Influence of bulk pre-straining on the size effect in nickel compression pillars

A.S. Schneider a,*, D. Kiener b, C.M. Yakacki c, H.J. Maier d, P.A. Gruber e, N. Tamura f, M. Kunz f, A.M. Minor g, C.P. Frick b

a INM—Leibniz Institute for New Materials, Campus D2 2, 66123 Saarbrücken, Germany
b University of Leoben, Department of Materials Physics, Jahnstr. 12, 8700 Leoben, Austria
c Department of Mechanical Engineering, University of Colorado Denver, Denver 80217, USA
d University of Paderborn, Lehrstuhl für Werkstoffkunde (Materials Science), 33098 Paderborn, Germany
e Karlsruhe Institute of Technology, Institute for Applied Materials, Kaiserstr. 12, 76131 Karlsruhe, Germany
f Advanced Light Source (ALS), Lawrence Berkeley National Laboratory (LBNL), 1 Cyclotron Road, Berkeley, CA 94720, USA
g Department of Materials Science and Engineering, University of California, Berkeley, and National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
h University of Wyoming, Mechanical Engineering Department, 1000 East University Avenue, Laramie, WY 82071, USA

Article history:
Received 17 May 2012
Received in revised form 9 August 2012
Accepted 13 August 2012
Available online 19 August 2012

Keywords:
Nickel
Plasticity
Dislocations
Size effect
Micropillar compression
Electron microscopy

A R T I C L E  I N F O

1. Introduction

It is well known that microstructural features which influence dislocation motion dictate mechanical strength in metals [1]. However, as specimen dimensions approach the micron and nanometer regime, metals that are manufactured such that they have a pristine microstructure demonstrate yield strength values near the theoretical strength [2–6]. In such cases, it is believed that these ultra-high stress values are associated with dislocation nucleation in an otherwise defect-free crystal [3,5–8]. Often it is observed that once this strength has been reached, the stress required to continue plastic deformation decreases drastically [2]. However, fabrication processes that produce relatively defect-free metals represent only a fraction of the possible microstructures over this size scale. A wide range of small-scale manufacturing techniques continue to emerge, which inherently produce microstructural features likely to dominate dislocation nucleation and motion.

A powerful technique used for small-scale deformation studies is the compression of focused ion beam (FIB) manufactured micro/nano-pillars (e.g. see recent reviews [9–11]). To date, this technique has primarily been used to test single crystal samples cut from bulk face centered cubic (FCC) metals, ranging in diameter from tens of microns down to hundreds of nanometers [12–17]. Experimental results have clearly shown mechanical strength values well below the theoretical strength, although a strong correlation to the pillar diameter has been observed. As diameter decreases, the relationship that best fits the collective yield strength data is approximately \( \sigma_y \sim d^{-0.6} \) [9]. However, deformation characteristics inherent to FIB manufactured compression...
pills make yield strength measurements extremely sensitive to the specific measurement technique used. Deformation behavior in the micron and sub-micron regime demonstrate intermittent strain bursts directly observed to be correlated to dislocation motion [18,19] both in load-controlled [13–17] and displacement-controlled [18–21] compression. Furthermore, studies have identified relatively low strain hardening rates or even negative strain hardening for individual compression tests [15,22,23], as well as drastically increased size dependent strain hardening behavior with decreasing diameter [14,16–18,24].

Several theories have been proposed to explain the compressive behavior of FIB machined pillars. The first explanation developed by Greer and Nix assumes that dislocations leave the pillar at the surface before can occur multiplication, thus requiring high stresses in order to nucleate new dislocations [15,22]. This theory has been substantiated by two-dimensional discrete dislocation simulations (2D-DDS) [25,26], and in situ transmission electron microscopy (TEM) results that show dislocations exiting 150–300 nm diameter [111] oriented Ni pillars upon loading [19].

The results were explained as a competition between the dislocation annihilation rate on the pillar surface relative to the dislocation activation/nucleation rate, leading to a progressive exhaustion of dislocation sources and therefore a hardening response. An alternate explanation for the size effect was originally proposed by Parthasarathy et al. [27] based on the availability of dislocation sources. For small-scale samples, dislocation sources were assumed to be controlled by the largest average distance between internal pinning points and the free surface. Both in situ TEM [28,29] and 3D-DDS studies [30–32] have illustrated the feasibility of such single arm sources. Furthermore, Rao et al. [30] introduced the term exhaustion hardening, which refers to the hardening response when a dislocation source stops operation due to typical forest hardening processes. One key characteristic of exhaustion hardening is that dislocations are not required to leave the pillar [33]. A small amount of dislocation accumulation is expected [30], which has been indirectly substantiated by post-mortem TEM investigation [17,34].

However, it is important to note that dislocation starvation and exhaustion hardening are not necessarily mutually exclusive [28], and the possibility exists that microstructural properties of the pillar, the pillar size, and/or the testing parameters may dictate which mechanism is dominant [23]. For example, in situ TEM investigation of submicron single crystal aluminum demonstrated that the dislocation density remained constant over a large period of strain and increased only with a sudden increase in strain rate [29]. Copper compression pillars with diameters of 125 nm or below demonstrated a strain-rate sensitivity, explained by a transition from a single-arm source dominated exhaustion hardening regime for micron pillars to the starvation/surface-source dominated regime for nano-pillars [35]. Small-scale aluminum pillars which had been coated, filled, or contained a grain boundary demonstrated a suppression in strain bursts, thought to be caused by preventing the dislocations from annihilating on the free surface, allowing for accumulation and multiplication [36,37].

The initial dislocation density and its arrangement is believed to have a significant effect on the size effect, although the vast majority of experimental studies have used well-prepared samples with bulk dislocation densities thought to be on the order of 10^{12} m^{-2} [38]. Pre-strained [001] oriented Au pillars showed that a pre-existing dislocation density as high as approximately 10^{15} m^{-2} prior to FIB manufacture did not influence the strength results for pillars with aspect ratios larger than 2:1 [23]. It was proposed that plasticity was controlled by dislocation nucleation on the pillar surface, and that pre-existing dislocation networks unravel, eventually becoming starved of mobile dislocations. Pre-straining of FIB manufactured Mo pillars, followed by further FIB cutting and re-compression illustrated no significant influence on the size effect [39]. In this study by Schneider et al. [39], the strain hardening behavior typical for small-scale pillars was erased upon re-FIB cutting, clearly demonstrating that pillar hardening behavior cannot be caused by forest hardening mechanisms, although no post-mortem TEM was shown to directly demonstrate the post-deformation dislocation density. Further, a recent 2.5 DDS study explicitly designed to investigate the effect of initial dislocation density observed a suppression of the size effect for high densities [40]. It was concluded that both specimen size and initial source density caused a shift from forest hardening to exhaustion hardening dominated behavior.

The purpose of this research is to systematically investigate the effect of increasing initial dislocation density on the size dependent strength behavior of FIB manufactured micro/nano-pillars for nickel, which is one of the most extensively studied FCC materials [14,17,19]. A large [111] oriented Ni specimen was sectioned into four cylindrical samples, and compressed to increasing strains (5%, 10%, 15% and 20%). The bulk pre-strained specimens were carefully sectioned, electropolished, and small-scale compression pillars were milled into the surface. Results show that yield strength and strain hardening rate is strongly correlated to the amount of pre-straining. For pillars in the micron regime, pre-straining increases the strength as a function of pre-straining magnitude. For pillars in the sub-micron regime, strength values for all samples converge. TEM and micro-Laue diffraction investigation of the bulk specimens revealed an inhomogeneous dislocation cell structure, with average dislocation density scaling with the pre-straining amount. In situ TEM on sub-micron and micro-Laue diffraction on micron sized compression samples demonstrated that the dislocation density does not change drastically during compression. Results of this study are discussed in context with previous small-scale pillar work.

### 2. Experimental methods

A high-purity, single crystal Ni sample with [111] crystallographic orientation was produced by the Czochralski method and subsequently sectioned into four equally sized compression samples of approximately 20 mm height and 7 mm diameter. These samples were compressed to engineering strains of 5%, 10%, 15% and 20% with a mechanical tester (Instron 5567, Norwood, MA) equipped with a 30 kN load cell. Samples were compressed to the desired strain levels at a strain rate of 0.001 s^{-1} with data collection at 10 Hz. Samples were then unloaded to zero stress at the same strain rate with the exception of the 15% strained sample, which was manually unloaded using the instrument controller. After compression, thick slices were prepared out of the center of each deformed sample using electron discharge machining, and were subsequently mechanically and electrochemically polished in order to remove the damage layer induced by the cutting procedure. The orientation of the final surface was measured using electron back-scattered diffraction (EBSD). It was observed that the orientation of the final planar surfaces were several degrees off from the initial [111] direction. Therefore, in order to account for the slightly different bulk orientations, the yield stresses as well as the strain hardening rates determined from the pre-strained samples were in the later analysis resolved on the slip system with the highest Schmid factor as determined by EBSD.

To conduct the ex situ compression experiments, pillars with diameters ranging from 200 nm to 6 µm were machined on the surfaces of the as-prepared samples using a FEI Strata 235 focused ion beam microscope (FIB) operated at 30 keV. The final milling current during annular milling was 10 pA. During FIB machining...
the sample surface was held normal to the ion beam, which produced pillars with a taper of approximately 2–3° and a length to diameter aspect ratio of about 3:1. The pillars were compressed in load-controlled mode using an MTS XP nanoindenter equipped with a 10 μm diamond flat punch. All tests were performed with a constant stress rate of 20 MPa s⁻¹. For consistency with previous work on as-received Ni [17], the pillar top diameter was used to calculate engineering stress–strain data. To get an accurate measure of strain, the elastic deformation of the base material and the indenter was estimated using elastic contact theory, and subsequently subtracted from the overall displacement. The testing methodology and the data analysis are identical to that used in previous work [17].

In order to investigate the dislocation arrangements and densities within the material, 1.5 mm thick disks were cut from the 5% and 20% pre-strained Ni samples using a low-speed diamond wheel. Next, the disks were ground down to 0.6 mm thickness, and electron-transparent foils were finally obtained using a chemical method. For TEM, bright-field imaging two-beam conditions were employed whenever possible and standard textbook approaches [41] were used to determine the dislocation densities.

For the in situ TEM experiments, a small piece of 2 mm × 2 mm was cut from the 5% pre-strained Ni single crystal and electrochemically thinned to a wedge. On top of this wedge pillars with top diameters ranging from 180 nm to 2000 nm and aspect ratios (length: diameter) > 5:1 were milled. Sample testing was performed using a Hysitron Picoindenter with a Perfomech controller in situ in a Jeol 3010 TEM operated at 300 keV. Compression tests were run under displacement control with nominal displacement rates of approximately 1 nm s⁻¹ resulting in strain rates of about 10⁻³ s⁻¹. During the tests the load-displacement data were collected with 1 kHz and a video was recorded using a Gatan CCD camera at 30 frames per second. A detailed description of the setup used and the data evaluation based on a correlation between the recorded in situ testing video and the measured mechanical data can be found in Refs. [18,42].

Micro-Laue diffraction experiments were performed at the micro-diffraction beamline 12.3.2 of the Advanced Light Source (ALS) at the Lawrence Berkeley National Laboratory. The experiments were carried out using a polychromatic X-ray microbeam with energies between 5 and 22 keV. The full width at half maximum (FWHM) of the beam was around 0.9 μm in vertical and 1.0 μm in horizontal direction. Details of the beamline and data analysis are described elsewhere [43,44]. All samples were scanned in the X-ray beam with a step size of 1 μm. The region of interest was defined using fluorescence signal maps, which have been recorded prior to the collection of the Laue patterns. Laue diffraction patterns of the bulk material were taken for the 0%, 5%, and 15% pre-strained material. Additionally, Laue diffraction patterns of an individual pillar with 5% pre-strain and a top diameter of 1 μm were recorded. This pillar was identified based on a map of the integrated intensity of the complete individual Laue diffraction pattern. Using this method, the silhouette of the individual pillar can be detected and discriminated from the base material. The micro-Laue diffraction experiment on the individual pillar was performed before and after compression testing.

3. Results

3.1. Bulk testing & analysis

Single crystal Ni samples were pre-strained in a macroscopic compression test in order to increase the dislocation density in the bulk material. The stress–strain curves of four samples loaded to increasing strain are shown in Fig. 1. Relative to the other bulk samples loaded to nominally 5%, 10% and 15% strain, the sample deformed to 20% shows a decrease in strain hardening starting at approximately 13%. This was caused by plastic instability of the sample during the test. The top of the sample sheared relative to the bottom half, diminishing the cross sectional area, and thereby decreasing the load required for further deformation. Therefore, stress–strain values of this sample were not used for calculating a dislocation density. However, considering that the purpose of this pre-straining was to systematically increase the dislocation density, results from the 20% strained sample were utilized for further micro-pillar testing. This is justified by TEM results which show a dislocation network similar to samples deformed to lower strains (discussed later in the text).

The stress–strain data of the bulk tests demonstrate significant strain hardening, which is generally related to the accumulation and interactions of dislocations [45–47]. The parabolic shape of the macroscopic stress–strain curves is typical for multislip orientations [48], which is indicative of the [111] orientation as confirmed by EBSD and Laue diffraction (discussed later in the text). From the data shown in Fig. 1, the strain hardening rate (SHR) and the dislocation density required for the observed hardening were determined using the Taylor relation [49] and are listed in Table 1:

\[ \rho = \left( \frac{\sigma - \sigma_0}{2M\mu} \right)^2 \] (1)

where \( \sigma \) is the flow stress, \( \sigma_0 \) is the yield stress of the bulk single crystal (50 MPa, as determined from the stress–strain behavior of the bulk Ni samples), \( \alpha = 0.3 \) [50] is a numerical constant, \( \mu = 76 \) GPa is the shear modulus [51], and \( b = 2.49 \times 10^{-10} \) m [51] is the parabolic shape of the macroscopic stress–strain curves is typical for multislip orientations [48], which is indicative of the [111] orientation as confirmed by EBSD and Laue diffraction (discussed later in the text).

![Fig. 1. Stress–strain behavior of [111] oriented bulk Ni crystals. Samples were pre-strained to 5% (black), 10% (red), 15% (green) and 20% (blue).](image)

<table>
<thead>
<tr>
<th>Sample</th>
<th>SHR (GPa)</th>
<th>SHR ( \times f_t^2 ) (GPa)</th>
<th>( \rho ) (m⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5%</td>
<td>3.8</td>
<td>0.28</td>
<td>( 5.9 \times 10^{13} )</td>
</tr>
<tr>
<td>10%</td>
<td>3.4</td>
<td>0.25</td>
<td>( 9.2 \times 10^{13} )</td>
</tr>
<tr>
<td>15%</td>
<td>4.7</td>
<td>0.35</td>
<td>( 3.2 \times 10^{14} )</td>
</tr>
<tr>
<td>20%</td>
<td>4.7</td>
<td>0.35</td>
<td>–</td>
</tr>
</tbody>
</table>
the magnitude of the Burgers vector, $M$ is the reciprocal Schmid factor, and $\rho$ is the total dislocation density. The SHR of the macroscopic samples is defined as the slope between the yield stress (50 MPa) and the stress at 5% strain. For later comparison, also shown in Table 1 is the SHR resolved on the most favorable slip system. It is calculated by multiplying the SHR with the Schmid factor ($f_S$) squared (0.27 for the [111] orientation).

The Ni single crystals exhibit resolved SHR that range from 0.25 to 0.35 GPa. These SHRs are typical for crystals initially oriented for multiple slip ($\mu/300 \sim 0.25$ GPa) such as the [111] orientation, which has six slip systems with equal Schmid factors. With regards to the dislocation density, values from $5.9 \times 10^{13}$ to $3.2 \times 10^{14}$ m$^{-2}$ were calculated using Eq. (1). As expected, the dislocation density increases with increasing strain.

In order to quantify the dislocation arrangement within the bulk material in more detail, two approaches were used to characterize the microstructure prior to pillar compression: TEM analysis of random sections taken from the bulk pre-strained samples and Laue diffraction on the surface of the bulk crystals. Fig. 2 shows representative bright field TEM micrographs of 0%, 5%, and 20% pre-strained bulk specimens. The undeformed material microstructure consists of several dislocation bundles spaced apart from one another. After deforming to a pre-strain of 5%, dislocation cell structures with relatively thick and cloudy cell walls are discernible. This is typical for a deformed FCC single crystal with multiple slip compression axis [52]. The higher pre-strain of 20% is not observed to obviously affect the cell diameter, but leads to well defined and narrow cell walls, with a much higher dislocation density within the cell walls. An analysis based on an intersection concept [41] and correcting for the fraction of invisible dislocations was performed on multiple micrographs to quantify (1) the average cell diameter, (2) the dislocation density inside the cells, and (3) the dislocation density of the cell walls. Based on the average area fractions of cell wall and cell interior, a total dislocation density was calculated as well. The results are summarized in Table 2. By comparing Tables 1 and 2 it can be

<table>
<thead>
<tr>
<th>Sample</th>
<th>Cell diameter (µm)</th>
<th>Dislocation density in cells (m$^{-2}$)</th>
<th>Dislocation density in walls (m$^{-2}$)</th>
<th>Total dislocation density (m$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0%</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>$8.4 \times 10^{13}$</td>
</tr>
<tr>
<td>5%</td>
<td>0.67 ± 0.23</td>
<td>$9.7 \times 10^{13}$</td>
<td>6.5 $\times 10^{14}$</td>
<td>3.7 $\times 10^{14}$</td>
</tr>
<tr>
<td>20%</td>
<td>0.76 ± 0.50</td>
<td>2.0 $\times 10^{13}$</td>
<td>1.1 $\times 10^{15}$</td>
<td>4.5 $\times 10^{14}$</td>
</tr>
</tbody>
</table>

Fig. 2. TEM images showing the initial dislocation arrangement in the 0% (a), 5% (b) and 20% (c) pre-strained Ni bulk samples. The dislocation density in the 0% sample is already fairly high. In the 5% and 20% pre-strained samples dislocation cells with a size in the submicron range are observed. The cells are more evolved in the 20% pre-strained samples. Especially the dislocation density within the cell walls is much higher for the 20% pre-strained sample.
seen that the dislocation densities determined from the TEM images are similar and follow the same trend as those calculated from the stress–strain data.

In Fig. 3 Laue diffraction patterns of 0%, 5% and 15% pre-strained Ni bulk materials are shown. The Laue spots of the unstrained material exhibit an almost symmetric circular peak shape, which is typical for an undeformed single crystal containing a fairly high density of statistically stored dislocations, in qualitative agreement with the TEM observations shown in Fig. 2. For the pre-strained samples the peak shape changes drastically. Here, clouds of diffraction spots are observed, reflecting the mosaic-like arrangement of dislocation cells and walls within the deformed single crystals. The heterogeneous structure and angular width of the diffraction clouds increases only slightly from 5% to 15% pre-strain. The angular width of the clouds is about $8.4\pm 0.5^\circ$ for 5% pre-strain and about $8.7\pm 0.5^\circ$ for 15% pre-strain.

3.2. Micro-pillar compression

Fig. 4 shows representative stress–strain curves of pillars with diameters ranging from 200 nm to 5 μm, machined onto the well-prepared surface of the 5% and 20% pre-strained Ni samples. These pre-straining values were chosen to represent the deformation behavior of the Ni pillars over the pre-strain range tested in this study. Stress–strain data for the unstrained material was published previously [14,17]. The 5% pre-strained pillars show features indicative of load-controlled pillar compression: size-dependent yield strength and strain hardening, as well as intermittent plastic burst behavior [10]. Pillars with diameters larger than approximately 1–2 μm demonstrate relatively continuous stress–strain curves, while smaller pillars exhibit frequent strain-bursts separated by nearly elastic loading segments. The 20% pre-strained pillars exhibit similar shapes for the stress–strain curves, with slightly larger and less frequent strain bursts, but the size dependence of the yield strength and the strain hardening behavior is different compared to those of the 5% pre-strained pillars.

From the stress–strain data shown in Fig. 4 it appears that for small pillar diameters the hardening is more pronounced for the

---

**Fig. 3.** Laue diffraction patterns of the bulk of Ni single crystals with 0%, 5% and 15% pre-strain. The sample with 0% pre-strain exhibits mainly radial broadening, while samples with 5% pre-strain or more show clouds of diffraction spots. This reflects the formation of a mosaic cell structure during pre-straining. The angular width of the cloud only slightly increases from 5% to 15% pre-strain. Note that the Laue pattern of the sample with 0% was taken with a different detector.

**Fig. 4.** Representative stress–strain curves for [111] oriented Ni pillars with diameters in the range of 200 nm to 5 μm machined in the 5% (a) and 20% (b) pre-strained Ni samples.
5% bulk pre-strained sample. This is quantified in Fig. 5, which shows the strain hardening rate (SHR) of the Ni pillars as a function of pillar diameter. Due to the intermittent flow of the smaller pillars, the SHR is defined as the slope between 2.5% and 10% strain, consistent with previous work on small-scale pillar studies [16,17]. For comparison, data from unstrained Ni pillars with [111] orientation were added. In order to take into account the different orientations of the samples, the strain hardening rate was resolved onto the most highly stressed slip system for the respective crystal orientation. Irrespective of the amount of bulk pre-straining, larger pillars show SHRs close to the values calculated for the bulk crystals (Table 1). Below approximately 1 μm the SHR increases significantly to values several times larger than bulk. The highest SHR values are observed for the 0% and 5% strained samples, while the 20% strained samples are least affected.

Fig. 5. Strain hardening rate (SHR) versus pillar diameter for pre-straining values of 5%, 10%, 15% and 20%. For comparison, representative data for micro-pillars without pre-strain are shown as well, taken from [17]. In order to take into account the different orientations of the samples, the SHR was multiplied by $f_s^2$.
pre-strained samples. All pillars deformed predominately on a single slip system; however, the activation of less favorable additional slip systems was also detected.

In order to quantify the effect of bulk pre-straining on the size dependent plastic strength of the Ni pillars, Fig. 7 shows the critical resolved shear stress (CRSS) at 2.5% strain as a function of pillar diameter for the different bulk pre-strains. For comparison, representative data for micro-pillars without pre-strain are shown as well, taken from Refs. [14,17]. It can be seen that the scaling relationship between CRSS and sample diameter, as well as the strength level of the Ni pillars, is affected by the amount of bulk pre-straining. In particular, for pillars with diameters above approximately 1 μm the strength increases with the amount of pre-strain, while for pillars with smaller diameters, the bulk pre-straining has only a little effect on their flow strength. Consequently, the size dependence decreases with the amount of pre-strain. If only the size dependence well in the submicron range is considered, with the limited data available it seems that the size dependence of the pre-strained pillars approaches those of the 0% pre-strained pillars.

3.3. In situ experiments

Fig. 8 shows SEM images and the peak shape of the (22-6) Laue diffraction peak for a 1 μm diameter Ni [111] pillar, prepared by FIB milling from 5% pre-strained Ni, before and after microcompression testing. Already in the uncompressed state, the peak shape is very complex, resultant of the pre-straining and FIB preparation. The mosaic nature and angular width of the peak is certainly smaller than for the bulk material, due to the smaller volume that is cut out of the bulk material and thereby probed by the white X-ray beam. Still, the pillar interior consists of different
cells and a network of cell walls. This is the likely reason that the peak shape resembles a horseshoe and is not as wide and crescent-shaped as the diffraction pattern of the pre-strained bulk material (Fig. 3). Interestingly, the shape of the (22-6) diffraction peak does not significantly change after the micro-compression test. The individual spots have become slightly more diffuse, indicating that the complex dislocation arrangement within the dislocation walls may have been slightly modified during pillar deformation. The SEM image of the deformed pillar clearly shows well-defined slip traces extending through the top portion of the pillar, indicating that the pillar deformation has been carried predominantly by slip along single slip systems. As no additional streaking can be detected, it seems that slip on these slip systems is rather unconstrained and no dislocations are stored on these slip systems [54].

Fig. 9 shows TEM bright field and dark field images of a 190 nm diameter Ni [111] pillar which was 5% pre-strained prior to FIB fabrication before and after in situ compression using the $g_{110}$ diffraction condition. The 190 nm pillar will not contain a complete dislocation cell; however, given the high aspect ratio of 5:1, it will in any case contain at least a fraction of a dislocation cell wall. Unfortunately, the high FIB dislocation density of approximately $10^{15}$ m$^{-2}$ near the sample surface prevents a detailed analysis of the internal dislocation arrangement. However, the high defect density...
present in the initial sample is not mechanically annealed [19] during loading but remains to a large extent inside the tested volume. Besides the two distinct strain bursts resulting in the formation of two slip steps indicated in Fig. 9, the stress-displacement data reveal numerous smaller events at increasing stresses, indicating several operative dislocation sources. The full video is provided as supporting online material in compressed form. Samples with pre-strain larger than 5% were not attempted for in situ testing because the internal stress due to the dislocation networks made both, Laue diffraction and TEM imaging, very difficult.

4. Discussion

4.1. Bulk dislocation density

The aim of this experimental study was to observe the influence of bulk pre-straining on the size effect of FIB manufactured pillars. Bulk samples were compressed to increasing pre-strain values from 5% to 20%, from which small-scale compression pillars were fabricated. The microstructure of the bulk pre-strained specimens was evaluated using both TEM (Fig. 2) and micro-Laue diffraction (Fig. 3), with specific emphasis on characterizing the dislocation arrangement. Both techniques clearly demonstrate that even 5% pre-straining drastically alters the dislocation microstructure. From the streaking of the Laue diffraction spots it is clear that the dislocation arrangement changes significantly from the undeformed to the 5% pre-strained state, then changes only slightly with further increasing pre-strain. Laue diffraction findings are mirrored by results from TEM micrographs that show that a dislocation cell network forms already after 5% pre-straining. Careful analysis of TEM images demonstrate a cell structure approximately 500 nm in diameter, typical for deformed single crystal Ni [55]. Increasing the pre-straining from 5% to 20% does not substantially change dislocation cell size, but the density within the cell walls increases significantly (Table 2). Use of the Taylor relation (Table 1) predicts dislocation densities on the same order of magnitude as observed via TEM; however, it does not manage to capture the actual arrangement of the dislocations in a cell wall structure.

4.2. Effect of pre-straining on micro-pillar compression

In contrast to the bulk material, the pre-strained pillars show the key deformation features of load controlled micro-pillar compression tests. With decreasing diameter, discrete bursts in strain are observed more frequently, which have convincingly been shown to be caused by dislocation motion [18,19,29]. This general understanding is directly confirmed in this study using in situ TEM compression (Fig. 9), which shows dislocation events during compression of a 190 nm diameter, 5% pre-strained pillar. During in situ testing, the nanoindenter was operated in a displacement controlled mode, and therefore dislocation bursts correlate to a drop in load. Stress–strain curves of micro-pillars reveal no obvious systematic change in the burst behavior as a function of increasing pre-strain (Fig. 4), and SEM images post compression show clear slip traces (Figs. 6 and 8). It appears that the higher dislocation density in the pre-strained pillars did not suppress the intermittent dislocation flow of the microcrystals. This is directly substantiated by in situ TEM results shown in Fig. 9. It was reported for just FIB prepared Ni pillars tested with the same equipment by Shan et al. [19] that the samples became completely dislocation free (a phenomenon termed ‘mechanical annealing’) during early loading. Our experiments were performed on pre-strained Ni with similar pillar size and orientation, but using a different imaging condition. There is some loss of near surface dislocations originating from FIB preparation during initial loading, but a substantial defect density remained in the crystal. The two large plastic events resulting in the formation of surface slip steps were triggered by dislocations nucleated from sources near the pillar top in the vicinity of the pillar surface. The internal dislocation structure evolves during compression, and several dislocations annihilate at the surface. However, the residual dislocation density is similar to the initial one, indicating there are multiple sources in the specimen [29].

The micro-Laue diffraction measurements, also from a 5% pre-strained pillar as in the in situ TEM experiment, but with a larger diameter of around 1 μm, compliment the submicron in situ results. Diffraction analysis also reveals that the dislocation density and arrangement does not substantially change during pillar compression, as no distinct streaking and peak broadening were observed in Fig. 8. The slightly more diffuse Laue spots after micro-compression testing indicate that the dislocation walls may have been slightly deformed due to pillar compression and the curvature and entanglement of the dislocations may be changed. This suggests that the slip events, which carry the deformation of the pillar, do not substantially alter the dislocation arrangement introduced by bulk pre-straining. Lattice rotations observed in previous in situ micro-diffraction of FCC pillars [56–58] were not explicitly observed in this work; however, this is likely due to the localized slip deformation near the pillar top (Fig. 8).

For bulk material it is well known that the flow strength increases during straining due to the multiplication and interactions of dislocations. Our results show that as size scale decreases, there is a transition from a bulk-like behavior, to a behavior characteristic for small-scale metal structures. This transition might be the result of different dislocation processes dominating the deformation in (1) micron and (2) sub-micron regime: (1) For pillars in the micron regime, it can be assumed that their strength is likely controlled by cell walls acting as obstacles for dislocation motion. With increasing bulk pre-strain the dislocation density in the cell walls increases significantly. Therefore, these walls become more effective barriers for mobile dislocations, and thus start to control the strength of the pillars. For the highest pre-strains (15% and 20%) the strong cell walls lead to the highest and size independent flow stresses. For lower pre-strains, the obstacle strength is comparably lower, and therefore does not fully suppress the dislocation processes which usually govern the size dependent deformation of FIB prepared micropillars without pre-strain. (2) By contrast, for pillars with diameters well below 1 μm, the contribution of the cell walls becomes more difficult to interpret, since the sample size approaches the cell size and the yield strength values tend to converge, regardless of pre-straining (Fig. 7). As all the pillars tested in this study have aspect ratios larger than 3:1, it is unlikely that pillars were fabricated without intersecting at least one cell wall. However, when diameters decrease below 1 μm, it becomes increasingly likely that pillars in this size range contain only one cell wall or fragments of a cell. However, a description of the influence of pre-straining for sub-micron pillars can be attempted based on the in situ TEM experiments (Fig. 9). Dislocations responsible for the plastic deformation are for the most part nucleated near the pillar top. Residual dislocations due to initial pre-straining further down the sample height remain largely unaltered during compression. This indicates that the initial deformation at very high stresses in the sub-micron range is not dominated by residual pre-straining dislocations.

4.3. Strain hardening rate (SHR)

For larger pillars and all investigated amounts of pre-strain, the SHR is close to the bulk value, strongly suggesting that bulk
dislocation processes, governed by the confined motion of dislocations within the cell interior, dominate the plastic deformation. In this case, any size dependence should relate to differences in the cell size, not the sample dimensions. With decreasing pillar size into the sub-micron regime, a dichotomy in strain-hardening rate is observed. For pillars with little or no pre-straining the SHR increases drastically, while for substantial pre-strain the SHR remains close to the bulk material (Fig. 5). The SHR starts to deviate from the bulk behavior at roughly the same diameter at which the yield stresses of the pillars become insensitive to pre-straining.

Previous in situ TEM studies of sub-micron samples [18,19] have shown that a high SHR is explained by an exhaustion-type hardening. Dislocations are observed to nucleate near the pillar surface in an area of high stress. This source will emit several dislocations until it becomes exhausted, and a less favorable source needs to be activated at a higher stress.

In the current study the situation is altered, since the sub-micron pillars still contain cell walls, as supported by the high dislocation density observed in the deformed sample in Fig. 9. These cell walls resemble a bundle of dislocation segments which can interact upon loading, forming pinning points to support operation of a spiral source. However, after a single operation, the revolving arm will again encounter the cell wall and might get blocked by dislocation interaction. The less dense walls in the 5% pre-strained samples contain fewer dislocations compared to the 20% pre-strained ones. Thus, fewer dislocation sources will be available in the 5% pre-strained pillars, leading to a more pronounced exhaustion hardening and consequently a higher SHR, as evidenced by the staircase hardening in Fig. 9(e). The denser cell walls in the 20% pre-strained material should result in a higher number of entanglements formed, and consequently a reduced exhaustion hardening contribution, as observed in 3D-DD simulations [30].

4.4. Relation to other work

Yield strength observations in this work are in agreement with the previous work by some of the present authors investigating small-scale compression pillars made from oxide dispersion strengthened (ODS) Ni superalloy [59]. In this case, ceramic particles were evenly dispersed throughout the Ni matrix, with spacing much smaller than the pillars tested (i.e. < 100 nm). Similar to the 15% and 20% pre-strained Ni samples used in this study, the ODS Ni demonstrated no size effect, while maintaining strain burst behavior. Similar results have been observed in plastically deformed micro-pillars fabricated from NiTi containing semi-coherent Ti$_3$Ni$_4$ precipitates with spacing and size approximately 50 nm [60,61], and in proton irradiated Cu containing closely spaced stacking fault tetrahedron [62]. It is clear that adding microstructural features that are smaller than the pillar diameter and effective at blocking dislocation motion have a dominating influence on the material strength [1]. The results shown in this study strongly suggest that once the dislocation cell walls created during pre-straining get dense enough to block dislocation motion, they become a sufficient barrier to dislocation motion, and therefore represent the dominant strengthening mechanism.

To fully understand the results from FIB manufactured compression pillars, it is important to reflect on pillars manufactured using alternate techniques. Bei et al. [3] produced initially dislocation free Mo alloy pillars by directional solidification and etching. These pillars exhibited whisker-like behavior during the initial deformation. Pre-straining the whiskers embedded in the metallic matrix and subsequent etching led to a significant weakening of the pillars [5]. For pre-strains ranging from 4%–8%, Bei et al. found jerky stress–strain behavior and size dependent yield stresses, as it is typically observed for FIB machined samples. Conversely, for 11% pre-strain their pillars demonstrated bulk-like flow with continuous stress–strain curves, one order of magnitude lower yield stresses than for the original whiskers, and size independent mechanical properties. The influence of the pre-straining on the Mo alloy pillars is quite different from the results shown in our study. It seems that the main reason for this difference and the significant weakening observed by Bei et al. is that their samples are initially dislocation free. For the unstrained Mo alloy pillars, dislocations have to be nucleated in a perfect crystal, which requires stresses close to the theoretical limit. However, once deformed, residual defects act as nucleation sources, effectively lowering the yield strength of the pillars. By contrast, the initial dislocation density in our pillars is significant due to the defects introduced by the FIB machining and those already present in the bulk material. Although the single crystal used for the present study was grown from a melt, the bulk dislocation density is fairly high as shown by the TEM images (Fig. 2), the radial broadening of the Laue peaks for the undeformed material (Fig. 3a), and the high yield stresses of the Ni single crystals (approx. 50 MPa) in the macroscopic compression test (Fig. 1). In addition, it appears that the pre-straining in the study of Bei et al. is more effective than in our experiments, because already at 11% pre-strain the burst behavior and the size dependent properties of their pillars were suppressed completely. By contrast, at 10% pre-strain we still observe size dependent yield stresses and discontinuous deformation. Only for pre-strains of 15% and 20% the Ni pillar exhibit size independent yield stresses, but still accompanied by strain bursts.

A possible reason for the more effective pre-straining in Bei et al. is that during this bulk deformation the Mo alloy pillars are embedded in a metallic matrix. It is likely that during the constrained deformation of the Mo pillars, the dislocations are multiplied and stored more effectively than in a conventional bulk material. The dislocations are hindered from leaving the pillars by the impenetrable interface enhancing dislocation storage in the material and by this the probability for dislocation multiplication and interactions. In fact, while their initial material is close to defect free, Phani et al. [63] observed an increase from approximately $10^{12}$ m$^{-2}$ at 4% pre-strain to approximately $10^{13}$ m$^{-2}$ for 16% pre-strain, which is more pronounced than in the present case (Table 2). Further, this embedded pre-straining creates surface steps on the pillars that facilitate dislocation nucleation [64]. In addition, the dislocation arrangements introduced by bulk or small-scale fiber pre-straining are completely different. For our bulk material, most of the dislocations are trapped in dislocation cell walls. Therefore, even if the overall dislocation density is high, there is only a small number of mobile dislocations, mainly in the cell interior. As a consequence, new dislocations need to be nucleated from dislocation sources to maintain plastic deformation, explaining the jerky stress–strain behavior of the Ni pillars. Conversely, for the Mo pillars the dislocations were less entangled and only loosely organized [63]. As a consequence, the mobile dislocation density in these pillars is higher relative to the pre-strained Ni pillars. Therefore, in the Mo pillars interaction of mobile dislocations is more probable, leading to bulk-like stress–strain behavior.

In the study by Lee et al. [23] the effects of pre-straining and annealing on the strength of 300 nm Au pillars on MgO substrates were investigated. Two different sample geometries were chosen: high aspect ratio nanopillars with diameters of approximately 400 nm and puck shaped pillars with diameters of about 2100 nm. After pre-straining, the pillars were FIB machined to a final diameter of 300 nm and then mechanically tested. A significant reduction of the yield stress was observed for the pillars machined from the pre-strained pucks, while pre-straining of the nanopillars had no effect on the mechanical properties. Further, it
was found that the samples would deform easier with increasing amount of pre-strain, whereas thermal annealing of the pucks and plastic deformation of the final pillars reduced the stress difference between the pillars originating from puck shaped samples and nano-sized pillars.

The different influence of pre-straining, depending on whether a puck shaped or nanosized pillar was pre-strained, was explained based on the starvation theory [15,65]. Accordingly, pre-straining of nanosized pillars has no influence on the mechanical properties, as the nucleated dislocations are not maintained in the pillar. This is not the case for the puck shaped samples, where dislocation accumulation and storage is more likely due to the larger sample volume. Therefore, the pillars machined out of the puck shaped samples have initially a higher dislocation density than similar sized pillars machined out of nanopillars. In pillars which would otherwise suffer from a lack of mobile dislocations, these additional dislocations might reduce the yield stress by acting as new sources or by reducing the activation stress of pre-existing sources through their stress field.

In our experiments a weakening of the pillars post pre-straining was not observed. For the nanopillars pre-straining had a negligible effect and quite to the contrary for Ni micro-pillars the pre-straining led to significant higher flow stresses. The missing weakening effect for the Ni pillars in our study might be explained by a lower dislocation density realized by the bulk pre-straining compared to the puck compression. Similar to the study by Bei et al., the dislocation motion in the Au pillars is constrained by a hard MgO substrate enhancing the probability for dislocation multiplication and interactions. Consequently, even if the samples in our experiments and in the study by Lee et al. were pre-strained by similar amounts, the dislocation density in the Au pillars on the MgO substrate may still be much higher. Thus, it is not surprising that pre-straining has a more pronounced effect in the work of Lee et al. Furthermore, the unconstrained bulk pre-straining leads to a dislocation structure with volumes of very high and very low dislocation densities as high as 10^{15} m^{-2} [18,19,28]. This value matches the maximum dislocation density deduced from our experiments. The insensitivity of the smaller pillars against pre-straining might indicate that the defects introduced by the bulk pre-straining are not dominant compared to the defects caused by FIB milling. By contrast, for larger pillars the FIB-damage layer takes only a small volume of the total pillar and is therefore not important for their flow strengths [66,67]. Here the dislocation processes in the pillar are more relevant.

5. Conclusions

1. TEM and Laue diffraction analysis of representative sections of the bulk Ni specimens demonstrate that dislocation density increases as a function of applied compressive pre-strain. Dislocation cell structures are observed in all pre-strained specimens. Analysis of TEM images shows that the dislocation density remains in the same order of magnitude for 5% and 20% deformation, although the cell wall dislocation density is significantly higher for the 20% pre-strained specimens. Laue diffraction confirms that the dislocation structure remains relatively similar once a pre-strain of 5% is reached.

2. For pillar diameters larger than approximately 1 μm, pre-straining has a direct influence on the yield strength. The cell walls increase in dislocation density, and therefore become more effective obstacles for inhibiting mobile dislocation motion. As the diameter decreases into the sub-micron regime, which also corresponds to the size of the dislocation cells, pre-straining has little effect on the yield strength, and all specimens converge at approximately 200 nm diameter and a CRSS of approximately 300 MPa.

3. In situ TEM and micro-Laue diffraction of 5% pre-strained pillars demonstrate that the pillar dislocation density remains relatively constant during compression, although the nature of the dislocation structure is altered during deformation. Neither dislocation starvation or mechanical annealing is explicitly observed, nor would they be expected with increasing dislocation density from pre-straining.

4. The pillars tested in this study clearly show a transition in dominant strength mechanisms from the micron range to the nanometer range. For a bulk single crystal, it is well known that dislocation motion is heavily influenced by forest hardening. As sample size decreases from the micron into the sub-micron regime, a size-dependent mechanism begins to dominate, independent of dislocation density.

5. Despite the presence of dislocations within the pillar, in situ TEM shows that dislocations are nucleated near the surface for a sub 200 nm diameter pillar. Therefore, for FIB manufactured pillars, it is believed that defects created during fabrication dominate dislocation nucleation as the diameter decreases, and therefore dictate the pillar yield strength.

6. For lower dislocation densities or smaller diameter pillars, dislocations are expected to leave the pillar with regularity. Sources are observed to become exhausted and consequently less favorable surfaces sources are activated at higher stresses, leading to a size dependent strain hardening rate. However, dislocations become trapped in cell walls as the dislocation density is increased, providing favorable dislocation sources within the pillar. Therefore, the SHR is not drastically affected for the pre-strained material.

Acknowledgments

Special thanks to Arnold Waible from the Max Planck Institute for Intelligent Systems (formerly Max Planck Institute for Metals Research) for growing the Ni single crystal. CPF gratefully thanks Wyoming NASA Space Grant Consortium, NASA Grant no. NNG05G165H, for partial funding of this work. The in situ TEM experiments were performed at the National Center for Electron Microscopy (NCEM), Lawrence Berkeley National Laboratory (LBNL). Synchrotron experiments were conducted at the Advanced Light Source (ALS) at LBNL. Both NCEM and ALS are supported by the Director, Office of Science, Office of Basic Energy Sciences, US Department of Energy under contract number DE-AC02-05CH11231. DK gratefully acknowledges financial support of the Austrian Science Fund (FWF) through the Erwin Schrödinger fellowship J2834-N20.

Appendix A. Supporting information

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.msea.2012.08.055.

References
