Quantitative in situ TEM tensile fatigue testing on nanocrystalline metallic ultrathin films†

Ehsan Hosseinian and Olivier N. Pierron*

A unique technique to perform quantitative in situ transmission electron microscopy (TEM) fatigue testing on ultrathin films and nanomaterials is demonstrated. The technique relies on a microelectromechanical system (MEMS) device to actuate a nanospecimen and measure its mechanical response. Compared to previously demonstrated MEMS-based in situ TEM techniques, the technique takes advantage of two identical capacitive sensors on each side of the specimen to measure electronically elongation (with nm resolution) and applied force (with μN resolution). Monotonic and fatigue tests were performed on nanocrystalline gold ultrathin film specimens that were manipulated and fixed onto the MEMS device without the use of a focused ion-beam microscope (and therefore, importantly, without any associated surface damage). The major advantage of the technique is its capability to use TEM imaging solely for high magnification microstructural observations while the MEMS device provides continuous tracking of the material’s response, thereby expanding the capabilities of MEMS-based techniques towards more complex in situ TEM nanomechanical tests, such as fatigue tests.

1. Introduction

Nanocrystalline metallic thin films are routinely used in a large range of applications, from hard coatings on bulk components1 to structural components in micro/nano-electromechanical systems (MEMS/NEMS)2 and conductive layers in microelectronics devices.3 In all these applications, the nanocrystalline thin films undergo cyclic loading, either due to repeated surface contact, applied mechanical loading, or thermally induced stresses. As such, the fatigue properties of this class of material should be adequately characterized to ensure proper reliability of these components and devices.4-5 In addition, the underlying mechanisms should be understood so that fatigue resistant nanocrystalline thin films can be processed. While the monotonic plastic deformation behavior of nanocrystalline metals and the underlying length-scale effects have been the topic of intense investigation over the past two decades,6 there is so far little mechanistic insight into the length-scale effects in fatigue processes.7 Particularly, the current knowledge of plastic deformation in nanocrystalline metals precludes accurate fatigue modeling in terms of the mechanical irreversibility of the inelastic strains. Therefore, there is a need to systematically study the cyclic plastic deformation of nanocrystalline thin films leading to fatigue crack initiation, which will ultimately shed new light into the governing fatigue mechanisms in the nanocrystalline regime. In this paper, we present a novel MEMS-based experimental technique to perform quantitative in situ transmission electron microscopy (TEM) fatigue testing on ultrathin, nanocrystalline metallic films. Specifically, we demonstrate quantitative in situ TEM fatigue experiments using a MEMS tensile testing device that solely relies on capacitive sensing of both applied force and specimen elongation, thereby providing an additional level of sophistication to the current state-of-the-art MEMS-based techniques required to perform long fatigue experiments.

Early research on the fatigue behavior of nanocrystalline face centered cubic metals shows clear evidence of improved fatigue limit compared to their coarse grain counterparts (grain size, d > 1 μm), which is attributed to an improved resistance to fatigue crack initiation for high strength materials.8-10 This qualitative interpretation is consistent with the classical models predicting fatigue crack initiation life, N_{init} as a function of grain size (e.g., N_{init} ~ 1/d for Tanaka and Mura’s model).11 However, these classical models rely on a specific stress distribution within the complex persistent slip bands, which are too big to form in nanosized grains.7 Hence it is primordial to obtain a fundamental understanding of cyclic plasticity in nanocrystalline metals to provide a scientific basis for prediction of their fatigue behavior. Kraft and coworkers studied the dislocation structures developed under cyclic loading for ultrathin grain and coarse grain copper thin films.12-15 They observed a transition from “bulk-like” behavior (formation of persistent slip bands and persistent slip markings at the surface) for coarse grain films, to “small volume” behavior for ultrathin grain films, characterized by individual dislocations and interface-mediated...
damage behavior such as cracking and/or voiding along grain boundaries and/or twin boundaries.\(^1\)\(^2\) Cheng et al. observed a significant difference in cyclic behavior between large-angled-grain-boundary ultrafine grain and coarse grain nickel (Ni).\(^1\)\(^6\) Both cyclic hardening and softening were obtained for ultrafine grain Ni, and interpreted in terms of deformation activities absent in coarse grain Ni, such as deformation twinning, stacking fault formation, and disentangling of dislocations. Suresh and co-workers did not observe any dislocation debris or significant grain growth with TEM after fatigue deformation of nanocrystalline Ni (average grain size: 30 nm).\(^17\) The measured cyclic strain hardening was therefore explained by exhaustion of non-equilibrium dislocation sources. The cyclic deformation behavior of nanocrystalline Ni was also found to be frequency-dependent, suggesting the importance of time-dependent mechanistic processes. The authors hypothesized the occurrence of time-dependent, irreversible grain boundary processes such as grain boundary sliding. A more recent study highlighted the role of grain coarsening during cyclic loading for electro-deposited nanocrystalline nickel alloys, leading to the classical fatigue crack initiation mechanisms associated with persistent slip band formation.\(^18\)

One difficulty in interpreting the results and drawing general conclusions on the fatigue mechanisms of nanocrystalline metals is the lack of consistency between the experimental conditions from all the above studies. Also, post-mortem characterization makes the understanding of the complex microstructure evolution and defect formation during cyclic loading challenging. Quantitative in situ TEM testing provides an opportunity to observe the microstructure evolution during deformation of the specimen, while both the applied force and deformation are measured. This powerful technique has been employed extensively over the past 10 years to investigate the monotonic deformation of metallic nanospecimens and ultra-thin films.\(^19\) Among the existing in situ TEM testing techniques, the MEMS based techniques\(^20\) have evolved tremendously since the pioneering work of Haque and Saif.\(^21–23\) Notably, Espinosa and coworkers have demonstrated the use of MEMS components (thermal actuator, comb structures) for in situ TEM actuation and load sensing.\(^24–26\) These state-of-the-art MEMS based techniques are mostly suited for short experiments (such as monotonic loading to failure) given the time-consuming digital image correlation technique process of measuring apparent or local strain. These strains are measured based on relatively low magnification electron images of the entire specimen, during which the evolution of the microstructure cannot be observed in great details. Here we demonstrate truly quantitative in situ TEM tensile fatigue experiments of nanocrystalline ultrathin films, whereby the evolution of the stress-strain curve is monitored over thousands of cycles, thanks to capacitive sensing of both load and elongation. This versatile technique offers great advantages over the existing MEMS-based techniques,\(^24–27\) such as the possibility to use TEM imaging solely for microstructure evolution (high magnification images), and the possibility to perform the same fatigue tests outside of an electron microscope (ex situ testing) to probe environmental effects on the fatigue degradation properties.

2. Experimental section

2.1. Description of the MEMS material testing setup

Our MEMS-based technique for quantitative in situ TEM fatigue tests relies on a MEMS device that provides both actuation and sensing of the specimen via electrical signals,\(^28\) while TEM imaging provides information on the microstructure evolution during cyclic deformation. Fig. 1 shows the MEMS device (which is small enough, ~1 mm by 3 mm, to fit inside an electrical bating holder), with an accompanying schematic and lump model. These devices are fabricated with the SOIMUMPs process from MEMSCAP; see ESI S1\(^+\) for an overview of the fabrication process. They are made of monocrystalline Si (thickness: 10 μm) and comprise a thermal actuator, a specimen gap, two capacitive sensors located on each side of the specimen gap, and a load sensor. All these components are fabricated over a through-wafer window, for TEM imaging purpose.

The thermal actuator, based on Joule heating, consists of 10 pairs of beams inclined at 5°, providing a displacement \(X_a\) for an input voltage, \(V_{in}\), applied across the beams. The actuator can provide \(X_a \sim 1.6 \mu m\) for \(V_{in} = 4\) V without any appreciable increase in temperature (~10 °C) near the specimen, thanks to a

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**Fig. 1** (a) SEM image of a wirebonded MEMS device. (b) Corresponding schematic, with a description of its components and their displacements (e.g. \(X_a\): displacement of thermal actuator). (c) Lump model of the MEMS device with corresponding governing equations.
heat sink located next to the actuator and a large distance (>1 mm) between the actuator and the specimen.29 The first capacitive sensor, CS1, is rigidly connected to the thermal actuator (i.e., \(X_{CS1} = X_A\)) while electrically isolated from it,\(^{18}\) and consists of 21 pairs of “combs” on each side of the MEMS device’s symmetry axis. In the absence of a specimen (or after failure of the specimen), the output signal of the MEMS corresponds to the change in capacitance of CS1 (\(\Delta C_1\)) which can be directly related to \(X_A\) using the parallel plate capacitance formula (see Section 2.2). The specimen gap is \(-5 \mu m\), and consists of two adjacent large areas (50 \(\mu m\) by 200 \(\mu m\)) for proper clamping of the specimen onto the MEMS device (see Fig. 1(a) and (b) as well as Movies 6 and 7 for specimen manipulation and clamping in ESI†). The load sensor is made of 4 beams (length: 500 \(\mu m\)) deforming in bending mode. The stiffness of the load sensor, \(K_{LS}\), can be accurately calculated given the known properties of monocrystalline Si.\(^{39}\) The width of the beam mainly dictates \(K_{LS}\) that ranges from 100 N m\(^{-1}\) (width: 12.5 \(\mu m\)) to 480 N m\(^{-1}\) (width: 21 \(\mu m\)). The second capacitive sensor, CS2, is rigidly connected to the load sensor (\(X_{CS2} = X_{LS}\)), and is identical to CS1 (see Fig. 1(a)).

### 2.2. MEMS electrical signal measurements and calibration

The displacements of capacitive sensors CS1 and CS2 are related to a change in capacitance, \(\Delta C\), based on the following parallel plate capacitance formula:

\[
\Delta C = \epsilon_0 \epsilon_n k A \left( \frac{1}{d_1 - X} + \frac{1}{d_2 + X} \right)
\]

(1)

where \(\epsilon_0\) is a calibration constant, \(\epsilon_0 = 8.854 \times 10^{-12}\) F m\(^{-1}\) is the permittivity of free space, \(k\) is the relative permittivity of the dielectric material \((k \sim 1\) for air\)), \(A\) is the overlapping area of a comb structure, \(n\) is the number of comb structures, \(d_1\) and \(d_2\) are the initial gaps between the comb structures \((d_1 = 4 \mu m, d_2 = 9 \mu m\); see Fig. 1(a)), and \(X\) is the displacement of CS1 \((X_{CS1})\) or CS2 \((X_{CS2})\) (see Fig. 1). The precision in capacitance measurements is better than 0.1–0.2 fF, which corresponds to a precision in displacement of approximately 1–5 nm (see Section 3.1). This level of precision is very good and can be used to measure the mechanical properties of nanospecimens without requiring high magnification images. However, the accuracy of the displacement calculation relies on the accuracy of the simplified eqn (1) which, for example, ignores the fringing field effects. More importantly, parasitic capacitance can affect the measurements given the very low \(\Delta C\) values (fF level). Hence a calibration is performed for each MEMS device and consists of accurately measuring the displacement of CS1 for large values (~1.6 \(\mu m\), corresponding to \(V_{in} = 4 V\)) based on high magnification images (either optical for \(ex\) \(situ\) tests, or electron images for \(in\) \(situ\) SEM or TEM tests). The procedure to measure displacement with pixel accuracy was reported in ref. 39. The ratio between the measured displacement at 4V and the calculated displacement at 4V using eqn (1) (assuming \(\alpha = 1\)) is used as the calibration constant, \(\alpha\).

The capacitance change \(\Delta C_1 - \Delta C_2\) (or \(\Delta C_3\) in the absence of a specimen) is measured using a universal capacitive readout sensor (MS3110, Irvine Sensor). The output voltage \(V_0\) (in Volt) is related to the change in capacitance \(\Delta C\) via the following equation:

\[
V_0 = 5.13 \times \frac{C_2 - C_1 + C_{int} - C_{int}}{C_1} + 2.25
\]

(2)

where \(C_f\) is the feedback capacitor \((C_f = 399 fF)\), and \(C_{int}\) are pre-programmed internal capacitances (in fF). During a test, the driving voltage \(V_{in}\) is applied in incremental steps and the corresponding \(V_0\) is averaged after measurement durations ranging from 500 ms to 2 s at an acquisition rate of 1 kHz (for noise reduction). The capacitance change is therefore given by:

\[
\Delta C_1 - \Delta C_2 = C_f \times \frac{V_0(V_{in} = 0) - V_0(V_{in})}{5.13}
\]

(3)

where \(V_0\) represents the average \(V_0\) over the acquisition period.

### 2.3. Fabrication of ultrathin film specimens

Nanocrystalline Au thin films were fabricated using a process involving optical lithography (as opposed to electron beam lithography previously employed for the fabrication of Ni nanobeams),\(^{39}\) electron-beam evaporation of Au, a lift-off technique, and XeF\(_2\) etching of the Si substrate with a thin native oxide; see ESI S2† for details on the fabrication process. The specimens are 100 \(\mu m\) long, 1500 nm wide and 100 nm thick. At the end of the process, the specimens are free-standing cantilevers, connected on one side to a large island of Au as a support. Fig. 2 shows SEM (Fig. 2(a) and (b)) and TEM (Fig. 2(c)) images of the nanocrystalline gold (Au) thin film specimens. These images show a nanocrystalline microstructure for the 100 nm thick Au films with a wide distribution of grain size ranging from 10 to 400 nm. Selected area electron diffraction patterns (see example in Fig. 2(c)) reveal a heterogeneous microstructure without any strong out-of-plane or in-plane texture. The lack of a strong (111) out-of-plane texture (which is typically observed for thin Au films)\(^{33}\) may result from the deposition of the film on a Si substrate with the presence of a thin native oxide.\(^{40}\)

The critical step to minimize the line edge roughness of the specimens is the patterning of the photoresist used for the lift-off technique. Fig. 2(a) shows specimens with very smooth edge features. In contrast, the tested specimens shown in this manuscript were from a fabrication batch that had rougher edge features (if any) of line edge roughness on the fatigue initiation life, as is the case for macrocomponents with notches or scratches. However, recent studies have cast some doubts on the actual effects of nanonotchess on the fracture of nanocrystalline ultrathin films.\(^{41,42}\) The present technique can therefore offer an opportunity to establish the effects (if any) of line edge roughness on the fatigue initiation lives of our specimens.

### 3. Results and discussion

#### 3.1. Calculation of stress and strain

The force \(F\) applied onto the specimen (and therefore the applied stress, knowing the specimen cross-sectional area) can
be calculated if the load sensor displacement, $X_{LS}$, is known, using:

$$F = K_{LS} \times X_{LS}$$  \hspace{1cm} (4)$$

During a test, the difference in capacitance between $CS_1$ and $CS_2$, $\Delta C_1 - \Delta C_2$, is measured with a MS3110 chip (Irvine Sensor) (see Section 2.2), which is related to the elongation of the specimen, $X_S$. The stress and strain of the specimen can be calculated if both $X_{LS}$ and $X_S$ are known (the dimensions of the specimens are measured accurately with SEM). Therefore, two independent sets of measurements are needed to calculate the stress-strain curve: $\Delta C_1 - \Delta C_2$, vs. $V_{in}$ (measured during the test) and $\Delta C_1$ vs. $V_{in}$ (measured after the specimen is broken or before it is fixed onto the MEMS). The former essentially gives $X_S$ and the latter gives $X_A$, from which $X_{LS}$ can be calculated using:

$$X_A = X_S + X_{LS}$$  \hspace{1cm} (5)$$

However, for a given $V_{in}$, the thermal actuator displacement without a specimen (i.e., no applied force $F$ onto the actuator), $X_A^{0\circ}$, is different from the displacement with a deformed specimen (i.e., a force $F$ is applied onto the actuator), $X_A$. Both quantities can be related via the following equation:\textsuperscript{24}

$$\frac{X_A^{0\circ}}{X_A} = \frac{1}{1 + \frac{K_{LS}}{K_S} + \frac{K_{LS}}{K_A}}$$  \hspace{1cm} (6)$$

where $K_S$ is the stiffness of the specimen, and $K_A$ is the thermal actuator stiffness ($K_A = 3975 \text{ N\ m}^{-1}$).\textsuperscript{25} This equation can be used to predict $X_A^{0\circ}$ based on the calculated $X_A^{0\circ}$ from the measured $\Delta C_1$ vs. $V_{in}$. Once $X_A^{0\circ}$ is known, both $X_S$ and $X_{LS}$ can be calculated based on the measured $\Delta C_1 - \Delta C_2$ vs. $V_{in}$. Eqn (6) requires knowledge of $K_S$, which depends on the specimen’s elastic modulus $E$ ($K_S = EA/L$, with $A$ and $L$ being the cross-sectional area and length of the specimen, respectively) and is typically unknown. In that case, an iterative procedure based on the calculated $E$ value is used to calculate stress and strain.

As explained in Section 2.2, each MEMS device is calibrated to ensure accurate measurements of the various MEMS components’ displacement ($X_A$, $X_S$, $X_{LS}$). The parallel plate capacitance formula (see eqn (1)) is calibrated by measuring actual displacement of the capacitive sensor 1 ($CS_1$) at $V_{in} = 4 \text{ V}$ using either optical or electron images. Fig. 3(a)–(c) show excellent agreement, for 3 different MEMS devices without any specimen, between the calculated $X_A$ values based on electrical measurements and eqn (1), and the measured $X_A$ values at lower $V_{in}$ values, using also either optical or electron images. Fig. 3(d)–(f) also show excellent agreement between the calculated $X_A$, $X_{LS}$ and $X_S$ values for an actual in situ TEM test of a nanocrystalline Au specimen based on electrical measurements and eqn (1), (4)–(6) (see the above mentioned procedure), and the measured values based on TEM images (see ESI S3\textsuperscript{+} for the images). In addition to providing accurate measurements, this experimental setup is also very precise with a noise floor in the electrical signal of about 0.1–0.2 fF (see ESI S4\textsuperscript{+}), which corresponds

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**Fig. 2** (a) Top-down SEM images of our Au thin film specimens with large pads on each end for FIB-less manipulation and clamping onto a MEMS device. (b) Inclined SEM images showing 100 nm thick freestanding Au specimens attached on one side to large island of material. (c) TEM images of the Au specimens and selected area electron diffraction patterns showing no strong texture.
to 1–5 nm in displacement depending on the actual deformation of the capacitive sensors (see eqn (1)). As highlighted next, this MEMS-based nanomechanical testing technique presents unique opportunities to perform quantitative in situ TEM fatigue experiments.

3.2. Specimen manipulation and clamping

The manipulation and clamping of small scale specimens, including ultrathin films, is a notorious challenge in experimental nanomechanics.\(^{28}\) One previously employed approach to circumvent these issues is the co-fabrication of the thin film specimen along with the MEMS tensile machine.\(^{23,28,43}\) Such approach limits the MEMS device to a single use as well as the choice of materials to be tested. Another approach often employed for the testing of nanowires relies on the use of a micromanipulator inside a dual focused ion beam/scanning electron microscope (FIB/SEM) to manipulate the nanowire onto the testing platform. In that case, clamping of the nanospecimen is performed with electron or ion beam induced deposition of platinum (Pt). This technique also suffers several drawbacks, including possible compliance of the Pt clamps, Pt overspray covering the surface of the specimen, and ion-beam damage of the specimen’s surface (see ESI S5†). While the compliance of the Pt clamps can be accounted for if strains are measured based on digital image correlation of high magnification images,\(^{27}\) the issues of Pt overspray on the specimen and Ga implantation damage associated with the FIB manipulation may result in surface damage which can significantly affect the fatigue results.

Based on the above considerations, we developed a “FIB-less” method to manipulate and clamp our ultrathin film specimens onto the MEMS device. Specifically, we modified the geometry of the ultrathin film specimen from a simple nanobeam™ to a dogbone-shaped specimen (see Fig. 2). This specimen consists of a nanobeam in the center (width ~1.5 µm, length ~20 µm, thickness ~100 nm), with two large pads (~50 by 50 µm\(^2\)) on each end. The large pads allow manipulation of the specimen under a probe station and can be attached to the MEMS device with a UV-curable adhesive Dymax Light Weld 425 (see details below), thereby eliminating the FIB manipulation step and providing stiff clamps (given the large contact area for gluing). The thickness of the specimen should not exceed 150–200 nm to provide adequate TEM imaging. The length (~10 µm, depending on the exact location of the glue, which can be measured accurately with SEM images) and width (~1 µm) were chosen such that \(K_S\) is commensurate with \(K_{LS}\) to enable its testing to failure (which cannot occur if \(K_S \gg K_{LS}\)). As such, the volume of material that is tested under fatigue is approximately 1 µm\(^3\), resulting in the ability to perform in situ TEM fatigue tests on nanocrystalline thin films made of a large number of grains ranging from ~10\(^5\) to ~10\(^6\) grains.

Movies of the procedure for manipulation and clamping of the dogbone-shaped specimens onto the MEMS devices are available in the ESI (see Movies 6 and 7†). Briefly, the small strip of material connecting the specimen to the substrate (see Fig. 2(a)) is severed using a probe tip fitted to a micromanipulator, under a probe station (see Movie 6). The specimen, whose large pad is in contact with the probe tip, is then transferred to the MEMS’ gap, where UV curable epoxy has been dispensed (but not cured) on each side. The micromanipulator allows proper in-plane alignment of the specimen with respect to the MEMS’ gap, after which the specimen is lowered until it touches the epoxy (see Movie 7†). The epoxy is then cured using UV light. Overall, the yield associated with this procedure is very good, and specimen misalignment (such as the specimen shown in Fig. 5, one of the earlier specimens manipulated with this technique) can be minimized with proper experience.
3.3. **Ex situ** monotonic tests to failure

The capability of this MEMS-based technique was first demonstrated with two *ex situ* monotonic tests to failure of our Au specimens in laboratory air; see ESI S6† for details. Such tests cannot be performed with the previously demonstrated MEMS based techniques given the requirement of high magnification images (SEM or TEM images) to precisely measure the deflections of the MEMS components. The ultimate tensile strengths of the two specimens are 1.3 and 1.2 GPa, for a total strain to failure of 2.5 and 2.3%, and an estimated plastic strain to failure of 0.5 and 0.6%, respectively. Both specimens failed along the gauge length, away from the clamps. No obvious apparent deformation of the epoxy clamps could be observed.

3.4. **Ex situ** fatigue test

The above results, along with the *in situ* SEM tests described in ESI S7† and shown in the accompanying movies, highlight the apparent effectiveness of the FIB-less specimen manipulation and clamping onto the MEMS device, as well as of the electrical sensing scheme for stress and strain calculation. We now demonstrate the capability of this MEMS-based technique for fatigue testing. Fig. 4 and 5 show the results of an *ex situ* fatigue test of another nanocrystalline Au specimen performed in laboratory air ($K_{LS} = 480 \text{ N m}^{-1}$ for this experiment). The driving voltage $V_{in}$ was cyclically varied from 0.4 to 1.8 Volt in increments of 0.1 Volt for 11 125 cycles, after which the test was interrupted. This corresponds to a fatigue test with a load ratio $R = \sigma_{min}/\sigma_{max} \approx 0$, and a maximum applied stress of $\sim 0.6$ GPa, at a frequency of 0.07 Hz. Continuous acquisition of the $\Delta C_1 - \Delta C_2$ vs. $V_{in}$ data was performed throughout the entire test, which allows the calculation of the stress–strain curves for all cycles (using an automated procedure).

Fig. 4(a) shows five stress–strain curves for selected cycles, while Fig. 4(b)–(d) show the evolution throughout the fatigue test of the maximum applied stress, $\sigma_{max}$, permanent strain under no applied stress, $\epsilon_{\sigma=0}$, and $E$, respectively. $E$ remains constant during the test ($\sim 75$ GPa) and is calculated based on the initial portion of the unloading curves. A ratcheting behavior is observed, with the maximum applied strain, $\epsilon_{max}$, increasing from 1 to 1.4% throughout the test (see Fig. 4(a)) and a slight decrease in $\sigma_{max}$ from 0.62 to 0.54 GPa. Also, $\epsilon_{\sigma=0}$ increases from $\sim 0$ to 0.6%, with a sudden increase after 6000 cycles. Positive $\epsilon_{\sigma=0}$ values mean that the specimen is under compression below these values, as shown in Fig. 4(a), which is a result of specimen elongation. While some amount of buckling may occur due to the thinness of the specimens (see for example ESI S5†), the elongated specimens (with stiffnesses ranging from 300 to 1500 N m$^{-1}$ depending on the actual specimen dimensions) may “push” the load sensing beams ($K_{LS} = 100$ or 480 N m$^{-1}$), resulting in measured negative stresses.

It is important to note that this evolution is not an artifact of the MEMS device, given that the corresponding changes in the MEMS’ output signal are about 5–10 times larger than the noise level, and given the measured long-term stability of the MEMS device over long periods of time (see ESI S4†). This technique can therefore be used to probe minute changes in the material’s response that may be indicative of fatigue-related microstructural changes. Fig. 5 shows SEM images of the specimen and
after interruption of the test ($N = 11\,125$ cycles). No significant changes in the microstructure (such as grain coarsening) were observed, as illustrated with the high magnification images of the same area before and after the fatigue test. However, the use of post-mortem SEM images can only give limited information regarding the development of fatigue damage. Instead, quantitative in situ TEM fatigue tests provide the opportunity to observe fatigue-related microstructural evolution, as demonstrated next.

3.5. **In situ** TEM fatigue test

A fatigue experiment was carried out inside a HF 2000 TEM (acceleration voltage: 150 kV), using a Hummingbird electrical biasing holder with 6 leads (see Table of contents entry). The driving voltage $V_{in}$ was cyclically varied from 0 and 4 volt in increments of 0.4 V. This corresponds to a fatigue test with a load ratio $R = 0$, and $\sigma_{max} \sim 0.95$ GPa, at a frequency of 0.25 Hz ($K_{LS} = 100$ N m$^{-1}$ for this experiment). The specimen failed after 6995 cycles. TEM observations were performed during the first 500 cycles, after which the electron beam was turned off (the specimen was kept in the TEM chamber for the entire duration of the test). No effect of the beam on the material’s response was observed. Fig. 6(a) shows five stress–strain curves for selected cycles, while Fig. 6(b)–(d) show the evolution throughout the fatigue test of $\sigma_{max}$, $\varepsilon_{\sigma=0}$, and $E$, respectively. Ratcheting behavior is also observed (see Fig. 6(a) and (c)), with $\sigma_{max}$ increasing from 1.8 to $\sim$2.5% (similar to the fracture strain measured for the monotonic tests shown in Fig. S7†) and $\varepsilon_{\sigma=0}$ increasing from 0 to 1.1%. TEM and SEM images of the specimen after fatigue failure (see Fig. 7 and 8, respectively) reveal the formation of a fatigue nanocrack ($\sim$300 nm long) near the fracture surface. A large amount of dislocation-based plastic deformation at the tip of this fatigue crack is observed in Fig. 7(c) and (e). Fig. 7(e) also reveals embryos of surface cracks near the fatigue nanocrack (which can also be seen in Fig. 8(e)). The area near the fracture surface is also associated with large densities of dislocations (see Fig. 7(c) and (d)). A comparison of the SAED patterns away from the fracture surface and near the fracture surface (see Fig. 7(f) and (g)) suggests that near the fracture region a number of neighboring grains rotated into strong diffracting conditions for either the (111), (200) or (220) planes. The fatigue-related microstructural events leading to the formation of this nanocrack and fracture of the specimen were not captured given that the beam was turned off when failure occurred. However, the microstructural evolution was observed during the first 500 cycles. Fig. 9 shows a series of bright-field TEM images of a small portion of the specimen at different applied stresses at $N = 400$ cycles, along with a comparison of the same portion at the beginning of the fatigue test. Continuous changes in contrast could be observed for many grains during cyclic loading in bright-field TEM observations, implying that dislocation-mediated processes are active.

**Fig. 6** Selected stress–strain curves for an in situ TEM fatigue test for five different cycles ($N = 1, 10, 2000, 5000$, and 6990). (b) Evolution of the maximum applied stress, $\sigma_{max}$, as a function of cycles. (c) Evolution of the permanent strain under no applied stress, $\varepsilon_{\sigma=0}$, as a function of cycles. (d) Evolution of elastic modulus, $E$, as a function of cycles.

**Fig. 7** (a) Low magnification TEM image of the specimen before fatigue testing. (b) Low magnification TEM image of the same specimen after fatigue failure. (c)–(e) High magnification TEM images of the fracture surface and fatigue nanocrack. (f) SAED pattern of the specimen away from the fracture surface after fatigue testing. (g) SAED pattern of the specimen near the fracture surface after fatigue testing.
in this grain-size regime. Dislocations can also be observed in a large grain in the center, as highlighted with the zoomed area in Fig. 9(f). Fig. 9 also shows twins and stacking faults (see red arrows in Fig. 9(a) and (b)). The occurrence of stacking fault formation and mechanical twinning can be expected for a material like nanocrystalline Au, given its relatively large ratio $\gamma_{usf}/\gamma_{sf} = 2.2$ (unstable stacking fault energy for Au, $\gamma_{usf} = 92$ mJ m$^{-2}$, stacking fault energy $\gamma_{sf} = 41.6$ mJ m$^{-2}$) and its low ratio $\gamma_{usf}/\gamma_{usf} = 1.09$ (unstable twin fault energy, $\gamma_{usf} = 100.6$).

This in situ TEM fatigue test represents the first observation of the formation of nanoscale fatigue cracks in nanocrystalline ultrathin films. A study by Kumar et al. did not observe any fatigue crack formation in nanocrystalline Al films (after more than 10 million cycles) under tension–tension fatigue for maximum applied strains of 0.4% (compared to ~2% strains to failure). In contrast, this Au specimen failed after less than 7000 cycles, which is likely due to the larger maximum applied strains (see Fig. 6(a)).

### 3.6. Outlook

As evidenced with the above results, this experimental technique is well suited to investigate the fatigue properties of these nanocrystalline ultrathin films, especially their fatigue crack nucleation properties. Further quantitative in situ TEM experiments of the Au specimens can be performed to capture the details of the fatigue crack nucleation process, including, importantly, the nature of the irreversible cyclic plastic deformation mechanisms. The quantitative nature of the technique allows investigation of important governing parameters on the material’s fatigue response (evolution of the stress–strain curves, fatigue crack initiation lives), such as the maximum applied stresses and plastic strains, frequency, and load ratio $R$. Another critical aspect of the technique is its ability to perform ex situ fatigue tests (and interrupt them with in situ TEM observations), such that the environmental effects (including the effects of the native surface oxide for reactive nanocrystalline metals) on fatigue crack nucleation can be quantified.

Our technique represents a significant advance in the state-of-the-art of the MEMS-based nanomechanical testing techniques. In essence, the presence of the two capacitive sensors eliminates the constraint of the existing techniques to measure the evolution of the specimen gap to calculate strain (as well as stress) and the deformation of the stiffness beams using relatively low-magnification electron images. Further refinements in the sensing technique should be pursued. For example, it would be more straightforward to directly measure $\Delta C_1$ and $\Delta C_2$ during the test to calculate stress and strain, instead of the current sensing scheme ($\Delta C_1 - \Delta C_2$ with and without a specimen) which requires an iterative procedure to accurately calculate $X_f$ and $X_{1.5}$. Appropriate modifications in the sensing circuit may allow these simpler measurements, which in turn would facilitate the development of fatigue testing schemes with closed feedback control. This technique can also be used for high testing frequencies (up to the kHz regime) or high strain rates (for monotonic tests) given the inherent fast thermal response of the actuator.
4. Conclusions

We employed a MEMS-based tensile testing setup to measure the monotonic and fatigue properties of ultrathin nanocrystalline Au thin film specimens. The novelty of the technique lies in its ability to accurately measure both specimen elongation (with nm resolution) and applied force (with μN resolution) electronically using two identical capacitive sensors, making it a promising quantitative in situ TEM nanomechanical characterization tool. Particularly, the demonstrated technique provides a significant advance with respect to the current state-of-the-art for MEMS-based nanomechanics, such as the capability to use TEM imaging solely for high magnification observations, the capability to perform quantitative in situ TEM fatigue tests with continuous tracking of the material’s response, and the capability to perform similar ex situ fatigue tests to probe environmental effects. A FIB-less manipulation and clamping technique was developed to secure the specimens onto the MEMS device without inducing ion-beam-related damage. The 100 nm thick, 1.5 μm wide nanocrystalline Au specimens have a tensile strength of \( \sim 1.2 \)–\( 1.3 \) GPa, and a total elongation to failure of \( \sim 2.5\% \). The specimens exhibit ratcheting behavior under tension–tension cyclic loading in near stress-controlled conditions. Nanoscale fatigue cracks were observed after nearly 7000 cycles for \( \sigma_{\text{max}} \sim 0.95 \) GPa. The quantitative in situ TEM technique is well suited to study the fatigue crack nucleation mechanisms in these nanocrystalline ultrathin films. This is also a promising technique to investigate the mechanical properties and degradation mechanisms of a large range of nanomaterials.

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References

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