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Permalink

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Journal

Applied Physics Letters, 109(8)

ISSN

0003-6951

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Publication Date

2016-08-22

DOI

10.1063/1.4961683

Peer reviewed

Local and transient nanoscale strain mapping during *in situ* deformation

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(Received 17 June 2016; accepted 15 August 2016; published online 26 August 2016)

The mobility of defects such as dislocations controls the mechanical properties of metals. This mobility is determined both by the characteristics of the defect and the material, as well as the local stress and strain applied to the defect. Therefore, the knowledge of the stress and strain during deformation at the scale of defects is important for understanding fundamental deformation mechanisms. Here, we demonstrate a method of measuring local stresses and strains during continuous *in situ* deformation with a resolution of a few nanometers using nanodiffraction strain mapping. Our results demonstrate how large multidimensional data sets captured with high speed electron detectors can be analyzed in multiple ways after an *in situ* TEM experiment, opening the door for true multimodal analysis from a single electron scattering experiment. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4961683>]

Improving the structural properties of metals such as strength, toughness, and resistance to degradation in extreme environments depends on our ability to understand and control defects that determine the mechanical response of metals.^{1–3} The theoretical upper strength of a material is typically orders of magnitude higher than the global stress required to move an individual defect.² The actual local stress required to move an individual defect is not a well-characterized parameter, but by measuring the elastic strain field surrounding a defect as it starts to move this should be possible. In a traditional mechanical test, a load is applied to a sample, causing the sample to deform. Initially, at stress levels below the yield stress, the sample experiences only elastic strain. However, once the sample yields, the recorded strain is composed of both plastic and elastic components, representing the summation of stretched atomic bonds as well as many defects moving in multiple directions. Thus, in typical mechanical testing experiments there is not a clear connection between the stress required to move defects and the applied stress required to deform a bulk material.

TEM analysis has previously shown the ability to accurately measure the strain field around static dislocations at the nanoscale.^{3,4} In addition to the *ex situ* analysis of defects, *in situ* deformation in the TEM has provided great insight into the fundamental mechanisms occurring during deformation.⁵ The development of *in situ* deformation holders allowing for high-resolution measurements of the load-displacement relationship with sufficient sensitivity to detect transient phenomena such as the nucleation of bursts of dislocations has enabled quantitative correlation with real-time diffraction contrast images revealing these mechanisms.^{6,7}

The development of compression and tension geometries with well-defined gauge sections has further enabled the measurement of time-resolved global stresses and strains from the load-displacement curve.⁵ However, this measured plastic strain is not an intrinsic parameter of a material, but rather is a summed response of a volume of material based on a specific set of defects. Elastic strain is fundamentally the displacement of individual atoms from their relaxed positions; but a robust way to measure the local elastic strain, which subsequently drives defect propagation during deformation, has been lacking. There have been efforts to do so with X-ray diffraction that combine orientation mapping and strain mapping with *in situ* experiments,^{8,9} however, the spatial resolution is not at a level of individual defects and thus is still in essence a volume averaged measurement.

In the present work, we combine nanometer strain-mapping with *in situ* deformation in the TEM, allowing to record the local nanoscale transient strain in the sample. Figure 1 shows the experimental setup used to achieve this result, with an expanded schematic of the full analysis shown in [supplementary material](#), Figure 1. In a conventional *in situ* TEM mechanical test, a sample is deformed in the TEM and a video is recorded during deformation using a pre-selected bright-field or dark-field condition. In a STEM experiment, an image is formed by scanning a probe over the sample and collecting the scattered electrons serially with a monolithic integrating detector, such as an annular dark field detector (ADF). In the present paper, we acquire diffraction patterns (DPs) on a pixelated detector for each beam position with a small convergence angle. The rich diffraction-based dataset then contains all of the structural information of the sample for a given crystallographic orientation, including the reconstruction of images with arbitrarily shaped virtual apertures generated with image processing software.¹⁰ Alternatively, the precise measurement of diffracted peak positions allows for the determination of local strain in the material using Bragg's law. Nanodiffraction strain mapping is not only a

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FIG. 1. Schematic of the experimental procedure. In a conventional *in situ* TEM deformation experiment (top left), a sample is deformed and a video is acquired using a chosen bright- or dark-field condition. Using the diffraction mapping technique, the video comprising of ADF-STEM images is acquired while at the same time diffraction patterns are recorded for each beam position. The series of diffraction patterns contains all the information and allows for reconstruction of virtual dark-field videos and strain maps by determining the distance between diffracted spots.

very accurate technique¹¹ but also very robust;¹² while techniques based on high-resolution electron microscopy or electron holography have stringent sample requirements such as perfect zone-axis or the presence of an unstrained reference region in the field of view, nanodiffraction strain mapping is able to measure strain as long as the corresponding diffracted spots are visible and is therefore well suited for *in situ* deformation where some bending is inevitable. Nanodiffraction strain mapping is largely immune to dynamical scattering effects from large sample thicknesses because the measurement is only sensitive to the position of diffraction discs, not the details of fine intensity structure within the discs. The strain measurement is carried out within each individual diffraction pattern; therefore, the determined strain values are not limited by sample movement.

For the present work, STEM diffraction mapping was carried out using a FEI Titan whose three independent condenser lenses allow forming a sub-nanometer electron probe with a semi-convergence angle of 1.5 mrad using a 40 μm condenser aperture at 300 kV. Strain mapping was carried out by cross-correlation of a template with the diffracted spots for every electron probe position and time. The location of the maximum of the squared cross-correlation images was deduced and reference values for the diffracted spots were computed by averaging over the unstrained sample. For each diffraction pattern, a transformation-matrix was calculated and converted to strain values.¹² The strain maps present an average projection through the TEM foil.

Scanning nanobeam diffraction has previously been limited by the speed of conventional CCD cameras, where recording diffraction data over a 500 nm² field of view with 1 nm² sampling would take almost 3.5 h at 20 diffraction frames per second. While it is possible to perform quasi-static experiments using scanning nanobeam diffraction over these relatively long periods of time,¹³ it is not

experimentally practical for continuous *in situ* TEM experiments that must be performed without stopping the experiment. Recently, the use of millisecond acquisition times of direct electron detectors has been shown to retain the precision of the strain measurement¹⁴ and we have recently demonstrated that it can be used to acquire strain maps of a large field of view.¹² Here, we take advantage of a Gatan K2 IS direct electron detector operating at a frame rate of 400 f/s. The increase in speed by a factor of 20 $\times$ enables us to record diffraction maps *in situ* without pausing or stopping the experiment. In the current setup, the a typical *in situ* experiment takes only a few minutes, whereas collecting the same data set with a conventional CCD camera would take more than an hour. The result is a five-dimensional data set (a time series of 4-dimensional acquisitions consisting of 2-dimensional diffraction patterns recorded for each point in a 2-dimensional raster scan).

To demonstrate the feasibility of our method of *in situ* strain mapping, we deformed an aluminum alloy Al 5754 (nominally Al-3 at% Mg) in tension. Aluminum is not only a good model material as it has a large ductility with a large number of dislocations generated during deformation but aluminum alloys are also important lightweight structural materials. To achieve a well-defined tensile specimen with minimal damage, a single-crystalline specimen was lifted out from the edge of an electropolished TEM specimen via Focused Ion Beam (FIB) machining. An FEI Strata 235 FIB/SEM equipped with an Omniprobe micromanipulator was used to cut a rectangle (about 8 $\times$ 2 μm in size) from the electron transparent region of the sample and transfer it to a Hysitron Push-to-Pull device. The Push-to-Pull Device transforms the compressive motion of an indenter into an *in situ* tensile test.¹⁵ The sample was attached using a Pt-deposition system and cut into a miniaturized dogbone for *in situ* TEM tensile tests with a width of 260 nm, a length of 560 nm, and a thickness of 140 nm (cf. Figure 2(a)). Figure 2(b) shows a bright-field image of the tensile specimen with contrast resulting from bend contour and dislocation structures.

Upon loading, the tensile specimen shows purely elastic deformation followed by a plastic deformation and hardening regime. During plastic deformation, dislocations in the

FIG. 2. Tensile specimen. (a) SEM image of the dog-bone shaped Al alloy specimen attached to a Push-to-Pull Device using Pt-clamps. (b) TEM bright-field image of the gauge section of the tensile specimen.

aluminum alloy form and move in a jerky manner. The dislocations frequently stop and interact. A representative video of a similar tensile sample deformed *in situ* and recorded only in TEM bright-field mode is shown in the [supplementary materials](#). A high density of mobile dislocations is visible; however, the local strain cannot be determined from the video formed using a single imaging condition (supplementary movie 1).

To analyze the local transient strain during deformation, the sample was deformed in tension at a strain rate of 0.2 nm/s. Figure 3(a) shows the stress-strain curve recorded with a Hysitron Picoindenter during deformation. The engineering stress-strain curve was calculated from the force-displacement curve taking into account the sample geometry without taking into account the machine compliance and other sources of compliance. The sample shows a region of mainly elastic deformation followed by a plastic deformation regime reaching a plateau of around 0.7 GPa and finally fracturing at a strain of about 10%. A simultaneous movie was acquired comprising 22 strain maps having a size of 64×64 pixel and a spatial resolution of 2.8 nm. Each strain map, determined from over 4000 diffraction frames, took only 10 s for acquisition. A schematic drawing of the tensile bar

with the indicated region where the experiment was carried out is shown in Figure 3(b). To correlate the strain maps during the deformation, each strain map was followed by five fast STEM images acquired to obtain a more continuous video. Figures 3(c) and 3(d) show two different time steps extracted from the experiment; the full video can be found in the [supplementary materials](#) (supplementary movie 2).

Figure 3(c) shows the sample towards the beginning of deformation. The strain-map shows strain inhomogeneities due to the presence of dislocations. The corresponding ADF image shows complex contrasts due to bending and the high density of defects in the sample. The mean diffraction pattern (DP) obtained by summing all the DP shows that the sample has a [112] zone axis-orientation. Figure 3(d) shows the same series taken from a time step towards the end of the experiment, when the sample is under greater strain. Deforming the sample leads to an increase of the elastic strain in the sample and to variations in the inhomogeneities in the strain field due to defect motion. After fracture, the strain goes down again, but inhomogeneities in the strain field persist due to the presence of remaining dislocations.

FIG. 3. *In situ* strain mapping during tensile testing. (a) Engineering stress-strain curve deduced from the Hysitron Picoindenter (green solid line) overlaid with a color-coded histogram of the local stresses as calculated from the measurements of the local strains (blue to red). The position of each histogram is determined by the time of the scan. Fracture of the sample occurred just after 10% strain. (b) Schematic drawing of the tensile bar, the region studied by *in situ* strain mapping is indicated. (c) and (d) Two frames extracted from the *in situ* strain mapping movie; the time is indicated in the stress-strain curve. In addition to the color-coded strain map, an ADF image and the average DP are shown (c) Initial state of deformation. (d) Upon tension the mean strain increases.

To compare the global stress with the local transient stress in the sample, the local strain was converted to a stress assuming linear elasticity and using the modulus of elasticity of single crystalline pure aluminium in $\langle 110 \rangle$ direction = 72.6 GPa.¹⁶ It is important to point out that the local stresses were calculated from the local elastic strains given in the strain maps. A histogram of local stress values was computed for every diffraction strain map. In Figure 3(a), the color coded histogram is overlaid on top of the global stress-strain curve measured using the indentation holder, with each scan synchronized by time with the global stress-strain curve. The mean of the local stresses follows the global stress measured by the indentation holder well but does not fully represent the wide spread in the actual local stresses responsible for generating and initiating motion of defects. While the spread increases during deformation due to formation of dislocations there is already a considerable spread before deformation due to a high density of preexisting dislocations. The spread in the local strain corresponds to around 0.3%. Taking into account that the precision of the nanodiffraction strain mapping technique is around 0.1%,¹¹ it can be concluded that this spread is considerably larger than the statistical error. The agreement of the mean local stress with the global stress demonstrates the applicability of the presented *in situ* strain mapping method and the ability to measure sample stresses without the need of a quantitative deformation holder and independent of the sample geometry. The spread in the local stresses caused by the high density of dislocations shows the importance of measuring the local strain field.

To study an initially defect free material, a second *in situ* test was carried out. For this experiment, a cube corner indenter was used to indent a polycrystalline pure Al thin film evaporated onto silicon wedge substrates.⁷ The resulting strain maps shown in the [supplementary materials](#) (Figure S3) reveal nanoscale strain fluctuations, demonstrating that in the case of the *in situ* nanoindentation, similar nanoscale strain measurements can be carried out. While the combination of elastic and plastic strain can be measured from the elongation of the sample or more precisely using digital image correlation, the knowledge of local elastic strain is important for interpreting the mechanical response of a material as it is typically not possible to separate elastic from plastic strain in the plastic regime. While the plastic strain is simply an indication of the accumulated deformation of the sample, the local elastic strains are an accurate measure of the local stress fields associated with defects and the fundamental processes driving the deformation that cause failure. The technique demonstrated here is able to provide time-resolved local elastic strains and thus the local time-resolved stresses in the sample with a few nanometer resolution. Our results also demonstrate the power of acquiring large datasets and analyzing them after the experiments. There is efficiency in only recording a single pre-selected diffraction condition during an *in situ* test, but there is great power in recording all of the diffraction conditions and deciding which one(s) to analyze after the test. Synchrotron experiments have already combined large data processing with *in situ* deformation or even tomography,^{8,9} but only TEM offers the resolution needed to analyze the fundamental

deformation processes happening at the scale of individual defects. The strain-maps shown in the present case have a spatial resolution better than 3 nm. As has been shown by prior work, nanodiffraction strain mapping with convergent beams can reach resolutions better than 1 nm, with the strain-measurement precision better than 10^{-3} .¹¹ In addition, the resolution of nanodiffraction strain mapping can be easily tuned to the specific need with the only constraint being the signal/noise of each individual diffraction pattern.

In conclusion, we have presented an experimental method to determine the local transient strain during *in situ* TEM deformation. Nanodiffraction patterns are recorded for each beam position during STEM acquisition and the strain field is calculated from the peak positions. We applied this method to an Al alloy deformed in tension and pure Al during nanoindentation with nanometer resolution. While the mean of the local stresses calculated from the strain measurements during tension shows a good correspondence with the global stress measured with the deformation holder, there is a large spread in local stresses associated with the dislocations. The local strain field represents a transient state during deformation and we carried out the measurements without stopping or pausing the experiment. The time resolved strain maps acquired during deformation show that the elastic strain varies considerably at the nanoscale, showing the importance of local *in situ* strain measurements in the TEM. The current result demonstrates the potential for research that enables the measurement of the transient local strain associated with fundamental deformation processes occurring on the nanoscale, such as dislocation interactions. A clear understanding of quantitative stresses and strains at the nanoscale will allow the comparison with theoretical models and thus facilitate the understanding and design of advanced materials.

See [supplementary material](#) for supplemental methods, materials and videos.

The authors acknowledge support by the Austrian Science Fund (FWF): [J3397], the National Science Foundation CMMI/MoM program under GOALI Grant No. 1235610, and the Molecular Foundry at Lawrence Berkeley National Laboratory, which is supported by the U.S. Department of Energy under Contract No. # DE-AC02-05CH11231. We would also like to thank U. Dahmen for helpful discussions.

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