Tensile deformation of electroplated copper nanopillars

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First published on: 20 August 2010
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(Received 17 November 2009; final version received 26 June 2010)

The results are presented of uniaxial tensile testing of single crystalline electroplated copper nanopillars with diameters between 75 nm and 165 nm fabricated without the use of a focused ion beam (FIB). The experiments were performed in an in situ nanomechanical instrument, SEMentor, and reveal that the pillars' ultimate tensile strengths follow a similar power law dependence on diameter as reported for microcompression studies on fcc metals fabricated with and without FIB. Further, these pillars are characterized by limited or non-existent initial homogeneous deformation, immediately followed by necking in the top portion of the pillar. The particular deformation attributes are discussed in the context of hardening by dislocation starvation. Site-specific transmission electron microscopy microstructural analysis of as-fabricated nanopillars indicates the presence of scarce twin boundaries in some specimens. We comment on the potential for mechanical effects due to the presence of twins.

Keywords: copper; nanoscale plasticity; electroplating; single crystal; nanopillar; uniaxial tension; dislocation

1. Introduction

In the last five years, significant work has been performed on studying mechanical properties of materials at reduced dimensions through compression of cylindrical, micro- and nanopillars fabricated by the focused ion beam (FIB) [1–24]. A key advantage of this technique over other small-scale testing methodologies is the removal of strong strain gradients during mechanical testing. These experiments have been primarily focused on uniaxial compression of pillars with diameters ranging from ~250 nm up to several microns [1–4,6,8–10,12–15,17,19,21–25]. Reports of nano- and microscale tensile experiments, however, are significantly scarcer as they require custom-made instrumentation designed specifically for this purpose [7,11,13,14,26]. Similar to the compression experiments, the majority of the existing tensile experiments have also been performed on samples machined from bulk single crystals through the use of FIB. Significantly, all FIB-fabricated fcc metallic pillars exhibit nearly identical size dependent power-law strengthening with size in both compression and tension, the physical origin of which is still being actively pursued and discussed. Recently, the present authors reported that single crystalline

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electroplated Cu pillars, which have never been exposed to FIB, also obey the same power-law strength vs. size dependence as FIB-manufactured fcc pillars, and that this size dependence is a direct result of initial dislocation density and microstructure [27]. In this work, we present the results of tensile tests on 75–165 nm diameter single crystalline Cu samples with non-zero initial dislocation densities, fabricated by e-beam patterning followed by electroplating [28], a process nearly identical to the one used to create the compressive specimens in [27]. Through \textit{in situ} mechanical tests, we correlate physical deformation phenomena with the uniaxial stress–strain response. Transmission electron microscopy (TEM) analysis of pillar microstructure revealed the occasional twin boundary in some pillars. We comment on the geometry of the twin boundaries as well as the complexities of dislocation–twin boundary interactions.

2. Experimental

Tensile samples were fabricated by electroplating copper into the electron beam patterned holes in a PMMA (polymethyl methacrylate) matrix with intentional overplating to enable subsequent gripping of the pillar tops. Specific details of this fabrication method can be found in [28]. Unfortunately, these samples do not adhere well to the seed Au film initially evaporated on the Si substrate [28], and initial tests on these as-fabricated nanopillars resulted in the pillars fully detaching from the substrate at minute loads. Therefore, in order to perform successful tension tests, we effectively glued the pillar bottoms to the underlying film by using a focused electron (rather than ion) beam with a tungsten hexacarbonyl, W(CO)$_6$, source (FEI Nova 200) to locally deposit a thin layer of organometallic tungsten at the pillar–substrate interface, similar to the procedure described by Richter et al. [29]. TEM images of this e-beam deposited W confirm this coating to be amorphous (as evidenced by the diffuse rings in the diffraction pattern in Figure 7), suggesting its deformation mode to be of brittle character, without exhibiting any appreciable plasticity prior to fracture.

Figure 1a shows a scanning electron microscope (SEM) image of an as-fabricated 150 nm Cu pillar with nearly perfectly vertical sidewalls. As a result of tungsten deposition (Figure 1b), the bottom ~600 nm of the pillar becomes non-uniformly wider than its initial diameter due to this coating. In order to deposit the W, rectangular electron beam patterns (1 $\mu$m × 100 nm) were defined at 52° tilt in the SEM. The top of the rectangle was positioned several hundred nanometers away from the pillar base, and the deposition was performed at 0.54 nA and 10 kV in a FEI Nova 200. This procedure was then repeated on the opposite pillar side to ensure adequate adhesion. Tensile tests were performed in the SEMentor, an \textit{in situ} mechanical deformation instrument comprised of an SEM and the dynamic contact module (DCM) nanoindenter arm (Agilent Corp.) [14]. Whereas the nanoindenter is an intrinsically load controlled instrument, all of our experiments were performed at a nominally constant displacement rate through a custom-written, feedback loop based software method. In this work, both tension and compression tests were performed at nominal displacement rates of 1–2 nm/s. The diamond indenter tip was custom machined by FIB such that the tops of the copper pillars can be conformally
gripped during tensile testing (Figure 1c). Figure 1d shows a glued pillar in the SEMentor prior to testing, exemplifying the presence of W coating towards the pillar bottom and an unaffected top part of the pillar reflecting no added W in this region. A schematic of the tungsten glue geometry is shown in Figure 1e.

In order to evaluate whether the presence of W layer contributes to the measured tensile strength, we also performed compression tests on electroplated Cu pillars with and without the glue. The as-fabricated compressive pillars had aspect ratios between 3:1 and 6:1, whereas that for the glued pillars was closer to 3:1 after ~600 nm of length was subtracted from the overall height since plastic deformation occurred only in the non-coated section of the pillar. Contact stiffness was monitored throughout the experiments by utilizing a continuous stiffness measurement (CSM) option with the harmonic displacement amplitude of 1 nm at 75 Hz oscillation frequency in order to validate the quality of compression tests. Only those pillars for which contact stiffness matched theoretical prediction were included in the analysis. Recently, it has been shown that dynamic vs. static loading can have an effect on
deformation during nanoindentation tests [30], however we believe that this effect is marginal in these tensile experiments since the data under both conditions looks nearly identical. Samples for TEM microstructural analysis were prepared by the now-standard liftout procedures [31].

3. Results and discussion

Single crystalline copper nanopillars fabricated by the procedure described above are oriented such that their loading axis is close to \((111)\) orientation [28]. The locus of close-to-(111) orientations produced by this process is shown in the conventional stereographic triangle (Figure 1f) and illustrates that in tension, the slip planes will rotate towards the double slip condition [32]. As a result, the deformation path for these pillars is mixed: some pillars fail by double slip and necking; whereas others undergo single slip before necking. Figure 2 shows a progressive series of SEM images for uniaxial tension of two \(\sim150\) nm diameter representative nanopillars deforming via different mechanisms: single slip followed by necking (Figures 2a–2d) and by necking only (Figures 2f–2j). Their corresponding, annotated stress–strain curves are shown in Figures 2e and 2j, respectively. Figure 2c shows the pillar

Figure 2. Progressive snapshots and engineering stress–strain curves of two nanopillars during tensile experiments deforming via different mechanisms: single slip (a–e) and necking (f–j). (a) 155 nm pillar prior to loading with W deposited at the bottom, (b) the pillar immediately prior to slip, (c) the extent of deformation after the major burst, and (d) final extent of the plasticity. The annotated tensile curve (e) correlates each snapshot with its corresponding location in the stress–strain response. (f–j) A similar strain series is for a 164 nm pillar that deforms by nearly instantaneous necking. Noticeably, most of the plastic strain is accommodated by one major burst; subsequent strain of the deformed region is relatively small. These 150 nm pillars exhibit \(\sim30\)% engineering strain. The true strain in the neck of both of these pillars is \(\sim250\)%.
initially forming single slip offsets; however, after the initial single slip burst, these slip planes rotate towards the loading axes such that multiple slip is activated, as evidenced by the formation of a very thin, on the order of 35 nm, neck (Figure 2d). On the contrary, a 164 nm pillar undergoes minimal homogeneous extension, as evidenced by the instantaneous vertical displacement of the pillar head in favor of forming a neck, quickly reaching its final extension of 33% (Figures 2f–2i). We estimate true strain in the neck to be between 236% and 296% by utilizing the reduced area

\[ \varepsilon_T = \ln \left( \frac{A_i}{A_0} \right) \]

This somewhat large range for strain is a result of ambiguity in the neck diameter determination due to limited SEM image resolution at the working distance suitable for the mechanical experiments. Nevertheless, this localized deformation is very large and not surprising, as single crystalline pillars oriented for double slip have been shown to draw down to a point in the necked region resulting in very high local strains [14,33].

Figure 3 shows the pre- and post-deformation images of an 80 nm tension sample, as well as its engineering stress–strain curve. As with the larger pillar shown

![Figure 3](image)

**Figure 3.** (a) Pre- and (b) post-deformation SEM images of an 80 nm pillar (diameter measured in the top section as representing the site for subsequent neck formation). Inset in (b) shows a zoomed-in region of the neck. (c) Engineering stress–strain curve showing that the pillar reaches ultimate tensile strength (UTS) at \( \sim 1.7 \) GPa and then rapidly forms a neck, as evidenced by a pronounced strain burst.
in Figures 2e–2h, this 80 nm pillar necks instantaneously. During the initial loading up to \( \sim 1\% \) strain, the pillar ‘head’ slightly tilts to conform to the grips due to their less-than perfect initial contact. The remaining loading segment reflects nearly elastic response of the whole pillar. At 2.5\% engineering strain there is a small amount of plastic deformation followed by further loading until reaching the ultimate tensile strength (UTS) at 1730 MPa, where the top-most region of the pillar instantaneously necks. Immediately after this neck formation, the pillar was unloaded in order to preserve the neck region for subsequent visual analysis, as shown in the inset of Figure 3b.

The lack of homogeneous deformation alludes to the absence of strain hardening in these pillars. This can be shown through the necking criteria: 
\[
\frac{\Delta\sigma}{\sigma} \leq -\frac{dA}{A}
\]
which means that when the fractional increase in stress through strain hardening is less than or equal to the fractional decrease in the cross-sectional area, the pillar will form a neck [33]. When this condition is not satisfied the pillar will deform homogeneously. All of the pillars tested here show little to no homogeneous deformation prior to necking, a behavior very different from many previous tensile tests on FIB-fabricated single crystalline micro-tensile samples oriented for multiple slip [14]. In those experiments, necking occurred only in some cases, and only at significant strains of \( \sim 20–30\% \). This discrepancy is likely due to the predominantly micron- rather than nano-sized specimens used in previous reports, as they were limited by the ion beam resolution during FIB fabrication steps such that the smallest attainable pillar diameter was \( \sim 250 \) nm. Significantly, all pillars tested in this study are below 200 nm, where predominant plasticity mechanism is expected to be hardening through dislocation starvation or “mechanical annealing” [3,4,21]. In these nano-sized samples, the gliding dislocations have to travel only very short distances before annihilating at a free surface; furthermore, as the pillar necks the deforming diameter further decreases, thereby amplifying this phenomenon. Previous tensile studies specifically on copper micropillars oriented for single slip reported homogeneous deformation throughout their gauge length [7]. The smallest pillar diameters tested in that work, however, were 500 nm, significantly larger than the largest tension tests reported here, 165 nm.

It should also be noted that surface roughness, a source of local stress concentrations, may play a non-trivial role in a pillar’s ability to deform homogeneously. In order to maintain homogeneous plastic deformation, any stress concentrator must be neutralized through strain hardening, otherwise the pillar will neck at the stress concentration. A complete analysis requires a combined experimental and computational approach, which we are currently exploring.

Figure 4 shows engineering stress–strain plots for different initial pillar diameters. Each curve shares many similarities: after the initial loading segment, very limited homogeneous deformation takes place prior to reaching the UTS, followed by a single large burst. This burst corresponds either to the nearly instantaneous necking or to the initial single slip followed by slip plane rotation into a necking condition. Unlike larger pillars (Figure 2), which show extended deformation after the initial burst, during which the neck is further thinned, pillars with diameters of \( \sim 100 \) nm and smaller do not plastically deform after the initial burst (Figure 3). Instead, the initial strain burst represents the full extent of plastic deformation, with any subsequent increase in applied load resulting in immediate fracture in the neck.
In order to study the developed microstructure in the necked region, we made several attempts to arrest the tests on smaller pillars prior to their fracture, a challenging task since the neck is very fragile and tends to break soon after the major burst. Such a representative neck with the final diameter of $\sim 35$ nm prior to fracture is shown in the inset of Figure 3b. Studying plasticity in these very thin necks is appealing because at these length scales, surface effects begin to seriously affect several properties, including thermodynamic, i.e. melting temperature. Efforts focusing on investigating these effects are currently underway.

It is apparent from the figure that the initial loading has some variability, which we believe is a result of the imperfect initial alignment between the pillar ‘heads’ and the grips, generating small elastic bending and possibly torsional moments. However, those pillars that undergo either torsion or bending are easily identified during in situ experiments as revealed by the rotation of the anisotropic pillar head. Pillars with identified torsional or bending activity were excluded from the data. Small bending moments are frequently unavoidable due to the roughness of the pillar tops. Noticeably, after the settling process the loading slope is consistent across tensile tests. We are currently investigating ways to improve the smoothness of electroplated pillars.

The stress–strain curves in Figure 4 clearly show the presence of a size effect, where pillars with the smaller diameters yield at the higher UTS. To examine the size effect, we have plotted the tensile and compressive data from this work on a log–log graph in Figure 5. Here, the tension samples are marked by closed circles; whereas, compression pillars are open squares and triangles. The stresses for both types of deformation appear to obey a single power-law dependence with the exponent of $-0.63$, a value nearly identical to the previously reported one for electroplated copper pillars in compression only [27] and for all other FIB-machined fcc metals to date [22].

This plot also shows that there is no tension–compression asymmetry in the range of diameters tested (75 nm to 165 nm): both compression and tension samples of the
same diameter appear to deform at similar stresses, as expected for fcc metals. Of note is the difference in size between the smallest compression and tensile samples: 105 nm and 75 nm, respectively. This difference is due to the $\sim 1^\circ$ taper angle present in the smallest pillars. As plastic deformation in tension is dominated by localized necking, the diameter is recorded as the pre-deformation diameter at the site of necking; whereas, in compression, the average diameter along the gage length is used. Furthermore, compression tests were performed on pillars ~300–600 nm tall to avoid buckling; whereas, tensile samples were ~1150 nm tall, the height of the PMMA matrix. The associated taper angle causes the shorter compression samples to have slightly larger diameters than the taller, thinner tensile samples.

In order to identify any strengthening effects, the tungsten glue may cause in tensile tests, we compressed several samples that were glued to the substrate with the same procedure as for tensile testing and compared their strength with the as-fabricated compression samples. Recently Ngan et al. [34] used ion rather than electron beam deposition of a similar tungsten compound to completely coat Al micropillars with the purpose of trapping dislocations inside the specimens during compressive testing. They report an increase in strength by 50–300% depending on the micropillar diameter, as well as a change in the deformation signature: from discrete, burst-ridden curves to continuous, constantly-increasing stress–strain behavior.

Figure 6 shows the pre- and post-deformation images of a 155 nm diameter Cu pillar with glue at the bottom. The deformation behavior of these pillars is similar to that reported by Lee et al. [15] on the compression of single crystal Au pillars on stiff MgO substrates rather than to that of the coated pillars by Ng and Ngan [34]. As evidenced from the images, the bottom of the pillar is constrained by the W glue and remains undeformed, serving the role of a stiff substrate while the unconstrained top
deforms as typical for nanopillar deformation. We postulate that the presence of the W coating effectively raises the substrate and reduces the initial pillar height, as illustrated by the fiducial line in Figure 6. Figure 6c shows the representative true stress–true strain curves for 155 nm diameter pillars with and without the tungsten glue. Noticeably, the pillar strengths calculated at 10% flow strength are equivalent.

In Figure 5, the 150 nm compression samples are separated into open squares and open triangles reflecting those pillars with and without glue at the pillar substrate interface. The log–log plot shows that both glued and non-glued pillars sustain the same compressive flow stresses at 10% strain, as well as the UTS of 150 nm tensile pillars. Figure 6d shows the corrected contact stiffness as compared with the theoretical stiffness for a pillar with this geometry, showing very good agreement. The corrected curve is the measured stiffness of the pillar taking into account the Sneddon correction of the pillar itself acting as a flat punch indenter on the substrate [3]. The theoretical stiffness curve is determined by assuming the conservation of volume during plastic deformation with the height defined as the difference between the original pillar height and the extent of the glue, i.e. the plastically deforming region. The theoretical and corrected stiffnesses deviate from one another at the larger strains due to the top and bottom constraints. Perhaps most convincingly, unlike the stress–strain signature present in Ng and Ngan’s [34] coated pillars, the
compression pillars here with the glue at the interface exhibit no strain hardening and generally do not differ from the stochastic behavior of their non-glued counterparts. In Figure 6c, the unglued pillar shows a larger initial burst than the glued sample. This is not a characteristic feature of glued versus non-glued pillars, but rather is a result of the natural statistical sample-to-sample variation, as these differences are also present among the as-fabricated samples. We observed one pillar, with the aspect ratio below 3:1 above the glued region, exhibit hardening, however in this case, the entire pillar, including the region covered with glue deformed, and a stress–strain curve similar to that seen by Ng and Ngan was observed.

TEM analysis has previously revealed that most electroplated samples are single crystalline; however, we have also observed the presence of nanoscale twins. Figure 7 shows several dark field images of a pillar containing a few twins. The diffraction pattern in Figure 7d suggests that this pillar has coherent twin boundaries (CTBs)

![Figure 7](image)

Figure 7. (a–c) Dark field TEM images of a Cu nanopillar clearly showing twin boundaries. (d) Diffraction pattern associated with dark field images in (a–c) indicating the presence of coherent twin boundaries across \{111\} planes. The protective layer around the Cu pillar is an amorphous W layer; it was deposited in the same manner as is used to glue pillars to the substrate. The diffuse rings in the diffraction pattern show the lack of long-range structure and, therefore, amorphous nature of W layer.
across the \{111\} planes, which are also dislocation glide planes \cite{35}. Recently, CTBs in copper thin films have become an interesting topic due to their ability to increase both ductility and strength of the metal \cite{36}. The structure of the twins observed here is different than the twins in Cu films as they are stacked along the \{111\} growth direction in the latter, such that most of the twin boundaries are perpendicular to the growth direction. The twin boundaries in the pillars, as evidenced by Figure 7, exist on the inclined \{111\} planes rather than only those orthogonal to the loading direction. This variation of the CTB inclination with respect to the loading direction may have important consequences for the dislocation reactions at these CTBs, especially in the presence of nearby free surfaces.

Several atomistic simulations have begun to identify the complex reactions between dislocations and CTBs in order to describe the increased strengths of nano-twinned metals. Zhu et al., for example, used simulations based on transition rate theory to evaluate the athermal stress for transmission and absorption of a screw dislocation at a CTB and found that the required stress is dependent on the dislocation storage inside the twin boundary \cite{37}. Furthermore, several MD simulations have been performed to evaluate the complex dislocation reactions with twin boundaries in both bulk \cite{38-40} and in nanopillar geometries \cite{41-44} and demonstrated that dislocation reactions are dependent on the dislocation character as well as on the loading condition (pure shear versus tension). Image stress calculations for a twin boundary in Cu showed that a perfect screw dislocation is repelled by the twin boundary \cite{45} suggesting that dislocations in nanopillars containing twin boundaries may be even more attracted to the free surface, amplifying the tendency for dislocation starvation. We are currently engaged in the analysis of the mechanical effects of CTBs in copper nanopillars, which will be reported in a separate manuscript.

4. Conclusions

We have reported the results of \textit{in situ} uniaxial tensile tests on \sim\{111\}-oriented single crystalline copper nanopillars fabricated without the use of the focused ion beam. In order to perform these tests, we developed a procedure to ‘glue’ the samples to the substrate with a W compound. We have compared the ultimate tensile strengths (UTS) with the compressive strengths of nanopillars with and without the same glue and conclude that the glue process does not substantially contribute to pillar strength. The tensile behavior is characterized by limited homogeneous deformation prior to reaching UTS, followed by a plastic burst. We attribute this lack of homogeneous ductility to the annihilation of dislocations at free surfaces before they have the opportunity to interact. We report size-dependent strength, which scales in a nearly identical power-law fashion with the sample diameter as previously reported size effects in both electroplated and FIB machined face-centered cubic pillars. Whereas most pillars tested are single crystalline, dark field TEM images reveal the presence of several twins in some samples. Efforts are currently underway to investigate the effects of twin boundaries in nanopillars on their deformation behavior and strength.
Acknowledgements
The authors gratefully acknowledge the financial support of the NSF CAREER award (DMR-0748267). We also thank J.-Y. Kim for tip fabrication and discussions, M.J. Burek for electroplating materials and helpful discussions, and C.M. Garland for TEM discussions and assistance.

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