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A tale of two mechanisms: Strain-softening versus strain-hardening in single crystals under small stressed volumes ☆

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ABSTRACT

Pre-straining defect-free single crystals will introduce heterogeneous dislocation nucleation sources that reduce the measured strength from the theoretical value, while pre-straining bulk samples will lead to strain hardening. Their competition is investigated by nanoindentation pop-in tests on variously pre-strained Mo single crystals with several indenter radii (~micrometer). Pre-straining primarily shifts deformation mechanism from homogeneous dislocation nucleation to a stochastic behavior, while strain hardening plays a secondary role, as summarized in a master plot of pop-in strength versus normalized indenter radius.

Engineering materials in most structural applications exhibit strengths that are one or several orders of magnitude lower than the theoretical strength. As a large fraction of the Young's or shear modulus, the theoretical strength can only be achieved or approached when the material is pristinely clean and free of any defects, such as in uniaxial tests of carefully grown, micrometer-sized metallic whiskers or fibers [1–3]. For ductile metallic materials, sample preparation and prior thermomechanical treatment usually introduce at least a moderate density of dislocations, and the material strength is governed by the evolution of the dislocation microstructure through a variety of nucleation, propagation, and multiplication processes [4]. A multitude of small scale mechanical experiments have been conducted in recent years to understand the size-dependence of the material strength, which will ultimately bridge the above theoretical strength limit and the bulk flow limit [5–16]. Experimental efforts using micro-pillars machined by focused ion beam (FIB) are mainly focused on searching the scaling relationship between the yield strength and the pillar diameter [5–7]. These studies suffer two major drawbacks, one being the potential sample surface damage due to FIB treatment [6] and the other being the large variation of data that prevent a meaningful simple scaling [7–9]. An alternative testing method that avoids the cumbersome FIB process or the whisker/fiber growth is the use of instrumented nanoindentation at sub-micron scales [11–16]. For carefully electro-polished surfaces (thus removing surface oxides and other contaminating surface layers), the load–displacement curves usually exhibit sudden displacement bursts (or called pop-ins), and the pop-in strength is an indication of the material yield strength in the corresponding small stressed volumes.

Similar to the extensively studied uniaxial tests, the pop-in strength has two asymptotes as shown in Fig. 1(a). When the stressed volume is free of any pre-existing defects, the pop-in corresponds to the homogeneous nucleation of dislocation at the theoretical strength. At the bulk limit, the pop-ins are difficult to observe and the deviation from the elastic load–displacement curves is governed by the bulk yield stress of the material. In the intermediate stage between these two asymptotes, the pop-in strength shows a significant scattering. Primarily because of the random nature of the distribution of pre-existing defects, chances exist that the stressed volume may contain no or a small number of pre-existing defects, so that the pop-in strength can vary from theoretical strength to the bulk stress. In principle, it should be noted that any type of mechanical tests in the intermediate scale regime should find similar scattering, which is however rarely studied systematically in micro-pillar tests because the tedious

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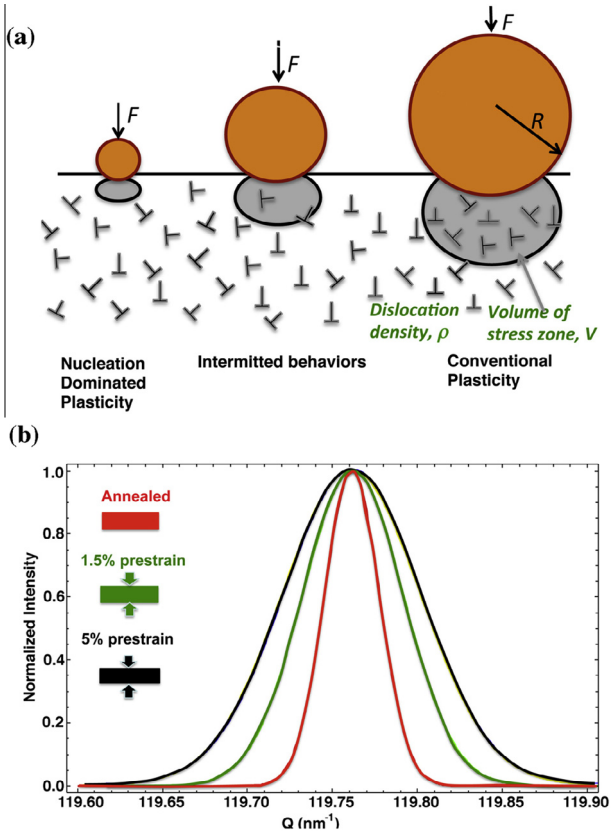

Fig. 1. (a) Schematic diagrams showing different dominant mechanisms for nanoindentation pop-ins, being homogeneous nucleation at small stressed volumes, stochastic behavior at intermediate sizes, and conventional plasticity at large sizes. (b) Synchrotron X-ray results of the peak broadening of {006} Mo reflection in the three samples, from which the dislocation densities can be calculated as given in Table 1.

sample preparation by FIB prevents such statistical measurements. In contrast, nanoindentation pop-in tests [3,12,13,15] can be conveniently done in a small surface area and thus be utilized to study the transition of governing mechanisms with respect to the change of stressed volume size, defect density, and other parameters.

One question that arises naturally from observing the deformation characteristics in Fig. 1(a) is what the effects of pre-strains are on the material strength. At the bulk limit, pre-strain will lead to the increase of dislocation density, and correspondingly the yield stress will increase – this is the *strain hardening mechanism* when the dislocation density increases [4]. Near the theoretical stress limit, however, the pre-existing defects introduced by pre-straining before the pop-in tests will reduce the pop-in stress. These defects will act as the weakest links that change the homogeneous dislocation nucleation (that requires the theoretical stress) to a heterogeneous dislocation nucleation mechanism (such as Frank-Read source that requires a low stress). This is a *strain softening mechanism* when dislocation density increases. Although the above two mechanisms operate at different limits, they both contribute in the intermediate scale regime and in principle one can tune their relative significance by tuning the degree of pre-strain. Consequently, in this paper, a synergy among nanoindentation pop-in tests, synchrotron X-ray measurements (for the characterization of dislocation density), and theoretical modeling will be employed to identify the dominant regimes of these two mechanisms and to study whether and how they can be separated.

Molybdenum single crystals used in this study (from Alfa Aesar) had been electron beam floating zone refined and had a purity of 99.99%. The typical interstitials in electron beam refined Mo are carbon (~0.5 ppm) and oxygen (5 ppm) [17]. All specimens for nanoindentation tests were cut from the same Mo single crystal rod (~11 mm in diameter and 100 mm in length) by using electron-discharge-machining. Before cutting, the Mo single crystal rod was homogenized at 1600 °C for 4 h in a vacuum furnace. Nanoindentation specimens were disk-shaped, of 2 mm thickness, and with surface normal in ⟨100⟩ directions. Three disks were compressed in room temperature with pre-strains (ratios of the reduction of thickness to the initial thickness) of 1.5%, 5%, and 13%. Together with no pre-strained disk (0%), these specimens were mounted in epoxy, ground and polished with standard metallographic procedure. The final polishing step was conducted electrochemically at ~10 V in a 12.5 vol.% H_2SO_4 methanol solution. In order to cover the whole range of deformation behavior in Fig. 1(a), nanoindentation tests were performed in a Nanoindenter XP (MTS Nano Instruments, Oak Ridge, TN) by using a number of indenters, including two Berkovich diamond indenters with effective tip radii R of 115 and 210 nm, five diamond spherical tips with effective radii R of 0.58, 1.5, 3.66, 6.9, 18 μm. The tip radii were calibrated by the method in [18], which considers the contribution of the machine stiffness. All tests were performed in the continuous stiffness mode (CSM) with a constant rate of $\dot{P}/P = 0.05\ s^{-1}$. A total number of 36 indents were made with each indenter so as to achieve sufficient statistical variations, and these indents were placed far from one another to avoid mechanical interference.

Before presenting our nanoindentation pop-in measurements, we identify the relationship between pre-strain and dislocation density in these samples by the polychromatic X-ray micro-diffraction (PXM) technique. Data collection with PXM was carried out at the beamline ID-34-E at the Advanced Photon Source (Argonne, IL), using a modified Laue diffraction technique. It allows for true 3D mapping of crystalline phase, orientation, elastic strain and plastic deformation with less that 0.5 μm spatial resolution [19–21]. Laue patterns from pre-strained Mo single crystals consist of both streaked and broad Laue spots, with the former governed by the formation of geometrically necessary dislocations (GNDs) and deviatoric strain and with the latter depending on the total dislocation density. Measurements were performed in several locations for appropriate statistical sampling. The broadening of the {006} Mo reflection is shown in Fig. 1(b) for annealed, 1.5% pre-strained, and 5.0% pre-strained samples, where Q is reciprocal to the spacing of {006} lattice planes. The method in [19] was employed to calculate the total dislocation density, as given in Table 1. The 13% pre-strained sample was not utilized because it was found that the dislocation arrangement became inhomogeneous – dislocations organized into specific patterns with different kind of dislocation walls with high dislocation density and some part of randomly distributed dislocations between them.

Table 1
Nanoindentation modulus and hardness obtained by experiments, dislocation density measured by the polychromatic X-ray micro-diffraction (PXM) technique, and the pre-existing defect density and strength (ρ_{defect}, τ_{defect}) obtained by the unified model in Eqs. (3) and (4) for annealed and pre-strained Mo (100) single crystals. The theoretical strength is τ_{th} = 16.1 GPa.

Pre-strain	0%	1.5%	5.0%
Modulus (GPa)	327	327	327
Hardness (GPa)	2.22 (±0.02)	2.54 (±0.01)	2.67 (±0.02)
Disl. density (cm^{-2}) (PXM)	1.047×10^7	5.349×10^{10}	1.599×10^{11}
Disl. spacing (μm) (PXM)	3.09	0.0432	0.0250
ρ_{defect} (μm^{-3}) (Eqs. (3) and (4))	0.0053	17.5	55
Defect spacing (μm) (Eqs. (3) and (4))	5.7	0.38	0.26
τ_{defect}	0.04 τ_{th}	0.046 τ_{th}	0.048 τ_{th}