Thickness dependent critical strain in submicron Cu films adherent to polymer substrate

R. M. Niu, G. Liu, C. Wang, G. Zhang, X. D. Ding, and J. Sun[a)]
State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China

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For the polymer-supported metal thin films that are finding increasing applications, the critical strain to nucleate microcracks (ε_C) should be more meaningful than generally measured rupture strain. In this letter, the ε_C values of polymer-supported Cu films are simply but precisely determined by measurements of both electrical resistance and statistical microcrack density changes on the film surface. Significant thickness dependence of ε_C, i.e., the thinner the film the lower the ε_C, has been revealed for the Cu films with a thickness range from 700 down to 60 nm, which is suggested to result from the constraint effect of refining grain size on the dislocation movability. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2722684]

Metal thin films are extensively used in interconnects in multilevel metallization of ultralarge scale integrations. As the dimension of integrated circuits continues to shrink, it is becoming ever more crucial to understand the mechanical properties of metal thin films with size in submicron scale,[1] in order to ensure the reliability of interconnects and favor the design and fabrication of integrated circuits. The strength of metal films, as well known, has attracted numerous investigations and some measurement methods, such as tensile testing, indentation, microbeam bending, and bulge testing, have been developed[2–5] to determine the thickness dependent yield strength of the films. Recently, the method of tensile testing the metal thin films on compliant substrates[6,7] has also been suggested to determine the yield strength of metal films, where the force of highly elastic compliant substrates could be subsequently subtracted from the total measured force of the compound to yield the real one of films. This provides a more precise measurement on yield strength for thin film used especially as flexible electronics that are being widely used in many applications, including paperlike display[8] and sensitive skins.[9]

Besides the yield strength, ductility is another important parameter in the mechanical property of the metal films. Measurement on freestanding films by using simple tensile test could provide result closer to intrinsic ductility of the metal films, although this method involves too much complicated sample preparation and handling and is also not applicable for the films with a thickness less than about 3 μm. When the freestanding metal films due subjected to a tensile strain, the films can deform plastically but cannot harden as their bulk counterparts because the dislocations in metal films are ready to escape due to the limited thickness constraint. This means that a neck that is induced preferentially at a local spot could cause further intense localized deformation, resulting in fast rupture. In other words, the rupture strain of freestanding films is close to the strain needed to nucleate microcrack or neck, because of the low hardening capability and small thickness-to-length ratio of the films. This is responsible for the previous reports[10,11] that the rupture strain of freestanding films is much lower than the corresponding bulk metals, such as only 0.6% for 5.77-μm-thick Au film and only 1.3% for 4.28-μm-thick Cu film.[10]

On the other hand, when the metal film is deposited on a polymer substrate, the strain localization in the film could be suppressed by the substrate due to geometrical constraint. Both numerical simulations[12] and experimental investigations[13–15] have clearly shown that the polymer-supported metal films, if well bonded to the substrate, could deform uniformly to a large strain. Two methods have been used to estimate the deformation capability of the films. One is to observe the microcracks in the films with applied strain. For example, it was found[13] that the Cu films well bonded on polymer substrate could sustain strain up to 10% without appreciable cracks and up to 30% with only discontinuous microcracks, far larger than its freestanding counterpart. The other one is to make real-time examinations on the change in electrical resistance of the films because the damage or failure in microstructure could be sensitively detected by the macroscopic property of electrical resistance. For example, Cr thin films, deposited on polymer substrates with granular In layer as buffer layer, were found[15] to sustain strain up to 38% with electrical resistance changed by a factor of less than 2.

These crude measurements with no comparability between each other, however, were qualitatively used to present how the rupture strain of the films could be achieved. Actually in the polymer-supported metal films, the rupture strain is much larger than the critical strain (ε_C) for nucleating microcrack or neck, and ε_C should be more meaningful for applications because this parameter indicates the beginning of damage in microstructure and weakening in serve properties. Besides, the determination of ε_C could make the different measurements comparative. These make the systematic works on ε_C urgently needed especially for the applications in electronic fields, where the lifetime prediction of components is very important in order to avoid sudden failure. In this letter, the ε_C values of polymer-supported Cu films are simply but precisely determined by measurements of both electrical resistance and statistical changes in the microcrack density of the films. Significant thickness dependence of ε_C

[a)]Author to whom correspondence should be addressed; electronic mail: junsun@mail.xjtu.edu.cn

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has been clearly revealed for Cu films with a thickness range of 700 down to 60 nm, for which explanations were presented.

The submicron Cu films used in the present work, with a thickness range from 700 down to 60 nm determined by Rutherford backscattering, were deposited on dogbone-shaped polyimide substrate (4×20 mm^2) by magnetron sputtering. The sputter current was 0.45 A and the bias voltage −80 V, which resulted in a deposition rate of about 5.9 nm/min. This low-energy slow deposition rate was experimentally determined to have almost no effect on the mechanical properties of the 125-μm-thick polyimide due to its good temperature endurance. Prior to deposition, the polyimide was firstly cleaned by Ar ion bombardment. The as-deposited films were *in situ* annealed at 100 °C for 2 h to eliminate the residual stress. X-ray diffraction measurements revealed that all the Cu films were polycrystalline and no obvious texture was found even in the thinnest 60 nm film. Based on quantitative microscopy, the average grain size of the films was determined using transmission electron microscope.

Uniaxial tensile testing was performed using a Micro-Force Test System (MTS) microtensile tester with a 50 N force transducer at a constant strain rate of 1×10^{-4} s^{-1}. The force and displacement/strain were automatically recorded by machine and a high-resolution laser detecting system, respectively. In order to obtain the intrinsic strain-stress curve of Cu films, pure polyimide with the same width and length as those of the Cu film/polyimide system was also tested and the strain-force data were recorded and subtracted from those of the Cu film/polyimide system, following the treatments in Ref. 6. When the intrinsic strain-stress curve of Cu films is obtained after subtraction, the yield strength is determined as the 0.2% offset.

In order to determine the critical strain (ε_C) for nucleating microcrack or neck, both electrical resistance (ER) and statistical microcrack density (SMD) were employed for comparison. During tensile testing, the real-time electrical resistance (R) of the films was measured and the relative change in resistance [$\Delta R/R_0$, where $\Delta R = R - R_0$ and $R_0 = R(\varepsilon = 0)$] was subsequently depicted with respect to strain (ε). Two distinct regions could be clearly seen in the $\Delta R/R_0$ vs ε curves. In the first region, the films will elastically deform with conserved volume. However, the change in the film form (lengthening and decrease of cross section) leads to a change in $\Delta R/R_0$ with strain. Because no damage exists, $\Delta R/R_0$ increases linearly with ε in this region. With the further increase in strain, damage will be induced and the existence of microcracks and voids will increase the electrical resistance significantly, causing the $\Delta R/R_0$ vs ε curve to deviate from the linear stage and go into the second nonlinear stage. The critical strain, where the $\Delta R/R_0$ vs ε curves transform from linear first stage to the nonlinear second stage, is defined ε_C in the electrical resistance change. On the other hand, in the statistical microcrack density measurement, the density of microcracks (total length of the microcracks per unit area, denoted as S) in the films was *ex situ* determined after different strain testing. Extrapolation treatment was subsequently employed to put the S vs ε curves back to the point of $S=0$, where the strain was regarded as ε_C. The physical meaning of ε_C in the statistical microcrack density change is that, at this critical strain, microcracks begin to form. The two methods for determining ε_C are typically illustrated in Figs. 1(a) and 1(b), respectively.

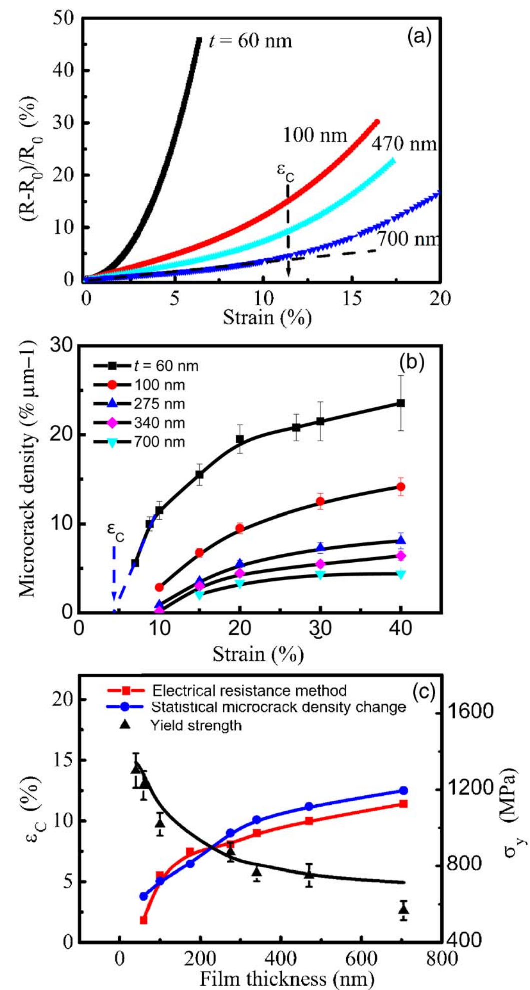

FIG. 1. (Color online) (a) Dependence of electrical resistance change [$\Delta R/R_0 = (R - R_0)/R_0$] on strain for different thick films. (b) Changing in microcrack density with strain for different thick films. (c) Thickness dependent SMD-determined and ER-determined critical strains, as well as yield strength. Illustrations for determining the critical strain by using both electrical resistance method and statistical microcrack density changes are shown in (a) and (b), respectively.

The grain size is experimentally found to decrease with decreasing film thickness t, while the yield strength enhances with decreasing t [Fig. 1(c)]. These trends are well consistent with previous results.[16–18] The yield strength was found to be dependent on the grain size, which follows the expression of $\sigma_y = \sigma_0 + k(d)^{-0.5}$, where σ_0 is the lattice friction stress (116 MPa for Cu), and k is a constant of 362 MPa μm$^{0.5}$.

Some $\Delta R/R_0$ vs ε curves for Cu films with different thicknesses are typically shown in Fig. 1(a). One can find that the thicker films should undergo somewhat longer linear first stage than the thinner ones, which means that the thicker films should have larger ε_C. All the films exhibit an enhanced electrical resistance with increasing strain, i.e., a monotonic increase in $\Delta R/R_0$ with ε, and the thinner films have more remarkable promotions in $\Delta R/R_0$ than the thicker ones at a given strain. When the film thickness decreases down to several tens of nanometers such as 60 nm, the $\Delta R/R_0$ vs ε curve of film is intensely sharp. This means that the metal films with a thickness of only several tens of nanometers may be more ready to fail in tension, resulting in a serious increase in electrical resistance, which should have an impact effect on the further shrinking in dimension of integrated circuits. On the other hand, Fig. 1(b) presents statistical changes of

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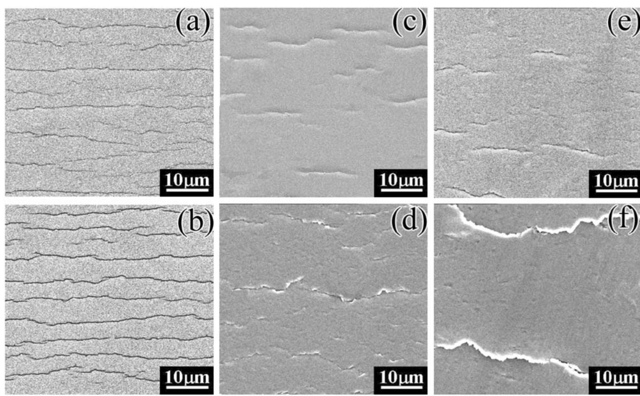

FIG. 2. Microstructural evolution with strains in 60-nm-thick [(a) and (b)], 340-nm-thick [(c) and (d)], and 700-nm-thick [(e) and (f)] Cu films, where (a), (c), and (e) are under the strain of 20% and (b), (d), and (f) are under the strain of 30%. All the tensile directions are vertical and normal to the microcracks.

the microcrack density S with strains for the different thicknesses of Cu films. It is demonstrated that S raises with the strain and the thinner the film the larger the S values, i.e., the higher the microcrack densities in the films at the same strain level, which is essentially similar to what most recently observed in polymer-supported brittle metal Ta films.[19] Hence it is very clear that the less ε_C values were obtained for the thinner films by extrapolating the S vs ε curves back to the point of $S=0$.

In Fig. 1(c), the SMD-determined and ER-determined ε_C are depicted with respect to the film thickness as circle dots and square dots, respectively. The SMD-determined ε_C is found to be very close to the ER-determined ε_C and both exhibit significant thickness dependence. This dependence, i.e., ε_C decreasing with film thickness decreasing, is just contrary to that of yield strength, similar to the general law that strength and ductility are mutually exclusive. The constraint effect of fine grains on the dislocation movability may be responsible for the present thickness dependence of both σ_y and ε_C values.

From Fig. 1(a), it could be further revealed that thinner films exhibit much more increase in $\Delta R/R_0$ than the thicker films at the same increment of the strain, especially in the second nonlinear stage. This is also related to the microstructural evolution. As seen in Fig. 2 where the respective microstructural evolution of 60-nm-, 340-nm-, and 700-nm-thick Cu films is typically presented, although all the three films show increasing microcracks with strain, denser microcracks or higher crack density is observed in the thinner films at any strain. Besides, the microcracks in the 60-nm-thick films are apt to traverse the entire gauge width of the specimens, while in the thicker films such as the 700-nm-thick films, the microcracks are not only much less but also much shorter even at the strain up to 20%. The lower hardening capability of the thinner films, caused by the less dislocation accumulation, should be responsible for the easy propagation of the microcracks.

In summary, we deposited submicron Cu films with a thickness range of 60–700 nm on compliant polyimide and investigated the thickness dependent critical strain by using both electrical resistance method and statistical microcrack density changes. The two methods yield similar results that the critical strain decreases as the thickness decreases, contrary to the dependence of yield strength on the film thickness. The two methods are applicable and feasible to determine the critical strain of supported metal films.

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