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Size effects on intergranular crack growth mechanisms in ultrathin nanocrystalline gold free-standing films

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ABSTRACT

This study investigated the combined effects of thickness (30 vs 100 nm) and average grain size (40 vs 70 nm for the thicker films) on the crack propagation mechanisms in ultrathin nanocrystalline gold microbeams, using a microelectromechanical system device to perform in situ transmission electron microscope (TEM) tensile experiments. Monotonic tensile tests of the two types of microbeams show similar strength levels (~400 MPa) and ductility (~2%). However, the thicker specimens exhibit a much more ductile behavior under repeated stress relaxation experiments, which the in situ TEM experiments revealed to be related to differences in intergranular crack propagation mechanisms. The governing crack growth process is in both cases dominated by grain boundary dislocation activities leading to grain boundary sliding. For the thinner specimens, secondary nanocracks are generated (as a result of grain boundary sliding) ahead of the main crack and coalesce together. Instead, secondary nanocracks do not form ahead of the main crack for the thicker specimens; the main crack extends as a result of sustained grain boundary sliding at the crack tip.

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1. Introduction

The understanding of plastic deformation mechanisms in nanocrystalline (nc) metals and the related grain size effects has greatly improved over the last twenty years [1–5] but many aspects remain controversial. For example, atomistic simulations can predict a material-dependent grain size below which transgranular dislocation activities become negligible compared to grain boundary (GB) mediated processes [6] while some experiments have confirmed [7] or inferred [8] such transition for very small grains. In situ experiments can also quantify or identify the respective role of each mechanism in monotonic deformation [9] or even dynamic recovery [10]. In comparison, the mechanisms responsible for cracking in nc metals are not as well understood, due in part to the complications associated with the increased stress field near the crack tip. Atomistic simulations on nc Ni (grain sizes ranging from 5 to 10–12 nm) revealed that the amplified stresses ahead of the crack tip trigger transgranular dislocations that would otherwise not occur in such small grains [11,12]. Artificial or natural stress concentrators such as notches and cracks in free standing nc films also demonstrated stress-assisted grain growth at room temperature [13–15]. This effect was also encountered in larger nc Ni specimens [16]. The large stresses also result in grain boundary decohesion and nano-void formation that may coalesce with the main crack, leading to intergranular fracture (in contrast to the typical transgranular cracking in coarse-grained metals) [11,12]. However, simulations on nc Al (grain size: 5 nm) ahead of a “blunt” crack (or nano-notch, with a root radius of 5 nm) highlighted the role of grain rotation (similar to the Ashby-Verral mechanism [17]) ahead of the notch, which resulted in a homogeneous (gradient free) stress field and led to the conclusion that nc metals may be insensitive to stress concentration [18]. Experimentally, grain rotation near a crack tip in nc Al has been observed at room temperature [19], and then refuted [20]. Explored in a more quantitative way recently, it was found that grain rotation is a mechanism that participates to plastic deformation in nc Al thin films, but is not the main stress-relaxation mechanism [21]. From these studies, stress-assisted grain growth (probably carried out by shear-coupled migration [22–25]) appear as the dominant mechanism of stress relaxation in nc metals [15], while grain rotation only accommodates local grain incompatibilities. Such discrepancies highlight the challenges in capturing the governing mechanisms in nc metals and especially those associated with crack

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growth.

A few experimental studies have been carried out to specifically unveil the crack propagation mechanisms using in situ TEM observations on ultrathin films (<200 nm) [26–35]. These studies may not be representative of bulk nc metals, due to the presence of free surfaces, and other thin film effects such as a lower confinement of the grains [29,36]. Nonetheless, in situ TEM experiments provide a unique opportunity to observe the deformation mechanisms at the crack tip and directly compare them with atomistic simulations, and are relevant for assessing the reliability of ultrathin films used as structural components in micro/nano-electromechanical systems (MEMS/NEMS) and other emerging applications [37]. Most of these in situ TEM observations revealed intergranular crack growth via nucleation and growth of nanovoids at triple junctions and/or grain boundary nanocracks, and their coalescence to the main crack, as predicted by the simulations of Farkas et al. [11,12], even though the experimental grain sizes are significantly larger (electroplated Ni with an average grain size of 30 nm and narrow grain size distribution (unknown thickness) [29], 100-nm-thick pulsed laser deposited Ni with grain size ranging from 5 to 30 nm [27], 125-nm-thick sputter-deposited Al with grains ranging from 35 to 420 nm (average grain size: 130 nm) [26], and 60-nm-thick sputter-deposited Ag with grains ranging from 10 to 120 nm (average grain size: 40 nm) [35]). These studies mainly observed transgranular dislocation activities that led to the coalescence events, sometimes due to necking of the connecting regions. Most studies did not observe GB dislocations or grain rotation in the cracking process [26,27,29]. In the present study, we clearly show that intergranular crack growth mechanisms rely on GB dislocations. We further demonstrate that the GB dislocation mechanisms result in a cracking process that depends on grain and film size, with nanocracks developing favorably ahead of the main crack in thinner films where the average grain size is also smaller.

2. Experimental methods

2.1. Specimen fabrication and characterization

Fig. 1(a) shows the dog bone-shaped Au thin film specimens that were used in this study. The same batch fabrication process was employed to fabricate both types of specimen, the only difference being the thickness of the deposited Au layer. First, high resolution photo-lithography was performed with a negative resist deposited being the thickness of the deposited Au layer. First, high resolution photo-lithography was performed with a negative resist deposited on each side of the specimen gap, and a load sensor. A heat sink, located next to the actuator, help minimizing the temperature increase near the specimen to less than 10 °C [44]. All these components are fabricated over a through-wafer window, thereby allowing TEM imaging. The first capacitive sensor, CS1, is rigidly connected to the thermal actuator (i.e., their displacements X are equal, XCS1 = XA) using non-conductive epoxy glue which electrically isolates the thermal actuator from the capacitive sensors. The second capacitive sensor, CS2, is rigidly connected to the load sensor (XCS2 = XLS), and is identical to CS1. The load sensor is made of 4 beams (length: 500 μm) deforming in bending (stiffness, KLS, of 100 or 480 N/m depending on the beams width [44]). The specimen gap is ~5 μm, and consists of two adjacent large areas for clamping of the specimen onto the MEMS device. The specimen is clamped to the device using UV curable glue, as shown in Fig. 2(d), and a micromanipulator (see details in Refs. [40–42]).

A test consists of applying a displacement XA by controlling VIn, resulting in the elongation of the specimen (XS) and the displacement of the load sensor (XLS). The applied force is given by:

\[
F = K_{LS}X_{LS} = K_{LS}X_{CS2}
\]  

(1)

while the elongation of the specimen, Xs, is obtained via the difference between the displacement of the two capacitive sensors:

\[
X_s = X_A - X_{LS} = X_{CS1} - X_{CS2}
\]  

(2)

The applied stress is obtained by dividing F by the specimen’s cross sectional area, and the strain by dividing Xs by the specimen’s gauge length.

2.2. Ex situ tests

The tensile stress-strain curve were obtained for both types of specimens by performing ex situ tests. The change in capacitance of sensors CS1 and CS2, ΔCS1 and ΔCS2, were independently measured.
using commercially available CMOS chips MS3110 [40,45]. These chips provide an output voltage $V_{out}$ proportional to the difference between two input capacitances: $C_S^1$ (or $C_S^2$), and a constant capacitor. The change in capacitance, $\Delta C$, is given by:

$$\Delta C_1 = \frac{V_{out}(V_{in} = 0) - V_{out}(V_{in})}{\lambda_a}$$

$$\Delta C_2 = \frac{V_{out}(V_{in} = 0) - V_{out}(V_{in})}{\lambda_b}$$

where the superscripts $a$ and $b$ refer to the two MS3110s respectively, $\lambda_a$ and $\lambda_b$ are the proportionality constants of the two MS3110s. $V_{out}$ is the averaged value of $V_{out}$ over measurement durations ranging from 500 ms to 2s at an acquisition rate of 1 kHz. The displacement $X_{CS1}$ or $X_{CS2}$ (and therefore stress and strain using Eqs. (1) and (2)) is then given by:

$$\Delta C = a \varepsilon_0 \varepsilon_0 A \left[ \frac{1}{d_1} + \frac{1}{d_2} + \frac{1}{X} \right] - \left[ \frac{1}{d_1} + \frac{1}{d_2} \right]$$

where $a$ is the calibration constant, $k$ is the relative permittivity of air ($k=1$), $\varepsilon_0 = 8.854 \times 10^{-12} \text{ Fm}^{-1}$ is the permittivity of free space, $n (= 42)$ is the number of comb structures, $A$ is the overlapping area of the comb structure, $d_1$ (nominal value: 2.5 $\mu$m) and $d_2$ (nominal value: 10.5 $\mu$m) are the initial gaps between the comb structures which are actually measured using SEM images of the device typically after or before the test. The capacitive sensors are calibrated before each test to determine the calibration constant $a$, using the procedure explained in Ref. [40] consisting of optically
measuring the displacement $X_A$ at a large $V_{in}$ while measuring the corresponding $\Delta CS$.

2.2.3. In situ TEM tests

In situ TEM experiments were carried out inside a JEOL 2010 transmission electron microscope operated at 200 kV in order to observe the crack growth mechanisms. The MEMS was glued into a specimen carrier fitted to an electrical biasing holder (see Fig. 2(c)), allowing actuation of the MEMS by applying $V_{in}$. Unfortunately, the electronic sensing was not operational for these experiments. Hence $X_A$ and $X_{LS}$ (and therefore $X_S$) could not be measured electronically. Instead, in a few instances, the elongation of the specimen $X_S$ and $X_{LS}$ could be measured based on TEM images. Otherwise, for a given $V_{in}$, $X_A$ was approximated as following, in order to give an estimate of the total machine’s displacement. The MEMS device was first calibrated in the absence of a specimen, by measuring $X_A$ in the TEM for a given $V_{in}$. Under an applied force (i.e., when there is a specimen being pulled), $X_A$ can be approximated as following [46]:

$$X_A = \left( \frac{1}{1 + \frac{K_S}{K_A}} + \frac{1}{1 + \frac{K_S}{K_{LS}}} \right) X_{F=0}^A$$  \hspace{1cm} (6)

where $K_S$ is the stiffness of the specimen that could be approximated using the nominal elastic modulus of gold (~75 GPa) and geometric dimensions of the sample ($K_S = 2000$ N/m for the 100-nm-thick specimens and 610 for the 30-nm-thick specimens), $K_A$ (3760 N/m) is the stiffness of the thermal actuator [44] and $K_{LS}$ (480 N/m) is the load sensor stiffness. Using these numerical values, Eq. (6) gives: $X_A = 0.9 X_{F=0}^A$ (respectively $0.93 X_{F=0}^A$) for the 100-nm-thick (resp. 30-nm-thick) specimens.

3. Results and discussion

3.1. Stress-strain curve and fracture surface comparison

Fig. 3 shows the stress-strain curves to failure of two 30- and 100-nm-thick specimens, using the procedure described in Section 2.2.1 and 2.2.2 (ex situ tests). The strain rate was $4.7 \times 10^{-4}$ s$^{-1}$ (resp. $9.4 \times 10^{-4}$ s$^{-1}$) and the free-standing gauge length was 18.6 µm (resp. 18.2 µm) for the 100-nm-thick specimen (resp. 30-nm-thick). The 30-nm-thick specimen was first tested to a plastic...
strain of 2.2%, then unloaded. During the unloading, the specimen buckled due to its small cross-section. During the reloading portion, the stress is therefore 0 for the first 2.2%. The apparent elastic moduli, $E$, for both specimens are 42 (100-nm-thick) and 47 GPa (30-nm-thick). These low values (reported $E$ values for Au thin films are in the 50–60 GPa range, while $E$ for bulk polycrystalline Au is ~80 GPa) have been showed to be related to the finite compliance of the clamps (using finite element models) along the fillet region of the specimens and along the section of the gauge length that is glued (see Fig. 2(d)) [45]. Nevertheless, the stresses in these regions are much lower than the stress in the freestanding region, and therefore the measured plastic strains are accurate and relate directly to the plastic deformation of the free-standing gauge length [45]. Both curves show a maximum stress of 400 MPa, and limited ductility of 2–3% (defined as plastic elongation at failure). The thinner specimen, with a smaller average grain size, is expected to have a larger yield stress [47], which is not apparent from these stress-strain curves. As will be shown below, the in situ TEM tests reveal a different cracking behavior for both film thicknesses, with easier susceptibility to cracking for the 30-nm-thick film. It is therefore possible that cracking occurs at stresses lower than the film's yield stress for the thinner film.

The fracture surfaces of the two tested specimens, shown in Fig. 4, are notably different. The fracture surface is nearly perpendicular to the applied load for the thinner film (Fig. 4(d)-(f)), while it is inclined at an angle of 30–40° with respect to the loading axis for the thicker film (Fig. 4(a)-(c)). While the amount of measured ductility (see Fig. 3) is similar between the two specimens (whose low values are confirmed from Fig. 4), the fracture surface of the 100-nm-thick specimen is reminiscent of a ductile, shear-dominated failure, while that of the 30-nm-thick specimen is more consistent with brittle failure. This difference is confirmed and even amplified for fracture surfaces of specimens that underwent several stress relaxation tests. Fig. 5 compares the fracture surface of a 30-nm-thick specimen (see (a) and (b), test consisting of twelve successive holds at constant displacements $x_h$, increasing from 250 nm ($V_{in} = 1.5$ V) to 460 nm ($V_{in} = 2.2$ V) nm, each hold lasting up to 10 min) to a 100-nm-thick one (see (c) and (d), test consisting of a series of fifteen successive holds at constant displacements $x_h$, increasing from 855 nm ($V_{in} = 3$ V) to 1295 nm ($V_{in} = 3.6$ V), each hold lasting between 2 and 10 min). Fig. 5(b) shows an intergranular crack that propagated from the right edge through the entire 30-nm-thick specimen’s width, along a plane perpendicular to the loading axis in average. Despite two others embryos that can be seen on the left of Fig. 5(b) (see red arrows), only one main crack propagated along that specimen, along GBs...
that are oriented $20^\circ$–$70^\circ$ away from the tensile direction. Instead, several cracked regions developed in the 100-nm-thick specimen (see Fig. 4(d)), some of them ultimately joining each other. The main crack, along with another crack below, is oriented at an angle ($\sim30^\circ$–$40^\circ$) with respect to the loading axis, also in contrast to the thinner specimen. The specimen’s elongation is $\sim30\%$, with locally a decrease in width of up to $\sim35\%$ (see Fig. 5(d)), whereas the decrease in width is only $\sim15\%$ for the thinner specimen (see Figs. 5(b) and 6). The notable difference in fracture surface highlights an apparently different crack growth behavior between the two specimens, with an apparent plasticity-governed failure for the thicker film and an apparent decohesion-like failure (with limited plasticity) for the thinner one. However, as highlighted in the next sections thanks to the in situ TEM crack growth observations, the governing crack

![Fig. 5. (a)–(b) TEM images of a 30-nm-thick specimen before and after fracture (in situ TEM test) (c)–(d) TEM images of 100-nm-thick specimen 1 before test and after failure (in situ TEM test).](image1)

![Fig. 6. TEM images of a 30-nm-thick specimen (a) before and (b) after fracture (in situ TEM test). The red line in (a) highlights the crack path seen in (b). Limited grain growth is observed along the crack path. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)](image2)
growth mechanisms are in fact very similar in nature between the 30 and 100-nm-thick specimens, and differ mainly in the fact that secondary nanocracks form easily ahead of the main crack of the thinner films (that also have smaller grains) but not of the thicker films (that also have larger grains).

3.2. In situ TEM crack growth observations in 30-nm-thick Au specimens

The test performed on a 30-nm-thick Au specimen inside the TEM (see Fig. 5(a) and (b) and Fig. 6 for pre- and post-test TEM images) consisted of twelve successive holds at constant displacements \( X_A \), increasing from 255 nm (\( V_{in} = 1.5 \text{ V} \)) to 460 nm (\( V_{in} = 2.2 \text{ V} \)) nm, each hold lasting up to 10 min. At the location of the crack, the width decreased by 15\%, indicative of localized plastic deformation in the crack region, which is also consistent with the evidence of limited grain growth surrounding the crack tip shown in Fig. 6. The in situ TEM observations clearly reveal that the intergranular crack propagates mainly via the growth of nanocracks that follow each other along the sample gauge. Secondary nanocracks generate just ahead of the main crack and eventually coalesce with the main crack. This leads to an apparent GB decohesion that occurs during the loading steps or at the very beginning (i.e. within the first minute) of the holding steps during which large amounts of GB dislocation activities can be observed in a localized region ahead of the main crack. Fig. 7(a) shows the crack after initiating from the right edge at \( X_A = 255 \text{ nm} \ (V_{in} = 1.5 \text{ V}) \). A nanocrack is observed roughly 200 nm ahead of the crack tip, along the GB of a large grain. At the end of the loading step to \( X_A = 325 \text{ nm} \), rapid GB opening occurs and the nanocracks grow by ~50–60 nm (see blue arrow in Fig. 7(b)). During the hold at \( X_A = 325 \text{ nm} \), the main crack blunts (probably because of the larger grain present at its tip), and the nanocrack grows by ~60 nm over 10 min (see Fig. 7(c)–(e)). The blunting of the main crack and the increased opening of the nanocrack most likely results in a decrease of the local stresses. Hence, dislocation activities drop rapidly 10 min into the hold segment at \( X_A = 325 \text{ nm} \), except for the grain that is ahead of the nanocrack (see Movie 1). Movie 2 shows clear evidence of secondary cracking at a GB ahead of the main crack and coalescence to the main crack tip during a loading step to \( X_A = 360 \text{ nm} \ (V_{in} = 1.9 \text{ V}) \). Towards the end of the loading step, large amounts of dislocation activities are observed in a region ~200 nm ahead of the main crack, associated in some cases to grain growth but mainly leading to crack grow along the GB when these dislocations are confined in GB. These activities appear to slow down quickly (within a minute), in part because of the local unloading occurring due to GB crack opening. Very similar events occur during the later loading steps; see Movies 3 and 4 (loading to \( X_A = 375 \text{ nm} \ (V_{in} = 1.96 \text{ V}) \) and 415 nm \( V_{in} = 2.07 \text{ V} \), respectively). The nanocrack that can be seen at the end of Movie 4 blunts, probably again because a large grain is present ahead of the crack tip, imposing the deviation of GB dislocations in directions that are less favorable and limiting their propagation along the initial crack direction, as can be seen in Movie 5 (corresponding to the loading to \( X_A = 420 \text{ nm} \ (V_{in} = 2.09 \text{ V}) \)). Three minutes into the hold at \( X_A = 420 \text{ nm} \ (V_{in} = 2.09 \text{ V}) \), Movie 6 shows GB shearing at the nanocrack opening from the left edge of the specimen. The remaining ligament is observed to fail by plastic deformation during the loading to \( X_A = 460 \text{ nm} \ (V_{in} = 2.2 \text{ V}) \); see Movie 7.

Supplementary video related to this article can be found at https://doi.org/10.1016/j.actamat.2017.10.004.

3.3. In situ TEM crack growth observations in 100-nm-thick Au specimens

As mentioned in 3.1, the fracture surface of the 100-nm-thick Au specimens, although also intergranular, is distinctly different from that of the 30-nm-thick specimen. As for the 30-nm-thick specimen, GB dislocation activities are abundant and lead to grain boundary sliding. However, compared to the thinner films, grain boundary sliding is more profuse and occurs much further ahead of the main crack tip of the 100-nm-thick specimens, affecting many grains. In addition, nanocrack formation ahead of the main crack does not occur. As a result, the main crack extends in a continuous manner and spreads along several different GBs. This may cause the development of secondary cracks at other locations along the gauge.

**Fig. 7.** (a)–(e) Series of TEM images extracted from a video of the in situ TEM test of the 30-nm-thick specimen shown in Figs. 5 and 6, showing the formation and growth of a secondary nanocrack (indicated by blue arrows) ahead of a main crack (indicated by red arrows) that formed on the right edge of the specimen. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
length, each contributing to the total larger elongation of the specimen (Fig. 5(d)). This behavior was clearly observed during the in situ TEM tests of three specimens, as described next. As mentioned in 3.1, specimen 1 (see Fig. 5(c) and (d)) was tested with a series of fifteen successive holds at constant displacements \(X_A\), increasing from 855 nm \((V_{in} = 3 \text{ V})\) to 1295 nm \((V_{in} = 3.6 \text{ V})\), each hold lasting between 2 and 10 min. At \(X_A = 855 \text{ nm}\), a large \((220-\text{nm-long})\) angle GB, located in the center of the gauge section and oriented 35° with respect to the loading axis, undergoes sliding as a result of significant GB dislocation activities. A 200-nm-long grain boundary crack, oriented perpendicular to the loading axis, has formed on the right edge of the specimen, near the sliding large GB. On the left side of the specimen, slightly above \((-500 \text{ nm})\) the sliding GB, another grain boundary crack has formed. Over the successive holds until \(X_A = 1130 \text{ nm}\), the crack on the right blunted and branched out along adjacent GB’s at angles with respect to the loading axis. The crack on the left did not blunt and kept growing along GB’s, as illustrated in Fig. 8. Movie 8 clearly shows GB activities initiating at the crack tip, resulting in stable intergranular crack growth. At variance from the 30-nm-thick film, dislocations traversing grains near the crack tip can also be observed, likely triggered by the large stresses ahead of the crack and favored by the larger grain volumes (see Movie 9). Movie 10 shows GB dislocations flowing continuously from the crack tip to the sliding large GB, as the applied displacement is increased to \(X_A = 1230 \text{ nm}\). At the end of the test \((X_A = 1295 \text{ nm})\), a series of TEM images, shown in Fig. 9, clearly highlights GB sliding and its prominence on the resulting strain. The sliding path, shown by the dashed yellow line, corresponds to a facet in a \((111)\)-oriented grain. Stereographic projections prove that G1 and G2 have a relatively large misorientation angle of about 54°, close to the axis \((011)\). Comparison of the images shows a relative shear of 55 nm in a 10-min period with an angle of approximately 45° with respect to the tensile axis. By defining shear strain as the ratio of displacement along the GB over the diameter of G1 \((400 \text{ nm})\), this shear displacement corresponds to an increase in shear strain from 4 to 11.5% in 10 min, equivalent to a strain rate on the order of \(2 \times 10^{-4} \text{ s}^{-1}\).

Supplementary video related to this article can be found at https://doi.org/10.1016/j.actamat.2017.10.004.

Intergranular crack extension assisted by GB dislocations was also observed in another specimen (specimen 2). A 165-nm-long GB crack formed early during a 45-min-long hold at \(X_A = 550 \text{ nm}\) \((V_{in} = 2.45 \text{ V})\) \((\sigma \approx 400 \text{ MPa} \text{ based on TEM measurements of } X_{LS})\). Movie 11 shows GB dislocations flowing from the crack tip along a 210-nm-long GB that is inclined ~20° with respect to the loading axis, after 20 min at \(X_A = 550 \text{ nm}\) \((V_{in} = 2.45 \text{ V})\). A comparison of TEM images taken at an interval of 35 min shows a shear angle of 65° resulting from the GB sliding as shown in Fig. 10. Movie 12 shows increased GB dislocation activities as the displacement \(X_A\) was raised to 625 nm \((V_{in} = 2.6 \text{ V})\), which led to the fracture of the specimen shortly after.

Supplementary video related to this article can be found at https://doi.org/10.1016/j.actamat.2017.10.004.

In another instance (specimen 3), we observed cracking along an active slip band in a large grain \((-300 \text{ nm})\). During a hold at \(X_A = 360 \text{ nm}\) \((V_{in} = 1.94 \text{ V})\), a sustained slip band of dislocations in a \((111)\) plane, oriented 30° with respect to the loading axis, concentrates most of the deformation in the specimen. A GB crack forms on the left side of the specimen near the deforming grain \((X_A = 530 \text{ nm}, \sigma = 100 \text{ MPa})\), while a void forms at a triple point in the lower right of the deforming grain. As the applied displacement is increased to \(X_A = 540 \text{ nm}\), a continuous path of inter and transgranular dislocations flows from the triple junction void to the GB crack (see Movie 13). As \(X_A\) is further increased to 550 nm, a crack forms along the slip band (see Movie 14), linking the GB crack on the left side to the triple junction void. Crack extension further occurs as a result of GB dislocation motion, as clearly evidenced in Movie 15 \((X_A = 565 \text{ nm})\) and Movie 16 in which the crack extends...
by ~30 nm due to GB sliding during the increase of $X_A$ to 615 nm.

Supplementary video related to this article can be found at https://doi.org/10.1016/j.actamat.2017.10.004.

### 3.4. Discussion

By employing the same testing technique (in situ TEM MEMS-based tensile tests consisting of successive displacement holds), same specimen geometry, same material (99.99% Au), and similar surface roughness to thickness ratio (1.7% for the 30-nm-thick specimens, and 1.2% for the 100-nm-thick specimens), this study investigated the combined effects of thickness (30 vs 100 nm) and grain size distribution (see Fig. 1(f)) on the crack growth behavior in nc ultrathin films. Although the basic process (GB sliding) is the same, the results highlighted significant differences in the intergranular crack growth behavior between the two types of specimens. It should be noted that the presence of surface grooves at the grain boundaries (see Fig. 1(g) and (h)) leads to stress concentrations that can favor GB dislocation nucleation [48]. The similar surface roughness to thickness ratio between the two film
thicknesses (whether the rms roughness or deepest grooves are considered) implies similar stress concentrations at the GB grooves for both films. For the 30-nm-thick specimens having an average grain size of 40 nm and grains no larger than 100 nm in diameter, the crack growth process occurs mainly due to multiple cracking that remain aligned in average with the main crack. Upon a displacement impulse, very fast GB dislocation activities can be observed ahead of the main crack. If a GB is well oriented with respect to the applied and local stress (due to the main crack tip), a secondary nanocrack can open ahead of the main crack, also due to GB dislocation shear. The GB dislocation activities then slow down quickly (within a minute), likely as a result of local unloading due to nanocrack formation. Therefore, what appears like a GB decohesion on a micro scale is actually the result of GB dislocation activity that produce local GB shearing and sliding. Once a nanocrack is formed ahead of the main crack, the crack path is defined between this secondary crack and the main one. In contrast, for the 100-nm-thick specimens having an average grain size of 70 nm and grains as large as 350 nm in diameter, nanocrack formation ahead of the main crack is not observed. Intergranular crack growth occurs mainly as a result of sustained (i.e. even after a displacement hold of 30 min) GB dislocation activities. This process leads to GB sliding along the large (>200 nm) grain boundaries that are oriented favorably (typically at a large angle with respect to the loading axis) and intragranular dislocation activity can also be observed. For these thicker specimens, secondary GB shearing activities are spread out around the crack and can also be triggered away from it, initiating remote secondary cracks in the specimen. This multiple activity diffuses deformation instead of concentrating it, giving rise to apparent increased ductility.

The main difference between these two cracking behaviors is the formation of nanocracks ahead of the main crack for the 30-nm-thick specimens but not for the 100-nm-thick ones. Indeed, the zig-zag pattern observed for the 30-nm-thick films (see Figs. 4(e), 5(b) and 6), with an overall crack path that is on average perpendicular to the applied load, is a result of a series of nanocrack formation just ahead of the main crack followed by coalescence events. This crack growth behavior is also accompanied with lower amounts of plastic deformation (decohesion-like failure, as discussed in 3.2) because of the rapid unloading occurring ahead of the main crack as a result of nanocracks. We propose the following explanations for this thickness- and grain-size-dependent nanocrack formation. First, a simple geometrical explanation can be invoked: while GB shear is active in both 30- and 100-nm-thick films, the amount of shear, and thus the number of GB dislocations, to provoke grain decohesion, is \(3.3\) (i.e. the thickness ratio) times less for the thinner film. It is therefore much easier to nucleate a nanocrack in the thinner films as a result of GB dislocations. In addition, this GB shear is accompanied by a significant grain displacement or rotation in the case of the thicker film. This implies that the first GB dislocations activated may be less favorable for motion with respect to the applied stress once grains have rotated. GB shearing may thus be more favorable elsewhere and produce the multiple crack paths observed in the 100-nm-thick films.

Another explanation lies in the difference in grain size between the two types of specimens. A theoretical model predicted nanocrack formation ahead of blunted cracks due to the combined effects of large stresses ahead of the crack and the superposed stress field associated with GB dislocations at triple junctions [49]. The larger the grains, the less triple junctions are present in the high stress field ahead of the main crack, and the lower the local stress value at that triple junction (since farther from the main crack). Hence, that model predicted that larger grain sizes decrease the likelihood of nanocrack formation [49]. We believe a similar scenario occurs for our specimens, as GB dislocations are often produced at triple junctions [21]. For the thinner specimens, with a lower average grain size, there are statistically more triple junctions ahead of the main crack under large enough stresses where GB dislocations can be produced. As a result, GB sliding, carried out through gliding GB dislocations, and therefore decohesion, is more likely to occur within the favorably oriented GBs. This scenario is less likely to occur for the 100-nm-thick specimens with larger average grain size, as there are less triple junctions ahead of the main crack and as these triple junctions, being farther from the main crack tip, experience lower local stress values that may not be enough for nanocrack formation. Instead, GB sliding concentrates at the main crack tip.

Atomistic simulations performed on nc Ni (grain sizes ranging from 5 to 10–12 nm) showed a crack growth process consisting of nucleation of nanocracks (via GB decohesion) followed by coalescence with the main crack. These simulations revealed that large hydrostatic stresses are present ahead of the crack tip and trigger both transgranular dislocation activities and GB decohesion. The exact GB activities leading to GB decohesion/nanocrack formation could not be identified in that study, but involved the formation of vacancy clusters. Our in situ TEM study highlights the role of GB dislocations (see Movies 10–13, 15 and 16), resulting in GB shear and sliding, as a major component of crack nucleation and extension. For the 30-nm-thick Au films, the GB dislocations lead to nanocrack formation ahead of the main crack, whereas for the 100-nm-thick Au films, they lead to extension of the main crack without formation of nanocracks. Transgranular (partial or perfect) dislocation activities ahead of the crack tip were observed in several instances, (see Movies 8, 9, and 13), but are not the main deformation mechanism. Hence our results are in contrast with most previous in situ TEM crack growth studies that did not report GB dislocations or grain rotation in the cracking process [26,27,29]. This may be attributed to the fact that this mode of deformation is not easy to track, and that it has been evidenced only recently as a key mechanism [48]. Slip-induced GB sliding was clearly observed in 300-nm-thick Al films with an average grain size of about 250 nm [48]. In that study, GB dislocations as well as intragranular dislocations getting absorbed by GBs were observed irrespective of the GB orientations. The specific configuration of thin films for which a large proportion of GBs lies perpendicular to the free surfaces may also favor the nucleation, propagation and escape of GB dislocations. Overall, the present report emphasizes that different mechanisms can act in parallel in this range of grain size. Among those, GB dislocation activities are the most prominent ones, even at room temperature. They occur at stresses that are not significantly higher than for intra-granular dislocation activities that take place in large-grained metals.

4. Conclusions

MEMS-instrumented in situ TEM tests allowed the observation of crack propagation mechanisms in 30- and 100-nm-thick free-standing Au films with nc or sub-micrometer grain sizes. In this size range, several plasticity mechanisms were observed. The ones that led to crack extension were clearly identified to be GB shearing and sliding. Both mechanisms carried out by GB dislocation activity were extensive for both films. However, the combined effects of thickness (30 vs 100 nm) and grain size distribution (average grain size of 40 nm (resp. 70 nm) for the 30-nm-thick (resp. 100-nm-thick) films) led to a significant difference in the intergranular crack growth process, namely the nucleation of secondary cracks in the thinner films, which in turn led to an overall different fracture surface. Crack propagation in the 30-nm-thick films occurred both through the opening of a main crack and by the coalescence of secondary cracks generated in front of it, leading to a single crack.
path and limited ductility. In contrast, because nanocrack formation ahead of the main crack does not occur in the 100-nm-thick films, grain boundary sliding is more diffuse and occurs much further ahead of the main crack tip, affecting many grains. As a result, the main crack extends in a continuous manner and spreads along several different GBs.

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