an optical diffraction grating for the measurement of the strain. The yield stress and the rate of work hardening both increase strongly with decreasing layer thickness. These effects combine to raise the stress for ductile fracture up to 700 MPa and to lower the maximum plastic strain.

KEYWORDS

Thin films, multilayers, yield stress, Hall-Petch, work hardening, fracture stress, ductile fracture.

INTRODUCTION

The study of the mechanical properties of thin films and multilayers is of considerable technological interest. At the same time, the microstructural control provided by the deposition processes of these films makes it possible to study systematically the effects on mechanical behavior of two length scales, the grain size and the individual layer thickness. These are not independent: if the deposition temperature is sufficiently high to allow extensive surface diffusion, the grain size is similar to the thickness of the film (if single component) or of the individual layer thickness in multilayers [1,2].

The main difficulty in the mechanical testing of thin films is the accurate measurement of the strain, which should be carried out directly on the deforming sample [3,4,5]. In our laboratory, we developed a technique based on optical diffraction from a lithographically deposited grid [6]. It allows accurate determination of elastic strains as well as measurements of large plastic strains.

In this paper, we describe the technique and review the results obtained on Ag and Cu thin films, as well as Ag/Cu multilayers [7]. The dependence of the yield stress on the layer thickness is analyzed in terms of the Hall-Petch relation and compared to the results from hardness tests. The rate of work hardening strongly increases with decreasing layer thickness [2]. These results are used to interpret the trends observed in the fracture stress, the ductility and the fracture surface morphology.

EXPERIMENTAL METHODS

Thin film samples were deposited through a dogbone-shaped mask on glass substrates by electron beam deposition in high vacuum. Typical dimensions were a thickness of 3 μm, a gauge length of 10 mm and a width of 3 mm. The bilayer repeat length of the multilayers varied from 3 nm to 3 μm, depending on the number of layers. The thickness of the Ag and Cu layers was equal. Both the pure films and the multilayers had a strong <111> texture.

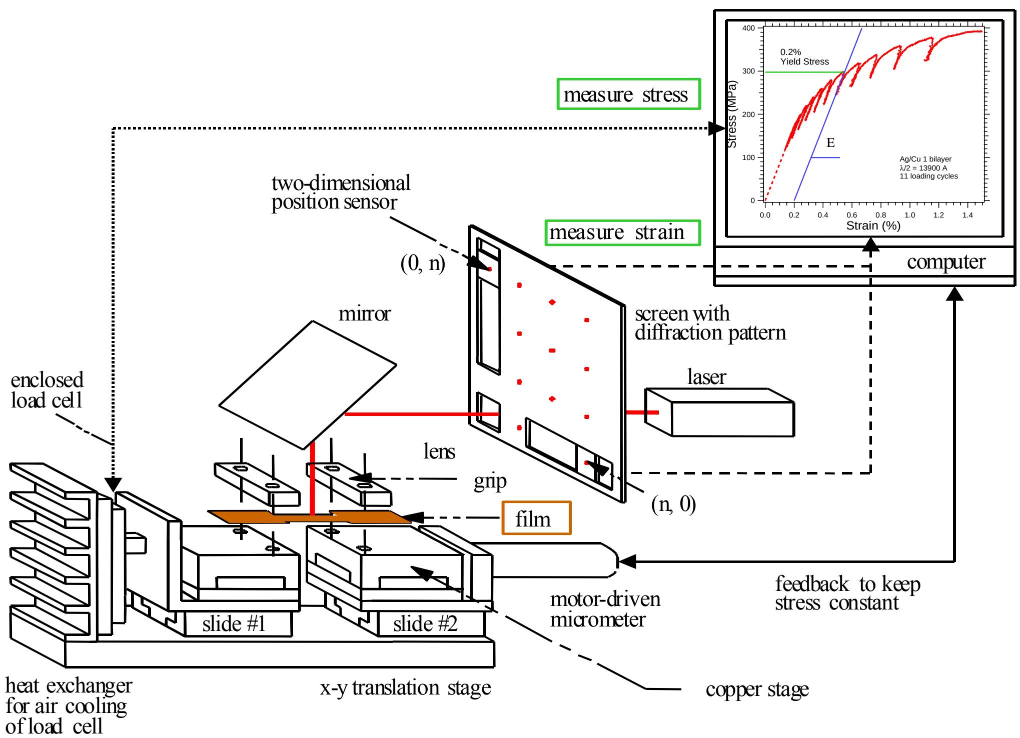


Figure 1: Schematic diagram of the tensile tester for thin film samples [7].

A thin, very compliant, two-dimensional square grid of photoresist dots (see Figure 4(a) for an oblique view) was deposited on the films by photolithography. The films were then removed from the substrate and placed in the grips of the tensile apparatus of Figure 1. The differences between the positions of two spots of the diffraction pattern produced by a

He-Ne laser, monitored by two-dimensional position-sensitive detectors, were used to determine the longitudinal and transverse strains. The transverse waviness that developed during plastic flow, however, made the measured transverse strains, and hence Poisson's ratio, unreliable.

The result of a typical test is shown on the computer screen of Figure 1. The initial stages of the tests did not yield meaningful data because it is impossible to mount the samples such that they are perfectly taut. It was therefore necessary to deform the sample first and to determine Young's modulus from partial unloading. The value of the modulus was then used to reconstruct the initial elastic loading line; its intersection with the zero-stress axis determined the origin for the strains. The yield stress, σ_y, was determined at the conventional value of 0.2% plastic strain. The full stress-strain curve is the outer envelope of the data. All tests were carried out at room temperature and at a strain rate of 1.1×10^{-5} s^{-1}.

RESULTS AND DISCUSSION

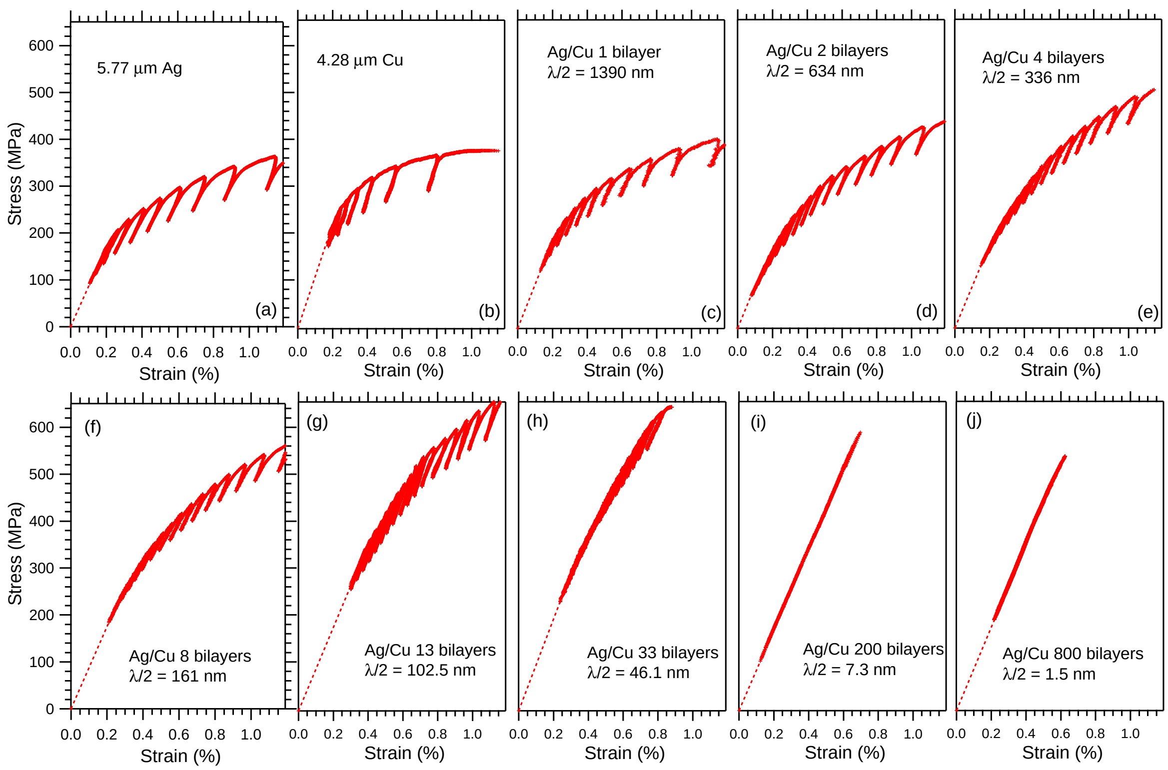


Figure 2: Stress-strain curves for films of pure Ag, pure Cu and Ag/Cu multilayers. The thickness of each film, or the number of bilayers and thickness ($\lambda/2$) of each layer are indicated [7].

Yield stress

Figure 2 shows the stress-strain curves for pure Ag, pure Cu and a series of Ag/Cu multilayers with decreasing bilayer repeat lengths, λ. For the shortest repeat lengths, no yield stress could be determined because no macroscopic plastic flow could be measured before fracture occurred (see Figure 2(i) ad 2(j)). The yield stress clearly increases with decreasing λ. Traditionally, this dependence has been analyzed using a power law:

$$\sigma_y = \sigma_o + k\, d^{\,n} \qquad (1)$$

where d is the grain size and σ_o is the yield stress for very large-grained samples. For n = –0.5, Eqn. 1 reduces to the Hall-Petch relation. The yield data from our tests are shown as a log-log plot of $(\sigma_y - \sigma_o)$ vs. grain size in Figure 3, together with nanoindentation data on the samples with the smallest repeat lengths [2] and microindentation data on coarse-grained copper [8]. The hardness data, H_v, were converted to yield stresses using the Tabor relation:

$$\sigma_y = H_v/3. \qquad (2)$$

σ_o for all data was 168 MPa, which was obtained from a Hall-Petch plot (σ_y vs. $d^{-1/2}$) of the microhardness data. The grain size in our samples was taken to be $\lambda/2$. The offset between the tensile and nanoindentation data on the multilayers arises from the inaccuracy of the Tabor relation for samples with strong work hardening.

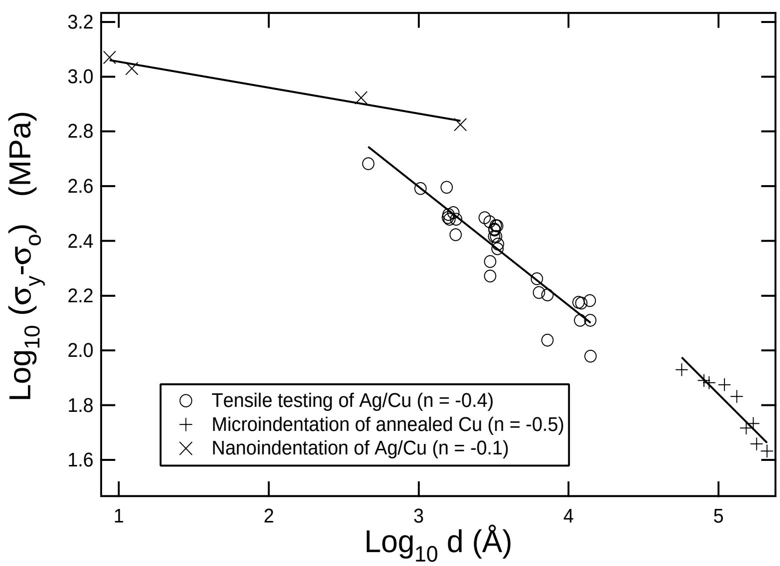


Figure 3: Comparison of the yield stresses of Ag/Cu multilayers (O) [7] with nanoindentation data on Ag/Cu multilayers (×) [2] and microindentation data on coarse-grained Cu (+) [8]. Hardnesses were converted to yield stresses by the Tabor relation ($H_v/3$). The value of σ_o, 168 MPa, was determined from a Hall-Petch plot of the microindentation data.

The slope of the lines in Figure 3 gives the exponent of Eqn. 1. For the microindentation data, n = –0.5. For the tensile data, n = –0.43, which is close to the classical Hall-Petch value. The slope of the line through the nanoindentation data, however, is n = –0.1. Such a deviation from Hall-Petch behavior has been seen in nanoindentation measurements on other metallic multilayers [9] and has been attributed to changes in the dislocation dynamics when the number of dislocations in the pile-ups near the grain boundaries becomes small [10]. Importantly, however, the yield stress and hardness continue to rise with decreasing grain size even at the smallest sizes; no softening is observed.

Work hardening

The work hardening rate, $\theta = d\sigma/d\varepsilon$, of the stress-strain curves for the multilayers in Figure 2 has been analyzed [2] using the formalism developed by Kocks [11]. The initial work hardening rate is 10 to 20 times higher than the maximum work hardening rate, θ_{II}, in single crystal silver. This can be explained by the introduction of an additional set of dislocation obstacles with an effective spacing, D.

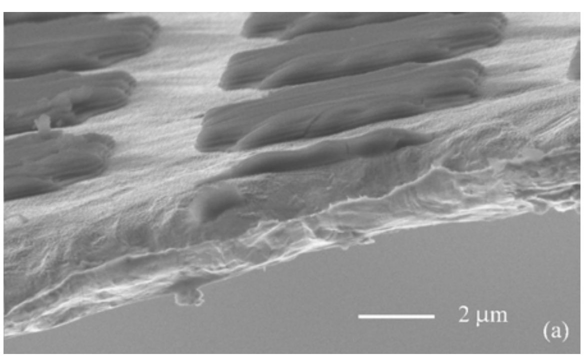
$$\theta = \theta_{II} + \frac{M^3 (\alpha G)^2 b}{2\sigma D} - \sigma\left(\frac{\theta_{II}}{\sigma_{sat}} + \frac{KM}{2D}\right) \quad (3)$$

where M is the Taylor factor, G the shear modulus, b the Burgers vector, and σ_{sat} the saturation stress of the single crystal; α and K are constants.

D decreases with decreasing grain size but is substantially smaller than d. The dislocation structures built up as a result of the presence of interfaces could decrease the effective obstacle spacing provided by the layers [2].

Fracture and ductility

As shown in Figure 2, the fracture stress of the multilayers increases with decreasing λ as long as macroscopic yield can be observed. Values as high as 700 MPa have been measured. The plastic strain at fracture decreases continuously with λ and is only 0.16% in the strongest samples. For the shortest repeat lengths (λ < 80 nm) no macroscopic plastic strain is observed. In a few samples, with large repeat lengths, plastic strains up to 2.3% have been measured.



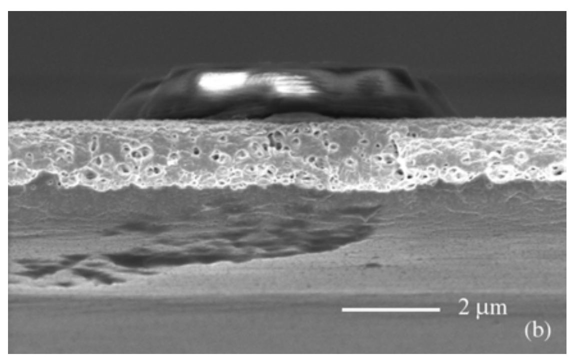


Figure 4: Scanning electron micrographs of the fracture surfaces of (a) a Cu thin film, (b) a Ag/Cu multilayer (bilayer repeat length 40 nm), both fractured in tension.

Figure 4(a) shows the fracture surface of a pure Cu film broken in the tensile tester. The overall ridge shape and the "vein" in the middle are clear indications of a simple, macroscopic ductile fracture. Figure 4(b) shows the fracture surface of a multilayer with a bilayer repeat length of 40 nm. Although the overall shape of the surface is more planar than that in Figure 4(a), there are many "vein"-like features present that indicate the occurrence of extensive plastic flow. The edges of the pits also indicate that they were formed by local plastic flow.

The fracture of the Ag/Cu multilayers is intrinsically ductile. The fracture stress increases with decreasing repeat length due to the increase in, first, the yield stress, and then the rate of work hardening. The decrease in the ductility is directly attributable to work hardening as well. The absence of macroscopic yielding and the lower strength in the multilayers with the shortest repeat lengths is most likely due to the samples being more sensitive to alignment in the apparatus when there is less plastic accommodation.

CONCLUSION

The interfaces in metallic multilayers can be very effective obstacles to dislocation motion, leading to substantial increases in yield stress as well as in the rate of hardening. These effects combine to raise the resistance to ductile fracture in these materials.

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