Modelling creep induced by internal stresses in freestanding submicron Cu film

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Abstract. In a lab-on-chip experiment, lithography and selective chemical etching are used to pattern microscopic tensile test samples within a thin metallic layer hosting large internal stresses. After partial release of the layer from the substrate on which it was deposited, the free-standing beam-like structures are stretched by the actuator to which they are connected. The lab-on-chip also comprises cantilever beams which shorten freely upon release from the substrate. Experimental observations of both the instantaneous and the delayed deformations in a 170 nm thick copper film were simulated using a theoretical model. The model properly reproduced the experiments only when accounting for both plasticity and significant kinematic hardening occurring already during the deposition of the polycrystalline film. Once released from the substrate, cantilever beams contracted well beyond the elastic range because the amplitudes of back-stresses were sufficient to cause reverse plastic yielding. Large tensile stresses inside the actuated beams led to delayed uniform elongations (creep) exceeding 16%. Such values are much larger than the uniform strain of 5-6% that was observed in beams that underwent necking as soon as the film was released from the substrate, i.e. directly after etching of the sacrificial layer.

1. Introduction
Reducing the grain size of metallic alloys contributes to increasing not only their strength, but also the heterogeneity of internal stresses and the strain rate sensitivity [1, 2, 3, 4]. Under tensile loads, rate-dependent viscoplastic processes, which are promoted among others in grain boundary regions [5, 6], tend to postpone necking and they may hence impede that the gain in strength results in significant loss of ductility [7]. Under cyclic loading, nanocrystalline alloys undergo elastic-plastic transients which demonstrate large kinematic hardening: backstresses due to dislocation pile ups cause early yielding upon load reversal [8, 9].

Mechanical testing of submicron-thick freestanding films [10, 11, 12, 13, 14, 15] allows a rather convenient characterization of the latter phenomena. The experimental results reported here were obtained using a testing method specifically designed in order to probe the creep/relaxation response of e-beam evaporated metallic films. Such a lab-on-chip technique, which is illustrated in figure 1 and described in Section 2, has already been applied to aluminum [16], palladium [17] and copper [18, 19]. Over the years, the technique has been improved for instance by annealing of the deposited films prior to release in order to tune the amplitude of internal stresses [20]. However, it is an early experiment, dating from 2014, that is revisited here because it demonstrated phenomena which ceased to operate with the new lab-on-chip designs.
Figure 1. Principle of the lab-on-chip experiment: (a) patterned tensile test structure still tied to the substrate; (b) partial release of the Cu film by chemical etching of the sacrificial layer; (c) unconstrained shortening of a cantilever beam after release; (d) Adjacent tensile test structures with (tapered) actuators of increasing lengths ensuring that the amplitude of post-release elongation of the narrow beams also increases.

In recent on-chip experiments, the ultimate tensile strength of the test material is much lower than the strength of the actuators which are made of silicon nitride, brittle at more than 5 Gpa. As a result, the actuator behaves elastically and interpretation of the measurements is rather straightforward [20]. In the experiment considered here, actuators were patterned directly in the Cu film that was tested. Both types of structural elements hence had the same (a priori unknown) non-linear elastic-viscoplastic response. The present contribution shows how to determine the latter material response by inverse modeling of such experiments, in which the post-release creep is dominated by significant backstress levels. The parameters of the viscoplastic model are identified iteratively, by repeated computational simulations aiming to reproduce the experimentally observed deformations of a large number of tensile test structures within the lab-on-chip. The theoretical model is described in Section 3 and a comparison of numerical predictions to experimental data is discussed in Section 4.

2. Description and main results of the lab-on-chip experiment
The fabrication of the lab-on-chip proceeded as follows, using a single mask. The first deposited layer was 1 µm thick and covered the whole surface of the silicon substrate. Such sacrificial layer consisted of a plasma enhanced chemical vapor deposited (PECVD) silicon dioxide layer. The precursor gases were composed of 100 sccm of SiH₄, 700 sccm of N₂O and 300 sccm of N₂. After deposition, the wafer was densified at 800°C for 20 min and then covered with a negative photoresist (AZ5214) allowing patterning by UV light. Copper was then deposited on
the patterned substrate at room temperature, by e-beam evaporation at a pressure of 0.1 mTorr and a deposition rate of 0.1 nm/s. The photoresist was then removed in acetone (lift-off) revealing copper tests structures still bounded to the sacrificial layer. The thickness of the Cu film was measured to be $170 \pm 10$ nm by profilometry.

The test structures were released by wet etching of the sacrificial layer using hydrofluoric acid (HF 73%), which ensures a high etching selectivity between PECVD SiO$_2$ and the Cu layer. This led to an etching rate of $0.181 \mu$m/s. As the release was performed in liquid, supercritical drying was performed to avoid sticking of the freestanding structures to the substrate underneath.

Once released, the cantilever beams shortened whereas the narrow beam of every auto-actuated tensile test structure was stretched. Such changes of lengths of the various beams were probed in the SEM by measuring the relative positions of two cursors which faced one another before etching of the sacrificial layer (figure 1a) but were then shifted due to the change of lengths of the beams (figure 1a and figure 2). Cursors patterned in regions of the Cu film still anchored to the substrate were considered to hold fixed positions. The displacements of the shifted cursors were measured using high magnification micrographs from a field emission gun scanning electron microscope (FEG-SEM). Displacements were measured with an accuracy of about 50 nm, limited by the photolithography process rather than by the image resolution achieved by FEG-SEM. The first observations of the deformed lab-on-chip were performed 7 hours after release. In order to probe not only the instantaneous, but also the delayed, response of tensile tests structures undergoing creep, electron microscopy observations of the lab-on-chip were performed 25 times, scheduled from one day up to four months after the release.

In the tensile test structures, the constant width of the beam was 1.5 $\mu$m, whereas the actuator had a tapered shape with minimum and maximum widths of 7.5 and 11.5 $\mu$m, respectively. The length of the narrow beam was systematically equal to 25 $\mu$m, whereas the length of the actuator varied from one structure to the next, ranging from 100 to 1200 $\mu$m. The initial lengths of freely contracting cantilever beams ranged from 100 to 400 $\mu$m.

Before the release, such patterned Cu films host uniform equibiaxial tensile stresses of unknown amplitude which, in the absence of an annealing treatment, vary a lot depending on the deposition conditions and the delay before release. After the release, equilibrium of axial force in the tensile test structures imposes that the stress in the narrow beam be 6.4 times larger than the average stress in the actuator (due to the different cross-section areas). This means that the actuator contracts and unloads elastically (figure 1b), whereas the beam undergoes plastic elongation. Its strain hardening causes tensile stresses much larger than the amplitude of the initial internal stress. During elastic unloading of the actuator, the change of length (measured by the cursor displacement) increases proportionally to the actuator length. Hence, mechanical equilibrium is reached for a cursor displacement, i.e. for an elongation of the test beam, that increases with the actuator length. On the other hand, as the unloading of the cantilever beams is unconstrained (figure 1c), their contraction is directly proportional to their length.

In the experiment on the 170 nm thick Cu film considered here, the instantaneous unconstrained contraction of cantilever beams was 0.4%. Necking and failure of the test beam were observed whenever the actuator was longer than 850 $\mu$m. In the measurements performed right after release, the largest unbroken beam was stretched by 5.1%. We may thus assume that the (yet unknown) ultimate tensile stress was reached for a uniform elongation lower than 6%.

Figure 3 shows the creep responses of four tensile test structures in which the actuator length was 100, 200, 400 and 800 $\mu$m, respectively. During the four months of measurements, the displacement $\Delta u$ of the cursor was found to increase linearly with time. Hence, the creep rate was constant. After four months, among the unbroken beams, the largest elongation among all actuated test beams was 16%. Surprisingly, free beams underwent creep strains too. After 1750 hours (73 days), free beams had contracted by -2.2% (whereas the largest contraction of an actuator with a test beam attached to it was -1.3%).
Figure 2. Micrographs showing two regions of the lab-on-chip three months after release. In the tensile test structures shown in (a), the actuator initial length increases from left to right and the test beam on the right underwent necking and failure. The initial lengths of the cantilever beams shown in (b) were 100, 200 and 300 $\mu$m, respectively. (c) Close-up of two cursors used to measure a beam contraction.

3. Description and main results of the model
The theoretical analysis is greatly simplified if we assume that all beams are subjected to a uniaxial stress once released from the substrate and if the deformation of each actuator is considered uniform (in spite of its tapered shape). Indeed, under uniaxial tension a scalar description of the stress-strain relationship suffices.

In reality, a uniaxial stress state is valid only when the beams are fully released from the substrate. Until full release, there is a transition from the equibiaxial stress state (resulting from the film deposition) to the uniaxial situation. Elastic unloading of the in-plane transverse stress causes a decrease of the axial stress in all tensile test structures. The latter axial stress is multiplied by $1 - \nu$ where $\nu = 0.3$ is the Poisson ratio of copper. As etching of the sacrificial layer underneath the beams starts from the edges, the central axis of the beams is released last and it is likely that the in-plane transverse stress is largely relaxed when the beam starts to deform along the longitudinal direction.

It is further assumed that the internal stresses were sufficiently large to induce plasticity (and kinematic hardening) already during the film deposition. Otherwise the amplitude of contraction of the free beams would not exceed that expected from elastic unloading. However, such pre-deformation is not modeled here. The initial state, just before the release, is characterized by a yield stress $\sigma_{y0}$ and a positive backstress $\zeta_0$ which are determined by inverse modelling.
Figure 3. Illustration of the delayed deformation of tensile test structures by the progressive shortening of actuators having lengths $L^a$ equal to 100, 200, 400 and 800 $\mu$m. For comparison, free beams underwent an instantaneous contraction of -0.004 and a delayed contraction of -0.022 after 1750 h.

3.1. Simulation of the instantaneous deformation

The instantaneous response, observed immediately after etching, is considered elastic-plastic in both the actuator and the stretched test beam (denoted, respectively, with superscripts $a$ and $b$). As the total length $L^a + L^b$ of the tensile test structure remains constant, the cursor displacement $\Delta u$ defines the axial strains of both components: $\Delta \varepsilon^a = -\Delta u/L^a$ and $\Delta \varepsilon^b = \ln(1 + \Delta u/L^b)$.

In the model, $\Delta u$ is computed by enforcing that $\sigma^b = 6.4 \sigma^a$, which ensures force equilibrium (Section 2). The value of $\Delta u$ is computed iteratively, leading to a different result in every tensile test structure since the actuator lengths $L^a$ are different. The response is linear elastic if

$\sigma^a = (1 - \nu) (\zeta_0 + \sigma_{y0}) + E \Delta \varepsilon^a > \zeta_0 - \sigma_{y0}$ in the actuator

$\sigma^b = (1 - \nu) (\zeta_0 + \sigma_{y0}) + E \Delta \varepsilon^b < \zeta_0 + \sigma_{y0}$ in the test beam

where $E$ is the Young modulus. Otherwise, plastic yielding occurs and the increments of equivalent plastic strain $\Delta p^a$ and $\Delta p^b$ must fulfill the following conditions:

$\sigma^a = (1 - \nu) (\zeta_0 + \sigma_{y0}) + E (\Delta \varepsilon^a + \Delta p^a) = \zeta^a (\Delta p^a) - \sigma_{y} (\Delta p^a)$ in the actuator

$\sigma^b = (1 - \nu) (\zeta_0 + \sigma_{y0}) + E (\Delta \varepsilon^b - \Delta p^b) = \zeta^b (\Delta p^b) + \sigma_{y} (\Delta p^b)$ in the test beam

Experimental measurements are best reproduced by relying on the following representation of strain hardening:

$\sigma_{y}(\Delta p \leq p_l) = \sigma_{y0} + H\Delta p$

$\sigma_{y}(\Delta p > p_l) = \sigma_{sat} - \left(\sigma_{sat} - \sigma_{y0} - Hp_l\right) \exp\left(\frac{p_l - \Delta p}{p_s}\right)$
This implies that the hardening rate $H$ is constant until $\Delta p = p_l$, after which it decreases until the yield stress reaches its saturation value $\sigma_{sat}$. Setting $p_s = (\sigma_{sat} - \sigma_0)/H - p_l$ ensures that the hardening rate decreases continuously.

The backstress evolves differently in the two components of the tensile test structure. Indeed, $\zeta^a$ undergoes a fast decay because yielding switches from elongation (before the release) to contraction. On the other hand, stretching of the beam causes $\zeta^b$ to increase beyond the initial value $\zeta_0$:

$$\zeta^a(\Delta p^a) = \zeta_0 - H \zeta \Delta p^a$$

$$\zeta^b(\Delta p^b) = \zeta_{sat} - (\zeta_{sat} - \zeta_0) \exp\left(-\frac{\Delta p^b}{p_s}\right)$$

(4)

The selected values of the various hardening parameters are listed in table 1. They ensure that the free contraction of cantilever beams is -0.4%, as observed experimentally, and also that the onset of necking of the stretched beam occurs at $\Delta p^b = 6\%$ according to the Consid`ere criterion.

| Table 1. Fitted values of the parameters of the elastic-plastic model |
|------------------------|--------|--------|--------|--------|--------|--------|--------|
| $\sigma_y0$            | 86 MPa | $H$    | 7 GPa  | $p_l$  | 0.028  | $p_s$  | 0.0126 |
| $\sigma_{sat}$         | 370 MPa| $\zeta_0$| 207 MPa| $H \zeta$| 54 GPa | $p \zeta$| 0.01   |
| $\zeta_{sat}$          | 222 MPa|

Figure 4 shows that the model closely reproduces the instantaneous contraction of the actuators (which was probed 7 h after release). The predicted stress strain curve of stretched beams is shown in figure 5a. For comparison, figure 5b shows the stress-strain curve that would be predicted if, instead of relying on the elastic-plastic model, one would consider that the unloading of all actuators is purely elastic, which would imply that the initial backstress $\zeta_0$ is much lower. The latter prediction of the Cu film response is unrealistic because the hardening of stretched beams is excessive, postponing necking and failure later than $\Delta p^b \simeq 6\%$.

### 3.2. Simulation of the delayed deformation

According to figure 3, the post-release displacement of the cursor occurs at constant velocity. This can be reproduced by the theoretical model if the behavior of the Cu film is considered viscoplastic. Inside the tensile test structures, the stress ratio $\sigma^b / \sigma^a$ increases beyond the initial value of 6.4 since the cross-sections of stretched beams decrease and the opposite applies within axially contracting actuators. As the response is highly viscous, large variations of the axial stress are impeded and the delayed deformation must be driven by constant creep rates inside the actuators. For simplicity, the present model considers that both the yield stress and the backstress hold constant values inside every actuator. The viscoplastic creep rate, corresponding to the slope of the linear creep strains reported in figure 3, is computed using the following power law relationship:

$$\dot{\varepsilon}_{vp}^a = \frac{-\dot{u}}{L^a} = -\dot{\gamma}_0 \left| \sigma^a - \zeta^a(\Delta p^a) \right|^{1/m} = -\dot{\gamma}_0 \left( \sigma_y(\Delta p^a) \right)^{1/m}$$

(5)

Predicted creep rates correspond best to experimental observations when $\dot{\gamma}_0 = 2.74 \times 10^{-32}/h$ and $m = 0.075$. The creep strains predicted within 24 actuators with different lengths are compared to experiments in figure 4.
Figure 4. Assessment the model prediction of the shortening of actuators resulting from the instantaneous deformation (probed 7 h after release) and the delayed deformation (probed 1750 h after release). The delayed viscoplastic response depends on the yield stress after release which is shown too.

The viscoplastic deformation of free beams is likely to be described by the same law since the only difference with regard to the actuators is that free beams underwent larger instantaneous contraction and hence larger reverse plastic yielding, leading to $\sigma_{fb}^y = 102$ MPa after release. The power law predicts that, after 1750 h, free beams had contracted by -3.4% whereas the experimentally measured value is only -2.2%. Such a discrepancy might be due to the fact that free beams tend to deflect and may undergo friction if they are in contact with the substrate.

4. Discussion

According to the inverse modeling of the lab-on-chip experiment, the amplitude of internal stresses prior to release is 293 MPa. This is in the lower range of the levels of internal stresses (300-450 MPa) which were deduced from the wafer curvature (Stoney method) after annealing of e-beam evaporated copper [20]. However, experience has shown that internal stresses may vary significantly after deposition unless the microstructure is stabilized by annealing.

Considering that the grain size is of the order of the film thickness (170 nm), the predicted value of the ultimate tensile stress (570 MPa) also is realistic. It compares well to previous investigations on free-standing Cu films with comparable thicknesses [8, 20].

The amplitude of contraction of both the free beams and the actuators is much larger than the amplitude expected in case of linear elastic unloading. The fact that the contraction continues at a constant rate long after the release can be explained only in the case of a viscoplastic process driven by the heterogeneity of internal stresses, which is commonly referred to as kinematic hardening. The backstress developed before the release which means that plastic yielding
ocurred already during the film deposition. Similar observations of reverse creep induced by a strong kinematic hardening were reported in other nanocrystalline films [8, 9].

In the present analysis, it was assumed that the strengthening of the beams stretched after release was due to an increase of the yield stress $\sigma^b_y$, whereas the backstress $\zeta^b_y$ rapidly reached a saturation value slightly larger than the initial backstress $\zeta_0$. In reality, the experiment does not allow determining which proportion of the axial stress should be considered a backstress. If the heterogeneity of internal stresses increases during the instantaneous stretching of the beams, the backstress also increases. Probing kinematic hardening in thin films is possible using other experimental set ups in which the material undergoes cyclic loading [8, 10].

Tensile test samples subjected to an instantaneous elongation of more than 5% underwent necking upon release, but a much larger elongation was achieved during creep. It is well known that viscous processes delay plastic instability [21, 22] and hence increase ductility. When relying on a viscoplastic power law, necking under uniaxial tension occurs after a uniform strain $\varepsilon_u = m \ln(f^{1/m} - 1)$ where $f$ represents the reduction of cross-sectional area due to an initial imperfection. Using $f = 0.99$ and $m = 0.075$ (as identified by inverse modeling), a uniform strain $\varepsilon_u = 15.5\%$ is obtained. This corresponds to the maximum strain measured in the stretched beams, whereas many of the stretched beams underwent necking at lower amplitudes of the creep strain, presumably due to larger initial imperfections.

In a previous study involving lab-on-chip experiments on free-standing Pd films with different thicknesses [23], the influence of grain size on the heterogeneity of internal stresses was investigated using a microscopic model accounting for the crystalline anisotropy of individual grains and for thermally-activated dislocation glide which reproduced the observed creep rates. Adapting the model to the Cu film investigated here is under progress.
5. Conclusion
When a patterned submicron thick Cu film is released from the substrate on which it was deposited, both the instantaneous (elastic-plastic) and the delayed (viscoplastic) responses are strongly influenced by the amplitude and the heterogeneity of the internal stresses resulting from film deposition. In a lab-on-chip experiment, some of the patterned beam-like elements contract whereas others are stretched by an actuator. In the experiment investigated here, the amplitude of the experimentally observed contractions was much larger than that expected from a purely elastic unloading of the initial biaxial tensile stress. Moreover most of the shortening of the beams occurred at constant rate for up to four months after film release, which is characteristic of a viscoplastic process driven by internal stresses. The same viscoplastic process was active in stretched beams, which demonstrated unexpectedly high levels of ductility.

Most of the experimental observations were properly reproduced by the theoretical model and the identification of parameter values by an inverse modeling analysis led to realistic predictions of the strength and strain rate sensitivity of the Cu film.

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