Detecting in-plane tension induced crystal plasticity transition with nanoindentation

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Abstract

We present experimental data and simulations on the effects of in-plane tension on nanoindentation hardness and pop-in noise. Nanoindentation experiments using a Berkovich tip are performed on bulk polycrystaline Al samples, under tension in a custom 4pt-bending fixture. The hardness displays a transition, for indentation depths smaller than 10nm, as function of the in-plane stress at a value consistent with the bulk tensile yield stress. Displacement bursts appear insensitive to in-plane tension and this transition disappears for larger indentation depths. Two dimensional discrete dislocation dynamics simulations confirm that a regime exists where hardness is sensitive to tension-induced pre-existing dislocations.

Keywords: nanoindentation; pop-in; dislocation dynamics; crystal plasticity; hardness

While there have been many local microscopic observational tools such as atomic force microscopy (AFM) \cite{1}, scanning electron microscopy (SEM) \cite{2}, or transmission electron microscopy (TEM) \cite{3,4}, it has been elusive to detect the precise onset of crystal plasticity. Most crystals deform plastically beyond the yield strength, but it is difficult to distinguish plastically yielded from unyielded crystalline locations. The principal reason is the fact that crystal plasticity is a subtle effect that involves the atomic-scale motion of defects, dislocations, that alter the local orientation and/or displacement of the crystal depending on
the microstructure \[5, 42, 7, 8, 9, 10, 11\]; in traditional microscopy techniques, the separation of the displacement and orientation contributions due to crystal plasticity represents a non-trivial inverse problem. Nanoindentation is the only existing technique that directly probes mechanical deformation at the nanoscale of an otherwise bulk material. While numerous experimental nanoindentation studies have been conducted to understand nano and micro scale plasticity \[12, 13, 14, 15, 16, 17, 18\], it has been elusive to use surface nanoindentation in order to detect the onset of crystal plasticity.

In a dislocation free region nanoindentation turns from elastic to plastic through a sudden burst on the load-displacement curve, called "pop-in". Such pop-in events have been used to reveal the fundamental mechanisms of the elastic-plastic transition in annealed crystals and they appear to provide a fingerprint of the nucleation of plastic yield of dislocation-starved microstructures \[19, 20, 21, 22\]. Micro and nano indentation have been used to investigate pre-stressed samples using a variety of approaches \[23, 24, 25, 26, 27\]. These studies reported that the hardness remains independent of plane tensile stress if the nanoindentation contact surface is properly accounted for \[28, 29\]. Pop-in event statistics was not investigated. In this paper, we focus on pre-stressed specimens using a custom 4-point bending fixture, and we concentrate only on very shallow nanoindentation depths. We observe a strong dependence of very shallow nanoindentation hardness on pre-existing stress near plastic yielding and consequently, we propose that nanoindentation can be used to detect the crystal plasticity transition.

Commercial aluminum bulk sputtering targets (99.99% purity, Plasma Materials Inc., US) were used in this study. Samples were prepared by the following sample preparation protocol. First, all samples were sliced by electric discharge wire machining (EDM) to a thicknesses of 3mm. The overall sample dimensions were 30mmx75mmx3mm. After EDM machining, the samples were electropolished (Materials Resources LLC, Dayton OH).

A custom-made 4-point loading fixture was designed to apply in-plane stresses at the center sample region prior to nanoindentation. The design details of the
Table 1: Measured total strain and calculated plastic strain and stress

| Deflection, µm | Total Strain, % | Plastic Strain, % | Max. Tensile Stress, MPa |
|---------------|----------------|------------------|-------------------------|
| 0             | 0              | 0                | 0                       |
| 97            | 0.031          | 0                | 19.1                    |
| 226           | 0.065          | 0                | 40.13                   |
| 316           | 0.11           | 0.04             | 41.86                   |
| 403           | 0.15           | 0.08             | 43.21                   |
| 493           | 0.2            | 0.127            | 44.6                    |
| 583           | 0.24           | 0.165            | 45.9                    |
| 717           | 0.31           | 0.233            | 47.12                   |
| 823           | 0.36           | 0.283            | 47.35                   |

4-point loading fixture can be found in the supplementary section. The applied in-plane stresses were calculated by finite element (ABAQUS) simulations according to the strain values on the sample surface measured by strain gauge and sample deflection. Our finite element simulations guaranteed that the top surface (at a range larger than $\sim 100\text{nm}$) is characterized by plane stress conditions. In strain gauge measurements, total strain values were recorded using uniaxial strain gauge with a resistance value of 320 Ω (National Instruments Inc., TX). The measured total strain values and their stress correspondence per screw rotation are listed in Table 1.

Nanoindentation experiments were performed with an iNano (Nanomechanics Inc., TN) nanoindenter with a Berkovich tip. Frame stiffness measurements of the 4-point loading fixture were carried out using a sapphire sample and measured as $1.516\times10^6 \text{N/m}$. It should be noted that the measured frame stiffness values were nearly constant at different stress values. For the purpose of detecting displacement bursts, during indentation the dynamic force oscillation was disabled. However, for the purpose of hardness measurements, this option was enabled and hardness was calculated by using the Oliver-Pharr continuous stiffness methodology (CSM) using a constant indentation strain-rate value of...
Displacement was measured with a differential capacitive sensor with 0.01 nm resolution, and typical values of the drift rate were maintained to less than 0.2 \( \text{nm/s} \).

For all nanoindentation experiments, the following experimental procedure was adopted. First, the aluminum test sample was placed in the 4-point loading fixture, and in-plane stress conditions were created using the motion of a setscrew with each screw rotation corresponding to 70 \( \mu \text{m} \) deflection, and verified with measured strain values. The samples were allowed to thermally equilibrate for an hour before running indentation cycles. The maximum indentation depth levels were set at 50 nm, and tests were performed with constant loading rate of \( \frac{dP}{dt} = 0.5 \text{mN/s} \). A total number of 5000 indents (two 50x50 square grids) were implemented at each of nine stress levels, so the total number indentation was kept at \( \sim 45000 \). All indentation locations were placed far enough from each other (2 to 10 \( \mu \text{m} \)) to avoid interference.

For collecting thousands of indentation points, one of the biggest challenges is experimentation time. In general, the required time to perform only one indentation is approximately 5 minutes resulting in a required instrument time of 3,000 hours for each dataset. This approach is impractical and inefficient for collecting large data sets for statistical analysis. To overcome this limitation, we implemented a new nanoindentation algorithm based on elimination of the unloading portion of the load-displacement hysteresis and scaling the indenter approach rate while keeping the initial contact rate the same as the standard indentation protocol.

In order to investigate the influence of the applied in-plane stresses on the load-displacement curves, we carried out indentation tests across a large surface area of 1x1 mm\(^2\) in the center of the specimens (\( \sim 5000 \) indentations at each stress levels given in Table 1) (Figure 1(a) to Figure 1(d)). In all cases, load-displacement curves show a continuous elastic response followed by multiple measurable displacement bursts. (see Figure 1(a)) The overall behavior is characterized by intrinsic material noise that is attributed to the stochasticity of plastic deformation, especially given that the samples were not thermally
Figure 1: (a) A representative sample of nanoindentation load-depth curves in Al non-stressed samples over a region of 1x1mm on top surface, (b) Detailed load-depth behavior at four randomly selected locations on non-stressed sample top surface, (c) Probability event distribution $P(S)$ as function of event size $S$ (described in text) for applied total strain of 0.031 (●) (blue), 0.065 (■) (green), 0.11 (▲) (red), 0.15 (●) (cyan), 0.2 (▲) (purple). The solid line is a guide to the eye and represents $y \sim x^{-1.6}$. (d) Probability distribution $P(V)$ of the “pop-in noise” intensity $V = \frac{dh}{dt}$ (described in text) (total strain shown in legend.)

annealed. Furthermore, the load-depth slope displays large variation (see Figure 1(b)) which is also attributed to nanomechanical behavior given the large statistical sampling over an otherwise highly homogeneous surface. The pop-in event probability distributions are plotted as a function of different in-plane stress conditions in Figure 1(c). Here, it is worth mentioning that, we define a new parameter, pop-in noise event size $S = \sum_{i \text{for } \delta h > h_{\text{thr}}} \delta h_i$, to analyze the displacement bursts in a quantitative manner, using the threshold value $h_{\text{thr}}$ to be equal to the machine noise threshold $h_{\text{thr}} = 0.2nm$. In Figure 1(c), $S$ is the magnitude of a single displacement burst, while $P(S)$ is the probability density. In Figure 2(d), the probability distribution of the local event intensity $P(V)$ is
presented as obtained data from depth vs. time \((h-t)\) curves.

Figures 2(a) to 2(d) show the variation of hardness as a function of indentation depth at different in-plane stresses. The effect of intermediate in-plane stresses on the depth dependent hardness was considered separately and can be found in the supplementary information. As shown in Figures 2(a-d), the hardness shows a strong dependence on depth for low and high in-plane stress conditions. Hardness values first increase with increasing indentation depth until it reaches a peak (approximately 2 GPa for zero stress and zero strain case) and then decreases towards a plateau at approximately 1 GPa consistent with prior studies [16, 17, 18]. For example, Feng et al. found a softening regime in the single crystal copper films in the depths less than 250 nm, attributing it to tip bluntness at early indentation stages [31].

Despite natural data disorder due to the small indentation depth, there is a strikingly strong correlation between the depth dependent hardness and the applied in-plane stress. At up to 43.21 MPa in-plane stresses, the hardness shifts towards lower values. As it can be seen in Figure 2c, the hardness below depths of 10nm shows a large variability, but with increasing in-plane stress to 47 MPa and total strain to 0.15%. The variation of depth dependent hardness has been a challenging concept for many years: Earlier studies [16, 17, 18] used the strain gradient plasticity approaches using geometrically necessary dislocations concept, however, further experimental investigations were not convincing enough to support this theory, particularly at nanometer scale depths [33]. Specifically, the Nix-Gao model overestimates the hardness values at very small depths [32]. Swadener et al. reported that the overestimation is due to the fact that the strong repulsion of the geometrically necessary dislocations (GND) at shallow depths causes an expansion on the effective volume of geometrically necessary dislocations [34]. Similarly, Nix and Feng reported similar observations in their later studies [35]. According to their study, significant number of dislocations spreading out from the plastic zone causes relaxation. Our experimental results suggest that none of these models cover the whole picture of the onset of the plasticity based on strain gradient plasticity.
Figure 2: Variation of hardness as a function of indentation depth for multiple samples at in-plane stress (a) zero and (b) 43.21 MPa (with 0.08% plastic strain (see Table 1)). For smaller (larger) applied stress, the behavior remains analogous to (a)-smaller applied stress ((b)-larger applied stress). In (c) we show the change of average and binned hardness with applied top-surface strain at various indentation depths (depths shown in legend), while in (d) we show the change of average and binned hardness with depth at various total applied strains (strains shown in legend).

To explore the physical phenomena that lead to the observed hardness deviation, we have used two dimensional discrete dislocation dynamics simulations to account for the effect of in-plane stress and indentation depth. A cylindrical indenter (circular in 2D with 1 µm radius) is utilized and simulation details can be found in a forthcoming publication [37]. A summary of our simulation results is shown in Figure 3. Figure 3a shows the variation of load with respect to depth for load-controlled indentation. We notice that for depths smaller than 10 nm, and holding the depth fixed, the increase of in-plane stress results in a sharp decrease of the indentation force.

Figure 3b shows the normalized hardness, defined as $H/H_0$, where $H_0$ is
Figure 3: 2D Dislocation dynamics simulation results of bulk Al. (a) Representative load-depth curves, (b) normalized hardness deviation with respect to applied in-plane stress. The bold line is the prediction from the theoretical model for indentation depth 5nm, (c) events statistics up to 40nm indentation depth, (d) effect of in-plane stress on total number of dislocations normalized by \( N_0 \) at different indentation depth. \( N_0 \) is the number of dislocations at zero in-plane stress, shown in inset.

the zero-in-plane-stress hardness measured for a specific depth. It is worth noticing that the effect of in-plane stress on hardness varies with depth: At small indentation depths, hardness decreases with increasing in-plane stress, and this trend disappears for indentation depths larger than 5nm. We identify the reason for this behavior in the dislocation density (number) evolution, shown in Figure 4d: when indentation depth is small (<10nm) dislocation nucleation is scarce and the dislocation density depends on the in-plane stress. Consequently, plasticity strongly depends on pre-existing dislocations.

Furthermore, one may apply small-scale plasticity considerations using a local yield stress picture framework. Gerberich et al. \(^{38}\) linked the indentation
size effect in the nanometer scale to a ratio between the energy of newly created surface and plastic strain energy dissipation. The hardness in the nanometer scale then follows, $H \approx \frac{1}{\left(\frac{S}{V}\right)^{2/3} \left(\frac{2\delta R}{l}\right)^{1/3}}$ for spherical indentation (tip radius $R$) where $S/V$ is the plastic surface area over volume ratio, $\delta$ is indentation depth. Based on the data of Au in table 2 of [38], $\frac{S}{V}\sqrt{\delta}$ decreases at small indentation depth. We define $\sigma_f$ as the local material flow stress, which is a function of the local dislocation density. In nanoindentation, the local flow stress should determine the measured hardness. The local flow stress is expected to have a complex non-monotonic dependence on the local dislocation density at the nanoscale, in analogy with early theoretical suggestions [39] as well as studies of metallic nanopillars [40]. Following Ref. [40], we suggest that the local flow stress during indentation is a function of the local dislocation density: $\sigma_f \propto \beta R \sqrt{\rho} + \alpha b \sqrt{\rho}$, where $\rho$ is the local dislocation density, $R$ is the nanoindenter’s radius, $b$ is the magnitude of the Burgers vector, and $\beta$, $\alpha$ are dimensionless fitting parameters which in our case take the values $1.76 \times 10^{-3}$ and 0.46, respectively. For the indentation depth of 5nm, our estimate of hardness vs. in-plane stress is shown in Fig. 3b (dashed line), which qualitatively agrees with our simulation and experimental findings.

In the light of our nanoindentation experiments and their agreement with our simple DDD simulations, we propose a novel qualitative explanation for hardness size effects: For indentation depths below 10nm, the pre-stress induced dislocation motion dominates the deformation. At that length-scale, initial dislocation density controls the deformation behavior of the sample. This behavior is analogous to the source-limited regime found in pillar compression [42, 41, 43]. For indentation depths above 10 nm, the dislocation density saturates and the system reaches to the critical GND density threshold, and is independent of the applied in-plain stress. As the indentation depth increases, the dislocation density is controlled by dislocation source nucleation, and the effect of dislocations generated by in-plane tension disappears at large indentation depth ($\leq$50nm). We believe that this transition may be used to detect the bulk plasticity tran-
sition in crystals.

In summary, we employed large arrays of nanoindentation tests on polycrystalline bulk aluminum at different in-plane stresses to investigate the incipient plasticity transition. The depth dependent hardness measurements show a clear transition at \( \sim 10\text{nm} \), as the applied in-plane stress increased to \( \sim 50\text{MPa} \) and the estimated in-plain plastic in-plane strain increased to 0.3\%. That is indicative of the high stochastic behavior as small indentation depths disappeared at high in-plane stresses, while the pop-in statistics indicate that displacement bursts are insensitive to in-plane stress. Our experiments are supported by 2D DDD simulations and a naturally applicable constitutive model. In the light of these findings, we propose that the bulk crystal plasticity transition is sensitive to nanoindentation measurements at depths smaller than 10nm.

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