2018

Detecting the Onset of the Bulk Crystal Plasticity Transition in Face Centered Cubic Metals Using Nanoindentation

Ryder Bolin

Follow this and additional works at: <https://researchrepository.wvu.edu/etd>

Recommended Citation

Bolin, Ryder, "Detecting the Onset of the Bulk Crystal Plasticity Transition in Face Centered Cubic Metals Using Nanoindentation" (2018). *Graduate Theses, Dissertations, and Problem Reports*. 5228.
<https://researchrepository.wvu.edu/etd/5228>

This Thesis is protected by copyright and/or related rights. It has been brought to you by the The Research Repository @ WVU with permission from the rights-holder(s). You are free to use this Thesis in any way that is permitted by the copyright and related rights legislation that applies to your use. For other uses you must obtain permission from the rights-holder(s) directly, unless additional rights are indicated by a Creative Commons license in the record and/ or on the work itself. This Thesis has been accepted for inclusion in WVU Graduate Theses, Dissertations, and Problem Reports collection by an authorized administrator of The Research Repository @ WVU. For more information, please contact researchrepository@mail.wvu.edu.

Detecting the onset of the bulk crystal plasticity transition in Face Centered Cubic metals using nanoindentation

Ryder Bolin

Thesis submitted
to the Benjamin M. Statler College of Engineering and Mineral Resources
as the West Virginia University

In partial fulfillment of the requirements for the degree of
Master of Science in
Mechanical Engineering

Stefanos Papanikolaou, Ph.D., Committee Chair
Vyacheslav Akkerman, Ph.D.
Kostas Sierros, Ph.D.
Department of Mechanical and Aerospace Engineering

Morgantown, West Virginia
May 2018

Keywords: Nanoindentation, Size effects, Roughness effect, Residual stress effect, Applied
stress effect, Order Parameter

Copyright 2018 Ryder Bolin

Detecting the onset of the bulk crystal plasticity transition in Face Centered Cubic metals using nanoindentation

Ryder Bolin

Abstract

The present thesis showcases the investigation of nanoindentation at ultra-low depths ($<500\text{nm}$), for the study of the origin and initiation of crystal plasticity in a number of Face Centered Cubic (FCC) metals: Nickel (Ni), Copper (Cu), and Aluminum (Al) in single crystalline or polycrystalline form. Nanoindentation was carried out using a Berkovich tip, and indentations were carried out in the nano-regime of polycrystalline FCC metals and single crystalline Cu and Al. Two custom-built four-point bending apparatuses were used to apply in-plane tension on all samples during indentation. Samples were indented using both displacement and load controls, in order to both carefully estimate the hardness and plasticity noise as a function of depth and applied in-plane tension. The noisy character of crystal plasticity manifests itself through nanoindentation pop-in events, which are abrupt and stochastic plastic displacements during the initial stage of plasticity. I discuss and demonstrate indentation size effects induced and/or promoted by the in-plane stress in FCC materials. The hardness analysis shows a statistically inverse dependence on in-plane stress for small depth indentation and I discuss the consistency of the effects in both the hardness and pop-in events among the various FCC crystals studied. Finally, I discuss a novel approach to detect the onset of pop-in noise in a realistic sample without special treatment and demonstrate its consistency for Cu single crystalline samples.

Acknowledgements

I want to thank Dr. Papanikolaou for giving me the opportunity to work on multiple projects under his leadership. I was honored to be given the role of Graduate Research Assistant and working on high valued research in indentation. I take great pride in the work and accomplishments I have accrued over the last two years. Dr. Papanikolaou provided me with top notch equipment and taught me advanced skills in code programming. In my projects, I was able to work on multiple aspects that included the beginning to the end material.

Finally, I would like to thank my family and friends for supporting me through these two years. Without them it would have been a struggle to complete my master's degree. They helped push me to make myself better each and every day and helped lift my spirits when I was feeling down. I want to especially thank Emily Widmeyer, who is my fiancée. She has been here from day one of my journey to get my Masters, and I could never have made it through this program without her support.

Table of Contents

Abstract.....	ii
Acknowledgements	iii
List of Figures.....	v
List of Tables	x
1. Introduction & Thesis Overview.....	1
1.1. Introduction.....	1
1.2. Thesis Overview.....	2
2. Literature Review/ Experimental Background	3
2.1. Nanoindentation/Plasticity	3
2.2. Expectations from Continuum Models.....	9
2.3. Indentation Size Effect.....	12
2.4. Residual stress detection through indentation.....	13
2.5. Roughness effect	15
2.6. Pop-in noise	15
3. Theoretical Explanations / Analysis.....	16
3.1. Expectations from Discrete Dislocation Dynamics	16
4. Nanoindentation Experimentation.....	19
4.1. Specimens.....	19
4.2. Experimental Procedures.....	25
5. Experimental Results/ Discussion	29
5.1. Description of data analysis tools.....	29
5.2. Polycrystalline Aluminum.....	31
5.3. Polycrystalline Copper.....	36
5.4. Polycrystalline Nickel	39
5.5. Single Crystal Aluminum.....	41
5.6. Single Crystal Copper.....	43
5.7. Construction of the Order Parameter.....	47
6. Conclusion.....	50
7. References	51

List of Figures

Figure 2.1: Schematic that gives a representation of the load versus displacement during loading and the unloading portion. $P_{\max}$ is the maximum amount of load during load, h_f is the final depth of the contact impression after unloading, and $h_{\max}$ is the max amount of displacement at the maximum loaded state. S is the calculated stiffness from the unloading portion [14].	6
Figure 2.2 is a schematic representation of the section through an indentation showing different qualities used in analysis [14].	7
Figure 2.3 shows the experimental parameters that are used in the analysis. The three key parameters in this schematic are the peak load, the depth in retrospect to the peak load, and the initial unloading contact stiffness. The contact stiffness is now only measured at the peak load and creates no restrictions on the unloading data being linear at any position during unloading [14].	8
Figure 2.4 is a schematic for two-dimensional compressive stresses being replaced by the hydrostatic pressure p and maximum shear stress k . This figure shows that $p = Q+k = P-k=1/2(P+Q)$ [64].	10
Figure 2.5 shows the slip-line pattern in ideally plastic metal deformed by a flat-punch with a diameter of d [64].	11
Figure 2.6 is a schematic representation of the indentation size effect (ISE) for geometrically self-similar indenters. Normally the hardness increases at shallow depths, but there are a few observations out there that show the reverse ISE. Continuum mechanics says there should be no dependence on depth for hardness, that gives us our conventional plasticity line [33].	13
Figure 2.7(a) shows the nucleation of dislocations caused by the flat-punch indenter tip. (b) shows the movement of pre-existing dislocations located in the bulk of the material.	16
Figure 3.1 Schematic of DDD simulations. The width is $w_p = 20\mu\text{m}$ and the height is $h_p = 10\mu\text{m}$. The red dots indicate the dislocation sources, and the blue dots indicate obstacles. These dots are randomly distributed on slip planes.....	17
Figure 3.2 2D DDD results for polycrystalline Al. (a) shows the load-depth curves with increased applied stress. (b) shows the normalized hardness and the dotted line shows the theoretical model for indentation depth of 5nm. (c) shows the event statistics for indentation depth up to 40nm. (d) shows how the in-plane stress effects the total number of dislocations normalized by N_0 . N_0 is the number of dislocations at zero in-plane stresses [68].	17

Figure 3.3 (a) shows the 3D indentation model, with Frank-Read sources whose initial density is $5 \times 10^{12}/\text{m}^2$. The top is a flat punch with a diameter of 250nm. (b) shows a tensile test done on (a) to determine the tensile pre-stress that should be assigned. (c) Shows the hardness vs applied stress with the 5 pre-stresses that we chose (0, 45, 90, 135, 180MPa). In Figure (c) we see clear hardness transition with increasing Applied stress at a depth of 5nm.	19
Figure 4.1(a) and (b) show the beginning schematics that were used to build the 4-pt bending systems. (a) is the 4-point bending system that was used on the single crystal samples since their smaller size could not fit in the bigger system. (b) was used on all the poly crystal samples. The empty space below in the center is where the metal plate that pushes against the samples is placed.....	21
Figure 4.2 (a-d) shows the materials sitting in the 4pt-bending systems. (a) shows the poly crystal Cu. (b) shows the poly crystal Ni. (c) shows the single crystal Al. (d) shows the single crystal Cu. (b-d) show samples after indentation while (a) gives a representation of the initial stage before applying the strain gauge.	22
Figure 4.3 is a visual representation of the process of indentation during a video calibration. The camera is the first component with the indenter tip in the second component.	26
Figure 4.4 is a landscape representation of the poly crystal Ni load protocol. The white space is filled with hardness protocol arrays. The different colors represent different strain levels.	29
Figure 5.1(a-b) are figures showing the raw data from the indentation readings. Different symbols/colors indicate different indentation test. Each figure has almost 200 indentations in them. (a-b) shows the decrease in the hardness as more in-plane tension was applied to the specimen. (a) Shows the hardness-depth curve for 0% strain and (b) shows the hardness curve at 0.45%.....	32
Figure 5.2 (a, b) show 5.1 (a, b) after the binning and removal of outliers. Figure 5.2 is binned at every 2.5nm. Different symbols/colors indicate different indentation test.....	32
Figure 5.3(a) and (b) show the average hardness-depth curves. Different symbols/ colors represent a different strain, and there are about 200 curves averaged in each strain. (a) shows the hardness is equivalent to a power law of the strain. (a) provides the full hardness average where (b) is zoomed in on the major area of focus. (c) shows the average hardness at each binned depth with respect to the strain.....	33

Figure 5.4 (a-c) are the load depth curves at different strains. Different symbols/colors indicate different indentation test. There are about 800 indentation tests in each figure. (a) shows indentations at 0.25% strain, (b) shows indentations at 0.4% strain.....34

Figure 5.5 (a-b) shows the statistically processed and binned load-depth data sets. By getting rid of the outliers we can now get a more accurate averaging of the load-depth. Different symbols/colors indicate different indentation test. The data is separated at a bin size of 2.5nm...34

Figure 5.6 (a) shows four load depth curves for a clearer representation of the pop in events. (b) shows the average load at each binned depth. In (b) about 800 indentation tests are averaged in each strain. (c) shows the probability density of pop-in events as a function of event size.....35

Figure 5.7 (a-b) are hardness curves for Cu as we applied more stress on the samples. Different symbols/colors indicate different indentation test. Almost 100 hardness measurements are in each strain plot. (a) is the copper at .101% strain and (b) is at 0.327% strain.....36

Figure 5.8(a) shows that hardness is equivalent to a power law of the strain. (a, b) different symbols/colors indicate different indentation strain. There are about 100 hardness plots averaged in each strain. (a) shows the hardness average per strain as a function of depth. (b) presents a zoomed in portion of the main area of analysis of (a). (c) shows the hardness transition from shallow depths to 10nm then (the light grey box line) we see this transition end. The transition for the poly crystal copper happens at faster rate than we see in the poly crystal aluminum.....37

Figure 5.9 (a-b) show load-depth data curves at different strains. Different symbols/colors indicate different indentation test. There are about 1000 indentation tests in each figure. The pop-in events are not as clear as they are in the poly crystal Al. (a) shows the load-depth curves for 0.101% strain and (b) shows the load-depth curves for 0.173% strain.....37

Figure 5.10 (a) shows 4 selected load-depth indentations for a better representation of individual curves. (b) shows the average load with respect to depth and is observed that the variability is not very high. There are about 1000 tests averaged per strain. (c) shows the pop-in probability density for 1000 tests for each strain.....38

Figure 5.11 (a-b) are hardness curves for Ni as we applied more stress on the samples. Different symbols/colors indicate different indentation test. Almost 100 hardness measurements are in each strain plot. (a) is the nickel at .030% strain and (b) is at 0.160% strain.....39

Figure 5.12 (a) shows that hardness is equivalent to a power law of the strain. (a, b) different symbols/colors indicate different indentation strains. There are about 100 hardness plots averaged

in each strain. (a) shows the full hardness average with respect to depth curve. (b) shows the area of interest and (c) shows us similar results to poly crystal copper. The nickel transition happens very fast as well at about the 6nm range and then after that the effect seems to dissipate.....40

Figure 5.13 (a-b) show load-depth data curves at different strains. Different symbols/colors indicate different indentation test. There are about 1000 indentation tests in each figure. The pop-in events are not as clear as they are in the poly crystal Al. (a) gives the load-depth curve for 0.012% strain and (b) shows the load-depth curve for 0.110% strain.....40

Figure 5.14 (a) shows the load-depth curves of 4 data sets to see a better representation of how one curve looks. (b) shows the variation in the average load across the strain levels. In (b) there are about 1000 tests averaged per strain (c) shows the probability density across 1000 tests per strain value.....41

Figure 5.15 (a-b) are figures showing the raw data from the indentation readings. Different symbols/colors indicate different indentation test. Each figure has almost 1000 indentations in them. (a-b) shows the decrease in the hardness as more in-plane tension was applied to the specimen. (a) Shows the hardness-depth curve for 0% strain and (b)shows the hardness curve at 0.100% strain.....42

Figure 5.16 (b) shows that hardness is equivalent to a power law of the strain. Different symbols/colors indicate different strains. Each figure has almost 1000 indentations averaged in each strain. (a) shows the full hardness average with respect to depth curve. (b) shows the area of interest and (c) shows us similar results to poly crystal aluminum. (a-c) shows the hardness at shallow depths transitions almost immediately, but the effect is still present till 12nm.....42

Figure 5.17(a) shows the load-depth curves of 4 data sets to see a better representation of how one curve looks. (b) shows the variation in the average load across the strain levels. In (b) there are about 1000 tests averaged per strain (c) shows the probability density across 1000 tests per strain value.....43

Figure 5.18 (a-b) are figures showing the raw data from the indentation readings. Different symbols/colors indicate different indentation test. Each figure has almost 500 indentations in them. (a-b) shows the decrease in the hardness as more in-plane tension was applied to the specimen. (a) Shows the hardness-depth curve for 0% strain and (b)shows the hardness curve at 0.140% strain.....44

Figure 5.19 Different symbols/colors indicate different strains. Each figure has almost 500 indentations averaged in each strain. (a) shows that hardness is equivalent to a power law of the strain (a) shows the full hardness average with respect to depth curve. (b) shows the area of interest and (c) shows us similar results to poly crystal copper. (a-c) shows the hardness at shallow depths transitions almost immediately, but the effect is still present till 15nm.....45

Figure 5.20 (a) shows the load-depth curves of 4 data sets to see a better representation of how one curve looks. (b) shows the variation in the average load across the strain levels. In (b) there are about 500 tests averaged per strain (c) shows the probability density across 500 tests per strain value. The pop-in statistics is consistent with various theories of avalanche phenomena [87]. However, this topic lies beyond the purpose of this work.....46

Figure 5.21 provides us with the AFM images that was done on the single crystal samples. (a) shows us the single crystal aluminum results and shows variations around 4nm. (b) shows the surface of the single crystal copper and the surface only has a surface roughness below 1nm. (b) has the least amount of roughness and results would be more influenced by the applied stress effect.....46

Figure 5.23 (a) experimental results of nanoindentation on (111), (110), and (100) copper crystal orientation. (b) shows the frequency histograms of the critical indenter force and displacement at the onset of displacement bursts [79].....48

Figure 5.24 (a) shows the plotting of the order parameter vs total % strain. Each line represents a different indentation depth. The shallower depths have the straighter lines, because close to 5nm is when the first pop-in happens. Meaning anything smaller than the 5nm should be almost completely flat. The deeper depths have a peak at 0.12 % strain which indicated the elastic to plastic transition. (b) shows the variability of the order parameter and is consistent with (a).....49

Figure 5.25 (a) shows the order parameter with respect to a certain load. The peaks in (a-b) are the showing the initiation of plasticity. The peak position is consistent with the locations in the annealed samples [79], as shown in Fig. 5.22.....49

List of Tables

Table 4.1: Polycrystalline Al	23
Table 4.2: Polycrystalline Cu.....	23
Table 4.3: Polycrystalline Ni	24
Table 4.4: Single Crystal Al	24
Table 4.5: Single Crystal Cu.....	24
Table 4.6 Nanoindentation array layout.....	28
Table 5.1 is a visual representation of how the data is set-up before analysis with python. The actual sets are substantially bigger then this little cut out. For the load analysis the two main columns are the Load and Depth columns.....	30

1. Introduction & Thesis Overview

1.1. Introduction

Over the last few decades it has well been established that plastic deformation of crystals shows fundamental differences between the micro- and nano-scale. Micro-scale plastic deformation has been a highly investigated field, but the nano regime has not yet been sufficiently researched. The reason is there are too many obstacles to get through in the accuracy of the data. Surface roughness, oxidation, and size effects invalidate the data. In order to understand the fundamentals of plasticity in the nano-regime, structured tests must be conducted, and the experimenter should be very meticulous. One widely used method to probe and measure the mechanical properties of metals is indentation [1]. Recent advances in the indentation technique have led to the development of a low depth/ low load sensing indentation technique called nanoindentation [2].

Nanoindentation is a non-destructive examination that gives one the ability to look at the local mechanical properties in extremely small regions of a material. With the increased tendency of manufacturing smaller specimens, there is also an increasing need for a better understanding of the mechanical behavior at smaller length scales. Also, investigating local properties at the micron and submicron levels for metals and ceramics is an important goal of material science. The ability of nanoindentation to probe such properties, has given it wide-spread popularity in the field. With the difficulty of the experimental design, conventional tensile/compressive tests are not applicable for small samples (at the millimeter to micron scale sizes) [3-11]. Nanoindentation is an experimental protocol that is able to meet such requirements.

Indentation displays an interesting phenomenon that happens to hardness measurements at the micron/sub-micron scale, in the form of indentation size effects (ISEs). These ISEs exhibit smaller depths with higher hardness values. Classical plasticity theories do not have the ability to predict size dependence of material behavior at the micron/sub-micron scale. Nix and Gao [12] found that the indentation size effects were related to the plastic strain gradient in the deformation field with geometrically necessary dislocations (GNDs) that accommodate this strain gradient.

Furthermore, in nanoindentation, it is observed that plasticity occurs with an abrupt transition from elasticity to plasticity, through a sudden burst in the indentation depth. These bursts are well known in the indentation community as “pop-ins”. For small tipped indenters (pyramidal Berkovich), the first bursts are likely to be in a dislocation free zone. This means that dislocations would be nucleated at the indenter tip’s contact area. Furthermore, with increasing tip radius

(equivalent to deeper indenter penetration), there is a higher chance that plasticity was affected by nearby dislocations.

One of the biggest challenges that effects the accuracy of the nanoindentation results is the roughness effect. The nanoindenter's greatest issue with this effect is due to the inaccuracy of recorded data from major inconsistencies in the surface of the samples. This leaves the indentation results being presented with some error with respect to the contact area. This is especially true as indentation depth gets shallower and shallower.

Another important effect that influences indentation is the presence / absence of residual stresses. Experimental studies have been conducted on plasticity at the micro and nano-scale. Tsui, Oliver, and Pharr used nanoindentation to study the effect of residual stress [13]. The experimental study used both uniaxial and biaxial stresses and results from finite element simulations [14]. Residual stresses affected the hardness in two ways: the hardness went up with compressional stresses and the hardness decreased with tensile stresses. The elastic modulus is affected in the same way as the hardness. Furthermore, it was shown that nanoindentation properties like the maximum indentation load, the profile of the loading curve, elastically recovered depth, the final depth, and the area of contact were all affected by the presence of residual stresses.

1.2. Thesis Overview

In this study, examination is done on FCC crystals such as Aluminum (Al), Copper (Cu), and Nickel (Ni). Each sample was placed in a 4pt-bending apparatus to apply in-plane tension. Through this study the only differences in testing were the in-plane stresses. Analogous to this logic, we expect the main difference in the experimental results are a consequence of the in-plane stress. Single crystal Copper and Aluminum samples are used to verify the in-plane stress effect. Single crystals do not have grain boundaries and the grain size is the sample. This provides us with a validating material where the in-plane stress effect would be more prominent. Both single crystal materials have the {100} crystal orientation. We acquired high amounts of data on each sample to successfully average out the surface roughness. We developed our own python code to process large data sets. The analysis is conducted on the hardness behavior across different applied stresses and then compared between the materials. The same will be done with the statistical analysis for pop-in events. We will introduce a new method for observing the plastic transition using an order parameter (dh/dp). These methods will hopefully lead to the connection of the pop-in and

the bulk yield stress. The experimental background will provide us with all the background information needed to understand this study.

2. Literature Review/ Experimental Background

2.1. Nanoindentation/Plasticity

Crystal plasticity studies have given us a better understanding of how crystals react under external loading [15-18]. Until recent years, it was not known that the mechanical properties of a material were tightly related to microstructural characteristics, which are highly dependent on sample dimensions. Indentation's main purpose is to measure the mechanical properties of metals, multiphase materials, thin films, and/or ceramics. Many studies have been conducted on the study of mechanical properties of thin films. Sub-micron indentation devices are widely used for these studies, with the focus on micro-scale hardness measurements [15]. With the advancement of technology, a new nondestructive examination for low loads and low penetration depth sensing has been developed called nanoindentation. Nanoindentation allows readings to be gathered at extremely small regions and is used for probing localized mechanical properties of a given material [19-22].

Indentation's main objective is to create plasticity in a small region to investigate the mechanical properties of a given sample. One of the most interesting properties is hardness, that shows how resistant solid matter is to an applied force. This gives a direct relationship to plasticity. Knowing hardness values allows for a much better understanding on the effect of plastic deformation as the transition of elastic to plastic occurs in a specific region on a sample. Hardness is calculated by the maximum indentation load over the contact area of the indentation impression [21, 22].

One of the most conventional hardness measurement tests involves indentation on a material that was then paired with atomic force microscopy (AFM) [23, 24] and scanning electron microscopy (SEM) [15, 25-27] to get the contact area measurement of the imprint that was left behind after the indenter tip was removed. This is called the Vickers hardness test which was developed in 1921 by Smith and Sandland. This process is simpler and easier than the Brinell method [28]. Although indentation hardness has been around for a century, only in the last decade have we been able to use a tool that had high resolution depth-sensing and to be able to continuously measure the force and displacement as indentations are implemented [29, 30].

Boussinesq and Hertz first looked at the elastic contact problem in the late 1900's and provided us with the importance in the analysis procedure of the data [31, 32]. Boussinesq developed a method used to compute the stresses and the displacements in an elastic body loaded with an axisymmetric indenter. Hertz analyzed the problem of the elastic contact between two spherical surfaces with different radii and elastic constants. This is used in classic solutions that are the basis for a lot of experimental and theoretical work. Sneddon developed a relationship between the load, displacement, and the contact area [33, 34]:

$$P = \alpha h^m, \quad (2.1)$$

where P is the load that the indenter is putting on the material, h is the elastic displacement of the indenter, and α and m are mathematical constants. It was mathematically calculated that for flat cylinders $m = 1$, for cones $m = 2$, and for $m = 1.5$ for spheres and for paraboloids of revolution.

Showing the effects of plasticity through the modeling of the indentation contact area and overall geometry, is a highly complicated problem. The constitutive equations are highly nonlinear, and a majority of the material parameters must be part of this model to describe the material behavior. Experimentation and the finite element simulations are the building blocks used in today's understanding of the plasticity in the indenter contact problem.

Tabor[1] first used the experimental procedure of load and displacement sensing to measure the mechanical properties of metals. In his study, he focused on indentation applied on a variety of metals deformed by hardened spherical indenters. Tabor realized that the shape of the indent impression from indentation at unloading will allow for a portion of the material to elastically recover. An analysis of the results showed that in metals the impressions left behind from a spherical indenter had spherical shape with a slightly larger radius than the indenter, and a similar situation was observed in the conical indenter where they saw a slightly larger tip angle. Looking at the interpretation of the elastic unloading data, the effects of plasticity can be seen by the shape of the indentation impression on the unloaded surface. Tabor used this experiment to show the shape of the entire unloading curve and the total amount of the recovered displacement can be accurately correlated to the elastic modulus and the residual size of the contact impression for conical and spherical tips [1, 2].

Three important observations can be made from the Tabor studies. The first was the diameter of the contact impression in the surface from the conical indenters did not recover during the unloading stages, while the depth was the only thing that recovered. The second observation was

that the indentation process must be cyclically loaded and unloaded a few times before a reversible load-displacement behavior can happen. The third observation is that the effects of non-rigid indenters on the load-displacement behavior can be considered by the effective elastic modulus also known as the reduced elastic modulus:

$$\frac{1}{E_{eff}} = \frac{(1-\nu^2)}{E} + \frac{(1-\nu_i^2)}{E_i} , \quad (2.2)$$

where E_{eff} is the effective modulus. E and E_i are the Young's modulus of the material and the indenter tip respectively. ν and ν_i are the Poisson's ratio for the material and the indenter tip respectively. Alekhin, Bulychiev, and Shorshorov [35-37] investigated load-displacement data using microhardness testing machines. From Fig. 2.1, an experimental equation for the stiffness was derived:

$$S = \frac{dP}{dh} = \frac{2}{\sqrt{\pi}} E_{eff} \sqrt{A} . \quad (2.3)$$

In this equation, they found stiffness S to be $S = \frac{dP}{dh}$ for the upper portion of the unloading data. A is the projected area of the elastic contact. They reported that the measuring of the initial unloading stiffness with the assumption that the contact area was equal to the optical measured area for the indentation impression. Then, the modulus can be derived.

Figure 2.1: Schematic that gives a representation of the load versus displacement during loading and the unloading portion. P_{max} is the maximum amount of load during load, h_f is the final depth of the contact impression after unloading, and h_{max} is the max amount of displacement at the maximum loaded state. S is the calculated stiffness from the unloading portion [2].

The previously discussed experimentations were used as a basis for a new method that was introduced in 1992. This method was able to continuously calculate the hardness and elastic modulus from the indentation load-displacement data. This method is known today as the Oliver-Pharr continuous stiffness method (CSM) [2]. The two main mechanical properties that are the most sought after are the elastic modulus, E , and the hardness, H . Load-depth sensing was commonly used to get data of a complete cycle of loading and unloading [38]. The unloading data is analyzed according to the highly used model for the deformation of an elastic half space. This method uses an elastic punch which relates the contact at the peak load to the elastic modulus. Then, AFM or SEM was used to estimate the contact area. From the images it was possible to measure a separate E and H value. The Oliver-Pharr method eliminated the need of optical imaging by introducing a method that could measure the hardness and elastic modulus continuously during indentation.

The Oliver-Pharr method is a major focus of this thesis, because the nanoindenter we use in the experimentation is applying this method. Figure 2.2 shows the cross section of indentation and

also shows the parameters that were used for this method's analysis. At any time during the application for this method, h is the total displacement:

$$h = h_c + h_s, \quad (2.4)$$

where h_c was the vertical distance that the indenter is in contact with the material. This parameter is also known in the indentation community as the indentation contact depth. h_s is the displacement of the surface at the perimeter of the contact.

Figure 2.2: Schematic representation of the section through an indentation showing different quantities used in analysis [2].

Figure 2.3: The experimental parameters that are used in the analysis. The three key parameters in this schematic are the peak load, the depth in retrospect to the peak load, and the initial unloading contact stiffness. The contact stiffness is now only measured at the peak load and creates no restrictions on the unloading data being linear at any position during unloading [2].

There are observable differences between Figs. 2.1 and 2.3., namely the experimental parameters that are used in calculation of the hardness and modulus. These changes are shown in the load-displacement schematics. The analysis starts with equation (2.3):

$$E_{eff} = \frac{\sqrt{\pi}}{2} \frac{S}{\sqrt{A}} . \quad (2.5)$$

This change is made and holds true for any indenter tip that has a body of revolution of a smooth function and is not limited to a specific geometry. This leads to the new projected area equation:

$$A = F(h_c) . \quad (2.6)$$

The function F , is established through the experimentation prior to analysis. When determining the contact depth from the experimental data, an equation is derived from equation (2.4):

$$h_c = h_{max} - h_s , \quad (2.7)$$

where h_{max} can be directly measured during indentation. The key parameter to calculate is h_s . Oliver-Pharr used Sneddon's expression for the shape of the surface outside the area of contact and obtained the following equations:

$$h_s = \frac{(\pi-2)}{\pi}(h - h_f) , \quad (2.8)$$

$$(h - h_f) = 2 \frac{P}{S} . \quad (2.9)$$

Combining Eqs. (2.8) and (2.9) and keeping in mind that the contact area of interest is that of the peak load, one can derive the following equation:

$$h_s = \epsilon \frac{P_{max}}{S} , \quad (2.10)$$

where the geometric constant ϵ for conical indenters is given by $\epsilon = 0.72$ or:

$$\epsilon = \frac{2}{\pi}(\pi - 2) . \quad (2.11)$$

When similar arguments are made, equation (11) is the same for different geometric constants. For flat punch, $\epsilon = 1$, and for a paraboloid of revolution, $\epsilon = 0.75$. Finally, after successfully calculating the modulus, the data obtained from the experiment can be used to determine the hardness H :

$$H = \frac{P_{max}}{A} , \quad (2.12)$$

where A is the projected area of contact at peak load, evaluated from (2.6). All the aforementioned parameters, are calculated during indentations [2]. This is the experimental set-up that is used in this study.

2.2. Expectations from Continuum Models

Theoretical solutions have been used to study the connections between material yield strength and hardness. Continuum models, exploit Tabor's law [39], which has been the main connection between material yield strength and hardness measurement. The relation between load and the size of indentation for spherical indenters tips can be expressed by several empirical relations, and the first is the Meyer's law [40].

Specific conditions are needed for two-dimensional plastic flow. The flow satisfies the Huber-Mises criterion when the maximum shear stress equals a critical value of the shear stress k , where $2k = 1.15Y$. Y is the yield stress. The other condition is that in a region in which the plastic strain has larger values compared to the elastic, we use the idea that the deformation is determined primarily the plastic properties of a material.

Figure 2.4: Schematic for two-dimensional compressive stresses being replaced by the hydrostatic pressure p and maximum shear stress k . This figure shows that $p = Q+k = P-k = 1/2(P+Q)$ [39].

P and Q are the compressive stresses on a two-dimensional body and can be replaced with the hydraulic pressure p and shear stress k . The lines of maximum shear stress are called slip-lines, but they should not be confused with slip-lines or the slip-bands detected with the microscope. The maximum shear stress is at 45 degrees to P and Q and has a magnitude $1/2(P - Q)$. If the maximum shear stress is flowing plastically, $2k = 1.15Y$ or Y depending on the which criterion is used. The Huber-Mises criterion states that $P = Q+2k$. In figure 2.4, P can now be replaced. Each term constitutes a hydrostatic pressure $p = Q+k$. Two families of slip-lines α and β cover the whole domain of plastic flow, with each of them cutting the other orthogonally. If the hydrostatic pressure at any point is known the following equations can be deduced (Fig. 2.4):

$$P = p + k, \quad (2.13)$$

$$Q = p - k. \quad (2.14)$$

If, however, there is any curvature of the slip-line by an angle θ with respect to a fixed direction, then:

$$p + 2k\theta = \text{constant along } \alpha, \quad (2.15)$$

$$p - 2k\theta = \text{constant along } \beta. \quad (2.16)$$

Figure 2.5: Slip-line pattern in ideally plastic metal deformed by a flat-punch with a diameter of d [39]

Figure 2.5 has four isosceles triangles, CMA, ANE, ESB, and BTG. These triangles show the areas in which the slip-lines are straight lines making an angle of 45 degrees with the face of the metal and flat-punch. The triangles from either side of the punch are connected to those beneath the flat punch by curved slip-lines. A and B are the centers of the triangles with straight slip-lines going to the edges of the triangle. These slip-lines are all in the plastically flowing region and the boundary is shown in Fig. 2.5. Starting at G_1 you can see that the normal principal stress Q at this point is zero, which means $p = k$. The equation along the slip-line is written as:

$$p + 2k\theta = k. \quad (2.17)$$

There is no change p since G_1T_1 is a straight line, but once the line reaches T_1 the slip-line begins to curve until the line reaches S_1 . The slip-line goes through an angle of $\theta = -1/2\pi$, so the equation at S_1 is:

$$p - 2k\frac{1}{2}\pi = k. \quad (2.18)$$

There are no changes in θ as the slip-line is traversed from point S_1 to E_1 , so at E_1 we get the following equation:

$$p = k + 2k\frac{1}{2}\pi. \quad (2.19)$$

Using the flat-punch tip it was found that the hardness is closely related to the pressure across the face of the tip, and the following relationship was obtained through extensive quantitative expansions and analysis starting with (13-19) to get:

$$P_m = 2.6Y \text{ to } 3Y, \quad (2.20)$$

where P_m is the pressure across the face of the tip. After extensive examinations it was realized that this equation holds true across all tip shapes with the proportionality constant varying right around 3.

In general, these facts have formed the foundation of how hardness is understood. The principal expectation is that hardness is proportional to yield stress, as stated in continuum mechanics, and thus depth independent. However, these investigations hold in the millimeter scale, and they may not accurately probe the micro and nano-scale. In the micro and nano-scale, one can see clear deviations from Tabor's model in the form of ISEs, showing that hardness is size dependent.

2.3. Indentation Size Effect

The increase of yield and/or flow strength has been observed in the uniaxial compressed test specimens with a reduced size [41-44]. This size-dependent strength increase is due to a unique deformation that is observed to be only in materials with dimensions that approach the average dislocation spacing and the plastic deformation which is controlled by a limited number of defects. The scale-dependent behavior is only observed when the hardness measurements are at small depths: This in turn gives us the indentation size effect (ISE). The observation of the indentation size effect is most often seen in materials that are indented with a geometrically self-similar indenter like pyramids and cones [45-48].

Normally, the indentation size effects come in as 'smaller is harder', but also, the opposite may occur [44, 49]. In this thesis, both cases will be introduced, but it should be noted that the experimental work is focused on the normal indentation size effects. Figure 2.6 shows a schematic of hardness vs depth curves. In this plot three possibilities are given: The normal ISE, conventional theory of hardness (continuum mechanics), and the reverse ISE. In the bottom of the figure, we show two images: The first, shows a basic schematic on how the hardness is calculated during indentation, while the second shows a basic image of what a Berkovich indenter tip looks like with a tip shape angle of 65.3 degrees on all three sides. The reverse indentation size effect is most often observed to occur in materials in which the plastic deformation is predominant or when the tip is not sharp [50, 51].

Figure 2.6: Schematic representation of the indentation size effect (ISE) for geometrically self-similar indenters. Normally the hardness increases at shallow depths, but there are a few observations out there that show the reverse ISE. Continuum mechanics says there should be no dependence on depth for hardness, that gives us our conventional plasticity line [42].

Nanoindentation gives a better representation of elastic to plastic deformation in the nano regime. Since the 1990's and the introduction of the Oliver-Pharr method (see section 2.1), the material science community has the ability to look at the indentation impressions from the load-depth curves. This has given researchers the ability to try and characterize what in the material is causing the indentation size effect at small depths [16, 26, 52-54].

2.4. Residual stress detection through indentation

The processing of materials and engineering components has many factors that affect the microstructure. However, the impact that is on the components cannot completely disappear even when the factors disappear. Residual stress is defined as the internal stress that is present in an object in the absence of an external load. The observation of residual stress state is different for

examinations and material processing routes. The residual stress can be broken up into two types: The macro-residual stress and micro-residual stress [55, 56]. Being able to measure the residual stress is important because it does not disappear in manufacturing processes and the residual stress effects the material's mechanical properties.

The hardness of metals and alloys measured by conventional techniques is dependent on the stress state of the material. The effects of residual stress on hardness measurements was first observed by Kokubo and Kostron in the 1930's [57, 58]. Two decades later, Sines and Carlson did research on these effects and suggested results that could be utilized to measure the local residual stresses of the surface for a given metal [59]. Sines and Carlson's data showed three key results. The first is that the effects of the stress are relatively small showing hardness variations no more than ten percent. The second is that the hardness increases with applied compressive stresses and decreases with applied tensile stresses. The third was that if the stresses were uniaxial than the influence of stress was higher in tension than it was in compression.

A great number of studies have been applied over the years to experimentally examine the association between hardness and residual stress through Brinell, Rockwell, and Vickers hardness testing methods [60-63]. These studies suggested that the hardness measurements may be a valuable way for characterizing residual stresses in materials. In a Tsui, Oliver, and Pharr study, they used nanoindentation to investigate the effect of applied stress on hardness [13]. The data revealed that the effect of applied stress on nanoindentation hardness measurements were similar to the results in the conventional hardness tests. This provides with evidence that nanoindentation can be used to investigate residual stresses. This study was followed up by Suresh and Giannakopoulos, who created general methodology for determining the pre-existing residual stress [64]. They created a set of formulas that could be used to solve for the mechanical properties for continuous shape indentation tests for a range of cases.

The residual stress effect has been further studied in elastically strained generic materials [65-67]. Hou and Jennet focused their residual stress investigation on a parameter that was being affected, which is the indentation contact area [68]. The indentation contact area has an increase in pile-up when compressive stress is present or an increase in sink-in with the presence of tensile stress. This study shows that the use of the elastic modulus rather than the hardness of material is a way to map the residual stress and correct the hardness values.

2.5. Roughness effect

One of the crucial requirements in nanoindentation is the knowledge of the exact indenter tip geometry. At low penetration depths, a slight deviation from the ideal tip geometry may seriously affect the results if deviated [69, 70]. The roughness effect dominates at shallow depths and promotes inaccuracies in your data, because of the variations in the sample surface. The roughness effect can cause major miss reads during the process of nanoindentation. Therefore, it is imperative that proper measurements are taken to combat this effect. In this study, we use high-throughput indentation to average out the roughness effect.

2.6. Pop-in noise

The transition from elastic to plastic behavior during nanoindentation in well annealed crystals happens from a distinct displacement burst during a continuous load-displacement curve and this is called a ‘pop-in’. When the load-controlled nanoindentation protocol is used, a horizontal plateau is observed on the load-displacement curve. This pop-in effect shows the exact moment the area under the indenter goes from elastic to plastic. The pop-in is an important part of many investigations of plasticity using indentation [71-74]. Pop-in events can happen in one of two ways. The first way is by the nucleation of dislocations on the surface caused by the indentation tip, as shown in Fig. 2.7. The second way is by the movement of pre-existing dislocations in the bulk material, as shown in Fig. 2.8. Figures 2.7 and 2.8 are simulations showing surface and pre-existing bulk dislocations. It is important to point out that the dislocations that are nucleated at the surface are insensitive to applied-stresses, but the dislocations that are pre-existing in the bulk of the material are very sensitive to the applied stresses. In this thesis we are going to experimentally investigate pop-ins across different stresses and try and relate it back to the yield stress [21, 72-75].

Figure 2.7: (a) the nucleation of dislocations caused by the flat-punch indenter tip and (b) shows the movement of pre-existing dislocations located in the bulk of the material.

3. Theoretical Explanations / Analysis

3.1. Expectations from Discrete Dislocation Dynamics

In a crystalline material, dislocations are line defects that show permanent deviations of atoms from their original crystallographic structure [58]. Dislocations move in slip systems of a crystal, which creates plastic deformation. The understanding of the nature of dislocations is needed when it comes to modeling the plasticity of a crystalline material [76-82]. Since continuum mechanics cannot capture the nano regime phenomena, different modeling techniques are required to get a full understanding of the hardness depth relations. Two- and three-dimensional, discrete dislocation dynamics (2D/3D-DDD) has been used to investigate the hardness deviation, affected by the in-plane stress and indentation depth. In the following, we will present our results of the 2D-DDD simulation model that was studied in Dr. Papanikolaou's group, that was used for polycrystalline aluminum study, and provides significant insights towards understanding experimental data [68]. The model is shown in Fig. 3.1. A sample with length of $1000\mu m$ and a thickness of $50\mu m$ is indented by a circular indenter with a radius of $R = 1\mu m$. In-plane stresses are applied to the material prior to indentation. The finite element mesh is extremely refined where the indentations are taking place. The element size in this region is $0.5nm$.

Figure 3.1: Schematic of DDD simulations. The width is $w_p = 20\mu\text{m}$ and the height is $h_p = 10\mu\text{m}$. The red dots indicate the dislocation sources, and the blue dots indicate obstacles. These dots are randomly distributed on slip planes [78].

Figure 3.2: 2D DDD results for polycrystalline Al. (a) shows the load-depth curves with increased applied stress. (b) shows the normalized hardness and the dotted line shows the theoretical model for indentation depth of 5nm. (c) shows the event statistics for indentation depth up to 40nm. (d) shows how the in-plane stress effects on the total number of dislocations normalized by N_0 . N_0 is the number of dislocations at zero in-plane stresses [78].

2D-DDD simulations are shown in Fig. 3.2 and compared with experimentation, where the simulation is a simplified model of the experiment, since only 2D planes are examined. The simulation field can be controlled and examined to the last detail, in contrast with experiments, where the controllable variables are limited. Nevertheless, this simplified model can accurately describe experimental procedures since it is based on theoretical models.

Moreover, three-dimensional, discrete dislocation dynamics (3D-DDD) were also used to investigate the hardness transition with increasing applied stress. A flat-punch tip was used for this simulation for the investigation of hardness transition. Figure 3.3 (a) shows the random pre-existing dislocations a copper sample. Figure 3.3 (b) shows a tensile test implemented on the copper sample, to determine what pre-stresses we should assign before indentation. Figure 3.3 (d) shows that the dislocation densities increase with applied stress. For this sample, we picked (0, 45, 90, 135, 180 MPa) as our pre-stresses. 0 and 45 MPa are the applied stresses that stays in the elastic regime. 90, 135, and 180 MPa are applied stresses that are in the plastic regime. It is clear from the results (Fig. 3.3 (c)) that the hardness transitions with increasing applied stress. 3D-DDD is an ideal way to investigate this problem, but there are time constraints on this type of simulations. 3D-DDD simulations also have problems with implementing boundary conditions and the codes are not as optimized as in the 2D-DDD simulations.

Furthermore, a dislocation density model can be used for examining small-scale plasticity considerations. The indentation size effect in the nano scale was linked to a ratio of the surface energy and the plastic strain dissipation by Gerberich and colleagues [83]. The hardness equation for spherical indentation tips becomes:

$$H \cong \frac{\sigma_f}{\left(\frac{S}{V}\right)^{\frac{2}{3}}} * \frac{1}{2\delta R^{\frac{1}{3}}}, \quad (3.1)$$

where H is the hardness, S/V is the plastic surface area over volume ratio, δ is the indentation depth, and R is the tip radius. $S/V * \sqrt{\delta}$ decreases at small indentation depth. We define σ_f as the local material flow stress as a function of the local dislocation density. The local flow stress should have a complex non-monotonic dependence on the local dislocation density at the nanoscale [84]. We then suggest that the flow stress is a function of the local dislocation density during indentation:

$$\sigma_f = \frac{\beta}{R\sqrt{\rho}} + \alpha b\sqrt{\rho}, \quad (3.2)$$

where ρ is the local dislocation density, R is the indenter tip radius, b is the magnitude of the Burgers vector, and β and α are dimensionless fitting parameters, and for this circumstance they are 1.76×10^{-3} and 0.46. In figure 3.2 (b) the dashed line at 5nm agrees with our simulation and experimental results.

Figure 3.3: (a) the 3D indentation model, with Frank-Read sources whose initial density is $5e12/m^2$. The top is a flat punch with a diameter of 250nm. (b) a tensile test done on (a) to determine the tensile pre-stress that should be assigned. (c) the hardness vs applied stress with the 5 pre-stresses that we chose (0, 45, 90, 135, 180MPa). In Figure (c) we see clear hardness transition with increasing Applied stress at a depth of 5nm. (d) dislocation density of the material increases with applied horizontal stress (MPa).

4. Nanoindentation Experimentation

4.1. Specimens

The testing materials for this project are from the FCC family of crystalline structures (Cu, Ni, Al). Commercial aluminum and copper bulk sputtering targets (99.99% purity, Plasma Materials Inc., US), commercial copper, aluminum bulk single crystals (100), and polycrystalline Nickel (99.99% purity, MTI Corporation., US), were the materials used in this study. The Single crystal

and polycrystalline Nickel were polished on purchase. The polycrystalline Al and Cu were electro-polished using the standard protocols at the third-party materials supplier (Materials Resources LLC, Dayton OH). The polycrystalline Cu and Al samples both have the same dimensions (in length x width x height) (50mm x 25mm x 2mm). The polycrystalline Ni has the dimensions (50mm x 25mm x 1mm). The single crystal samples dimensions are (10mm x 10mm x 1mm).

The testing samples were all put into a custom built 4-pt bending apparatus. Two apparatuses were designed for the specifications of the poly and single crystal samples. The apparatuses are shown in Fig. 4.1. Each apparatus has a screw in the center that, when turned, would move the metal stage upward and apply vertical stress on the samples, which causes the top surface to experience a tensile force. These structures were used to push the material into the yielding and plastic regions while we investigated the surfaces with nanoindentation. Figure 4.2 shows the single crystal Cu and Al and polycrystalline Cu and Ni in the 4-pt bending system. During the surface indentation, this was the experimental setup used to apply in-plane tension.

Figure 4.1: (a) and (b) show the beginning schematics used to build the 4-pt bending systems. (a) is the 4-point bending system that was used on the single crystal samples since their smaller size could not fit in the bigger system. (b) was used on all the poly crystal samples. The empty space below in the center is where the metal plate that pushes against the samples is placed.

Figure 4.2: (a-d) shows the materials sitting in the 4pt-bending systems. (a) shows the poly crystal Cu. (b) shows the poly crystal Ni. (c) shows the single crystal Al. (d) shows the single crystal Cu. (b-d) show samples after indentation while (a) gives a representation of the initial stage before applying the strain gauge.

While the top surface is under tension, the total strain values were recorded using uniaxial strain gauges that had a resistance value of $320\ \Omega$ (National Instruments Inc., TX). The in-plane stresses were calculated by fine element simulations (ABAQUS) using the strain gauge values and the sample deflection. Tables 4 (1-5) gives the measured deflection and total strain, and the calculated plastic strain and tensile stress. The single crystal aluminum sample only had enough applied stress to reach 0.1% strain, before clear surface steps started forming. For the same reason in the single crystal Cu, we only reach 0.16% strain. Figure 4.7 (a) shows a representation of the beginning stages of the experiment, and Fig. 4.7 (b-d) shows the samples after in-plane tension has been applied. Figure 4.7 (b-d) shows the strain gauges that were attached before experimentation.

Table 4.1: Polycrystalline Al

Deflection, μm	Total strain, %	Plastic strain %	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

Table 4.2: Polycrystalline Cu

Deflection, μm	Total strain, %	Plastic strain %	Tensile Stress, MPa
117	0.101	0	111
139	0.133	0.0004	129
193	0.173	0.004	139
249	0.242	0.01	155
271	0.279	0.125	161
316	0.327	0.167	172
360	0.378	0.021	190
405	0.435	0.025	191
449	0.495	0.03	195
539	0.597	0.04	203
560	0.629	0.043	205

Table 4.3: Polycrystalline Ni

Deflection, μm	Total strain, %	Plastic strain %	Tensile Stress, MPa
14	0.012	0	20
34	0.03	0	52
59	0.049	0.014	70
66	0.067	0.02	70
99	0.11	0.07	70
121	0.16	0.11	70
145	0.203	0.15	70
169	0.252	0.19	71
237	0.355	0.3	72

Table 4.4: Single Crystal Al

Deflection, μm	Total strain, %	Plastic strain %	Tensile Stress, MPa
2.3	0.005	0	3.5
9.3	0.05	0.009	20
11.2	0.1	0.04	20.11

Table 4.5: Single Crystal Cu

Deflection, μm	Total strain, %	Plastic strain %	Tensile Stress, MPa
13	0.04	0.02	33
17	0.08	0.045	33.4
22	0.12	0.09	33.5
25	0.14	0.1	33.6
26.7	0.16	0.13	33.7

4.2. Experimental Procedures

The 4-pt bending apparatus, using a sapphire sample, had a frame stiffness value of 1.516×10^6 N/m and the value stayed constant throughout all the procedures. The Nanoindentation was carried out by an iNano that was made by Nanomechanics Inc., TN [6, 79]. The nanoindenter was equipped with a diamond Berkovich tip. The dynamic force oscillation was disabled for detecting the displacement bursts and the feature was enabled throughout the hardness tests. The indenter uses the Oliver-Pharr continuous stiffness method (CSM), as mentioned in Sec. 2.1, to measure the hardness of a material.

Figure 3.3 shows the nanoindenter set-up during a video calibration. Video calibrations in the iNano are important to keep precision between the camera and indenter. System calibrations were also done to help the indenter distinguish the tip it was using. The last test was an operational calibration. The actuator, which is the component the tip is installed to, controls displacement, force, and spring stiffness. The operational calibration is used to make sure the actuator is working properly, and the inner components are not broken. Each calibration was done in between samples for optimal testing.

After the machine is properly calibrated, the hardness and the load-control protocol indentations were carried out. Hardness protocol used a constant indentation strain-rate of $0.2 s^{-1}$. The dynamic displacement was measured to be at $0.3 nm$ that had a standard deviation of $0.1 nm$. The frequency used for the hardness and loading protocols was at $100 Hz$. The displacement was measured with a differential capacitive sensor which can measure up to a resolution of $0.01 nm$, but the environmental noise was consistently measured to be around $2 nm$ consistently. Environmental noise is one of the toughest things to filter out of in experimentation. To fight the environmental noise the following steps were taken: The iNano indenter was set up in a laboratory where external noise gets buffered out by the rooms walls, the nanoindenter was placed on an advanced vibration-free table. These steps provided the optimal environment for the machine to run. During every test the thermal drift rate was sustained at less than $0.2 nm/s$.

The biggest challenge for collecting thousands of indentations is the experimental time. The time required for the standard load-control protocol ranges from 5-10 minutes. For example, 10,000 indentations would require a minimum run time of 70 days. This approach is unreasonable for collecting large data sets for statistical analysis. The above standard protocol was edited in the following ways: Firstly, we disregarded the unloading portion of the load-displacement process.

Secondly, we scaled the indenter approach rate. The new fast-protocol indentation time took no longer than 15s. When approaching the surface, the indenter does soft taps to obtain the surface of the material. Once the indenter tip makes contact with the surface, the force and displacement readings at that moment become the zero load and depth values. Even with the increased speed, the time frame for acquisition of this data took a little over a year.

Figure 4.3 visual representation of the process of indentation during a video calibration. The camera is the first component with the indenter tip in the second component.

The samples, prior to indentation, were vacuum sealed to protect from oxidation. Before indentation, the samples were removed from the seal and allowed one hour to thermally equilibrate. The experimental set-up for all testing materials had the following nanoindentation procedures: All the FCC samples were placed in the 4-pt bending apparatus, and in-plane stress conditions were applied. The polycrystalline samples were placed in the apparatus and stress was applied through the motion of a set-screw that had a $70\mu\text{m}$ deflection and was verified through the measured strain values.

Table 4.6 shows the experimental set up for all materials that were used in this study. The first row shows the set maximum depth for all tests. The second row shows the indentations per stress level. The third row shows the grids for each section done in the stress level. For example, poly crystal Cu had total number of 1000 indentations with a 10x10 grid size. This means that there were 10 different grids per stress level. Figure 4.9 presents the landscaping for the load control protocol over different strains. The loading rates for the poly crystal samples were set to 0.5 *mN/s*. It can be observed that for the single crystal samples the loading rate was set at 0.2 *mN/s*. The single crystal samples are extremely soft, which makes the load needed to reach the target depths substantially lower than the needed load for the polycrystalline samples. Using this experimental framework, the only difference in the experimental parameters were the in-plane stresses, therefore, the difference between experimental results of the data sets are expected to be a consequence of the in-plane stress.

As mentioned in section 2.4, the use of high-throughput indentations was done to statistically average out the roughness effect. In table 4.6, the poly crystal samples have a higher indentation value. This is mainly due to the need of our averaging statistics. The single crystal materials were much flatter with less deviation on the surface. Knowing this, we did not need as many indentations for accurate statistical analysis.

Table 4.6: Nanoindentation array layout

	Poly crystal Al	Poly crystal Cu	Poly crystal Ni	Single crystal Al	Single crystal Cu
Maximum Depth, nm	50	100	100	150	150
Number of indentations per stress level	5000	1000	1000	1000	500
Grid Size	50x50	10x10	10x10	10x10	10x10
Loading Rate (dP/dt) mN/s	0.5	0.5	0.5	0.2	0.2
Number of Stress levels	9	12	10	10	5
Total number of indentations	45000	12000	10000	5000	2800

Figure 4.4: landscape representation of the poly crystal Ni load protocol. The white space is filled with hardness protocol arrays. The different colors represent different strain levels.

5. Experimental Results/ Discussion

5.1. Description of data analysis tools

In this section, we are going to discuss the procedures that were taken to analyze the data. After the indentation runs were finished, the data was exported into CSV files (Comma Separated Values) and transferred to a computer setup to do data analysis using Python. Table 5.1 shows a sample of the basic structure of the data obtained from the loading protocol. Hardness data sets, also include indentation modulus and the hardness values, which are not present in the load representation.

	A	B	C	D	E	F	G	H	I	J
1	Markers	(dP/dt) / P	d(Disp)/d(Tir	Depth	Load	SurfaceZPosi	Time	Time On Sam	X Axis Positi	Y Axis Positi
2		%/s	nm/s	nm	mN	Åµm	s	s	Åµm	Åµm
3	Surface Index		152.513063	0	0	8162.12136	12.542	0	45453.5858	43576.5416
4		570.788827	148.721358	-0.1629643	8.42E-05	8162.12136	12.544	0.002	45453.5858	43576.5416
5		225.608102	142.966982	-0.1305238	0.00021305	8162.12136	12.547	0.005	45453.5858	43576.5416
6		138.500994	137.113431	-0.3260565	0.00034704	8162.12136	12.55	0.008	45453.5858	43576.5416
7		100.981872	130.859104	0.39020656	0.00047598	8162.12136	12.553	0.011	45453.5858	43576.5416
8		79.0743338	124.946148	0.32481954	0.00060785	8162.12136	12.556	0.014	45453.5858	43576.5416
9		69.1698958	121.188786	0.16195232	0.00069489	8162.12136	12.558	0.016	45453.5858	43576.5416

Table 5.1: visual representation of how the data is set-up before analysis with python. The actual sets are substantially bigger then this little cut out. For the load analysis the two main columns are the Load and Depth columns.

Once the acquisition was completed, we started the data analysis. The data was imported using Pandas. Pandas is a python library used primarily for data science and analysis. It allows the user to keep the structural basis of the csv file in a Dataframe. This importation method is user friendly that makes the process of analyzing data easier. After importation, the data was grouped by its strain level. Furthermore, due to the inordinate number of tests per strain level, it was found that the measurements had to be statistically processed. As you will see in the raw data figures presented in the experimental section, analyzing such data can be extremely difficult.

Two key steps were taken for statistical processing: Measurements were grouped by a 5nm depth range (arbitrarily chosen), where the range is called a bin, and an averaging technique was used to remove outliers. Data binning was needed to accurately average the data for our investigation purposes. For both the hardness and load data, a running average was implemented for the removal of outliers. The removal process was simple. For each strain level, we grouped all the data and averaged each of the binned depths. This is how we created a running average for each strain level. The averaging technique was implemented to the data and if the data deviated by 5GPa or 5mN (depending on the nanoindentation protocol) the data point was removed.

Through this process, we were able to “clean” the data sets and obtain figures that clarified our investigation. Hardness’s main focal point for analysis is the changes in the hardness values across the in-plane stresses. Load’s main focal point for analysis is the changes in the pop-in statistics across the in-plane stresses. The following procedures were done on the hardness protocol data: The “cleaned” data sets were averaged along with the calculation of the standard deviation of each bin. For the process of looking at transitions, two important figures were made. Firstly, the hardness vs. depth curves, where we plot the average point along with the standard deviation as the error bar. The standard deviation error bar was calculated by:

$$\sqrt{\frac{\frac{R^2}{N} * (\frac{R}{N})^2}{N}} * \sqrt{\frac{N}{N-1}}, \quad (5.1)$$

where R is the hardness or load (depending on the protocol), and N is the total amount of data points in the bin.

This process was done to each strain level and plotted together. The second was the hardness vs. strain at variable depths. Both figures are presented in the experimental section. The same procedures were implemented on the load protocol data, but additional measures needed to be taken to examine the pop-ins. For the analysis of the pop-in noise, the code used on the loading protocol was extended in the following ways. The pop-in noise event in our study is defined as $S = \dot{a}_{\text{if } dh > h_{\text{thr}}} dh_i$, to evaluate the displacement bursts in a quantitative manner, using the threshold value h_{thr} to be equivalent to the machine noise threshold $h_{\text{thr}}=0.2nm$. S is equal to the magnitude of one single displacement burst, while $P(S)$ is the probability density of an event taking place. The probability density vs. the magnitude, $P(S)$ vs. S , was plotted to look at the probability event distribution. In the experimental sections, you will see the FCC materials raw data, statistically analyzed data, and a comparison among the crystals results. At the end of the section, we will also present some conclusions drawn from our findings.

5.2. Polycrystalline Aluminum

In order to get an accurate investigation of the influence of the applied stress on the curves, the indentation depths were kept in a square surface area of 1 mm x 1 mm in the center of the materials. This was to create an indentation that would hit the surface with the same strain reading as the strain gauge. As we move away from the center of the sample, the strain readings are less accurate. In the following figures, our analysis will consist of the hardness-depth curves and then we will follow with the analysis on the load-depth curves.

Figure 5.1: the raw data from the indentation readings. Different symbols/colors indicate different indentation test. Each figure has almost 200 indentations in them. (a-b) shows the decrease in the hardness as more in-plane tension was applied to the specimen. (a) Shows the hardness-depth curve for 0% strain and (b) shows the hardness curve at 0.45%

Figures 5.1 (a-b) present us with clear variations of hardness as the in-plane stress is increased along with the strain. In 5.1 (a) the hardness is increasing with increasing indentation depths, until it reaches a peak at about 4 GPa and then dropping off and leveling out at about 1 GPa. Even as the peak decreases with increasing strain it is observed that they all level out to about 1 GPa. High statistical variations can also be observed in the figures. In figure 5.2 (b) we see that the peak reaches about 2 GPa and levels out at about 1 GPa. The indentation size effect is clearly shown in the polycrystalline aluminum data at depths just below 12 nm and dissipating after that.

Figure 5.2: Counterpart of 5.1 after the binning and removal of outliers. Figure 5.2 is binned at every 2.5 nm. Different symbols/colors indicate different indentation test

Figure 5.2 shows us the hardness-depth curves, after a noise reduction technique has been applied to the datasets. The outliers seen in Fig. 5.1 are gone, and the data is separated into bins, for increasing depths. For analysis purposes, it is almost impossible to just analyze the original

curves, so binning and then smoothing the data allows a more precise analyzation of the applied stress effect. We then record the average and the standard deviation of each bin.

Figure 5.3: the average hardness-depth curves. Different symbols/ colors represent a different strain, and there are about 200 curves averaged in each strain. (a) shows the hardness is equivalent to a power law of the strain. (a) provides the full hardness average where (b) is zoomed in on the major area of focus. (c) shows the average hardness at each binned depth with respect to the strain.

Figure 5.3 (a-c) show clear transition in hardness as the strain is increased. (c) shows how the effect is represented until it reaches about the 12.5nm depth. Once it reaches this depth it begins to level out with the increase in strain. Even though the natural disorder is due to small indentation depths. We can observe a convincing association between the depth-dependent hardness and the applied in-plane stress. As observed in Fig. 5.1 the hardness peak goes to almost 4GPa in (a) and almost falls below 2GPa by the time it reaches (b).

Figure 5.4: (a-c) are the load depth curves at different strains. Different symbols/colors indicate different indentation test. There are about 800 indentation tests in each figure. (a) shows indentations at 0.25% strain, (b) shows indentations at 0.4% strain.

For all tests, the load-displacement curves present a continuous elastic response immediately followed with numerous measurable displacement bursts. The displacement bursts or “pop-ins” (section 2.5) can be clearly observed in Fig. 5.4. Figure 5.5 shows how the loading data was binned before analysis. The approach for averaging the load data is similar to that of the hardening data. For pop-in, an analysis was done on the raw data from Fig. 5.4.

Figure 5.5: (a-b) shows the statistically processed and binned load-depth data sets. By getting rid of the outliers we can now get a more accurate averaging of the load-depth. Different symbols/colors indicate different indentation test. The data is separated at a bin size of 2.5nm.

Figure 5.6: (a) four load depth curves for a clearer representation of the pop in events. (b) shows the average load at each binned depth. In (b) about 800 indentation tests are averaged in each strain. (c) shows the probability density of pop-in events as a function of event size.

As observed in Fig. 5.6 (a), all the load depth curves across material show continuous elastic response with an immediate measurable displacement burst, and then followed by more displacement bursts. The overall behavior in the load-depth curves is characterized by the intrinsic material noise that is attributed to the surface roughness and stochasticity of plastic deformation. This is especially true since the samples were not thermally annealed. The load-displacement curves observed in Fig. 5.6 shows a large deviation which can be clarified by the nano-mechanical behavior given the large statistical testing over a highly homogeneous surface. The pop-in statistics of the probability distribution are plotted as different strain values in Fig. 5.6(c). Figure 5.6 (c), where S is the magnitude depth of a single displacement burst, and $P(S)$ is the probability density.

In this section we covered and explained all the data and the observations. The other materials are done and analyzed the same way, so we will just quickly look at all the results and see if the same effects are happening across all materials.

5.3. Polycrystalline Copper

As the polycrystalline copper results are presented the comparison with polycrystalline aluminum will be made. The polycrystalline copper was indented after the aluminum tests were finished.

Figure 5.7: (a-b) are hardness curves for Cu as we applied more stress on the samples. Different symbols/colors indicate different indentation test. Almost 100 hardness measurements are in each strain plot. (a) is the copper at 0.101% strain and (b) is at 0.327% strain.

Figure 5.7 (a-c) shows clear hardness transition with more applied stress, but when we look at the averaging statistics in Fig. 5.8 the effects aren't as clear. This is due to the variability in each of the test sequences and the roughness of the material. The polycrystalline Cu samples have rougher surface compared to the other materials. The poly crystal Cu peaks at about 25 GPa compared to the much smaller peak of 4 GPa for the polycrystalline Al. The polycrystalline Cu still shows the indentation size effect at depths below 10nm. This transition is happening faster than the transition of poly crystal Al. The effect on the Al lasts till 12.5nm unlike the smaller 10nm for the Cu.

Figure 5.8: (a) that hardness is equivalent to a power law of the strain. (a, b) different symbols/colors indicate different indentation strain. There are about 100 hardness plots averaged in each strain. (a) shows the hardness average per strain as a function of depth. (b) presents a zoomed in portion of the main area of analysis of (a). (c) shows the hardness transition from shallow depths to 10nm then (the light grey box line) we see this transition end. The transition for the poly crystal copper happens at faster rate than we see in the poly crystal aluminum.

Figure 5.9: (a-b) load-depth data curves at different strains. Different symbols/colors indicate different indentation test. There are about 1000 indentation tests in each figure. The pop-in events are not as clear as they are in the poly crystal Al. (a) shows the load-depth curves for 0.101% strain and (b) shows the load-depth curves for 0.173% strain.

Figure 5.10: (a) shows 4 selected load-depth indentations for a better representation of individual curves. (b) shows the average load with respect to depth and is observed that the variability is not very high. There are about 1000 tests averaged per strain. (c) shows the pop-in probability density for 1000 tests for each strain.

The polycrystalline Cu pop-ins are substantially smaller than the pop-ins that are observed in the polycrystalline Al data. One explanation that can be made is that the copper is a harder material and the indentation pop-ins are more subtle, and since the material is harder that may contribute to a smaller magnitude in the pop-in. It can also be observed that at 50nm, the max load, needed to reach that depth is 0.45mN for Cu and 0.2mN for the Al. Even though the aluminum is a softer material, it is interesting to see that the copper sample needed over twice the load of aluminum to reach the same depths. The last thing that can be noticed is the probability density of the pop-in in the copper has a smoother curve than the aluminum, but the probability is much lower which makes sense because the pop-ins are smaller.

5.4. Polycrystalline Nickel

As we present the Nickel data we will detail the comparisons between all three FCC crystals. The set-up was the exact same for all three testing materials. Now we will dive into how they all compare together.

Figure 5.11: (a-b) hardness curves for Ni as we applied more stress on the samples. Different symbols/colors indicate different indentation test. Almost 100 hardness measurements are in each strain plot. (a) is the nickel at .030% strain and (b) is at 0.160% strain.

The nickel curves are identical to the copper curves seen in Figs. 5.7 and 5.11. The applied stress effect is observed, but it seems to be subtle and dissipate at about 6nm unlike the copper which dissipates at 10nm copper. While the copper and nickel look similar it can be shown that the nickel has an even more subtle effect than the copper. This can be observed in 5.8 and 5.12. One key reason for this is the difference in the strain readings. If you compare the Figs. 5.8 and 5.12, it will be noticed that the nickel and copper have close to the same hardness readings at shallow depths, but as the depths increase, the nickel levels out to about 5GPa compared to the Cu's 3GPa. The biggest thing to bring away from this comparison is that the applied stress effect is apparent in all three FCC crystals.

Figure 5.12: (a) shows that hardness is equivalent to a power law of the strain. (a, b) different symbols/colors indicate different indentation strains. There are about 100 hardness plots averaged in each strain. (a) shows the full hardness average with respect to depth curve. (b) shows the area of interest and (c) shows us similar results to poly crystal copper. The nickel transition happens very fast as well at about the 6nm range and then after that the effect seems to dissipate.

Figure 5.13: (a-b) show load-depth data curves at different strains. Different symbols/colors indicate different indentation test. There are about 1000 indentation tests in each figure. The pop-in events are not as clear as they are in the poly crystal Al. (a) gives the load-depth curve for 0.012% strain and (b) shows the load-depth curve for 0.110% strain.

Figure 5.14: (a) shows the load-depth curves of 4 data sets to see a better representation of how one curve looks. (b) shows the variation in the average load across the strain levels. In (b) there are about 1000 tests averaged per strain (c) shows the probability density across 1000 tests per strain value.

The pop-in effect for nickel can be observed similarly copper with smaller pop-ins than the Al. The probability density shows this with a lot lower values than the aluminum and just slightly smaller values than the Cu. Now that we have observed the poly crystal data curves, we are going to take a look at the results of the single crystal testing. The purpose of the single crystal testing was to make sure the effect we are seeing was not caused by the grain size or grain boundary. In single crystal, the applied stress effect should be more prominent with the absence of these effects. After we finish the single crystal analysis, we will review and compare data the for single crystal and poly crystal.

5.5. Single Crystal Aluminum

As mentioned in the previous section, purpose of the single crystal results was to verify the results observed in the poly crystal samples. The same procedures and analysis was done to both the single crystal Al and single crystal Cu. However, it needs to be mentioned that the single crystal samples are extremely soft and yield exceptionally fast, meaning to reach and surpass the yielding stress the required strain was lower than the polycrystalline materials.

Figure 5.15: (a-b) are figures showing the raw data from the indentation readings. Different symbols/colors indicate different indentation test. Each figure has almost 1000 indentations in them. (a-b) shows the decrease in the hardness as more in-plane tension was applied to the specimen. (a) Shows the hardness-depth curve for 0% strain and (b) shows the hardness curve at 0.100% strain.

Figure 5.16: (b) shows that hardness is equivalent to a power law of the strain. Different symbols/colors indicate different strains. Each figure has almost 1000 indentations averaged in each strain. (a) shows the full hardness average with respect to depth curve. (b) shows the area of interest and (c) shows us similar results to poly crystal aluminum. (a-c) shows the hardness at shallow depths transitions almost immediately, but the effect is still present till 12nm

Figure 5.16 shows the applied stress effect is present in the single crystal aluminum with increasing in-plane tension. Figures 5.15 and 5.16 give evidence that the applied stress does have

an effect on the bulk surface hardness tests. From these results, we can see that the hardness decreases with the increase in applied strain but is only in the range below 12nm .

Figure 5.17: (a) shows the load-depth curves of 4 data sets to see a better representation of how one curve looks. (b) shows the variation in the average load across the strain levels. In (b) there are about 1000 tests averaged per strain (c) shows the probability density across 1000 tests per strain value.

Figure 5.17 shows us the loading analysis for the single crystal Al. The probability statistics give a similar curve to the poly crystal Al. The pop-ins are distinctive and can be seen in the black curve in Fig. 5.17(a). This data gives us the evidence we need for verification in the polycrystalline Al.

5.6. Single Crystal Copper

In this section we will be comparing the results of the single crystal copper and polycrystalline Cu.

Figure 5.18: (a-b) are figures showing the raw data from the indentation readings. Different symbols/colors indicate different indentation test. Each figure has almost 500 indentations in them. (a-b) shows the decrease in the hardness as more in-plane tension was applied to the specimen. (a) Shows the hardness-depth curve for 0% strain and (b) shows the hardness curve at 0.140% strain.

Figure 5.19 gives us similar results to the poly crystal copper. The applied stress effect dissipates at around 8nm instead of 10nm which was observed in the poly crystal copper. The single crystal copper and aluminum both show the evidence needed to verify the applied stress effect. Figures 5.17 and 5.20 shows evidence of the residual size effects in the nano regime, which also gives us verification of the poly crystal results.

Figure 5.19: Different symbols/colors indicate different strains. Each figure has almost 500 indentations averaged in each strain. (a) shows that hardness is equivalent to a power law of the strain (a) shows the full hardness average with respect to depth curve. (b) shows the area of interest and (c) shows us similar results to poly crystal copper. (a-c) shows the hardness at shallow depths transitions almost immediately, but the effect is still present till 15nm

Figure 5.20: (a) shows the load-depth curves of 4 data sets to see a better representation of how one curve looks. (b) shows the variation in the average load across the strain levels. In (b) there are about 500 tests averaged per strain (c) shows the probability density across 500 tests per strain value. The pop-in statistics is consistent with various theories of avalanche phenomena [85]. However, this topic lies beyond the purpose of this work.

Figure 5.21: provides us with the AFM images that was done on the single crystal samples. (a) shows us the single crystal aluminum results and shows variations around 4nm. (b) shows the surface of the single crystal copper and the surface only has a surface roughness below 1nm. (b) has the least amount of roughness and results would be more influenced by the applied stress effect.

The surface roughness plays a huge contribution in the accuracy of data acquisition. The single crystal samples were taken to AFM to make sure that the roughness would not greatly affect the statistical analysis. In the single crystal experiments, because of the limitations on the grain size, grain boundary and surface roughness, the effect of applied stress is more prominent. The noise statistics in the single crystal copper are similar except for the 0.16 strain. The 0.16 strains difference can be explained by the occurrence of surface steps from the quicker yielding. This makes the data accuracy drop and can explain the variation in the pop-in statistics analysis.

5.7. Construction of a Elasticity-Plasticity Transition Order Parameter

In this section, we will be introducing a new method for investigating plasticity transition. The idea revolves around pop-in events. It is well known that pop-in events are the initiation of plasticity [61, 63, 67]. If we analyze dh/dp at a pop-in, the value should be going toward infinity, because dp would approximately be equal to zero. We believe there will be variance in the order parameter (dh/dp), but at the moment of transition from elastic to plastic there should be a peak in this order parameter. The order parameter was a big goal for this research, and to show how useful it can be, we are going to look at a recent study trying to investigate the pop-in in a similar way.

A study was conducted in 2012 on the pop-ins by Wang, Zhong, Lu, Lu, McDowell, and Zhu [86]. In this study, they did experimentation on three single crystal copper samples using nanoindentation with Berkovich tip. The single crystal samples were grown by the Bridgman technique, and cut into rectangles by spark erosion. To get the results seen in Fig. 5.23, the single crystal samples were annealed in a vacuum chamber for six hours and the temperature was set 800 degrees Celsius. The surface roughness for the single crystals was below $3nm$. The single samples used in our study had a surface roughness of about $2nm$. Figure 5.23 (a) shows the pop-in results for the three single crystal materials. The most important line in this figure is the purple line (crystal orientation $\{100\}$), because that matches our single crystal copper. In contrast with our results, a

pop-in event is easily seen (where load plateaus in the region of 10-14nm). This is a consequence of them annealing of the sample for six hours.

Figure 5.22: (a) experimental results of nanoindentation on (111), (110), and (100) copper crystal orientation. (b) shows the frequency histograms of the critical indenter force and displacement at the onset of displacement bursts [86].

In our study, we believe these steps are not needed and can be replaced with an order parameter that allows for investigation with indentation pop-in. We obtained the order parameter in the following way:

$$\frac{dh}{dp} = \frac{x(i) - x(i-3)}{y(i) - y(i-3)}, \quad (5.2)$$

where $x(i)$ is equal to the depth and $y(i)$ is equal to the load in a current measurement i along the curve. We examine depths and load at three measurements apart ($i, i-3$) in order to avoid environmental noise.

Figure 5.24 (a-b) shows the order parameter and the variance of the order parameter with respect to strain. From this figure you can see a clear transition happening at 0.12 % strain. Figure 5.24 (a-b) are important, because this shows the validity of our order parameter. In figure 5.23 (a-b), you see that the pop-in for the orientation {100} happens around 25-30 μN or 0.025-0.03mN.

From figure 5.25 you see that the peak pop-ins happen at about $0.025mN$. This is important because this opens the door for different types of analysis on plasticity.

Figure 5.23: (a) the plotting of the order parameter vs total % strain. Each line represents a different indentation depth. The shallower depths have the straighter lines, because close to 5nm is when the first pop-in happens. Meaning anything smaller than the 5nm should be almost completely flat. The deeper depths have a peak at 0.12 % strain which indicated the elastic to plastic transition. (b) shows the variability of the order parameter and is consistent with (a).

Figure 5.24: (a) the order parameter with respect to a certain load. The peaks in (a-b) are the showing the initiation of plasticity. The peak position is consistent with the locations in the annealed samples [86], as shown in Fig. 5.22

The order parameter is a new technique that is equal to the inverse stiffness and can be used to investigate plasticity and yield stress. What has been presented in this thesis, is one of the beginning stages of our investigations. The order parameter needs to be processed, refined, and understood further. The next stages in this problem is connecting the order parameter to the Bulk hardness, if we can connect these to parameters we will be able to solve for the yield stress through nanoindentation. Tabors law $P_m = 2.6Y$ to $3Y$ (20) can also be written as:

$$H_{Bulk} = 2.6\sigma_y \text{ to } 3\sigma_y, \quad (5.3)$$

where H_{Bulk} is equal to the hardness of the bulk of the material. This equation is a part of the total hardness:

$$H = H_{Bulk} + H_{Surface}, \quad (5.4)$$

where $H_{Surface}$ is the hardness of the material on the surface. If we take nanoindentations at the zero stress that should give us $H_{Surface}$ and then measure the hardness with applied stress that would give us H , and if we subtract these to curves it should give us the Bulk hardness curve. If this works, we will be able to connect continuum mechanics to the nano-scale and create a non-destructive way to find the yield stress of a given material.

6. Conclusion

This study provides clear evidence of the applied stress effect in all the FCC crystal materials. In all experiments, ISE's were observed and affected by the in-plane stresses. The comparisons between copper and nickel are interesting, because they share similar results in the hardness and pop-in statistics, in contrast with the hardness and pop-in statistics in aluminum. The aluminum polycrystalline depth-dependent hardness measurements, show a clear transition close to $10nm$ as the applied in-plane stress is increased. Polycrystalline copper depth-dependent hardness measurements, show clear transition close to $10nm$. The nickel depth-dependent hardness measurements, show clear transition close to $6nm$. This indicates high stochastic behavior at small indentation depths, which disappears as in-plane stresses increase. The differences in the single crystal and their respective polycrystalline materials are a consequence of grain boundary and the grain sizes in the poly crystal samples. The pop-in statistics in all the tested samples show no clear difference across applied strains in the displacement bursts, which means pop-ins are unaffected by the in-plane stresses. Absolved of the limitations in any poly crystal material, our single crystal experiments clearly show the effect of the applied stress, which verifies our findings in the polycrystalline materials results. The order parameter worked for the analysis of single crystal copper, but still needs a lot of refining before the next stages in the research.

In summary, we employed large amounts of nanoindentation on polycrystalline materials (Al, Cu, Ni) and single crystal materials (Al, Cu) at different in-plane stresses, to investigate the incipient plasticity transition across the FCC crystals. Both single crystal copper and single crystal aluminum experiments solidify our findings on the in-plane stress effect in polycrystalline samples. The characteristics from the results of the average hardness plots are consistent with the findings in the polycrystalline samples, validating that the polycrystalline results were not influenced by the grain boundary or the grain size. There was a thought to add single crystal nickel indentations, but we already have two single crystal samples verifying our results. Nanoindentation

experiments on a third single crystal material would be redundant. An interesting observation is why the pop-ins for the aluminum were so much greater than the nickel and copper. The biggest jumps can range over 10nm in the polycrystalline Al, but in Cu and Ni the biggest noticeable jumps are around 1 to 2nm. This observation can be answered in the future with the pairing of simulations. This research also becomes a stepping stone for future nanoindentation experiments. Since our materials had a {100} crystal orientation, future work could include in-depth studies by EBSD on the effect of orientation in single grain/crystal materials. We believe that the order parameter is the beginning step going to be used in creating theoretical equations that will connect the hardness and yield stress in the nanoscale.

7. References

- [1] Tabor, D. (1948). A simple theory of static and dynamic hardness. *Proc. R. Soc. Lond. A*, 192(1029), 247-274.
- [2] Oliver, W. C., & Pharr, G. M. (1992). An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments. *Journal of materials research*, 7(6), 1564-1583.
- [3] Pharr, G. M., & Bolshakov, A. (2002). Understanding nanoindentation unloading curves. *Journal of Materials Research*, 17(10), 2660-2671.
- [4] Li, J., Van Vliet, K. J., Zhu, T., Yip, S., & Suresh, S. (2002). Atomistic mechanisms governing elastic limit and incipient plasticity in crystals. *Nature*, 418(6895), 307.
- [5] Schuh, C. A., Mason, J. K., & Lund, A. C. (2005). Quantitative insight into dislocation nucleation from high-temperature nanoindentation experiments. *Nature Materials*, 4(8), 617.
- [6] Pharr, G. M. (1998). Measurement of mechanical properties by ultra-low load indentation. *Materials Science and Engineering: A*, 253(1-2), 151-159.
- [7] Oliver, W. C., & Pharr, G. M. (2004). Measurement of hardness and elastic modulus by instrumented indentation: Advances in understanding and refinements to methodology. *Journal of materials research*, 19(1), 3-20.
- [8] Minor, A. M., Asif, S. S., Shan, Z., Stach, E. A., Cyrankowski, E., Wyrobek, T. J., & Warren, O. L. (2006). A new view of the onset of plasticity during the nanoindentation of aluminium. *Nature materials*, 5(9), 697.

- [9] Gao, H., Huang, Y., & Nix, W. D. (1999). Modeling plasticity at the micrometer scale. *Naturwissenschaften*, 86(11), 507-515.
- [10] Jarausch, K. F., Kiely, J. D., Houston, J. E., & Russell, P. E. (2000). Defect-dependent elasticity: nanoindentation as a probe of stress state. *Journal of Materials Research*, 15(8), 1693-1701.
- [11] Khan, M. K., Fitzpatrick, M. E., Hainsworth, S. V., & Edwards, L. (2011). Effect of residual stress on the nanoindentation response of aerospace aluminium alloys. *Computational Materials Science*, 50(10), 2967-2976.
- [12] Gao, H., Huang, Y., Nix, W. D., & Hutchinson, J. W. (1999). Mechanism-based strain gradient plasticity—I. Theory. *Journal of the Mechanics and Physics of Solids*, 47(6), 1239-1263.
- [13] Tsui, T. Y., Oliver, W. C., & Pharr, G. M. (1996). Influences of stress on the measurement of mechanical properties using nanoindentation: Part I. Experimental studies in an aluminum alloy. *Journal of Materials Research*, 11(3), 752-759.
- [14] Bolshakov, A., Oliver, W. C., & Pharr, G. M. (1996). Influences of stress on the measurement of mechanical properties using nanoindentation: Part II. Finite element simulations. *Journal of Materials Research*, 11(3), 760-768.
- [15] Doerner, M. F., & Nix, W. D. (1986). A method for interpreting the data from depth-sensing indentation instruments. *Journal of Materials research*, 1(4), 601-609.
- [16] Nix, W. D., & Gao, H. (1998). Indentation size effects in crystalline materials: a law for strain gradient plasticity. *Journal of the Mechanics and Physics of Solids*, 46(3), 411-425.
- [17] Roters, F., Eisenlohr, P., Hantcherli, L., Tjahjanto, D. D., Bieler, T. R., & Raabe, D. (2010). Overview of constitutive laws, kinematics, homogenization and multiscale methods in crystal plasticity finite-element modeling: Theory, experiments, applications. *Acta Materialia*, 58(4), 1152-1211.
- [18] Uchic, M. D., Dimiduk, D. M., Florando, J. N., & Nix, W. D. (2004). Sample dimensions influence strength and crystal plasticity. *Science*, 305(5686), 986-989.
- [19] Schuh, C. A., & Lund, A. C. (2004). Application of nucleation theory to the rate dependence of incipient plasticity during nanoindentation. *Journal of Materials research*, 19(7), 2152-2158.

- [20] Kelton, K., & Greer, A. L. (2010). *Nucleation in condensed matter: applications in materials and biology* (Vol. 15). Elsevier.
- [21] Kelton, K., & Greer, A. L. (2010). *Nucleation in condensed matter: applications in materials and biology* (Vol. 15). Elsevier.
- [22] Wang, L., Bei, H., Gao, Y. F., Lu, Z. P., & Nieh, T. G. (2011). Effect of residual stresses on the hardness of bulk metallic glasses. *Acta Materialia*, 59(7), 2858-2864.
- [23] Butt, H. J., Cappella, B., & Kappl, M. (2005). Force measurements with the atomic force microscope: Technique, interpretation and applications. *Surface science reports*, 59(1-6), 1-152.
- [24] Bhushan, B., & Koinkar, V. N. (1994). Nanoindentation hardness measurements using atomic force microscopy. *Applied physics letters*, 64(13), 1653-1655.
- [25] Rzepiejewska-Malyska, K. A., Buerki, G., Michler, J., Major, R. C., Cyrankowski, E., Asif, S. S., & Warren, O. L. (2008). In situ mechanical observations during nanoindentation inside a high-resolution scanning electron microscope. *Journal of Materials Research*, 23(7), 1973-1979.
- [26] McElhaney, K. W., Vlassak, J. J., & Nix, W. D. (1998). Determination of indenter tip geometry and indentation contact area for depth-sensing indentation experiments. *Journal of Materials Research*, 13(5), 1300-1306.
- [27] Lu, L., Schwaiger, R., Shan, Z. W., Dao, M., Lu, K., & Suresh, S. (2005). Nano-sized twins induce high rate sensitivity of flow stress in pure copper. *Acta materialia*, 53(7), 2169-2179.
- [28] Smith, R. L., & Sandly, G. E. (1922). An accurate method of determining the hardness of metals, with particular reference to those of a high degree of hardness. *Proceedings of the Institution of Mechanical Engineers*, 102(1), 623-641.
- [29] Pethica, J. B., & Oliver, W. C. (1988). Mechanical properties of nanometre volumes of material: use of the elastic response of small area indentations. *MRS Online Proceedings Library Archive*, 130.
- [30] Loubet, J. L., Georges, J. M., Marchesini, O., & Meille, G. (1984). Vickers indentation curves of magnesium oxide (MgO). *Journal of tribology*, 106(1), 43-48.
- [31] Boussinrsq, M. J. (1885). *Applications des potentiels*.
- [32] Hertz, H. (1882). J. reine und angewandte Mathematik 92, 156. *MATH Google Scholar*.

- [33] Love, A. E. H. (1929). The stress produced in a semi-infinite solid by pressure on part of the boundary. *Philosophical Transactions of the Royal Society of London. Series A, Containing Papers of a Mathematical or Physical Character*, 228(659-669), 377-420.
- [34] Love, A. E. H. (1939). Boussinesq's problem for a rigid cone. *The Quarterly Journal of Mathematics*, (1), 161-175.
- [35] Ternovskij, A. P., Alekhin, V. P., Shorshorov, M. K., Khrushchov, M. M., & Skvortsov, V. N. (1973). Mechanical testing of materials by micro pressing technique. *Zavodskaya Laboratoriya*, 39(10), 1242-1247.
- [36] Bulychev, S. I., Alekhin, V. P., Shorshorov, M. K., Ternovskij, A. P., & Shnyrev, G. D. (1975). Determination of Young modulus by the hardness indentation diagram. *Zavodskaya Laboratoriya*, 41(9), 1137-1140.
- [37] Shorshorov, M. K., Bulychev, S. I., & Alekhin, V. P. (1981, August). Work of plastic and elastic deformation during indenter indentation. In *Soviet Physics Doklady* (Vol. 26, p. 769).
- [38] Doerner, M. F., Gardner, D. S., & Nix, W. D. (1986). Plastic properties of thin films on substrates as measured by submicron indentation hardness and substrate curvature techniques. *Journal of Materials Research*, 1(6), 845-851.
- [39] Tabor, D. (2000). *The hardness of metals*. Oxford university press.
- [40] Meyer, E. (1908), *Zeits. d. Vereines Deutsch. Indenieur*, 52,645,740,835.
- [41] Swadener, J. G., George, E. P., & Pharr, G. M. (2002). The correlation of the indentation size effect measured with indenters of various shapes. *Journal of the Mechanics and Physics of Solids*, 50(4), 681-694.
- [42] Pharr, G. M., Herbert, E. G., & Gao, Y. (2010). The indentation size effect: a critical examination of experimental observations and mechanistic interpretations. *Annual Review of Materials Research*, 40, 271-292.
- [43] Mott BW. 1957. Microindentation Hardness Testing. London: Butterworths
- [44] Gane, N. (1970). The direct measurement of the strength of metals on a sub-micrometre scale. *Proc. R. Soc. Lond. A*, 317(1530), 367-391.
- [45] Westbrook, J. H. (1973). The science of hardness testing and its research applications.
- [46] Exadaktylos, G. E., Vardoulakis, I., & Kourkoulis, S. K. (2001). Influence of nonlinearity and double elasticity on flexure of rock beams—II. Characterization of Dionysos marble. *International Journal of Solids and Structures*, 38(22-23), 4119-4145.

- [47] Lam, D. C., Yang, F., Chong, A. C. M., Wang, J., & Tong, P. (2003). Experiments and theory in strain gradient elasticity. *Journal of the Mechanics and Physics of Solids*, 51(8), 1477-1508.
- [48] Mühlhaus, H. B., & Alfantis, E. C. (1991). A variational principle for gradient plasticity. *International Journal of Solids and Structures*, 28(7), 845-857.
- [49] Buckle H. 1959. Progress in microindentation hardness testing. "Metall. Rev. 4:49–100
- [50] Chen, C. C., & Hendrickson, A. A. (1973). Microhardness phenomena in silver. *The Science of Hardness Testing and Its Research Applications*, 21, 274.
- [51] Tabor, D. (1985). Indentation hardness and its measurement: some cautionary comments. In *Microindentation techniques in materials science and engineering*. ASTM International.
- [52] Greer, J. R., Oliver, W. C., & Nix, W. D. (2005). Size dependence of mechanical properties of gold at the micron scale in the absence of strain gradients. *Acta Materialia*, 53(6), 1821-1830.
- [53] Cheng, Y. T., & Cheng, C. M. (2004). Scaling, dimensional analysis, and indentation measurements. *Materials Science and Engineering: R: Reports*, 44(4-5), 91-149.
- [54] Liu, Y., & Ngan, A. H. W. (2001). Depth dependence of hardness in copper single crystals measured by nanoindentation. *Scripta Materialia*, 44(2), 237-241.
- [55] Withers, P. J., & Bhadeshia, H. K. D. H. (2001). Residual stress. Part 1—measurement techniques. *Materials science and Technology*, 17(4), 355-365.
- [56] Dieter, G. E., & Bacon, D. J. (1986). *Mechanical metallurgy*(Vol. 3). New York: McGraw-hill.
- [57] Kokubo, S. (1932). On the change in hardness of a plate caused by bending. *Science Reports of the Tohoku Imperial University*, 21, 256-267.
- [58] Kostron, H. (1933). Der Einfluß eines linearen Spannungszustandes auf die Kugeldruckhärte. *Metallwirtsch. XII*, 473.
- [59] Sines, G., & Carlson, R. (1952). Hardness measurements for determination of residual stresses. *ASTM Bull*, 180, 35-37.
- [60] Blain, P. A. (1949). The Effect of Internal Stresses on Hardness. *Sheet Metal Ind*, 26, 135-136.
- [61] Oppel, G. U. (1964). Biaxial elasto-plastic analysis of load and residual stresses. *experimental Mechanics*, 4(5), 135-140.

- [62] Blau, P. J. (1986). *Microindentation techniques in materials science and engineering: a symposium sponsored by ASTM Committee E-4 on Metallography and by the International Metallographic Society, Philadelphia, PA, 15-18 July 1984*(No. 889). Astm International.
- [63] LaFontaine, W. R., Yost, B., & Li, C. Y. (1990). Effect of residual stress and adhesion on the hardness of copper films deposited on silicon. *Journal of Materials Research*, 5(4), 776-783.
- [64] Suresh, S., & Giannakopoulos, A. E. (1998). A new method for estimating residual stresses by instrumented sharp indentation. *Acta Materialia*, 46(16), 5755-5767.
- [65] Lee, Y. H., & Kwon, D. (2003). Measurement of residual-stress effect by nanoindentation on elastically strained (1 0 0) W. *Scripta Materialia*, 49(5), 459-465.
- [66] Lee, Y. H., & Kwon, D. (2002). Residual stresses in DLC/Si and Au/Si systems: Application of a stress-relaxation model to the nanoindentation technique. *Journal of materials research*, 17(4), 901-906.
- [67] Kese, K., & Rowcliffe, D. J. (2003). Nanoindentation method for measuring residual stress in brittle materials. *Journal of the American Ceramic Society*, 86(5), 811-816.
- [68] Hou, X. D., & Jennett, N. M. (2017). A method to separate and quantify the effects of indentation size, residual stress and plastic damage when mapping properties using instrumented indentation. *Journal of Physics D: Applied Physics*, 50(45), 455304.
- [69] Herrmann, K., Jennett, N. M., Wegener, W., Meneve, J., Hasche, K., & Seemann, R. (2000). Progress in determination of the area function of indenters used for nanoindentation. *Thin solid films*, 377, 394-400.
- [70] Jennett, N. M., & Meneve, J. (1998). Depth sensing indentation of thin hard films: a study of modulus measurement sensitivity to indentation parameters. *MRS Online Proceedings Library Archive*, 522.
- [71] Morris, J. R., Bei, H., Pharr, G. M., & George, E. P. (2011). Size effects and stochastic behavior of nanoindentation pop in. *Physical review letters*, 106(16), 165502.
- [72] Bei, H., George, E. P., Hay, J. L., & Pharr, G. M. (2005). Influence of indenter tip geometry on elastic deformation during nanoindentation. *Physical review letters*, 95(4), 045501.
- [73] Leipner, H. S., Lorenz, D., Zeckzer, A., Lei, H., & Grau, P. (2001). Nanoindentation pop-in effect in semiconductors. *Physica B: Condensed Matter*, 308, 446-449.

- [74] Shim, S., Bei, H., George, E. P., & Pharr, G. M. (2008). A different type of indentation size effect. *Scripta Materialia*, 59(10), 1095-1098.
- [75] Wang, Z., Bei, H., George, E. P., & Pharr, G. M. (2011). Influences of surface preparation on nanoindentation pop-in in single-crystal Mo. *Scripta Materialia*, 65(6), 469-472.
- [76] Kucharski, S., Jarzabek, D., Piątkowska, A., & Woźniacka, S. (2016). Decrease of nano-hardness at ultra-low indentation depths in copper single crystal. *Experimental Mechanics*, 56(3), 381-393.
- [77] Durst, K., Backes, B., Franke, O., & Göken, M. (2006). Indentation size effect in metallic materials: Modeling strength from pop-in to macroscopic hardness using geometrically necessary dislocations. *Acta Materialia*, 54(9), 2547-2555.
- [78] Hakan Yavas, Hengxu Song, and Ryder Bolin. “Detecting the onset of bulk crystal plasticity using nanoindentation in pure Al” under review.
- [79] Shen, T. D., Koch, C. C., Tsui, T. Y., & Pharr, G. M. (1995). On the elastic moduli of nanocrystalline Fe, Cu, Ni, and Cu–Ni alloys prepared by mechanical milling/alloying. *Journal of Materials Research*, 10(11), 2892-2896.
- [80] Kirchlechner, C., Keckes, J., Motz, C., Grosinger, W., Kapp, M. W., Micha, J. S., ... & Dehm, G. (2011). Impact of instrumental constraints and imperfections on the dislocation structure in micron-sized Cu compression pillars. *Acta Materialia*, 59(14), 5618-5626.
- [81] Kiener, D., Motz, C., Grosinger, W., Weygand, D., & Pippan, R. (2010). Cyclic response of copper single crystal micro-beams. *Scripta Materialia*, 63(5), 500-503.
- [82] Chang, H. J. (2009). *Analysis of nano indentation size effect based on dislocation dynamics and crystal plasticity* (Doctoral dissertation, Institut National Polytechnique de Grenoble-INPG).
- [83] Gerberich, W. W., Tymiak, N. I., Grunlan, J. C., Horstemeyer, M. F., & Baskes, M. I. (2002). Interpretations of indentation size effects. *Journal of applied mechanics*, 69(4), 433-442.
- [84] El-Awady, J. A. (2015). Unravelling the physics of size-dependent dislocation-mediated plasticity. *Nature communications*, 6, 5926.
- [85] Papanikolaou, S., Cui, Y., & Ghoniem, N. (2017). Avalanches and plastic flow in crystal plasticity: an overview. *Modelling and Simulation in Materials Science and Engineering*, 26(1), 013001.

- [86] Wang, W., Zhong, Y., Lu, K., Lu, L., McDowell, D. L., & Zhu, T. (2012). Size effects and strength fluctuation in nanoscale plasticity. *Acta Materialia*, 60(8), 3302-3309.