A Quantized Crystal Plasticity Model for Nanocrystalline Metals: Connecting Atomistic Simulations and Physical Experiments

DESSERTATION

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ABSTRACT

Nanocrystalline (NC) metals, which consist of grains or crystallites with sizes less than 100 nm, have exhibited unique mechanical and physical properties, in comparison to coarse-grained (CG) counterparts. The appealing mechanical properties, for instance, include extremely high strengths, very extended elastic-plastic transitions, and unprecedented magnitudes of recoverable plastic strain. Further, footprints of intergranular stresses measured from diffraction experiments are distinct for NC metals vs. CG metals. In particular, recent in-situ synchrotron measurements reveal that residual lattice strains change rather modestly even after imposing macro plastic strains to ~1%. Remarkably, over the same regime, the corresponding residual peak widths decrease. These phenomena are in sharp contrast to CG metals, for which residual lattice strain and peak widths both increase with deformation.

In this dissertation, a quantized crystal plasticity (QCP) model is developed to explore the aforementioned unique NC features. The QCP model employs a crystallographic description of dislocation slip plasticity; in particular, single slip events across nano scale grains impart large (~1%) increments in grain-averaged plastic shear. Therefore, plasticity does not proceed in a smooth, continuous fashion but rather via strain jumps, imparting violent grain-to-grain redistribution in stress. This discrete feature is consistent with recent Molecular Dynamics (MD) simulations, which illustrate a dramatic jump in
grain-averaged shear strain when a dislocation spontaneously transverses a nano grain interior after depinning from grain boundary (GB) ledges. Finite element simulations implementing this quantized plasticity approach predict the experimental properties of enhanced strength, extended elastic-plastic strain, and recoverable plastic strain, as well as the trends in residual lattice strain and peak width mentioned, but only under certain conditions. First, the grain-to-grain distribution of critical stress for slip activation is very different from that for CG materials. In particular, no events occur below a rather large threshold stress $\sim 1/\text{grain size}$; and above this threshold, a very asymmetric distribution predominates, signifying that a relatively large number of easier-to-slip grains are balanced by a minority of harder-to-slip grains. Second, there exists a large residual stress state, which can be removed via post deformation.

The quantized crystal plasticity provides an alternate view of NC deformation, compared to hypotheses in the literatures that are centered on GB sliding or deformation of a GB phase separated from grain interior. The QCP model is capable of bridging the disparity in length and time scales between MD simulations and physical experiments, and as well establishes an insightful connection between them.
DEDICATION

THIS DOCUMENT IS DEDICATED TO MY PARENTS

Guicai Li and Hongying Shi
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Table of Contents

ABSTRACT ........................................................................................................................................... ii

DEDICATION ......................................................................................................................................... iv

ACKNOWLEDGMENTS ......................................................................................................................... v

VITA........................................................................................................................................................ vii

LIST OF FIGURES ............................................................................................................................... xiv

LIST OF TABLES .................................................................................................................................... xxiv

CHAPTER 1: INTRODUCTION ............................................................................................................... 1

1.1 Background Overview .................................................................................................................. 1

1.2 Featured Mechanical Properties of NC Metals ............................................................................. 2

1.2.1 Strength: Hall-Petch Breakdown .............................................................................................. 2

1.2.2 Limited Ductility: Strain Localization ....................................................................................... 5

1.2.3 Apparent Strain Hardening: Extended Elastic-Plastic Transition ........................................... 7

1.2.4 Plastic Recovery: Heterogeneous Plastic Deformation .............................................................. 8

1.2.5 Enhanced Strain Rate Sensitivity: Localized Dislocation Activities ....................................... 11

1.3 Underlying Deformation Mechanisms of NC Metals ................................................................. 13

1.3.1 Deformation Mechanisms indicated by MD simulations ......................................................... 13

1.3.2 Experimental Evidence of Deformation Mechanisms ............................................................ 15

1.3.2.1 Transmission Electron Microscopy .................................................................................... 15

1.3.2.2 In-situ Neutron and Synchrotron Diffraction .................................................................... 16
4.4 Prediction of the Calibrated QCP/FE Model

4.4.1 Fraction of the Grains Undergoing Forward and Reverse Slip

4.4.2 Characteristic of Grains with Backward Slip

4.4.3 Plastic Deformation and Residual Stress

4.5 Concluding Remarks

CHAPTER 5: QCP ON LATTICE STRAIN EVOLUTION OF NC Ni

5.1 Background on Lattice Strain Evolution

5.1.1 Lattice Strain Evolution of CG FCC Metals

5.1.1.1 Internal Stresses in Polycrystals

5.1.1.2 Canonical Evolution of $\bar{\varepsilon}_{<hkl>}$ in CG FCC Metals

5.1.2 Lattice Strain Evolution of NC FCC Metals

5.1.2.1 Distinct Observations of $\bar{\varepsilon}_{<hkl>}$ of NC Metals

5.1.2.2 Distinct Observations of Peak Width Evolution of NC Metals

5.1.3 Existing Models on Micro-stresses

5.1.3.1 Self-consistent Model

5.1.3.2 GB and Two-Phase Models

5.1.4 Controversial and Unsolved Issues

5.2 QCP/FE Simulations on Lattice Strains

5.2.1 In-situ X-ray Diffraction Geometry

5.2.2 QCP/FE Simulations

5.2.2.1 Diffraction-Group-Dependent $S_{x,mx}$

5.2.2.2 Diffraction-Group-Independent $\tau_c$

5.2.3 Model Calculation of Lattice Strain

5.3 QCP vs. In-situ XRD Results

5.3.1 Macro Stress-Strain Response of NC Ni
5.3.2 Mean Transverse Residual Lattice Strain, $\bar{e}_{T,hkl}$, for NC Ni.......................... 124

5.3.3 Deviation in Residual Lattice Strain, $s_{T,hkl}$, for NC Ni...................................... 126

5.3.4 QCP Simulations of XRD Peaks.............................................................................. 129

5.4 Insight into NC Deformation from QCP...................................................................... 131

5.4.1 A Physical View Based on Stress Redistribution................................................... 131

5.4.2 The Connection Between NC Properties and QCP Parameters............................ 133

5.4.3 NC vs. UFG Lattice Strain Evolution........................................................................ 139

5.4.4 Quantized Slip vs. Grain Boundary Deformation..................................................... 140

5.5 Concluding Remarks.................................................................................................. 141

CHAPTER 6: CONCLUSIONS.......................................................................................... 144

REFERENCES.................................................................................................................. 150
LIST OF FIGURES

Figure 1.1: Grain size dependent yield stress of Cu (yield stress vs. $d^{-1/2}$) [4]. Green dashed line is the extrapolation of Hall-Petch relation from experimental data for grain size larger than 1 μm. Blue solid lines are the fitting curves of experimental data in NC and UFG regime. The data sources are listed in the box below. Letters in the parenthesis after authors indicate the synthesis methods: B = bulk, EP = electro-deposition, VP = vapor deposition, and C = powder compaction; no letter means IGC (inert gas condensation).

Figure 1.2: (a) The stress-strain response of NC Au thin film with thickness 200 nm and an average grain size $d = 50$ nm undergoing two loading cycles, and an annealing process in between. (b) Time and temperature dependence of plastic strain recovery for NC Al thin film with thickness 200 nm and $d = 65$nm [41].

Figure 1.3: Deformation mechanism in NC FCC metals identified by MD simulations [60].

Figure 1.4: *In-situ* synchrotron measurement results: (a) stress-strain responses, (b) intragranular lattice strain index and (c) texture index for ED Ni of three grain sizes from nano to micro meters. Both indices are averaged over
the first three reflections \{111\}, \{200\}, and \{220\}. Strain index are calculated by \(|\bar{\epsilon}_{hkl,\text{exp}} - \bar{\epsilon}_{hkl,\text{cal}}|\) describing deviation from linear elastic behavior; texture index are given by \(|I_{hkl,\text{exp}} - I_{hkl,\text{ref}}|\) where \(I_{hkl,\text{ref}}\) is the peak intensity before loading [65].

Figure 1.5: Schematic plot for deformation mechanisms entailing activation volume of NC metals [48]. (a) Forest Dislocation localized at GBs. Multi-slip systems are activated in the vicinity of GBs resulting from stress concentration due to deformation anisotropy. (b) Dislocation nucleates at GB sources, the length of which are fraction of GB edges, and then bows into the grain interior [70].

Figure 1.6: Three types of microstructural models for NC metals: (a) two-phase model with grain size \(d = 26\) nm (see Section 1.4.2.1) [73]; (c) a dislocation slip model with partial dislocation emitting from GBs (see Section 1.4.2.2) [50]; (e) a heterogeneous GB sliding model (see Section 1.4.2.3). Experimental observations that contradict the model assumptions [84]: (b) HRTEM image of GB in ED NC Ni with \(d = 30\) nm [31]; (d) fraction of grains with deformation twins as a function of grain size [65]; and (f) evolution of FWHM of ED NC Ni with \(d = 30\) nm [38] (see Section 1.3.2.2 for details).

Figure 2.1: A full dislocation depins from GB ledge obtained by Molecular Dynamics simulations. (a) – (c) show a cut through a selective grain (G34) with the
atoms colored according to their local crystallographic order. Atoms with FCC environment are colored in grey, HCP in red; other 12 coordinate environment in green; and non-12 environment in blue. (d) shows the corresponding evolution of grain averaged shear $\gamma^*$ and resolved shear stress $\tau[10]$. ................................................................. 34

Figure 2.2: (a) A finite element model of a polycrystal with $10 \times 10 \times 10$ grains, each represented by an 8-node brick element. Uniaxial tension is applied along the $z$-direction. Each grain is colored according to $S_z^{\text{max}}$ the maximum Schmid factor among the 12 FCC slip systems in that grain. Numerical values are as shown in colorbar. (b) Microstructural information at a material point ................................................................. 41

Figure 2.3: The number of grains with a maximum Schmid factor $= S_z^{\text{max}} \pm 0.001$, out of $N_{\text{total}}$ grains. $N_{\text{total}} = 1000$ for (a) assigned to QCP model; $N_{\text{total}} = 20,000$ for (b), the set of random orientations obtained by the method described in [93]. (a) has the same mean value of $S_z^{\text{max}} (=0.45)$ as (b). (c) shows that (a) and (b) have the same probability density of maximum Schmid factor........................................................................................................... 44

Figure 2.4: {100}, {110} and {111} pole figure plots (stereographic projection) for (a) 1000-orientation QCP sample and (b) 20000-orientation sample. See caption of Fig. 2.3 for sample details ........................................................................................................... 45
Figure 2.5: Probability density distributions for $\tau_{\text{crit}}$, the critical shear stress for a slip event, for (a) a normal distribution with $\mu = 730$ MPa and $\sigma = 98$ MPa, (b) a gamma distribution with $k = 2$, $\theta \approx 165$ MPa, and $\tau_{\text{min}} = 400$ MPa, and (c) a gamma distribution with $k = 1$, $\theta \approx 330$, and $\tau_{\text{min}} = 400$ MPa. See Eqs. 2.12 and 2.13.

Figure 3.1: Shear strain and stress vs. global plastic strain, at an integration point in an interior grain, based on the elementary model (Table 3.1). The grain of interest has $S_{z}^{\text{max}} = 0.465$.

Figure 3.2: Evolution of plastic shear strain $\gamma^{(p)}$ with global plastic strain $\varepsilon_{\text{global}}^{p}$, at an integration point in an interior grain with maximum Schmid factor $S_{z}^{\text{max}} = 0.465$. Different values of $\gamma_{\text{target}}$ are considered, with $\tau_{\text{crit}} = 500$ MPa in all cases. Other input data is in Table 3.1.

Figure 3.3: Resolved stress vs. global plastic strain at an integration point in an interior grain, for $\gamma_{\text{target}}$ ranging from $1 \cdot 10^{-3}$ to $6 \cdot 10^{-3}$. Elementary model parameters (Table 3.1) are used and the grain of interest has $S_{z}^{\text{max}} = 0.465$. The vertical lines with arrows indicate the magnitude of local stress drop $|\Delta \tau^{\text{Eshelby}}|$ predicted from an Eshelby approach (Eq. 2.15), with $c = 1.2$. 

xvii
Figure 3.4: Global tensile stress-plastic strain response based on the *elementary model* (Table 3.1). \( \gamma_{\text{target}} \) ranges from \( 1 \times 10^{-3} \) to \( 6 \times 10^{-3} \) and \( \tau_{\text{crit}} = 500 \text{ MPa} \).

Figure 3.5: Global tensile stress-plastic strain response based on the *extended model*, and Type A, Type B, and Type C probability density distributions for \( \tau_{\text{crit}} \) (Fig. 2.5).

Figure 3.6: Tensile stress-strain data for electrodeposited (ED) Ni, for three different grain sizes (adapted from [110]).

Figure 3.7: The stress plastic strain responses from QCP predictions using the *extended model* data for Ni (see Tables 3.2, 3.3). Three types of \( \tau_c \) distribution are employed: Type A (symmetric normal distribution-Eq. 2.12), Type B (moderately asymmetric gamma distribution-Eq. 2.13 with \( k = 2 \)), and Type C (very asymmetric gamma distribution-Eq. 2.13 with \( k = 1 \)). Experimental \( \sigma_{\text{global}}^p, \varepsilon_{\text{global}}^p \) data (symbols) for ED Ni [110] is displayed for comparison.

Figure 3.8: The three types of critical stress \( \tau_c \) distributions used for \( d = 50 \text{ nm} \) in Fig. 3.7.

Figure 3.9: The fraction \( f_{\text{slipped}} \) of slipped grains vs. \( \varepsilon_{\text{global}}^p \) corresponding to the QCP results in Fig. 3.7. (a) Predictions for grain size \( d = 50 \text{ nm} \) and \( 300 \text{ nm} \) using Type A, B, and C distributions and (b) for \( d = 50 \text{ nm}, 150 \text{ nm}, \) and \( 300 \text{ nm} \) using the best fitting (Type C) distribution.
Figure 4.1: (a) Bright field TEM image of ED NC Ni. (b) The corresponding grain size distribution [117]................................................................. 78

Figure 4.2: Evolution of the local plastic strain $\gamma^{(\alpha)}$ and local resolved shear stress $\tau^{(\alpha)}$ on a specific slip system $\alpha$ in an arbitrary interior grain. The applied global strain rate reverses sign at 5s and 10s.............................................. 84

Figure 4.3: (a) The tensile stress-plastic strain response of an untextured polycrystal with $\tau_c = \text{asymmetric distribution}$, $\varepsilon_{\text{Ppre}} = -3\%$, 0, and 3\%, and $\tau_{\text{bias}} = 0$. Square symbols denote the experimental data for electrodeposited NC Ni with $d_{\text{mean}} = 50$ nm [110]; (b) the asymmetric $\tau_c$ distribution; (c) $\tau_{\text{ceff}}$ distribution after plastic pre-deformation $\varepsilon_{\text{Ppre}} = -3\%$ and (d) $\varepsilon_{\text{Ppre}} = 3\%$.................................................................. 87

Figure 4.4: (a) The tensile stress-plastic strain response of an untextured polycrystal for Case A: $(\tau_c, \varepsilon_{\text{Ppre}}) = (\text{asym}, 0)$ and Case S: $(\tau_c, \varepsilon_{\text{Ppre}}) = (\text{sym}, -3\%)$. $\tau_{\text{bias}} = 0$; (b) symmetric $\tau_c$ distribution; and (c) $\tau_{\text{ceff}}$ distribution after plastic pre-deformation $\varepsilon_{\text{Ppre}} = -3\%$. ............................................. 89

Figure 4.5: (a) The cyclic stress-plastic strain response of an untextured polycrystal at large strain, for Case A $(\tau_c, \varepsilon_{\text{Ppre}}) = (\text{asym}, 0)$ and Case S $(\tau_c, \varepsilon_{\text{Ppre}})$
Figure 4.6: (a) The cyclic stress-plastic strain response of an untextured polycrystal at small strain, for (a) Case A: \((\tau_c, \varepsilon_{P\text{pre}}) = (\text{asym}, 0)\); and (b) Case S: \((\tau_c, \varepsilon_{P\text{pre}}) = (\text{sym}, -3\%)\). \(\tau_{\text{bias}} = 0\). ................................................................. 92

Figure 4.7: Cyclic stress-plastic strain results for the best fitting QCP simulation (solid curve) vs. experimental data (dotted curve) for electrodeposited NC Ni with mean grain size \(d_{\text{mean}} = 30\) nm. The simulation uses \(\tau_c = \text{asym.}, \varepsilon_{P\text{pre}} = -0.4\%. \tau_{\text{bias}} = 0, 30, \) and 90 MPa depending on the imposed global plastic strain. See Table 4.3 for parameters.............................................. 93

Figure 4.8: Grain-to-grain distribution of the number of forward slip events \(q_f\), obtained from QCP simulations for electrodeposited nanocrystalline Ni with mean grain size \(d_{\text{mean}} = 30\) nm (see parameters in Table 3) at (a) \(\sigma_{\text{global}} = 800\) MPa (pt. 1’, Fig. 4.7); and (b) after unloading to \(\sigma_{\text{global}} = 0\) MPa (pt. 1, Fig. 4.7); (c) number of backward slip events \(q_{b,i}\) in grain \(i\) during unloading from pts. 1’ to 1, vs. the critical resolved shear stress \(\tau_{c,i}\) in grain \(i\); (d) \(q_{b,i}\) vs. the ratio \((q^{f\text{f}_{i,j}}/q_{f,i})_{800\text{MPa}}\) of the average number of forward slip events in grains that border grain \(i\) to the number of forward events in grain \(i\). The ratio is evaluated at \(\sigma_{\text{global}} = 800\) MPa (pt. 1’, Fig. 4.7)................................................................. 101
Figure 4.9: The distribution of residual stress $\sigma_{z,\text{res}}$ at different states in Fig. 4.7:

(a) initial state 0, prior to loading; (b) state 1, after unloading from $\sigma_{\text{global}} = 800$ MPa; and (c) state 3, after unloading from $\sigma_{\text{global}} = 1400$ MPa.

Figure 5.1: Schematic illustration of three type stresses in a randomly orientated polycrystalline assembly. Type I ($\sigma_I$) is macrostress, Type II ($\sigma_{II}$) is intergranular stress, and Type III ($\sigma_{III}$) is intra-granular stress [121].

Figure 5.2: (a) A schematic drawing of the experimental diffraction setup at PSI. (1) the incoming x-ray beam; (2) the tensile machine; (3) the CCD camera; (4) the diffracted beam, and (5) the micro-strip detector. (b) Diffraction geometry. The tensile axis is perpendicular to the viewing plane [128].

Figure 5.3: The probability density $\rho(S_{z,mx})$ for the polycrystal and various $<hkl>$ sub-groups. (See Table 5.1 for $S_{z,mx,hkl}$).

Figure 5.4: Asymmetric probability density $\rho(\tau_c)$ for the entire polycrystal and various $<hkl>$ sub-groups (See Eq. 5.2, Table 5.1).

Figure 5.5: $\sigma$-$\varepsilon_p$ response of electrodeposited (ED) NC Ni ($d_{\text{avg}} = 30$ nm) from (a) experiments [38] and (b) QCP simulations with asymmetric $\tau_c$ distribution (solid) vs. uniform $\tau_c$ (900 MPa) distribution (dashed). See Table 5.1 for other properties.
Figure 5.6: (a) Change in transverse residual lattice strain $\Delta \overline{\varepsilon}_{T<\text{hkl}>}$ vs. $\varepsilon_p$ for ED NC Ni ($d_{\text{avg}} = 30$ nm) [117] and UFG and CG Ni [65] from experiments. (b) $\Delta \overline{\varepsilon}_{T<\text{hkl}>}$ from QCP simulations of NC Ni (asymmetric $\tau_c$ distribution and $\Delta \gamma_{\text{p(max)}} = 1\%$) and UFG Ni (uniform $\tau_c = 380$ MPa and $\Delta \gamma_{\text{p}} = 0.2\%$). See Table 5.1 for other properties. ........................................................................................................... 125

Figure 5.7: (a) Experimental residual full width half maximum (FWHM) vs. $\varepsilon_p$ for ED NC Ni ($d_{\text{avg}} = 30$ nm) [117]. (b) Standard deviation in residual transverse lattice strain $\sigma_{T<\text{hkl}>}$ from QCP simulations of NC and UFG Ni. See Fig.5.6 caption for simulation parameters. ........................................................................................................... 127

Figure 5.8: QCP simulations of standard deviation in residual transverse lattice strain $\sigma_{T<\text{hkl}>}$ vs. $\varepsilon_p$ for ED NC Ni ($d_{\text{avg}} = 30$ nm) with properties in Table 5.1, except that the pre-strain $\varepsilon_{p(\text{pre})} = -0.5\%$ is achieved by transverse tension rather than axial compression. ........................................................................................................... 128

Figure 5.9: The distribution of $e_{T<220>}$ at $\varepsilon_p = 0\%$ (a), 0.5% (b), 1.0% (c), and 2.5%(d) from QCP simulations of NC Ni (asymmetric $\tau_c$ distribution and $\Delta \gamma_{\text{p(max)}} = 1\%$). ........................................................................................................... 130

Figure 5.10: QCP simulations of residual transverse lattice strain $e_{T<200>}$ vs. $\varepsilon_p$ for soft, hard, and all $<200>$ grains. Colored insets show the axial stress $\sigma_z$ in $<200>$ diffracting grains at different $\varepsilon_p$. $q = \#$ of slip events in a grain. Simulation parameters are in Table 5.1. ........................................................................................................... 132
Figure 5.11: $\sigma-\varepsilon_p$ response from QCP simulations of three supporting cases C (dashed line), C’ (dotted line), and C” (dot and dashed line). See Table 5.4 for QCP properties ................................................................. 136

Figure 5.12: (a) $\varepsilon_p$ vs. $\varepsilon_p$ from QCP simulations of CASE C (uniform $\tau_c$ distribution and $\Delta \gamma_p(\text{max}) = 1\%$). (b) $s_{T<200}$ vs. $\varepsilon_p$ from QCP simulations of CASE C and C”. See Table 5.4 for QCP parameters........................................................................................................ 139
LIST OF TABLES

Table 1. 1 Strain rates sensitivity \( m \) and activation volume \( v^* \) for CG, UFG, and NC Cu and GB-controlled processes.......................................................... 12

Table 3. 1: Elementary Model: Material and Computational Parameters .................... 53

Table 3. 2: Extended Model: Material, and Computational Parameters ...................... 60

Table 3. 3: Calibration of the Extended Model to Experimental ED NC and UFG Ni Data.................................................................................................................. 68

Table 4. 1: Notation in Chapter 4.................................................................................. 80

Table 4. 2: Multi-step Process to Calibrate QCP Simulations to Experimental Data.. 97

Table 4. 3: Best Fit of QCP Parameters to NC Ni \((d_{\text{mean}} = 30 \text{ nm})\) ....................... 97

Table 4. 4: Plastic Recovery in NC Ni \((d_{\text{mean}} = 30 \text{ nm})\) ........................................ 99

Table 5. 1: QCP Simulation Parameters for NC and UFG Ni ..................................... 118

Table 5. 2 QCP Simulation Parameters and Results for Diffraction Groups of NC Ni ......................................................................................................................... 125
Table 5.3: QCP Parameters vs NC Properties................................................................. 134

Table 5.4: QCP Simulation Parameters for Supporting CASE C, C’ and C” ............... 134
CHAPTER 1: INTRODUCTION

1.1 Background Overview

Nanocrystalline (NC) metals, by definition, are polycrystalline metals with an average grain size less than 100 nm. The remarkable properties of NC materials were first pointed out by Gleiter in 1989 [1]. A number of appealing mechanical properties and emergence of new deformation physics at nano-scale motivate tremendous research interest over the past twenty years [1-6]. The initial effort has been made on NC high strength for its potential application as a new structural material. With the development of Micro-Electoral-Mechanical Systems (MEMs), a comprehensive investigation of deformation at small scale has been carried out. In comparison to coarse-grained (CG) counterparts, NC metals exhibit several unique mechanical properties: (1) ultrahigh yield and fracture strength; (2) extended elastic-plastic transition regime; (3) large recoverable plastic deformation; (4) high strain rate sensitivity; but (5) limited elongation and toughness.

The underlying deformation mechanisms for NC metals have been investigated with considerable effort to explore the insight of the unique mechanical properties [7-17]. In stark contrast to CG metals, as grain size decreases into nano scale, intra-granular dislocation activities, such as operation of dislocation sources (e.g. Frank-Reed source) within grains, dislocation multiplication and intersection, are highly prohibited. Rather,
in NC metals, dislocations tend to nucleate at grain boundaries (GBs), traverse entire 
grains without impediment until absorbed by opposite GBs. The aforementioned 
dislocation scenario is identified after numerous Molecular Dynamics (MD) simulations 
[10]. However, given the small sample size of MD simulations and their extremely fast 
deformation rate, it is inappropriate to extrapolate MD pictures to what happens in real 
life. Therefore, mechanism-based models are required in order to bridge the gap in time 
and length scale and provide an insightful connection between atomistic simulations and 
experimental results.

In this opening chapter, featured mechanical properties of NC metals are first 
summarized in the following section. The underlying deformation mechanisms identified 
by MD simulations, and experimental observations are discussed in Section 1.3. The 
existing microstructural models are reviewed in Section 1.4 in terms of connecting 
identified deformation mechanisms and experimental observations. An open area of 
 microstructural modeling is identified in Section 1.5. This chapter is closed with the goal 
and outline of the dissertation in Section 1.6.

1.2 Featured Mechanical Properties of NC Metals

1.2.1 Strength: Hall-Petch Breakdown

The first appealing mechanical property of NC metals is the ultrahigh strength. Early 
experimental investigation have made great effort on the high strength, particularly its 
scaling with grain size reduction. The observed break-down or deviation from empirical
Hall-Petch relation in nano regime indicates a new grain size scaling, which may result from enhanced role of interface, \textit{i.e.} GBs, during deformation [18-20].

The Hall-Petch relation, which is followed nicely by CG metals, is an empirical description of grain refinement for strengthening [21, 22]. It is usually associated with stress concentration resulting from dislocation pile-up at GBs. The expression is shown as follows:

\[
\sigma_y = \sigma_0 + K_d \cdot d^{-1/2}
\]

where \(d\) is the grain size, \(\sigma_y\) is the yield strength, \(\sigma_0\) is the lattice friction stress and \(K_d\) is known as Hall-Petch constant, and indicates grain boundary strength in the pile-up theory. The square root reciprocal dependence on grain size in Eq. 1.1 predicts very strong NC metals. Figure 1.1 displays yield stress of Cu vs. \(d^{1/2}\) from micro to nano regime obtained from different sources with various synthesis methods. Two features can be noted. First, a reduction of grain boundary strength (\(K_d\) in Eq 1.1) in nano regime (increasing part of blue data-fitted line) in comparison with that in micro regime (green dashed line extrapolated from microcrystalline Cu). Second, a breakdown of Hall-Petch relation appears when \(d\) is less than 25 nm \((d^{1/2} = 0.2)\). The trend after \(d^{1/2} = 0.2\) becomes indeterminate[23]. For instance, some results plateau; while others even show a negative slope. The negative slope, indicating yield stress decreasing with the grain refinement, is termed as inverse Hall-Petch effect. It was first reported by Chokshi based on his compression test on NC Cu made by inert gas condensation (IGC) (blue filled circle in Fig. 1.1) [24]. He attributed this grain refinement softening to a switch of deformation mechanism from dislocation-mediated plasticity to Coble creep. However
these early experimental samples suffer from low quality, particularly in tens of nano regime. Further not much data is available for very small grain sizes (< 20nm), and corresponding microstructure often exhibits a very broad grain size distribution. Therefore, the existence of such an inverse effect is questioned, but a deviation from the conventional Hall-Petch relation seems to be present.

Figure 1.1: Grain size dependent yield stress of Cu (yield stress vs. $d^{1/2}$) [4]. Green dashed line is the extrapolation of Hall-Petch relation from experimental data for grain size larger than 1 µm. Blue solid lines are the fitting curves of experimental data in NC and UFG regime. The data sources are listed in the box below. Letters in the parenthesis after authors indicate the synthesis methods: B = bulk, EP = electro-deposition, VP = vapor deposition, and C = powder compaction; no letter means IGC (inert gas condensation).
The Hall-Petch breakdown in experimental observations has motivated simulations and modeling to reveal the underlying deformation mechanisms. Early MD simulations conducted by Van Swygenhoven [25, 26] and Schiotz [7, 8] on pure NC Cu indicate a critical grain size of 8 nm and 10-15 nm respectively, below which the plastic deformation is mainly accommodated by GB sliding. In addition, continuum-based models are proposed to explore the existence of a critical grain size. Conrad et al. developed an isostrain model (see Section 1.4.1) explaining softening behavior at grain sizes smaller than a material dependent critical value [27]. Argon and Yip proposed an isostress model by formulating a strongest size [28]. It indicates a grain size region between 10 and 20 nm, where a transformation takes place from intra-granular dislocation slip to GB mediated processes. It is noted that most of effort has been made to involve GB deformation as a significant contributor for explaining Hall-Petch breakdown. However, high resolution TEM (HRTEM) cannot find extended GB regions or randomly arranged atoms for grain sizes around 20 nm [3]. Therefore a transition of dislocation strengthening activities, rather than pile-up, should be investigated. In Chapter 3, a size dependence of dislocation propagation strength is proposed.

1.2.2 Limited Ductility: Strain Localization

In contrast to the ultrahigh yield strength, the ductility of NC metals after yielding is usually tremendously reduced (<10% tensile strain) in comparison with their CG counterparts [29, 30]. Lack of efficient strain hardening abilities is believed to be the key reason for the limited ductility. In NC metals, the grain size is too small to involve intra-
granular dislocation activities, e.g. dislocation multiplication and interaction. Consequently the conventional strain hardening capability, coming from the dislocation storage, is nearly prohibited. The localized deformation within the grains suffers from incompatibility at GBs, and results in very high stress concentration around GBs, especially at triple junctions. These highly stressed spots can generate nano-void and micro-crack, and finally cause failure. The typical fracture morphology of NC metals mixes ductile dimples and shear region [3, 31]. The tendency of localized shear limits NC ductility by triggering localized deformation mode, e.g. formation of shear band or necking in the early stage of deformation. In addition to the intrinsic limitations, low quality of sample counts for another factor.

In order to be a competitive candidate for new structural materials, in addition to ultrahigh strength, good ductility is also desired for NC metals and alloys. Engineering special material structures have shed light on achieving this goal. Firstly, a ‘bimodal’ grain size structure [32-34], which combines the high strength from nano grains and high ductility from micro grains. In micro regime, the reduction of grain size usually results in an enhancement in ductility, considering that a decrease of dislocation free path produces a large strain-hardening rate. As a result, substantial deformation strain can be provided by dislocation activities in the micro grains. On the other hands, nano grains remains deforming elastically, and act as load carrier. For such microstructure, a desirable spatial distribution of grain sizes is essential. It would be detrimental to strength if relative soft micro grains form a cluster, and become a load carrier, e.g. an isostress pattern. Another interesting engineering structure is nano-twin, particularly for Cu, which has lower stacking fault energy [29, 35-37]. It is proposed that the multi length scales associated
with twin structure benefiting the ductility. Along the twin plane, dislocation motion is relatively easy; on the other hands, perpendicular to twin plane, twin spacing is on the order of nano meter, which provides a large stress barrier for dislocation either pile up at or transmit through twin boundaries.

1.2.3 Apparent Strain Hardening: Extended Elastic-Plastic Transition

NC metals usually feature with an extended elastic-plastic transition regime, $\varepsilon_{p\text{(tran)}}$, which exhibits as apparent strain hardening in stress-strain response. In CG metals, the macroscopic yield stress is usually defined by the stress at which produces 0.2% of plastic strain. At this strain, it is assumed that majority (>90%) of grains in polycrystal are undergoing plastic deformation. In contrast, for NC metal 0.2% criterion is usually underestimated its macroscopic yield stress [38]. Extended $\varepsilon_{p\text{(tran)}}$, a strain regime between initial deviation from stress-strain linearity to macroscopic yielding, is widely observed in NC metals. First, Saada has conducted a scaling assessment of elastic-plastic transition regime in NC metals [39, 40]. Assuming a dislocation sweep through a gain with size $d$, this will produce a strain increment $b/d$ at grain level. Macroscopically, plastic strain

$$\varepsilon_p \approx \frac{n(\varepsilon_p) b}{N d} \quad (1.2)$$

where $N$ represents the total number of grains in polycrystals, and $n(\varepsilon_p)$ is the number of grains, which has undergone one dislocation slip event after macroscopic plastic
deformation $\varepsilon_p$. An assumption of macroscopic yielding is $\frac{n(\varepsilon_p)}{N} \geq 1$. Therefore, in NC sample with $d < 100b$, the extend of elastic-plastic transition may reach 1%. $\varepsilon_{p(\text{tran})}$ could be even larger if multiple events occur in one grain. On the other hands, for CG metals, $d > 1 \, \mu\text{m} (4000b)$, required for $\frac{n(\varepsilon_p)}{N} \geq 1$, $\varepsilon_p < 0.025\%$. Provided that dislocation free path is not as large as $d$ in CG metals, 0.2% is still a good estimation. Another possible explanations for extended $\varepsilon_{p(\text{tran})}$ will be highly inhomogeneous plastic deformation. Upon loading, plastic slips first take place at easy/soft grains, which results in deviation from elastic linearity, i.e. micro yielding. A gradual percolation of plastic deformation from easy/first yield grains to hard grains will require an increasing in stress, which gives an apparent strain hardening, till majority of the grains yield. Saif and his colleagues support this idea with experimental evidence and interesting experimental observation: recoverable plastic strain (discussed in Section 1.2.4)[41]. Both arguments seem reasonable; in fact, both of them are important. The large strain increment at grain level in NC metals, cannot produce $\varepsilon_{p(\text{tran})}$ if all the grains yield simultaneously. But the details of this extended regime still remain unclear. A quantitative investigation will be conducted in Chapter 3 and 4.

1.2.4 Plastic Recovery: Heterogeneous Plastic Deformation

NC metals challenge the conventional thinking of plastic deformation by an observation of recovering a significant fraction (50% to 100%) of plastic strain after unloading [41]. This recovery shows time and temperature dependence. Figure 1.2a displays the stress-
strain response of NC Au thin film with thickness 200 nm and an average grain size \( d = 50 \) nm undergoing two loading cycles. Four characteristic strains are identified. During the first unloading, an extended elastic plastic transition, I: \( \varepsilon_{\text{p(tran)}} \), is exhibited. 0.2% conventional definition is only half of \( \varepsilon_{\text{p(tran)}} \). In fact, their follow-up in-situ TEM work on a similar sample (NC Au with \( d = 70 \) nm) shows that even till \( \varepsilon_p = 0.6\% \) majority of small hard grains still remain deforming elastically. Upon unloading, 20% plastic deformation, \( \text{i.e. II: } \varepsilon_{\text{p(inst)}} = 0.15\% \) out of \( \varepsilon_p = 0.8\% \) recovers instantaneously. In addition, further annealing at 210C results in additional 35% of plastic recovery (III: \( \varepsilon_{\text{p(ninst)}} = 0.15\% \)). While reloading, the stress-strain curve joins the initial loading trace latter than the unloaded point, and produce IV: a reload gap \( \varepsilon_{\text{p(tran)}} \) as a result of recovery occurred during the cycle. The aforementioned stress-strain characteristics have not only been observed in Au but also Al NC thin films [42-44] and bulk NC Ni [38]. The deformation process is investigated by in-situ TEM study. Plastic recovery starts about the middle stage of unloading. Pervasive dislocation actives take place in large grains surrounding by small elastically deformed grains. Therefore, these reverse slips are assumed to be driven by large internal stresses built up among small/hard and large/soft grains. Further investigations suggest that the source of heterogeneous stresses is not only from grain size distribution. For instance, \( \varepsilon_{\text{p(inst)}} \) is dramatically reduced in a textured sample with almost the same grain size distribution. Though an iso-strain model is proposed to address the influence of texture. The simplistic assumption on the difference in Schmid factor between textured and non-textured samples overestimates the effect of grain orientations. A more physical assessment of heterogeneous deformation is still required.
Another aspect of the unique NC recovery process is its time and temperature dependence. **Figure 1.2b** shows the evolution of plastic recovery ($\varepsilon_{pr}/\varepsilon_p$) as a function of time and temperature for NC Al thin film with thickness 200nm and $d=65\text{nm}$ unloaded from $\varepsilon_p=0.55\%$. The results shows that ~40% recovery occurs very quickly at room temperature (RT) within about 400 s; whereas the remaining RT recovery (~40%) are much slower (~ $10^5$ s). An increase in temperature accelerates further recovery that saturate very soon. These observations suggest that the recovery process may involve a distribution of energy barriers. A conceptual model of thermally activated process: dislocation pinning-depinning from GBs is proposed. This is a process the activation volume of which agrees well with experimental measured strain rate sensitivity (see Section 1.2.5). In addition, a heterogeneous GB sliding and diffusion model is proposed to explain the rate dependent recovery (for details see Section 1.4.2.3). But the activation volumes of GB processes are usually smaller than experimental measurement for NC metals. In a word, this unique recovery phenomenon underscores the inherent heterogeneous nature in microstructure of NC metals.
Figure 1.2: (a) The stress-strain response of NC Au thin film with thickness 200 nm and an average grain size $d = 50$ nm undergoing two loading cycles, and an annealing process in between. (b) Time and temperature dependence of plastic strain recovery for NC Al thin film with thickness 200 nm and $d = 65$ nm [41].

1.2.5 Enhanced Strain Rate Sensitivity: Localized Dislocation Activities

NC metals have been identified to have higher strain rate sensitivity than CG materials. The study on strain rate sensitivity is appealing, since it is one of the key engineering
parameters that characterize deformation kinetics. The strain rate sensitivity, $m$, is typically defined as

$$m = \left. \frac{\partial \ln \sigma}{\partial \ln \dot{\varepsilon}} \right|_{\sigma, \dot{\varepsilon}}$$

(1.3)

where $\sigma$ is the flow stress and $\dot{\varepsilon}$ is the corresponding strain rate. Schwaiger [45], Dalla Torre [46, 47] and Wang [48] have systematically studied the strain rate effect on NC Ni by means of indentation and tensile tests (strain jump and stress relaxation). They report strain rate sensitivity of NC Ni is one order of magnitude higher than CG Ni, i.e. 0.01-0.03 for NC vs. 0.001-0.004 for CG counterparts. Table 1.1 summaries the value of strain rate sensitivity $m$ and activation volume $v^*$ of Cu over a broad regime of grain sizes. For comparison, also listed are theoretical values of GB sliding and Coble creep.

<table>
<thead>
<tr>
<th></th>
<th>CG Cu [49, 50] (&gt;$1 \mu m$)</th>
<th>UFG Cu [50-52] ($10^2 \sim 10^3 \text{ nm}$)</th>
<th>NC Cu [50-52] (10nm $\sim 10^2 \text{ nm}$)</th>
<th>GB sliding [53]</th>
<th>Coble creep [54]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m$</td>
<td>$0 \sim 0.004$</td>
<td>$0.014 \sim 0.018$</td>
<td>$0.024 \sim 0.06$</td>
<td>0.5</td>
<td>1</td>
</tr>
<tr>
<td>$v^*(b^3)$</td>
<td>$10^3 \sim 10^4$</td>
<td>$50 \sim 10^2$</td>
<td>$10 \sim 20$</td>
<td>1</td>
<td>$0.1 \sim 1$</td>
</tr>
</tbody>
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As grain size decreases from micro scale to nano scale, $m$ increases and $v^*$ decreases from $10^3b^3$ to $10b^3$ ($b$ is the magnitude of full Burgers vector). The reduced $v^*$ indicates more localized dislocation activity in NC metals (see Section 1.3.2.3 for details). On the
other hands, in comparison with GB-mediated plastic process \((v^* \sim 1b^3)\), \(v^*\) for NC Cu is still larger, which indicates that GB sliding or Coble creep is still not the dominant deformation mechanism for NC metals with grain size as small as 10 nm [6]. In summary, the distinct mechanical behaviors of NC metals from their CG counterparts indicate size dependence nature of dislocation-mediated deformation mechanisms.

1.3 Underlying Deformation Mechanisms of NC Metals

1.3.1 Deformation Mechanisms indicated by MD simulations

Deformation mechanisms of NC have been investigated intensively by MD simulations, and these atomic simulations provide the most widely accepted NC deformation pictures [10-17]. The identified deformation modes can be briefly categorized into two groups: (1) GB mediated and (2) dislocation mediated. Figure 1.3 is a schematic plot summarizing those observed deformation modes by MD simulations. First, GB activities become more popular. For instance, GB sliding (denoted as green arrows in Grain 4) is usually accompanied by atomic shuffling and stress-assistant free volume migration (in Grain 2). In contrary to GB sliding where local shear stress results in shear deformation along GB plane, a shear stress can also lead to normal motion of GB (in Grain 1) [55]. This deformation mode has been proposed to contribute to stress assistant grain growth during NC deformation at room temperature [56]. In addition, the triple junction can move and the complex interplay of GB motion can result in collective GB sliding (blue arrows in Grain 3) and grain rotation (Grain 4) in order to satisfy compatibility during deformation. In another aspect, dislocation activities still remain prevailing in nano regime. During a
lifetime, dislocations nucleate at GBs, propagate through entire grain interior, and in the end are absorbed by GBs (see Grain A-D). Apparently, the dislocation processes are dominated by its interaction with GBs rather than with other dislocations like in CG metals. Dislocation nucleation at GBs was first proposed by JCM Li in 1961[57]. MD simulations show that in FCC grains dislocation is nucleated through partials (Grain A) at GB stress concentrators. Stacking fault is formed between leading and trailing partials (Grain B & C). Also trailing partial is not necessary to be observed if simulated material has low stacking fault energy and simulated time is too short. This gives a chance to form twin fault where another leading partial nucleates and propagates in an adjacent plane (Grain a-c). Deformation twin is observed in NC Ni when deformation occurs at high strain rate and low temperature [58, 59]. Another interesting observation from MD simulations is that nucleated dislocation embryos vibrate around the nucleation sites, and are waiting for propagation. Consequently propagation becomes the most significant rate control step. In addition, during propagation, dislocation can be pinned by stress concentrator at GB, particularly GB ledge. The depinning process strongly depends on temperature and time, and tends to be a thermally activated process. Finally dislocation is absorbed by surrounding GBs, and leaves no dislocation network within grain interior.
The inherent limitation of MD simulations, e.g. unrealistic strain rate $>10^7$, and limited sample size, makes it remain unclear which of those identified mechanisms occur in realistic and which dominates. Therefore further direct experimental evidences are required.

### 1.3.2 Experimental Evidence of Deformation Mechanisms

#### 1.3.2.1 Transmission Electron Microscopy

Dislocation mediated plasticity is proved to continue playing dominate role by TEM observations. Ex situ TEM performed on deformed NC Ni ($d \sim 30$ nm) shows isolated dislocations and sporadic dislocation network. The dislocation density left, however, is not enough to accommodate the imposed large plastic strain [31]. This lack of

Figure 1. 3: Deformation mechanism in NC FCC metals identified by MD simulations [60].
dislocation debris on one hand could suggest dislocation may not be the key deformation carrier; on the other hands it supports the aforementioned dislocation lifetime (nucleation-propagation-absorption) identified by MD simulations. Therefore in-situ TEM evidence is necessary to address dislocation dynamics in nano grains. Shan performed combination in situ test of high resolution TEM and deformation on NC Ni [61]. During deformation, FCC full dislocations are frequently trapped in the vicinity of GBs in the grain even as small as 5nm. The further relaxation process results in dislocation recombination and annihilation. Though in-situ TEM has brought substantial details on deformation, the ultrathin TEM foils contain only a few grains on top of each other. The inevitable free surface may enhance dislocation activities, diffusion processes, and GB relaxation. As a result, the pervasive deformation processes observed in TEM may not occur that frequently in bulk sample.

1.3.2.2 In-situ Neutron and Synchrotron Diffraction

In-situ neutron and synchrotron diffraction are also employed to study deformation behaviors of NC metals. The strong penetration of neutron and high-energy synchrotron makes them capable of investigating deformation mechanisms at grain size level in bulk specimens. The first distinct observation is that the peak broadening induced during deformation of NC Ni ($d = 30\text{nm}$) is reversible after unloading at room temperature [62, 63]. This removal of heterogeneous lattice strain suggests that little dislocation network has been built up during this cycle, and thus aforementioned dislocation lifetime indicated by MD simulations can provide be a proper explanation. Furthermore, a comparison among peak width at several sequent unloaded states shows even a reduced
broadening (see Fig. 1.6f)[38]. This indicates that deformation can even erase the initial
locked heterogeneous structure. Recent effort involves addressing the distinct evolution
of lattice strain, $\bar{e}_{hkl}$, and texture development of NC metals vs the CG counterparts.

**Figure 1.4** shows the *in-situ* synchrotron measurement results: (a) stress-strain responses,
(b) intragranular lattice strain index and (c) texture index for ED Ni of three average
grain sizes from nano to micro meters. Completely different $\bar{e}_{hkl}$ and texture behaviors
can be identified between NC Ni ($d = 20$ nm) and UFG Ni ($d=100$nm and $1000$nm).

First, significant $\bar{e}_{hkl}$ (denoted by strain index in Fig. 1.4b) is built up in UFG Ni during
deformation; whereas little $\bar{e}_{hkl}$ develops in NC Ni up to 3% macroscopic strain. Second,
texture development (illustrated by texture index in Fig. 1.4c) linearly grows with
macroscopic strain in UFG Ni vs. stay constant in NC Ni. These extended elastic
behaviors of NC Ni after micro yielding promotes the hypothesis of GB mediated
deformation [64, 65]. The lack of solid evidence leaves these phenomena an interesting
unsolved issue to explore. Chapter 5 investigates this issue in details and provides a new
view on lattice strain evolution of NC metals.
Figure 1. 4: *In-situ* synchrotron measurement results: (a) stress-strain responses, (b) intragranular lattice strain index and (c) texture index for ED Ni of three grain sizes from nano to micro meters. Both indices are averaged over the first three reflections \{111\}, \{200\}, and \{220\}. Strain index are calculated by $|\varepsilon_{hkH,exp} - \varepsilon_{hkH,cal}|$ describing deviation from linear elastic behavior; texture index are given by $|I_{hkH,exp} - I_{hkH,ref}|$, where $I_{hkH,ref}$ is the peak intensity before loading [65].
1.3.2.3 Activation Volume in Rate-controlling Mechanisms

Activation volume $v^*$, obtained from macroscopic mechanical testing, is a kinetic signature of deformation mechanisms. Mentioned in Section 1.2.5, for NC metals $v^*$ is about $10 \sim 20b^3$, falling between $v^*$ for dislocation actives in CG metals and GB plasticity. Three scenarios, which may entail the right $v^*$, have been summarized by Wang [48]. First, mobile dislocations cut through the GB dislocations, which reside in the vicinity of GBs. This scenario is analogous to dislocation cutting though the forest dislocations in conventional FCC metals, but now the forest dislocations, are highly localized at GBs. The schematic picture is displayed in Figure 1.5a. Due to the decreasing of dislocation spacing, the activation volume is reduced. This process may occur in the UFG metals synthesized by severe plastic deformation, where a large amount of non-equilibrium GBs exists and provides excess GB dislocations [66]. However, in most NC metals produced by other methods (i.e. inert gas condensation, electro-deposition), high dislocation density layer has not been observed[3, 67-69]. In this context, the second model, dislocation emission from GBs, is proposed. And in this model, high dislocation density at GBs is not necessary. The activation length of this process, estimated as a fraction of grain edge (the edge length is on the order of $20b$ for a 15 nm Ni octagon grain in Fig 1.5b), results in a small activation volume ($\sim 20b^3$). Third, the propagating dislocations de-pin from GB obstacles, e.g. GB ledges, which is identified in MD simulations (Grain C in Fig. 1.3)[17]. When a dislocation is traveling through the grain interior, its two ends may move along the GBs. The GB ledges, can act as pinning points to hinder the motion of the dislocation, and the de-pinining process is thermally activated. This scenario provides an explanation of the observed storage of
dislocations inside NC grains at liquid nitrogen temperature but not at room temperature [58, 59]. The aforementioned deformation mechanisms all involve enhanced interactions between dislocations and GBs. Since as grain size decreases, it is more difficult for dislocation source within grains to operate; rather large fraction of GBs provide a high density of non-equilibrium GBs, GB dislocation sources and pinning sites.

Figure 1.5: Schematic plot for deformation mechanisms entailing activation volume of NC metals [48]. (a) Forest Dislocation localized at GBs. Multi-slip systems are activated in the vicinity of GBs resulting from stress concentration due to deformation anisotropy. (b) Dislocation nucleates at GB sources, the length of which are fraction of GB edges, and then bows into the grain interior [70]
1.4 Existing Models on NC Metals

1.4.1 Phenomenological Models

Models do not have crystallographic description of plastic deformation in grains are categorized as phenomenological model. Early phenomenological models were mainly aimed to explain breakdown of Hall-Petch effect in NC regime. Pande and Armstrong [19, 71] proposed a dislocation pile-up breakdown model. A simple argument of the pile-up break down is that there exists a critical grain size [18] (in the nano regime), wherein the number of dislocation at pile-up reduced to one. Consequently, no stress multiplication effect exists, and the H-P relation on grain refinement strengthening, in turn, does not hold to NC regime.

Later a set of rate-control formalization was proposed by Conrad and Narayan to addresses another feature of NC metals, strain rate sensitivity, in addition to Hall-Petch breakdown[27, 49]. In this formalization, deformation regime was divided into three according to the existing flow stress of Cu. Larger than 1µm is the conventional dislocation interaction controlled regime; while less than 10 nm, GB sliding is considered to dominate. In between, a mixture of dislocation interaction and GB sliding is attributed to rate dependence stress response. The grain size dependence in this crossover region comes from an imposed $d^{1/2}$ on the rate term. Conrad’s rate dependence formulation, particularly of grain boundary sliding, has been implemented to microstructural models for GB response.
1.4.2 Microstructural Models

Models described in this section employ a crystallographic description of plastic deformation. According to the unique deformation constitutive laws for NC metals, the models are categorized into three types: two phase, dislocation slip, and grain boundary.

1.4.2.1 Two-Phase Models

Two-phase models are promoted by the assumption that the volume faction of GBs increases as grain size decreases. Two competing deformation processes, dislocation and grain boundary mediated, are mixed with a size dependent weighted function.

In 1982, Meyers and Ashworth developed a core and mantle models (MA model) to study the size effect on yield stress [72], and this model was extended to nano regime by Benson and Fu in 2001 [73]. The schematic depiction of the MA model is shown in Fig 1.5a. The basic assumption is that GBs behave as work-hardened layers in the sense that elastic incompatibility results in stress concentration at GB wherein multiple slip systems are activated. By assuming that GB thickness $t$ is proportional to $d^{1/2}$, a new $d$ dependent term is added to the conventional H-P equation, which gives rise to a deviation from H-P relation in the small grain size. Fu and Meyers [74, 75] applied this model with the crystallographic description of plasticity on both grain interior and GBs. During deformation, the strain hardened grain boundary phase becomes stress concentrator, while plastic strain localizes in grain interior. This promotes the formation of shear localization that has been observed in experiments. But an unrealistic stress-strain response is predicted from this model. A strong work hardening rate continues up to 16%
true strain, while experimentally hardening exists to 4% at most, and in fact less than 2% for majority.

Wei and Anand [76, 77] have proposed a model to study the inelastic deformation and failure behavior of FCC NC metal. Cohesive elements along the grain boundaries are introduced to model GB evolution. These GB cohesive elements have evolution equations for both the normal and tangential tractions, and include both elastic and plastic components; whereas single crystal plasticity is applied to grain interior. The scaling with various grain sizes comes from (1) volume fraction of GB phase and grain interior; and (2) enhanced critical shear stress within grains. The simulation results of NC Ni suggest a transition of deformation mechanism from grain-interior to GB shearing as the grain size decreases from 50 nm to 10nm. In addition, the intra-granular failure due to GB shearing and resultant cavitation gives rise to low ductility of NC Ni. However, an increasing in ductility is predicted when grain size less than 60 nm, as a result of increasing volumetric fraction of GB phase. This prediction contradicts to experimental observation.

1.4.2.2 Dislocation Slip Models

Asaro et. al. adopted the idea that dislocation nucleates at grain boundary of NC metals and calculate the corresponding athermal critical nucleation stress for perfect as well as partial lattice dislocations [50, 78]. The process of creating lattice dislocations mainly involves depositing dislocation segments on surrounding GBs from existing GB dislocations. Consequently, the calculated critical stress has $1/d$ dependence on grain size,
and thus introduces a length scale into model. From the calculation, when grain size smaller than 30 nm, partial dislocations with stacking fault are more energetically favorable than full lattice dislocation for pure Ni. However, the experimental evidence is still sporadic. Stacking fault has been seldom observed in NC Ni even when gain size smaller than 30 nm [58, 59]. Deformation twin has only observed in NC Ni when deformation took place under complex loading state (indentation, rolling) at low temperature and high strain rate. Latter Zhu [79, 80] extended Asaro’s work by introducing a log-normal grain size distribution. The model underscores an important feature that the variance of grain size strongly affects the operating deformation modes. For NC Ni with a mean grain size as small as 20 nm but a large variation (~100), full dislocation emission, which is considered to occur in larger grains, dominates. On the other hands, NC Ni with a similar mean grain size but relatively small deviation involves more fraction of small grain mechanism, i.e. partial dislocation emission. The importance of microstructure heterogeneity (e.g. grain size) in nano regime has been illustrated to be essential for quite a few unique NC deformation responses, in particular plastic recovery (see Section 1.2.4). Unfortunately, the iso-strain assumption from Zhu’s work, excludes the spatial influence of deformation heterogeneity.

1.4.2.3 GB Sliding Models

The GB-mediated deformation mechanisms, e.g. GB sliding and GB diffusion, were proposed to dominate in NC regime with a intuitive impression that increasing volumetric fraction of GBs with grain size decreasing [24, 81]. However, these GB-controlled
mechanisms overestimated creep rate [82, 83] and strain rate sensitivity in comparison to experimentally measured values (see Section 1.2.5). It is widely accepted that GB-mediated deformation does not dominate even when grain size as small as 20 nm at least for bulk NC metals. Recently Wei YJ and Gao HJ proposes a GB mediated model with heterogeneous GB diffusion and sliding. This model is able to explain a tremendous plastic recovery observed in NC Al thin films [84, 85]. In the model, grain interior deforms only by anisotropic elasticity, and each grain boundary is assigned with one of two candidates of diffusivity and sliding viscosity (see Fig. 1.6c: GBs marked with square have diffusivity coefficient 1000 slower than other GBs). This heterogeneous feature is essential to obtain desirable amount of internal stress, which acts as a driving force of plastic recovery. However, the physical source of heterogeneity in GB diffusivity is unclear. The authors exclude the competing dislocation mechanism by arguing that experimentally only few grains have been observed containing dislocations, and de-pinning one dislocation in such a small group of grains can not produce the desirable amount of recovery. But what if several reverse slips occur in those plastically deformed grains? Another appealing outcome from the model is the rate dependent recovery. However, the initial stress-free sample cannot explain the high residual stress state existing in pre-test NC samples, indicating by a large FWHM from X-ray measurement (see Fig.1.6f). In Chapter 4, it is shown that desirable amount of plastic recovery can also be achieved by dislocation model with heterogeneous critical slip strength.

Figure 1.6 summaries the three types of microstructural models: (a) two-phase; (c) dislocation slip; and (e) GB sliding. Fig. 1.6 (b), (d), and (f) show the experimental
observations that do not consist with those model assumptions. Generally, a length scale is required to incorporate into the models for capturing the size effect as a result of grain size decreasing. For two-phase models, the length scale comes from a ratio, either one-dimensional ratio (Fu’s model) or three-dimensional ratio (Wei and Anand’s model), between GB and grain phases. Figure 1.6a shows the polycrystal aggregate employed in Fu’s model when grain size is 26 nm. It is noted that till this grain size, grain interior is even smaller than GB, which is in contradictory to the sharp and clear GB in TEM image in Fig. 1.6b. Further to achieve desirable size dependent deformation behaviors, different constitutive relations are applied to two phases. For instance, Fu has applied single crystal plasticity to both grain interior and GBs; whereas Wei and Anand treat GB as a soft amorphous layer. Enhanced and dominant GB phase deformation is predicted as grain size decreasing in these models. But majority of the experimental results suggest that above 10nm, NC deformation is still dominated by dislocation slips. In another aspect, Asaro focuses on the size effect from dislocation activity itself by means of activating partial dislocations below a critical grain size, e.g. 20nm for Ni (see Fig. 1.6c). This assumption would promote pervasive stacking fault, and deformation twin in NC metals. But the observation of deformation twin in NC metals strongly depends on loading condition, loading rate as well as material intrinsic properties, e.g. stacking fault energy and unstable stacking fault energy. In fact, recent observation shows that the density of deformation twins decrease with grain size, and UFG Ni tends to have a large twin density (see Fig. 1.6d).
Figure 1.6: Three types of microstructural models for NC metals: (a) two-phase model with grain size $d = 26\text{nm}$ (see Section 1.4.2.1) [73]; (c) a dislocation slip model with partial dislocation emitting from GBs (see Section 1.4.2.2) [50]; (e) a heterogeneous GB sliding model (see Section 1.4.2.3). Experimental observations that contradict the model assumptions [84]: (b) HRTEM image of GB in ED NC Ni with $d = 30\text{ nm}$ [31]; (d) fraction of grains with deformation twins as a function of grain size [65]; and (f) evolution of FWHM of ED NC Ni with $d = 30\text{nm}$ [38] (see Section 1.3.2.2 for details).
1.5 Open Area of Microstructural Modeling

Though a great many of deformation details of NC metals, particularly FCC metals, have been obtained by atomistic simulations, a connection between those identified details with unique macroscopic mechanical responses still remains fairly weak. To bridge the gap, mechanism-based models are required. One of the challenges is to appropriately involve size dependent deformation mechanism. This aim has been widely achieved by a size-weighted combination of CG dislocation activities with CG high temperature GB mechanisms. An enhancement of GB activities results in small size deformation responses. However, pervasive experimental evidences directly show or indirectly infers that dislocation behavior remain dominant in NC metals, at least till the grain size \( d = 10 \text{nm} \). Therefore, a length scale from dislocation slip itself should be paid more attentions.

One of such length scale can come from the discrete aspect of dislocation plasticity. As mentioned in Section 1.2.3, when grain size is as small as nanometer, a shear increment induced by one dislocation sweeping across a grain (~\( b/d \)) increases to the order of percent, which is compatible to macroscopic deformation. As known, very few studies have been done to investigate systematically the influence of this discrete plasticity enhanced by small grain size. Furthermore, deformation heterogeneity due to non-uniform microstructures, e.g. grain size, grain orientation, affects significantly the macro mechanical responses. Majority of the existing models have addressed the inhomogeneity coming from grain interior and GBs, but ignore the heterogeneity from grain to grain. Therefore a micro-structural model, which incorporates discrete plasticity and is capable of investigating grain-to-grain variation in deformation, would be valuable.
1.6 Dissertation Objectives and Outline

The objective of this dissertation is to (1) develop a mechanistic model, which incorporates the enhanced discrete feature of dislocation plasticity in nano regime; and (2) employ the model to explore the unique mechanical properties of NC metals. Therefore, the meso-scale model is able to bridge the disparity in length and time scales between MD simulations and physical experiments, and further to provide insightful connection between deformation mechanisms and experimental observations.

In Chapter 2, a quantized crystal plasticity (QCP) constitutive relation is developed within a large strain formulation scheme. This quantized/discrete feature is motivated, in part, by MD simulations showing that discrete changes occur in stress and strain at grain-averaged level during a dislocation slip in nano size grains. The developed QCP relation is implemented in commercial finite element code ABAQUS by a user subroutine UMAT. A rate independent crystal plasticity flow law is employed to achieve a quasi-instantaneous shear increment by dislocation slip, and an implicit time integration scheme is used. There are two key material parameters, which control QCP relations: quantized shear increment $\gamma_{\text{target}}$ for one slip event, and critical slip stress $\tau_c$. The relation between them is established through a dislocation prorogation condition.

In Chapter 3, an initial assessment of QCP relation is performed through an elementary model, which entails a spatially uniform distribution of critical slip stress $\tau_c$ and one slip increment $\gamma_{\text{target}}$ ($\sim b/d$). Furthermore, an extended model with a grain-to-grain variation in $\tau_c$ is investigated on the purpose of reproducing the monotonic stress-strain
characteristics of NC and UFG Ni. An asymmetric distribution of $\tau_c$ is identified and a physical rationale is provided based on dislocation propagation requirement in small grains. In addition this propagation requirement can be viewed as an alternate source of size-dependent strengthening.

In Chapter 4, a reverse slip relation is implemented into QCP model. Consequently two new key parameters are included: backward slip stress $\tau_{c,b}$, and the amount of residual plastic strain $\varepsilon_{p(pre)}$. The residual stress state significantly affects monotonic stress-strain response, and makes the solutions from Chapter 3 not unique. But a further investigation of cyclic responses of NC Ni validates the heterogeneous nature of slips induced by an asymmetric $\tau_c$ distribution. The QCP model capture the large reversible deformation observed in experiments by reversing slips in relatively soft grains with smaller $\tau_c$, which is driven by elastically deformed hard grains. An evolution of backward slip strength reflects, in part, the nature of grain boundaries that evolve with deformation.

In Chapter 5, QCP approach is further explored by explaining micro-strain features in NC metals. Combining with NC macroscopic stress-strain responses, seven NC properties are identified. A connection is established between these seven properties and three key QCP parameters: $\gamma_{target}$, an asymmetric $\tau_c$ distribution and an initial residual stress state $\varepsilon_{p(pre)}$. Finally, a new deformation insight of NC metals is provided from a quantized crystal plasticity point of view.

Dissertation is concluded in Chapter 6. The contribution to literature is discussed in terms of model development and applications. The unique discrete feature of the model
shed light to its implementations to other systems. The shortcomings of the model motivate future work required to understand more details of small-scale plasticity.
CHAPTER 2: QUANTIZED CRYSTAL PLASTICITY MODEL

2.1 Motivation From Molecular Dynamics Simulations

MD simulations have provided considerable insight into deformation mechanisms in FCC NC metals. A grain averaged analysis on resolved shear stress and strain obtained from MD simulations reveals quantized/discrete feature of dislocation slip events [10]. Figure 2.1d shows the evolution of a typical grain averaged resolved shear stress and strain from MD simulations in the course of a full dislocation propagating through a grain. Fig 2.1a-c illustrate a cut through one grain with atoms colored according to their local crystallographic order at three crucial snapshots during the process. Here a full dislocation loop has already been present for more than 25 ps (see Fig. 2.1a) before the grain-averaged stress $\tau$ reaches a critical value at Fig. 2.1b. Then this full dislocation depins from GB obstacles (Fig. 2.1b to c), and sweeps across the grain. Consequently, the grain-averaged shear $\gamma^*$ jumps by a discrete amount. And simultaneously the resolved shear stress $\tau$ drops abruptly due to load shedding (see Fig. 2.1d). This discrete feature of dislocation slip can be ascribed to the as small as nano size of grain $d$, which is comparable to the length scale of Burgers vector $b$ ($\sim 0.25$ nm). Thereby plastic strain induced by dislocation traversing a grain, approximately $b/d$, is on the order of percentage, and only takes quantized value of percentage. Two additional features are noted from MD simulations. First, dislocation mediated plasticity in NC metals is
dominated by propagation. The presence of dislocation embryos is predominant at small \( \tau \) (less than 300–400 MPa). Those embryos are waiting for propagating before \( \tau \) reaches a critical value (600–700 MPa). Therefore on grain-averaged level, dislocations in nano grains request a larger resolved shear stress to propagate than to nucleate, and thus slip process is controlled by propagation. **Second**, there exists a distribution of critical resolved shear stress to trigger slip events. The distribution shows an asymmetric feature, and peaks between 600 and 700 MPa. This is also a stress region where dislocation propagation mechanism dominates. At larger resolved shear stress (~900 MPa), dislocation slip precedes so fast (<10ps) that no separate stages of dislocation nucleation and propagation can be recorded. The jump in stress and strain indicates the occurrence of slip events. On the other hands, slip processes are seldom observed at averaged resolved shear stresses less than 500 MPa.

The quantized feature of dislocation slip observed from MD simulations motivates our microstructural model, termed as quantized crystal plasticity (QCP) model. The QCP model is aimed to capture the discrete features in shear stress and strain for a dislocation slip event at a grain-averaged level. With the quantized features, QCP model is employed to investigate unique NC mechanical behaviors at a strain realistic rate, and bridge the gap between atomistic simulations and experimental testing.
Figure 2. 1: A full dislocation depins from GB ledge obtained by Molecular Dynamics simulations. (a) – (c) show a cut through a selective grain (G34) with the atoms colored according to their local crystallographic order. Atoms with FCC environment are colored in grey, HCP in red; other 12 coordinate environment in green; and non-12 environment in blue. (d) shows the corresponding evolution of grain averaged shear $\gamma^*$ and resolved shear stress $\tau$[10].
2.2 Constitutive Relation of Quantized Crystal Plasticity

Constitutive relation associates total deformation increment at a material point to external stimulus, e.g. stress, with material properties at that point. The total deformation can be ascribed to elasticity and plasticity in current study. Crystal plasticity is motivated by plastic anisotropic response of crystal, that is the plastic deformation can only occur along some specific crystallographic orientations. Quantized crystal plasticity model employs the conventional crystal plasticity formulization [86-89], but features with shear stain induced by dislocation slip only taking some discrete/quantized values. This quantized feature results from decreasing grain size into nano scale.

A large deformation kinematical and constitutive framework is employed. Following [90], an infinitesimal vector $\mathbf{dX}$ in an undeformed (reference) configuration is distorted into a vector $\mathbf{dx} = \mathbf{FdX}$ in a deformed configuration. Here the deformation gradient

$$\mathbf{F} = \mathbf{F}^* \mathbf{F}^p$$

is multiplicatively decomposed into an elastic part $\mathbf{F}^*$ and a plastic part $\mathbf{F}^p$. The elastic strain is

$$\mathbf{E}^* = \frac{1}{2} (\mathbf{F}^{*T} \mathbf{F}^* - \mathbf{I})$$

The stress at a materials point is given by

$$\mathbf{T}^* = \mathbf{C} \mathbf{E}^*$$
Here $\mathbb{C}$ is a fourth-order elasticity tensor. For FCC materials, there are only three independent constants, which are traditionally denoted by $C_{11}$, $C_{12}$, and $C_{44}$. $\mathbf{T}^\ast$ is the symmetric Piola-Kirchoff stress, which is elastic work conjugate to $\mathbf{E}^\ast$. It relates with Cauchy stress $\mathbf{T}$ by

$$
\mathbf{T}^\ast = \det(\mathbf{F}^\ast) \mathbf{F}^{\ast-T} \mathbf{F}^{\ast-T}
$$

The time evolution equation for $\mathbf{F}^p$ is given by flow rule

$$
\dot{\mathbf{F}}^p = L^p \mathbf{F}^p
$$

with

$$
L^p = \sum_\alpha \dot{\gamma}^\alpha \mathbf{S}^\alpha_0, \quad S^\alpha_0 = \mathbf{s}^\alpha_0 \otimes \mathbf{m}^\alpha_0
$$

where the orthonormal vector pairs ($\mathbf{s}^\alpha_0$, $\mathbf{m}^\alpha_0$) define the slip direction and slip plane normal of the slip system $\alpha$. For FCC material, $\mathbf{S}^\alpha_0$ is chosen to be one of the twelve $\langle 110 \rangle/\{111\}$ slip systems. The shear rate $\dot{\gamma}^\alpha$ is determined by specific constitutive functions $\dot{\gamma}^\alpha = \tilde{\dot{\gamma}}^\alpha (\tau^\alpha, \tau_c^\alpha)$, where the resolved shear stress $\tau^\alpha$ is determined by

$$
\tau^\alpha = \mathbf{s}^\alpha_0 \cdot (\mathbf{F}^\ast \mathbf{F}^{\ast-T}) \mathbf{m}^\alpha_0
$$

and $\tau_c^\alpha$ is the critical stress for a slip event. For NC metals, at a material point $\tau_c^\alpha$ is constant with deformation, i.e. no strain hardening, and thus is the same for all twelve candidates, then denoted as $\tau_c$ for short.
The discrete jumps in local (grain-averaged) shear strain observed in recent MD simulations [10] are rationalized in terms of the increment in plastic strain at the grain level,

\[ \Delta \gamma^\alpha = \gamma_{\text{target}}^{\alpha} \otimes m_0^\alpha, \quad \gamma_{\text{target}} = \frac{A_s b}{V_g} = \frac{b}{d} \]

(2.8)
due to glide of a dislocation loop across a grain with cross sectional area \( A_s \) and volume \( V_g \). Here, the active slip system \( \alpha \) has Burgers vector magnitude \( b \). In principle, \( \gamma_{\text{target}} \) depends on the glide plane on which dislocation cuts through a grain, and the grain shape as well. These factors are taken care by geometric parameter \( g \). For instance, \( g = 1.5 \) for a center-cut glide plane of a spherical grain, and \( g = 1.2 \) for the maximum cross sectional area of a cubic grain with edge length \( d \). The discrete or quantized nature of slip in Eq. (2.8) is captured via a modification to conventional kinetic flow law proposed by Asaro and Needleman[88]. Specifically, the rate of plastic shear strain on the 12 FCC \( <110>/<111> \) slip systems is specified

\[
\dot{\gamma}^\alpha = \begin{cases} 
\dot{\gamma}_0 \frac{\tau^\alpha}{C(\tau_c)\tau_c} \left| \frac{1}{m} \right| \text{sign}(\tau^\alpha) & \text{inactive slip} \\
\dot{\gamma}_0 \text{sign}(\tau^\alpha) & \text{active slip} 
\end{cases}
\]

(2.9)

The *inactive slip condition* applies on slip system \( \alpha \) when \( \left| \tau^\alpha \right| < \tau_c \). Compared to the standard flow law formalism, a coefficient, \( C(\tau_c) = \tau_c/\text{MPa} \) has been inserted and also \( \tau_c \) (specified in MPa) remains constant with deformation. In this regime, \( \dot{\gamma}^\alpha < 10^{-28} \dot{\gamma}_0 \), and thus grains essentially deform by anisotropic elasticity. The *active slip condition* for slip
system $\alpha$ applies when $|\tau^\alpha| \geq \tau_c$ and it continues until $\gamma^\alpha$ increments by a quantized amount, $\gamma_{\text{target}} \text{sign}(\tau^\alpha)$, even though load shedding might make $|\tau^\alpha| < \tau_c$, prior to finishing the increment. The magnitude of $\tau^\alpha$ can become less than $\tau_c$ during slip propagating, but $\tau^\alpha$ is required to maintain the same sign in this process. This requirement is termed as propagation condition. The enforcement of propagation condition provides a lower bound to $\tau_c$ at which once a slip event occurs it completes as well. In this regime, $|\dot{\gamma}^\alpha| = \dot{\gamma}_0$, where $\dot{\gamma}_0 > 10\varepsilon_{\text{global}}$ to ensure relatively instantaneous slip.

2.3 Finite Element Implementation

The constitutive relation is implemented in the UMAT (User-Material) subroutine in ABAQUS/Standard Finite Element code [91]. A brief description of numerical implementation procedure is to calculate stress field $T$ and other field variables at the end of a time increment provided an estimated incremental displacement field $F$. The calculated $T$ needs to satisfy principle of virtual work (enforcing equilibrium and boundary condition in a weak sense) with the estimated $F$; otherwise a new $F$ will be estimated and restart the procedure. Accordingly, provided \{F$^p$(t), T(t)\} at time $t$ and a proposed new deformation gradient $F(\tau)$, there exist two goals: (1) to update the values of \{F$^p$(\tau), T(\tau)\} at some new time $\tau = t + \Delta t$; (2) to construct Jacobian matrix $\partial T / \partial E^*$ for revising estimated $F(\tau)$. Within the two goals, (1) is the key issue. A fully-implicit time integration procedure is employed, and follows Eq. (15) – (29) of Kalidindi [90]. The specific modifications in Eq. (2.9) produce
\[
\frac{\partial \dot{\gamma}^\alpha}{\partial \tau^\alpha} = \begin{cases}  
\frac{\dot{\gamma}^\alpha}{m \cdot \tau^\alpha} & \text{inactive} \\
\frac{\dot{\gamma}_0}{m \cdot \tau_c} \text{sign}(\tau^\alpha) & \text{active}
\end{cases}
\]  

(2.10)

\(\partial \dot{\gamma}^\alpha / \partial \tau^\alpha\) is used in Newton-Raphson iteration to find \(T(\tau)\) within fully-implicit scheme.

While slip is inactive, \(\partial \dot{\gamma}^\alpha / \partial \tau^\alpha \approx 0\) for negligible plasticity occurring (\(\dot{\gamma}^\alpha \sim 0\)). There is a dramatic increment in \(\partial \dot{\gamma}^\alpha / \partial \tau^\alpha\) when slip is turning on, since \(\dot{\gamma}^\alpha\) jumps from \(-0\) to \(\dot{\gamma}_0\) and \(\tau^\alpha = \tau_c \text{sign}(\tau^\alpha)\) in Eq. (2.10). Correspondingly, a reverse jump occurs when slip is turned off. A polynomial with power \(1/m\) (inactive slip case in Eq. 2.9) is applied to interpolate this abrupt change. But \(m\) cannot be too small to make stress iteration equation (Eq.(27) in Kalindindi[90]) too stiff, which will cause convergence problem.

A term \(C(\tau_c) \cdot \tau_c\) rather than \(\tau_c\) is employed while calculating \(\partial \Delta \dot{\gamma}^\alpha / \partial \tau^\alpha\) to construct Jacobian matrix (B.38 in [92]) in accordance with Eq. 2.9. In general, the coefficient \(C(\tau_c)\) decreases \(\partial \Delta \dot{\gamma}^\alpha / \partial \tau^\alpha\), the dependence of change in plastic increment on shear stress. This decrease is applicable to both inactive and active cases. The Jacobian matrix controls the rate of convergence, but does not influence solutions. Such a modification on Jacobian matrix gives a reasonable convergence rate.

A polycrystal is defined with \(N \times N \times N\) grains, each represented by an 8-node, 3D brick finite element (C3D8) using ABAQUS software [91]. Uniaxial strain rate \(\dot{\varepsilon}_{\text{global}}\) is imposed via a constant displacement rate \(\dot{u}_Z\) on the top face of \(Z\) direction with free surface boundary conditions on the X and Y faces shown in Figure 2.2a. Each FCC
grain is approximated by a cube and assigned an initial crystallographic orientation. Compatibility and equilibrium are then enforced throughout the polycrystal. Figure 2.2a shows the case for $N = 10$, with each grain (element) colored according to the maximum Schmid factor $S_{z}^{\text{max}}$ among the twelve $\frac{1}{2}<110>/\{111\}$ FCC slip systems in that grain. A 1000-grain model is sufficiently large to make the macro stress-strain response effectively independent of sample size. Smaller $N$ samples are prone to shear instabilities and larger $N$ samples can require excessive computation time without significant differences in stress-strain response. A typical analysis with 1000 grains strained to $\varepsilon^p \sim 1\%$ requires $\sim 7$ CPU hours with a Dell WorkStation PWS670 (Dual Xeon 2.80 GHz Processor and 2.00 GB RAM). The computational time has been dramatically shortened when running simulations at The Ohio Super Computer Center.
Figure 2.2: (a) A finite element model of a polycrystal with $10 \times 10 \times 10$ grains, each represented by an 8-node brick element. Uniaxial tension is applied along the $z$-direction. Each grain is colored according to $S_{z}^{\text{max}}$ the maximum Schmid factor among the 12 FCC slip systems in that grain. Numerical values are as shown in color bar. (b) Microstructural information at a material point.
2.4 Material Parameters

The microstructural information at a material point includes: (1) crystallographic orientation; (2) critical resolved shear stress for a slip event $\tau_c$; (3) grain size $d$, and thereby the quantized value of shear increment for one slip event $\gamma_{\text{target}}$ (ref. to Eq. 2.8); and (4) slip activity, e.g. quantized number of shear jump $q$, on each of 12 FCC slip systems. These are depicted in Figure 2.2b. The interactions between different slip systems at a material point and among different material points are captured. Our goal is to track the inhomogeneous development of slip activity with the influence of discrete/quantized plasticity.

2.4.1 Crystal Orientations

Crystal orientation affects both elastic and plastic anisotropy. It is the key material parameter for conventional crystal plasticity model. In the QCP study, only random orientation is considered, such that the influence from quantized slip, and slip strength can be well identified. In general, the Schmid factor $S_z^\alpha$ for a slip $\alpha$ dictates the stress taken on $\alpha$ from external load, which is imposed along Z-axis. $S_z^\alpha$ is calculated by

$$S_z^\alpha = s_z^{\alpha(g)} m_z^{\alpha(g)}, \alpha = 1 \text{ to } 12$$

(2.11)

where $s_z^{\alpha(g)}$ and $m_z^{\alpha(g)}$ are the z-components of the slip direction and slip plane normal for slip system $\alpha$, when $s^\alpha$ and $m^\alpha$ are represented in global coordinate systems.

Particularly the maximum Schmid factor, $S_z^{\text{max}}$, among the 12 FCC competing slip
systems at a material point, dictates the initial yielding. It varies from grain to grain and strongly affects the development of inhomogeneous deformation pattern. A set of 20,000 random orientations was obtained by the method described in [93]. Figure 2.3b displays $S_z^{\text{max}}$ of these orientations. From this set, 1,000 orientations are randomly selected and assigned to the 1,000 grains in QCP model. Sometimes, to improve small sample statistics, several random 1000-grain sets were selected from this 20,000-grain set (see Chapter 5). The resulting $S_z^{\text{max}}$ distribution of those 1000 orientations is shown in Fig. 2.3a and it replicates the 20,000-random-orientation set (Fig. 2.3b) and has the same probability density (see Fig. 2.3c), particularly the same mean value, $\bar{S}_z^{\text{max}} = 0.45$. The random nature of 1000 orientations is further confirmed by pole figures (Fig. 2.4)
Figure 2.3: The number of grains with a maximum Schmid factor $= S_{z}^{\text{max}} \pm 0.001$, out of $N_{\text{total}}$ grains. $N_{\text{total}} = 1000$ for (a) assigned to QCP model; $N_{\text{total}} = 20,000$ for (b), the set of random orientations obtained by the method described in [93]. (a) has the same mean value of $S_{z}^{\text{max}} (= 0.45)$ as (b). (c) shows that (a) and (b) have the same probability density of maximum Schmid factor.
Figure 2.4: \{100\}, \{110\} and \{111\} pole figure plots (stereographic projection) for (a) 1000-orientation QCP sample and (b) 20000-orientation sample. See caption of Fig. 2.3 for sample details.
2.4.2 Critical Stress for Dislocation Slip $\tau_c$

The critical stress for a slip event, $\tau_{\text{crit}}$ is the stress at grain level required to reach for completing a slip event. It is a larger value between stress to nucleate a dislocation and stress to expand/propagate a dislocation. In principle, $\tau_{\text{crit}}$ is spatially non-uniform and depends on numerous factors including slip plane orientation to the grain boundary, elastic anisotropy, grain boundary accommodation of incoming or outgoing dislocation content [94-100], and even grain size and shape. In addition to uniform $\tau_{\text{crit}}$ distribution, three types of $\tau_{\text{crit}}$ distributions from two probability density functions are employed. **Type A:** a normal distribution with probability density

$$
\rho(\tau, \mu, \sigma) = \frac{1}{\sigma \sqrt{2\pi}} \exp\left(-\frac{(\tau - \mu)^2}{2\sigma^2}\right)
$$

(2.12)

where $\mu$ is the mean value and $\sigma^2$ is the variance. **Type B and C** follows a gamma distribution,

$$
\rho(\tau, k, \theta) = (\tau - \tau_{\text{min}})^{k-1} \frac{\exp\left(-\frac{\tau - \tau_{\text{min}}}{\theta}\right)}{\Gamma(k) \theta^k}
$$

(2.13)

where the mean and variance are $\tau_{\text{min}} + k\theta$ and $k\theta^2$, respectively. **Figure 2.5** shows sample cases of these three types for which $\tau_{\text{crit}}$ has the same minimum (400 MPa) and mean (730 MPa). However, the normal distribution (Fig. 2.5a) is symmetric about the mean value whereas the gamma distributions are moderately (Fig. 2.5b, $k = 2$) and very (Fig. 2.5c, $k = 1$) asymmetric. In fact, as $k$ increases, gamma distribution approaches normal distribution.
Figure 2.5: Probability density distributions for $\tau_{\text{crit}}$, the critical shear stress for a slip event, for (a) a normal distribution with $\mu = 730$ MPa and $\sigma = 98$ MPa, (b) a gamma distribution with $k = 2$, $\theta \approx 165$ MPa, and $\tau_{\text{min}} = 400$ MPa, and (c) a gamma distribution with $k = 1$, $\theta \approx 330$, and $\tau_{\text{min}} = 400$ MPa. See Eqs. 2.12 and 2.13.

2.4.3 Quantized Plastic Strain Increment for Dislocation Slip $\gamma_{\text{target}}$

The quantized/jump $\gamma_{\text{target}}$ ($\sim b/d$, see Eq. 2.8) in shear strain is viewed as follows: one dislocation loop sweeps across a grain, and producing a magnitude of $b$ offset on slip plane. On grain-averaged level, such a $b$ offset results in a shear increment of $b/d$ (see Fig. 2.2). The discrete nature becomes significant when $\gamma_{\text{target}}$, the strain associated with one slip event, falls into the macroscopically investigated strain range, which is on the order of percent. Burgers vector $b \sim 0.25$ nm, and thus discrete nature needs to be considered when grain size $d$ is in the range of 10nm to 100 nm, the size of which is in the category of NC or UFG materials.
2.4.4 Relation Between $\tau_{c,\text{min}}$ and $\gamma_{\text{target}}$ from Propagation Requirement

The relation between $\tau_c$ and $\gamma_{\text{target}}$ is established through a *propagation/expansion condition*. This condition imposes a physical requirement that the resolved shear stress in a grain does not change sign during a slip event. The purpose is to ensure sufficient stress to complete expansion of the loop, which depends sensitively on line energy and local grain and dislocation geometry [101]. According to the Eshelby solution [102], a transformation strain $\gamma_{\text{target}}$ in an ellipsoidal region will induce a stress drop

$$\Delta \tau = c \Delta \tau^\text{Eshelby} = -c M \gamma_{\text{target}}; \quad M = \mu \cdot \frac{7 - 5\nu}{15 \cdot (1 - \nu)}$$

(2.14)

where $\mu$ and $\nu$ are, respectively, the elastic shear modulus and Poisson’s ratio of a homogeneous isotropic elastic medium. The scaling parameter $c$ is introduced to calibrate the numerical QCP results to cuboidal (non-ellipsoidal) regions and also to account for elastic anisotropy.

Using Eq. 2.14, the positive stress *expansion condition* requires that

$$\tau_{\text{crit}} \geq c M \gamma_{\text{target}} \quad \text{or} \quad \gamma_{\text{target}} \leq \tau_{\text{crit}} / c M.$$  

(2.15)

This condition is enforced QCP simulations by specifying $\gamma_{\text{target}}$ as the minimum of two quantities, i.e.,

$$\gamma_{\text{target}} = \min \left( \frac{\tau_{\text{crit}}}{c M}, 1.2 \frac{b}{d} \right)$$

(2.16)
Thus, grains with sufficiently small values of $\tau_{\text{crit}}$ require $\gamma_{\text{target}} < 1.2 \, b/d$ in order to satisfy the stress-based propagation/expansion condition. An interpretation is that in some grains, $\tau_{\text{crit}}$ is insufficient to drive events with $\gamma_{\text{target}} = 1.2 \, b/d$, but it is sufficient to drive smaller $\gamma_{\text{target}}$ events. From Eq. (2.8), smaller $\gamma_{\text{target}}$ events are those who have a smaller cross sectional area $A_s$. In QCP simulations, the variation in $\tau_{\text{crit}}$ on a grain-to-grain level introduces a grain-to-grain variation in $\gamma_{\text{target}}$, via Eq. 2.16. It is worth pointing out that the propagation condition be enforced during slip. That is to stop a slip event ($\gamma_{\text{target}}$ not yet reached) if the resolved shear stress drops to zero. Physically, this corresponds to dislocation sweeping part of a grain. However, this enforcement will depress the discrete nature of plasticity, and the physical picture does not consist with dislocation traversing the entire grain of NC metals.

2.5 Computational Parameters

The value of computational parameters are chosen to achieve two goals: (1) to capture quantized slip, i.e. relatively instant slip; (2) to have reasonable accuracy in producing quantized slip increment. For the first goal, the ratio between slip rate and macroscopic loading rate $\dot{\gamma}_0 / \dot{\varepsilon}_{\text{global}}$ is essential. The QCP simulations show that when $\dot{\gamma}_0 = 2 \cdot 10^{-2} / s$, the results are relatively insensitive to $\dot{\varepsilon}_{\text{global}}$, provided $\dot{\varepsilon}_{\text{global}} \leq 1 \cdot 10^{-3} / s$. Otherwise a numerical strain rate hardening will appear. This hardening results from activating less preferred slip systems since the current activating slips are not sufficient enough to accommodate the macroscopic deformation. Second, an accuracy representation of quantized slip $\gamma_{\text{target}}$ depends on $\dot{\gamma}_0 \cdot \Delta t_{\text{max}}$, where $\Delta t_{\text{max}}$ is the maximum time step
specified in FEM simulations. If $\Delta t_{\text{max}} = 0.1 \gamma_{\text{target}} / \dot{\gamma}_0$, this produces the shear increment for a jump between 0.9 to 1.1$\gamma_{\text{target}}$, which is acceptable. There is a balance between ‘instant’ and ‘accuracy’ of one slip event under the same computational consumption $\Delta t_{\text{max}}$. Finally, the choice $m = 0.1$ in Eq. 2.9 furnishes a plastic strain rate that is negligible when $\tau < \tau_{\text{crit}}$, and ensure stable, convergent solutions.

2.6 Concluding Remarks

A quantized crystal plasticity (QCP) model is developed within a large strain finite element formulation with four key microstructural features: (1) anisotropic elastic moduli; (2) a grain orientation distribution; (3) a grain-to-grain distribution of critical resolved shear stress $\tau_{\text{crit}}$; and (4) a grain-to-grain distribution of the largest possible jump $\gamma_{\text{target}}$ in plastic strain associated with a slip event [103, 104]. Several conclusions are drew:

- A quantized feature in QCP model is motivated by recent MD simulations showing that the discrete changes occur in stress and strain at the grain-averaged level during a dislocation slip event[105].

- The QCP model adopts the geometric scaling relation that a shear jump $\gamma_{\text{target}} \sim b/d$, where $b$ is the magnitude of Burgers vector, and $d = $ grain size (ref. to Eq. 2.8)

- The critical stress for a slip event, $\tau_{\text{crit}}$ is the stress at grain level required to reach for completing a slip event. $\tau_{\text{crit}}$ can be spatially non-uniform and depends on numerous factors including slip plane orientation to the grain boundary, elastic
anisotropy, grain boundary accommodation of incoming or outgoing dislocation content [94-100], and even grain size and shape. The calculated $\tau_c$ also entails a large spectrum in stress.

- A relation between $\tau_c$ and $\gamma_{\text{target}}$ is established through a *propagation/expansion condition* (ref. to Eq. 2.14). This condition imposes a physical requirement that the resolved shear stress in a grain does not change sign during a slip event.

Limitation of QCP model includes that: (1) GB deformation mechanisms, *e.g.* GB sliding, are not incorporated; (2) QCP constitutive relation is assumed to be rate-independent, though strain rate hardening may appear as a result of insufficient numerical implementation; (3) grain geometry is not well captured with FEM cuboidal representation, therefore the stress concentration at the GBs is not captured. But the aim of QCP model is to simulate the interaction of slip activity within and between material points under the influence of quantized plasticity; the details in stress concentration are not the main concern.
CHAPTER 3: QCP ON TENSILE RESPONSES OF NC AND UFG Ni

3.1 QCP with Homogenous Slip Strength $\tau_c$

3.1.1 Model Description and Material Parameters

The QCP with a uniform critical stress for slip $\tau_c$ and plastic shear increment $\gamma_{\text{target}}$, termed elementary model, are presented in this section. The featured model outcomes include (1) the quantized jumps in shear stress and shear strain by successfully implementing QCP constitutive law in Section 3.1.2; (2) the size dependence of those quantized jumps in features in Section 3.1.3; (3) grain size softening in stress-strain response predicted by this elementary model in Section 3.1.4.

The elementary model consisting of 1000 grains is investigated with material and computational parameters provided in Table 3.1
Table 3.1: Elementary Model: Material and Computational Parameters

<table>
<thead>
<tr>
<th>Elastic Moduli(^{(1)})</th>
<th>(C_{11}) (GPa)</th>
<th>(C_{12}) (GPa)</th>
<th>(C_{44}) (GPa)</th>
<th>(E_{\text{polycrystal}}) (GPa)(^{(2)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>246.5</td>
<td>147.3</td>
<td>124.7</td>
<td>236.5</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cryst. Texture</th>
<th>Distribution: (S_z)(^{\text{max}}) shown in Fig. 2.3(^{(3)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\tau_{\text{crit}})</td>
<td>500 MPa (uniform)</td>
</tr>
<tr>
<td>(\gamma_{\text{target}})</td>
<td>(g \cdot 10^{-3}) (uniform), where (g) ranges from 1 to 6</td>
</tr>
</tbody>
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<table>
<thead>
<tr>
<th>Computational Parameters</th>
<th>(\gamma_0) (s(^{-1}))</th>
<th>(m)</th>
<th>(\Delta t) (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2 \cdot 10^{-2}</td>
<td>0.1</td>
<td>0.01 for (\gamma_{\text{target}} = 1 \cdot 10^{-3}), 0.02 for others</td>
</tr>
</tbody>
</table>

\(^{(1)}\) From Huntington [106]; \(^{(2)}\) For Ni, \(\mu = 94.7\) GPa, \(\nu = 0.276\); \(^{(3)}\) \(S_z^{\text{max}} = 0.45\).

3.1.2 Quantized Jumps in Shear Strain and Stress on the Grain Level

The quantized jumps in shear strain and stress at the grain level are captured by QCP simulations. Figure 3.1 shows \(\gamma^{(\alpha)}\) and \(\tau^{(\alpha)}\) vs. \(\varepsilon^{p}_{\text{global}}\), for a slip system with a moderate Schmid factor \(S_z = 0.465\) in an interior grain. \(\varepsilon^{p}_{\text{global}}\) is computed as the difference between the total and elastic global tensile strain, where the latter is equal to \(\sigma_{\text{global}}/E_{\text{polycrystal}}\). The black stepped (lower) trace shows that \(\gamma^{(\alpha)}\) jumps successively by \(\gamma_{\text{target}}\) (= 4 \cdot 10^{-3} here). The gray lines labeled “B” and “C” indicate that the average slope of the trace increases with deformation. This is expected in regions where plasticity increases with \(\varepsilon^{p}_{\text{global}}\). The red (upper) trace confirms that \(\gamma^{(\alpha)}\) jumps when \(\tau^{(\alpha)} = \tau_{\text{crit}}\) (= 500 MPa here). During a jump, \(\tau^{(\alpha)}\) monotonically decreases, reaching a minimum at the completion of the jump. Subsequently, it monotonically increases via elastic deformation. Occasionally, \(\tau^{(\alpha)}\) drops abruptly (arrow A) or deviates from linearity.
(arrow A’), even though slip system α is not active. This is due to other nearby slip events.

Figure 3.1: Shear strain and stress vs. global plastic strain, at an integration point in an interior grain, based on the elementary model (Table 3.1). The grain of interest has $S_z^{max} = 0.465$.

3.1.3 Size Dependence of the Quantized Jumps

The magnitude of quantized jumps scales as $1/\text{grain size}$. Figure 3.2 shows the evolution of plastic shear, $\gamma^{p(\alpha)} \equiv \gamma_{ns}^{p(\alpha)}$, vs. global plastic strain $\varepsilon_p^{global}$, at an integration point in an
interior grain. As $\varepsilon^p_{\text{global}}$ increases, numerous jumps in $\gamma^{p(a)}$ occur in a step-like fashion, similar to the MD simulations (see Fig. 2.1). Also, the magnitude of the jumps increases with decreasing grain size according to Eq. 2.8. In general, several slip systems can contribute to plastic strain in a grain, provided the resolved shear stress on each of those systems reaches the critical value.

Figure 3. 2: Evolution of plastic shear strain $\gamma^{p(a)}$ with global plastic strain $\varepsilon^p_{\text{global}}$, at an integration point in an interior grain with maximum Schmid factor $S_z^{\text{max}} = 0.465$. Different values of $\gamma_{\text{target}}$ are considered, with $\tau_{\text{crit}} = 500$ MPa in all cases. Other input data is in Table 3.1.
Consequently the magnitude of quantized jumps in shear stress scales as $1/{\text{grain size}}$.

**Figure 3.3** shows corresponding traces of $\tau^{(\alpha)}$ vs. $e^{p}_{\text{global}}$ for four discrete values of $\gamma_{\text{target}}$: $1 \cdot 10^{-3}$, $2 \cdot 10^{-3}$, $4 \cdot 10^{-3}$, and $6 \cdot 10^{-3}$, with $\tau_{\text{crit}} = 500$ MPa. Several features are noted. First, the magnitude, $|\Delta \tau|$, of the stress drop tends to increase with increasing $\gamma_{\text{target}}$. There is good agreement with the Eshelby solution (see Eq. 2.14), when values of $\mu$ and $\nu$ for polycrystalline Ni (Table 3.1) are used with $c = 1.2$. Second, the number $n$ of oscillations increases with decreasing $\gamma_{\text{target}}$. This is reasonable since the product $n\gamma_{\text{target}}V_g/V_{\text{total}} \sim e^{p}_{\text{global}}$. Third, the non-uniform values of $\Delta \tau$ and nonlinear reloading of $\tau^{(\alpha)}$ following an event suggest strong interaction from neighboring slip events. Finally, in the limit of larger grain size, $\gamma_{\text{target}}$ diminishes and the QCP model reverts to a conventional elastic-perfectly plastic crystal plasticity model.
Figure 3.3: Resolved stress vs. global plastic strain at an integration point in an interior grain, for $\gamma_{\text{target}}$ ranging from $1 \times 10^{-3}$ to $6 \times 10^{-3}$. Elementary model parameters (Table 3.1) are used and the grain of interest has $S_z^{\text{max}} = 0.465$. The vertical lines with arrows indicate the magnitude of local stress drop $|\Delta\tau_{\text{Eshelby}}|$ predicted from an Eshelby approach (Eq. 2.15), with $c = 1.2$.

Figure 3.3 motivates two important microstructural connections. First, the propagation condition (Eq. 2.15) imposes a lower bound, $\tau_{\text{crit(min)}} \sim \gamma_{\text{target}}$. Since $\gamma_{\text{target}} \sim 1/d$ (Eq. 2.8),
this dictates $\tau_{\text{crit(min)}} \sim 1/d$, so that there is a source of strengthening with decreasing grain size. Second, the polycrystal is predicted to achieve a fully developed flow stress,

$$
\sigma_{\text{global}}^{\text{plateau}} \approx \frac{1}{\overline{S}_z^{\text{max}}} \left( \tau_{\text{crit}} - \frac{|\Delta \tau|}{2} \right)_{\text{mean}}
$$

(3.1)

where $\overline{S}_z^{\text{max}}$ is the maximum Schmid factor in a grain, averaged over all slipped grains in the polycrystal, and the subscript “mean” denotes an average over the numerous slip events in Fig. 3.3.

Eq. 3.1 suggests that $\sigma_{\text{global}}^{\text{plateau}}$ may increase or decrease with grain size $d$, depending on whether $\tau_{\text{crit}} (= \tau_c)$ is independent of $d$. When $\tau_{\text{crit}}$ is independent of $d$, $\sigma_{\text{global}}^{\text{plateau}}$ is predicted to decrease with decreasing $d$ since $|\Delta \tau| \sim 1/d$ (Fig. 3.3). Alternately, if $\tau_{\text{crit}} \sim 1/d$ is adopted as suggested by the propagation condition (Eq. 2.15), then $\sigma_{\text{global}}^{\text{plateau}}$ is predicted to increase with decreasing $d$.

3.1.4 Grain Size Softening with Homogeneous $\tau_c$

Grain size softening is predicted by elementary model, when $\tau_{\text{crit}}$ is independent of grain size $d$. Figure 3.4 shows the $\sigma_{\text{global}}-\epsilon_{\text{global}}^{\text{p}}$ response for discrete values of $\gamma_{\text{target}}$ ranging from $1 \cdot 10^{-3}$ to $6 \cdot 10^{-3}$. The elementary model parameters are used, so that $\tau_{\text{crit}} = 500$ MPa throughout the sample. Also, $\tau_{\text{crit}}$ is assumed to be independent of $d$. At larger $\gamma_{\text{target}}$, $|\Delta \tau|$ increases, and thus $\sigma_{\text{global}}^{\text{plateau}}$ is smaller, which is consistent with Eqs. 2.15 and 3.1. Further, strain softening occurs at larger strain, consistent with localization induced by large slip...
events. Yet, Fig. 3.4 is at odds with experimental trends that strength increases with decreasing $d$ down to $\sim 10$ nm [107]. The disparity suggests that a key assumption in Fig. 3.4— that $\tau_{\text{crit}}$ is independent of grain size—is inconsistent with experimental data.

Figure 3.4: Global tensile stress-plastic strain response based on the elementary model (Table 3.1). $\gamma_{\text{target}}$ ranges from $1 \cdot 10^{-3}$ to $6 \cdot 10^{-3}$ and $\tau_{\text{crit}} = 500$ MPa.
3.2 QCP with Heterogeneous Slip Strength $\tau_c$

3.2.1 Model Description and Material Parameters

The QCP model with a grain-to-grain variation in $\tau_{\text{crit}}$ (and $\gamma_{\text{target}}$), termed *extended model*, is presented in Section 3.2. Two featured aspects are addressed in the two subsections: (1) an asymmetric $\tau_{\text{crit}}$ distribution is able to capture the gradual transition from micro to macro plasticity (e.g. $\varepsilon_{\text{p,tran}}$ in Fig. 3.6) observed in NC metals; (2) an analytical analysis is performed to assess the relation between $\tau_{\text{crit}}$ distribution and stress-strain response.

Material and computational parameters for *extended model* are listed in Table 3.2.

| Table 3.2: Extended Model: Material, and Computational Parameters |
|----------------------|------------------|------------------|------------------|------------------|
| **Elastic Moduli**$^{(1)}$ | $C_{11}$ (GPa) | $C_{12}$ (GPa) | $C_{44}$ (GPa) | $E_{\text{polycrystal}}$ (GPa)$^{(2)}$ |
| Ni | 246.5 | 147.3 | 124.7 | 236.5 |
| **Cryst. Texture** | Distribution: $S_{z}^{\text{max}}$ shown in Fig. 2.3a$^{(3)}$ |
| $\tau_{\text{crit}}$ | Three possible distributions: |
| | Type A. Normal distribution (Eq. 2.12, Fig. 2.5a) |
| | Type B. Gamma Distribution (Eq. 2.13, Fig. 2.5b) |
| | Type C. Gamma Distribution (Eq. 2.13, Fig. 2.5c) |
| $\gamma_{\text{target}}$ | Distribution satisfying the expansion condition |
| | $\gamma_{\text{target}} = \min(\tau_{\text{crit}}/cM, 1.2 b/d)$ (Eq. 2.15)$^{(4)}$ |
| **Computational Parameters** | $\dot{\gamma}_{0}$ (s$^{-1}$) | $m$ | $\Delta t$ |
| Ni | $2 \times 10^{-2}$ | 0.1 | $\gamma_{\text{target,max}}/\dot{\gamma}_{0}$ (Eq. 2.16) |

(1) From Huntington [106]; (2) For Ni, $m = 94.7$ GPa, $n = 0.276$; (3) $S_{z}^{\text{max}} = 0.45$.

(4) For Ni, $M = 50$ GPa, $b = 0.25$ nm; and $c = 1.2$ in Eqs. 2.14 to 2.16.
3.2.2 Asymmetric $\tau_c$ Extends Elastic-Plastic Transition Regime

Figure 3.5 shows the predicted $\sigma_{\text{global}}$, $\varepsilon_{\text{global}}^p$ response for extended QCP models, in which $\tau_{\text{crit}}$ varies from grain-to-grain according to the Type A, B, and C distributions in Fig 2.5. In simulations, $\gamma_{\text{target}} = 6 \cdot 10^{-3}$, corresponding to $d = 50$ nm. All the three distributions have $\tau_{\text{crit(min)}} = 400$ MPa, so that the propagation condition (Eq. 2.15) is satisfied. Further, $\tau_{\text{crit(mean)}} = 730$ MPa, so that $\sigma_{\text{plateau}}^{\text{global}} = 1180$ MPa is estimated (Eq. 3.1). A key observation is that only the Type C distribution produces the large, extended elastic-plastic transition strain typical of NC metals.

Figure 3. 5: Global tensile stress-plastic strain response based on the extended model, and Type A, Type B, and Type C probability density distributions for $\tau_{\text{crit}}$ (Fig. 2.5).
3.2.3 Insight of Extended Elastic-Plastic Transition Regime

The extended elastic-plastic transition regime indicates a gradual transition of deformation from easy yield grains to hard, which is identified by \( \tau_{\text{crit}} \). Therefore the distribution of \( \tau_{\text{crit}} \) essentially determines the development of deformation, and thus stress-strain response. A simple analysis supports the observation that a Type C (very asymmetric) distribution promotes a large, extended transition response. First,

\[
de^{\text{p}}_{\text{global}} \approx \bar{S}_{z}^{\text{max}} \frac{dn_{\text{slip}}}{N_{\text{grains}}} \gamma_{\text{target}}
\]

where \( dn_{\text{slip}} \) is an increment in the number of plastic slip events and \( N_{\text{grain}} \) is the total number of grains. Second,

\[
d\sigma_{\text{global}} \approx \frac{1}{\bar{S}_{z}^{\text{max}}} \left( 1 - \frac{N_{\text{slipped}}}{N_{\text{grain}}} \right) d\tau_{\text{unslipped}}
\]

Thus, \( d\sigma_{\text{global}} \) is achieved via increments, \( d\tau_{\text{unslipped}} \), in the local stress within unslipped (elastically deforming) grains. This occurs since all of the \( N_{\text{slipped}} \) plastically deformed grains are assumed to not work harden but rather oscillate about a mean stress, \( \tau_{\text{crit}} = \frac{|\Delta \tau|}{2} \) (see Fig. 3.3). Finally, plastically deforming grains must, on average, deform compatibly with the surrounding polycrystalline material,

\[
de^{\text{p}}_{\text{slipped}} = de_{\text{global}} \Rightarrow \bar{S}_{z}^{\text{max}} \frac{dn_{\text{slip}}}{N_{\text{slipped}}} \gamma_{\text{target}} = de^{\text{p}}_{\text{global}} + \frac{d\sigma_{\text{global}}}{E_{\text{polycrystal}}}
\]
Here, $d n_{\text{reslip}}$ is an increment in the number of slip events in already-slipped grains. Eqs. 3.2 and 3.4 furnish the increment $df_{\text{slipped}} = (dn_{\text{slip}} - d n_{\text{reslip}})/N_{\text{grain}}$ in fraction of slipped grains and incorporating this into Eq. 3.5 gives

$$\frac{d \sigma_{\text{global}}}{d e^p_{\text{global}}} \approx \left[ \frac{(S_z^{\text{max}})^2 \gamma_{\text{target}}}{(1 - f_{\text{slipped}})^2} \frac{df_{\text{slipped}}}{d \tau} + \frac{f_{\text{slipped}}}{E_{\text{polycrystal}} (1 - f_{\text{slipped}})} \right]^{-1} \quad (3.5)$$

Eq. 3.5 is consistent with several features in Fig. 9. First, all three distributions produce $d \sigma_{\text{global}}/d e^p_{\text{global}} \to \infty$ initially, when $df_{\text{slipped}}/d \tau = 0$ and $f_{\text{slipped}} = 0$. Second, $d \sigma_{\text{global}}/d e^p_{\text{global}} \to 0$ as $f_{\text{slipped}} \to 1$. Finally, $d \sigma_{\text{global}}/d e^p_{\text{global}} \sim 1/[\gamma_{\text{target}} \cdot (df_{\text{slipped}}/d \tau)]$ during the early deformation stages when $f_{\text{slipped}} \ll 1$. This last statement suggests that the Type C distribution (Fig. 2.5) produces the most gradual increase in $\sigma_{\text{global}}$ because it has the largest $df_{\text{slipped}}/d \tau$ at small $f_{\text{slipped}}$. Indeed, that is observed in Fig. 3.5.

### 3.3 QCP Model Application to ED NC and UFG Ni

#### 3.3.1 Monotonic Stress-Strain Features

Figure 3.6 shows monotonic stress-plastic strain responses of NC and UFG Ni synthesized by electro-deposition (ED). As grain size decreasing from UFG (>100 nm) to NC (<100 nm) regime, three trends can be identified. First the flow stress of NC metals can be a factor of two or more times that of UFG materials. Second, an extended elastic-plastic transition strain ($\varepsilon_{\text{trans}}$) is observed, over which the flow stress increases from an initial value $\sigma_0$ to approximately 90% of the plateau stress ($\sigma_{\text{plateau}}$). This
apparent hardening is associated with a micro-plastic regime where not all grains are plastically deforming, even when $\varepsilon_p \sim 0.2\%$, a value typically associated with macro-plasticity in conventional grain size polycrystals [38-40]. Thirdly, although the strain rate sensitivity is often larger in NC metals, failure strains ($\varepsilon_f$) are often smaller, consistent with localized deformation [3, 4, 31, 69, 108, 109]. Admittedly, such generalizations can be difficult since various synthesis techniques produce different microstructures with unique processing-induced defects [3, 4]. Even for the same synthesis method, e.g. ED, a uniform microstructure is difficult to maintain at all the grains sizes (ref. to $d$ in Fig. 3.6). But as average grain size decreases, electro-deposition method is capable of producing NC Ni with more uniform grain size and more random texture [110]. This relatively simple structure enables the assessment to the influence of grain size on mechanical properties.
Figure 3.6 Tensile stress-strain data for electrodeposited (ED) Ni, for three different grain sizes (adapted from [110]).

### 3.3.2 Model Calibration to UFG and NC Ni Responses

The next step is to calibrate the extended model with properties for Ni (Table 3.2) to the experimental data in Fig. 3.6. The probability density for $\tau_{\text{crit}}$ is calibrated using $\sigma_{\text{global}(0)}$ and $\sigma_{\text{plateau}}$ values from Fig. 3.6. In particular, $\tau_{\text{crit(min)}} = \frac{S_c^{\text{max}}}{\varepsilon_f} \sigma_{\text{global}(0)}$ and $\tau_{\text{crit(mean)}}$ is obtained from Eq. 3.1, with $\Delta\tau$ from Eq. 2.14, $\frac{S_c^{\text{max}}}{\varepsilon_f}$ from Table 3.2, and $\gamma_{\text{target}}$ from Eq. 2.8 with $c = 1.2$. Table 3.3 lists these “Estimated” $\tau_{\text{crit(min)}}$ and $\tau_{\text{crit(mean)}}$ values for three
grain sizes related to Fig. 3.6. Optimal values of $\tau_{\text{crit(min)}}$ and $\tau_{\text{crit(mean)}}$ for distribution types A, B, and C are obtained by iterative QCP simulations to match the data in Fig. 3.6 as best as possible. **Figure 3.8 (a, b, c)** shows the optimal Type A, B, and C distributions for NC Ni with $d = 50$ nm.

A complication in the fitting process is that $\tau_{\text{crit(min)}} = 280$ MPa for $d = 50$, in order to match the experimental $\sigma_{\text{global}(0)}$ values. This violates the *propagation condition* (Eq. 2.15) that $\tau_{\text{crit}} > 360$ MPa. In principle, $\tau_{\text{crit(min)}}$ could be increased but the resulting $\sigma_{\text{global}}$-$\epsilon_{\text{global}}^{p}$ curves would not adequately capture $\sigma_{\text{global}(0)}$. Rather, $\gamma_{\text{target}}$ is decreased in select grains according to Eq. 2.16, to ensure each grain satisfies the *propagation condition*. Physically, this reduction in $\gamma_{\text{target}}$ corresponds to initial slip events that have a reduced area $A_{s}$ (Eq. 2.8). However, in the **Chapter 4** it is shown that such a modification is not necessary. The initial yielding can be adjusted by introducing residual stress state.

**Figure 3.7** shows the $\sigma_{\text{global}}$-$\epsilon_{\text{global}}^{p}$ predictions from the 1000-grain QCP model; the symbols are the experimental data from Fig. 3.6. The Type C (very asymmetric) distribution captures the $d = 50$ nm and 150 nm cases best. The other distribution types overestimate the slope, consistent with Eq. 3.5, and they do not capture the gradual transition from initial to fully developed yielding. With grain size increases, the differences reduce among asymmetric Types B and C and symmetric Type A. For instance, Type B can well capture the $d = 300$ nm data. This indicates that the asymmetric feature of $\tau_{\text{crit}}$ distribution decreases with grain size increases.
Figure 3.7: The stress plastic strain responses from QCP predictions using the extended model data for Ni (see Tables 3.2, 3.3). Three types of $\tau_c$ distribution are employed: Type A (symmetric normal distribution-Eq. 2.12), Type B (moderately asymmetric gamma distribution-Eq. 2.13 with $k = 2$), and Type C (very asymmetric gamma distribution-Eq. 2.13 with $k = 1$). Experimental $\sigma_{\text{global}}-\varepsilon_{\text{global}}^p$ data (symbols) for ED Ni [110] is displayed for comparison.
Figure 3.8: The three types of critical stress $\tau_c$ distributions used for $d = 50$ nm in Fig. 3.7.

Table 3.3: Calibration of the Extended Model to Experimental ED NC and UFG Ni Data

<table>
<thead>
<tr>
<th>average grain size $d$</th>
<th>50 (nm)</th>
<th>150 (nm)</th>
<th>300 (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\tau_{\text{crit}}$ distribution</td>
<td>$\tau_{\text{crit, min}}$</td>
<td>$\tau_{\text{crit, mean}}$</td>
<td>$\tau_{\text{crit, min}}$</td>
</tr>
<tr>
<td>(MPa) Estimated</td>
<td>283</td>
<td>700</td>
<td>173</td>
</tr>
<tr>
<td>Type C</td>
<td>280</td>
<td>700</td>
<td>160</td>
</tr>
<tr>
<td>Type B</td>
<td>280</td>
<td>650</td>
<td></td>
</tr>
<tr>
<td>Type A</td>
<td>280</td>
<td>625</td>
<td></td>
</tr>
<tr>
<td>$\gamma_{\text{target}}$ distribution</td>
<td>non-uniform (Eq. 8)</td>
<td>uniform</td>
<td>uniform</td>
</tr>
<tr>
<td>$\gamma_{\text{target, min}}$</td>
<td>$4.7 \cdot 10^{-3}$</td>
<td>$2.0 \cdot 10^{-3}$</td>
<td>$1.0 \cdot 10^{-3}$</td>
</tr>
</tbody>
</table>
3.3.3 Model Prediction of Strain Localization for NC Ni

A more localized deformation is predicted by QCP model of NC Ni than UFG Ni based on the fact that fewer grains deform to accommodate the same global plastic strain. Figure 3.9 shows corresponding results for the fraction of slipped grains, \( f_{\text{slipped}} \), vs. \( \varepsilon^p_{\text{global}} \). The Type C distribution produces a very gradual increase in \( f_{\text{slipped}} \) (Fig. 3.9a).

This trend is consistent with Eq. 3.5 and suggests that an extended transition in the \( \sigma_{\text{global}} - \varepsilon^p_{\text{global}} \) response correlates with a gradual increase in \( f_{\text{slipped}} \). At larger grain size \( (d = 300 \text{ nm}) \), the QCP simulations approach conventional crystal plasticity and \( f_{\text{slipped}} \) is predicted to increase rapidly regardless of whether a fitted Type A, B, or C distribution is employed.

The results underscore two key features of the QCP simulations. First, a 0.2% offset may capture \( \sigma_{\text{plateau}} \) at smaller \( \gamma_{\text{target}} \) \( (d = 300 \text{ nm}) \) but not at larger \( \gamma_{\text{target}} \) \( (d = 50 \text{ nm}) \). Second, the fraction \( f_{\text{slipped}} \) decreases for larger \( \gamma_{\text{target}} \) \( (d = 50 \text{ nm}) \). For example, at 0.2% strain, Fig. 3.9b shows \( f_{\text{slipped}} \sim 40\% \) for \( d = 50 \text{ nm} \) vs. >80% for \( d = 300 \text{ nm} \). This difference persists with increasing strain and suggests that materials with larger \( \gamma_{\text{target}} \) (smaller \( d \)) tend to localize plastic strain in fewer grains.
Figure 3.9: The fraction $f_{\text{slipped}}$ of slipped grains vs. $\epsilon_{\text{global}}^p$ corresponding to the QCP results in Fig. 3.7. (a) Predictions for grain size $d = 50$ nm and $300$ nm using Type A, B, and C distributions and (b) for $d = 50$ nm, $150$ nm, and $300$ nm using the best fitting (Type C) distribution.

3.4 Concluding Remarks

In this chapter, the developed quantized crystal plasticity constitutive relation is implemented into polycrystal FEM model. Initial assessment of QCP relation is performed through an elementary model, which entails a uniform distribution of critical slip stress $\tau_{\text{crit}}$ and one slip increment $\gamma_{\text{target}}$ ($\sim 1/d$). Furthermore, an extended model with a grain-to-grain variation in $\tau_{\text{crit}}$ is investigated on the purpose of reproducing the monotonic stress-strain characteristics of NC and UFG Ni. Several observations are made:
- A large $\gamma_{\text{target}}$ value (corresponding to small grain size $d$) produces violent stress redistributions (ref. to Fig 3.3).

- If the critical stress for slip events is independent of $d$, such violent redistributions cause polycrystalline strength to decrease (not increase) with decreasing $d$ (Fig. 3.5).

- An enhanced flow stress at small grain size is required to ensure a positive driving force to expand the slip event across a grain. In particular, an Eshelby construction shows that the minimum stress for slip events scales as $\tau_{\text{crit(min)}} \sim 1/d$ (Eq. 2.15).

- QCP simulations reproduce the unique extended plastic transition strain characteristic of NC Ni, provided a very asymmetric distribution of $\tau_{\text{crit}}$ is used (Type C, Fig. 3.8). In such cases, there is an abrupt transition stress at which the probability of slip events rapidly increases. This transition stress is estimated as 160 MPa for $d = 150$ nm and 280 MPa for $d = 50$ nm, based on a fit to $\sigma_{\text{global}} - \sigma_{\text{local}}$ data for NC and UFG Ni. Further, the ratio of $\tau_{\text{crit(max)}}/\tau_{\text{crit(min)}}$ present in these distributions is much larger than that due to experimental variation in $d$, assuming $\tau_{\text{crit(max)}}/\tau_{\text{crit(min)}} \sim d_{\text{max}}/d_{\text{min}}$ (Eqs. 2.8 and 2.15). This indicates that the heterogeneity in $\tau_{\text{crit}}$ cannot be fully ascribed to grain size inhomogeneity.

- An analytic model is developed to estimate the apparent hardening of NC metals in the early stage of plastic deformation, when slip events are isolated. An abrupt transition stress is shown to reduce apparent hardening and promote an extended plastic transition strain (Eq. 3.5).
QCP simulations predict that NC metals are more prone to localized plastic strain (Fig. 3.9). This stems, in part, from the more violent stress redistributions associated with slip events in NC metals.

QCP simulations predict that NC metals have a smaller fraction of plastically deformed grains for a given global plastic strain, compared to larger grain counterparts. For example, for Ni with $d = 50$ nm, only 40% of grains are predicted to have slipped at 0.2% plastic strain (Fig. 3.9). This is similar to predictions by Saada [39, 40].

The quantized nature of the QCP model can be assessed in light of MD simulations. The QCP model adopts a key feature from MD simulations [10] that grain-averaged shear strain can undergo abrupt changes associated with large slip events. However, only the largest possible slip events in a grain are modeled by the QCP approach, even though MD simulations show numerous smaller slip events.

The positive driving force condition, i.e. propagation condition, identified in the QCP model should be viewed as one of several sources of size-dependent strengthening. For example, the MD simulations that motivated the QCP approach show that for NC Al with a mean grain diameter of ~12 nm, the grain-averaged resolved shear stress for slip events is ~700 MPa, for $\dot{\varepsilon} = 10^6$/s and $T = 300$ K. During a slip event, this stress might drop by as much as ~400 MPa but even then, the positive driving force condition is not violated (see Fig. 2.1). Instead, dislocation slip process are controlled by the pinning and depinning of dislocation loops at grain boundaries [17], and this might produce size-dependent strengthening due to a smaller pinning distance at smaller $d$. MD simulations have not explicitly confirmed this scaling due to the limited range of possible grain sizes,
nor have MD estimates been extrapolated to experimentally relevant strain rates [9]. Another source of size-dependent strengthening might originate from the expense of depositing dislocation content at grain boundaries [50, 78], which can be framed in terms of the equilibrium angle made by an expanding loop as it intersects a grain boundary [101].

The QCP simulations suggest a unique feature of NC metals: an abrupt transition stress at which the probability of slip events rapidly increases. A hypothesis is that embryonic loops may be able to form at grain boundary stress concentrations, but these loops are not able to expand fully across a grain because a size-dependent propagation criterion is not yet satisfied. This size-dependent propagation is defined in the QCP model as originating from a positive stress (driving force) requirement. However, it could be related to a pinning/depinning phenomenon or to depositing dislocation content at grain boundaries. At larger grain sizes, the propagation criterion may be trivial to satisfy, so that slip events are controlled by a nucleation condition. At smaller (NC) grain sizes, the reverse may occur, meaning that the nucleation condition may be trivial to satisfy at many grain boundary sites, but the expansion condition is not. A detailed examination of this hypothesis is beyond the scope of the present QCP analysis.

Finally, the several limitations of QCP/FE simulations are worth mentioning. First, the FEM implementation considers a simple array of uniform cuboidal grains and thus cannot capture the complex nature of stress concentrations associated with realistic triple junctions. Second, QCP model does not include deformation-induced grain boundary migration or grain boundary slip that is observed in MD simulations [111, 112]. Third, the grain-averaged nature of the model does not consider many of these detailed aspects,
such as precise grain shapes, detailed distributions of grain boundary nucleation sites within grains, and grain size distribution. Nevertheless, QCP captures the intrinsic feature that on a grain-averaged scale, the plastic strain induced by single slip events increases with decreasing grain size, and this leads to more violent redistributions of stress associated with percolation of plasticity through the polycrystal. Such violent stress redistribution induces highly heterogeneous internal stress in NC metals. The next chapter will investigate the effect of the internal stress on NC mechanical behaviors.
CHAPTER 4:
QCP PREDICTIONS OF CYCLIC RESPONSE OF NC Ni

4.1 Experimental Motivation

4.1.1 Observations from Experiments and Atomistic Simulations

Under cyclic loading, NC metals exhibit new distinctive mechanical features compared to their CG counterparts [41-43]. Recent reports for NC Al and Au thin films show that more than 40% of plastic deformation is recoverable upon unloading (see Section 1.2.4)[41]. This extraordinary plastic recovery has been ascribed to large residual stress state, which is assumed to be originated from variation in microstructure e.g. grain size [44].

In addition, in situ X-ray diffraction studies reveal that peak broadening (represented by FWHM) is fully reversible upon unloading during room temperature deformation of ED NC Ni (d = 30 nm) [38, 63, 113]. This phenomenon illustrates that the inhomogeneous inter-granular stress induced during loading is entirely removed upon unloading. Consequently, no permanent dislocation network, which will induce internal stress heterogeneity, is expected to form during deformation cycle. Further, strain-dip tests performed on those samples show a very large value of internal stress, which is on the order of GPa; whereas for their CG counterparts, the internal stress is only ~MPa [114].
These tests also reveal negative creep when a stress drop exceeds 30% of its internal stress [114], which seems to suggest that dislocations run easily back into GBs upon unloading.

The MD simulations on NC Al with a mean grain size $d = 20$ nm have shown in details deformation activities in the course of loading-unloading cycle at room temperature [115]. In the loading stage, dislocations nucleate from GBs, and can traverse whole grains and reach neighboring GBs. Stacking faults, left behind partial dislocations, have been observed in grain interior. During unloading, traction of dislocation from opposite GBs reduces dislocation density, stacking faults, and thus lowers residual stresses. Reverse process by GB activities, such as GB sliding and diffusion, is also identified. However it is dislocation process that contributes to majority of the plastic recovery. In particular, at room temperature more than 90% of the recovery comes from dislocation activities. In addition, the kinetics of dislocation depinning from GBs is proposed to explain the recovery process upon unloading. The depinning process, identified by MD simulations during loading, promotes this assumption. However, MD simulations have not confirmed this picture yet due to restrictions in simulation time. The dislocation which is pinned during the forward propagation may not necessary be pinned again, since GB structures continue evolving. But an increase in depinning stress with straining is expected as a result of increase in pinning point density in GBs. Those deformation details from MD simulations can provide guanidine for modeling at higher level, such as QCP simulations [15, 17].

In this chapter, the Quantized Crystal Plasticity (QCP) model is further developed to incorporate reverse slip [116]. This enables an investigation of the combined
contribution from plastic pre-deformation ($\varepsilon_{\text{pre}}^p$), *grain-to-grain* distribution of critical strengths ($\tau_c$), and bias ($\tau_{\text{bias}}$) in the critical shear stress for forward vs. reverse slip. A premise is that these quantities can be determined via calibration of the QCP simulations to experimental $\sigma$-$\varepsilon$ data. An outcome is that both monotonic and cyclic $\sigma$-$\varepsilon$ data are needed to determine $\varepsilon_{\text{pre}}^p$, $\tau_c$, and $\tau_{\text{bias}}$. For ED NC Ni (30 nm), $\tau_c$ is described best by an asymmetric distribution, a noticeable $\tau_{\text{bias}}$ exists at small plastic strain, and a residual stress state exists prior to loading. The calibrated QCP model predicts enhanced plastic recovery upon unloading, due to a very heterogeneous stress state. In special cases, deformation is observed to reduce the residual stress state, as suggested by recent X-ray diffraction studies [38].

4.1.2 Experimental Sample Characterization

The QCP predictions are compared to data for ED NC Ni from two sources. The first is monotonic tensile data by Ebrahimi *et al.* [110] on material with a mean grain size, $d_{\text{mean}} = 50$ nm in Sections 4.2 and 4.3. The second is monotonic and cyclic uniaxial data by Van Swygenhoven *et al.* on material purchased from Goodfellow [38] with $d_{\text{mean}} = 30$ nm in Sections 4.3-4.4. The QCP application to Ebrahimi’s data has been described in details in Chapter 3. Here in first step, this fitted model is further investigated under cyclic loading in order to address the substantial effect of pre-strain, and the corresponding residual stress state. But due to the lack of cyclic data, the predicted cyclic behavior on Ebrahimi’s work is unable to be verified experimentally. Then the second
step is to calibrate the QCP model to Van Swygenhoven’s data with all the parametric characteristics identified by the first step.

The ED NC Ni used by Van Swygenhoven features with pore-free and relative narrow grain size distribution. Figure 4.1 shows the bright field TEM image of ED NC Ni and its corresponding grain size distribution. The number averaged grain size is 26nm with a standard deviation of 14 nm, which is obtained by analyzing 365 grains. The grain size distribution is narrow. For instance, only five grains have a grain size between 60 and 80 nm and two grains between 80 and 100 nm. The volume averaged grain size of the sample is 33.5nm with a standard deviation of 42nm. In QCP simulations, an average grain size of 30 nm is adopted.

Figure 4.1: (a) Bright field TEM image of ED NC Ni. (b) The corresponding grain size distribution [117].
4.2 QCP/FE Simulations with Reverse Slip

4.2.1 QCP Model with Reverse Slip

The Quantized Crystal Plasticity (QCP) constitutive relation is extended to include reverse slip during cyclic loading. **Table 4.1** summarizes the relevant notation. The nature of the QCP model is that on a grain-average scale, the plastic shear strain produced by an event on some slip system \( \alpha \) with slip plane normal \( \mathbf{n} \) and slip direction \( \mathbf{s} \) is

\[
\gamma_{\text{ns}}^{p(\alpha)} = q' \gamma_{\text{target}}
\]  

(4.1)

The coefficient \( q \) takes on integer values and \( \gamma_{\text{target}} \sim b/(\text{grain size}) \). A *forward* event increases the magnitude of \( q \) and is activated when the resolved shear stress satisfies

\[
\tau_{\text{ns}}^{(\alpha)} \cdot \text{sign}(\gamma_{\text{ns}}^{p(\alpha)}) \geq \tau_c.
\]

Likewise, a *backward* event decreases the magnitude of \( q \) and is activated when

\[
\tau_{\text{ns}}^{(\alpha)} \cdot \text{sign}(\gamma_{\text{ns}}^{p(\alpha)}) \leq \tau_{c,b}.
\]

The critical stress for forward vs. backward events may differ by an amount \( \tau_{\text{bias}} = \tau_c - \tau_{c,b} \). Upon activation during propagation, the plastic strain increments at a rate \( \dot{\gamma}_0 \) until \( q \) reaches an integer value, even if the activation condition is not satisfied continuously during the propagation process, due to stress redistribution. Eq. (4.1) and the activation conditions are implemented numerically as described in Section 2.2.
Table 4. 1: Notation in Chapter 4

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \dot{\varepsilon}_{\text{global}} )</td>
<td>macroscopic strain rate along ( z )-axis</td>
</tr>
<tr>
<td>( \sigma_{\text{global}} )</td>
<td>macroscopic stress along ( z )-axis</td>
</tr>
<tr>
<td>( \sigma_0 )</td>
<td>macroscopic stress for onset of the first slip event</td>
</tr>
<tr>
<td>( \tau^{(\alpha)}_{\text{ns}} )</td>
<td>local resolved shear stress, slip system ( \alpha )</td>
</tr>
<tr>
<td>( \gamma^{p(\alpha)}_{\text{ns}} )</td>
<td>local plastic shear strain, slip system ( \alpha )</td>
</tr>
<tr>
<td>( \gamma_{\text{target}} )</td>
<td>local shear strain produced by a slip event</td>
</tr>
<tr>
<td>( \dot{\gamma}_0 )</td>
<td>local plastic shear strain rate during a slip event</td>
</tr>
<tr>
<td>( \tau_c )</td>
<td>local critical resolved shear stress for a forward slip event</td>
</tr>
<tr>
<td>( \tau_{c,b} )</td>
<td>local critical resolved shear stress for a backward slip event</td>
</tr>
<tr>
<td>( \tau_{\text{bias}} )</td>
<td>( \tau_c - \tau_{c,b} )</td>
</tr>
<tr>
<td>( \tau_{\text{eff}} )</td>
<td>minimum additional ( \tau_{\text{ns}} ) to initiate a slip event</td>
</tr>
<tr>
<td>( \Delta \tau )</td>
<td>jump in local resolved shear due to a slip event</td>
</tr>
<tr>
<td>( \varepsilon_{\text{pre}}^p )</td>
<td>uniaxial plastic pre-strain along ( z )-axis prior to tensile testing</td>
</tr>
</tbody>
</table>
4.2.2 FEM Representation

A polycrystal is modeled by a $10 \times 10 \times 10$ array of 3D brick finite elements (C3D8) using ABAQUS software [91] as before. The eight material points in an element are assigned the same initial material properties: crystallographic orientation, $\gamma_{\text{target}}$, $\tau_c$, particularly $\tau_{c,bs}$, the critical stress for a backward slip event. Uniaxial loading and unloading is imposed via a macroscopic strain rate ($\dot{\varepsilon}_{\text{global}}^{p} = \pm 10^{-3}/s$) along the $z$-direction, with free surface boundary conditions on the $\pm x$ and $\pm y$ faces.

4.2.3 Material Parameters

The material parameters are consistent with local elastic behavior governed by anisotropic elastic constants of Ni, *i.e.* $C_{11} = 246.5$, $C_{12} = 147.3$, and $C_{44} = 124.7$ GPa respectively and random grain orientations consistent with an untextured polycrystal as in Chapter 3. Two distributions are considered for the non-uniform *grain-to-grain* values of $\tau_c$: an asymmetric distribution described by a Gamma function (Eq. 4.5a, Table 4.2) and a normal distribution (Eq. 4.5b, Table 4.2). Each of these has two free parameters that control $\tau_{c,\text{mean}}$ and $\tau_{c,\text{min}}$. In general, the most severe slip events are modeled by selecting $\gamma_{\text{target}} = \min(1.2 \frac{b}{d}, \frac{\tau_c}{60}$ GPa), where the Burgers vector magnitude $b = 0.25$ nm and a uniform grain size $d = 30$ or 50 nm is applied. The first expression for $\gamma_{\text{target}}$ corresponds to slip of a cubic grain of edge length $d$ on a glide plane through the center. The second expression is an estimate of the largest $\gamma_{\text{target}}$ that can be accommodated without reversing the sign of $\tau_{n}^{(\alpha)}$ during expansion of the loop across the grain. This is termed the *positive*
driving force or propagation condition [103]. The first expression normally applies, but there is typically a subset of ~15% grains with smaller $\tau_c$ values for which the second expression applies. In addition, a variety of $\tau_{bias}$ values are considered.

4.2.4 Computation Parameters

The primary computational parameters are the local strain rate $\dot{\gamma}_0 = 2 \cdot 10^{-2}$/s contributed by an active slip system and the macroscopic strain rate $\dot{\varepsilon}_{\text{global}}^p = 1 \cdot 10^{-3}$/s. The former is an order of magnitude larger so that deformation due to local slip is viewed as relatively instantaneous compared to macroscopic deformation. A maximum time increment $\Delta t = 2 \cdot 10^{-2}$s is specified to ensure that a quantized slip event is captured in several time steps. Prior studies [103] demonstrate that a 1000-grain model is sufficiently large so that the macro $\sigma$-$\varepsilon$ response is independent of the number of grains. A typical analysis with 1000 grains for 100 time steps requires ~ 4.5 CPU hours with the Glenn system at the Ohio Supper Computer Center [118].

4.2.5 Local Stress-Strain Evolution with Reverse Slip

Figure 4.2 shows the evolution of $\gamma_{ns}^{P(\alpha)}$ and (MPa) vs. time (s) for a specific slip system $\alpha$ at material point of a grain with $\gamma_{\text{target}} = 6 \cdot 10^{-3}$. Three periods are included: $\dot{\varepsilon}_{\text{global}}^p = +10^{-3}$/s from 0 to 5s; $\dot{\varepsilon}_{\text{global}}^p = -10^{-3}$/s from 5 to 10s; and $\dot{\varepsilon}_{\text{global}}^p = +10^{-3}$/s from 10 to 16s.
During the first period, $\tau_c$ is reached twice, so that $q$ increments from 0 to 1 and then 1 to 2. During the second period, $-\tau_{c,b}$ is reached twice, producing $q = 0$. In the third period, two forward events occur, producing $q = 2$. Thus, $\gamma_{ns}^{p(\alpha)}$ is quantized in units of $\gamma_{\text{target}}$.

Note that $\tau_c = 428.4$ MPa vs. $\tau_{c,b} = 228.4$ MPa, so that $\tau_{\text{bias}} = 200$ MPa. In Fig. 4.2, $\tau_{ns}^{(\alpha)}$ sometimes increases in a nonlinear fashion due to stress redistribution arising from slip events in neighboring grains. About 15 time steps (corresponding to 0.3 s) are required to fully implement a slip event, thereby ensuring sufficient computational resolution and shear jump accuracy.
Figure 4. 2: Evolution of the local plastic strain $\gamma^{(\alpha)}$ and local resolved shear stress $\tau^{(\alpha)}$ on a specific slip system $\alpha$ in an arbitrary interior grain. The applied global strain rate reverses sign at 5s and 10s.
4.3 Calibration of QCP/FE Model

4.3.1 Effect of Pre-strain

QCP simulations show that plastic predeformation (\(e_{\text{pre}}^p\)) alters the macro \(\sigma-\varepsilon\) response primarily at small strain (< 1%) but much less at larger strain. Figure 4.3a shows the results for \(e_{\text{pre}}^p = -3\%\), 0, and 3\%, assuming an asymmetric \(\tau_c\) distribution (Fig. 4.3b) and \(\tau_{\text{bias}} = 0\). Square symbols in Fig. 4.3a are the experimental results for electrodeposited NC Ni with \(d_{\text{mean}} = 50\) nm from Ebrahimi [110]. Pre-compression produces a “rounded”, gradual approach to the plateau stress (\(\sigma_{\text{plateau}}\)) while pre-tension generates more abrupt yield. A quantitative assessment is that the width of the transition is \(\Delta \varepsilon^p \approx 1.6\%\) for \(e_{\text{pre}}^p = -3\%\) vs. 0.05\% for \(e_{\text{pre}}^p = 3\%\), where \(\Delta \varepsilon^p\) is the plastic strain increment over which the flow stress increases from an initial value \(\sigma_0\) to approximately 0.9 \(\sigma_{\text{plateau}}\).

Fig. 4.3a also shows that the effect of pre-deformation diminishes with increasing strain. All cases approach \(\sigma_{\text{plateau}} = 1200\) MPa, regardless of \(e_{\text{pre}}^p\). This suggests that deformation history is ‘forgotten’ as plastic flow develops. An important caveat, however, is that the \(\tau_c\) distribution does not evolve with plastic deformation in these simulations. In reality, the \(\tau_c\) distribution may change as the density and strength of grain boundary pinning sites evolve with deformation.

Pre-deformation effect can be viewed in terms of the residual stress state it induces and how this stress state biases subsequent yield. Figure 4.3c.d show the distribution of.
defined as the additional resolved shear stress to initiate a slip event on the most favored slip system at an integration point. Among the $\alpha = 1$ to 12 systems, the most favored one renders $[\tau_c - \text{sign}(S_z^{(\alpha)}) \cdot \tau_{ns}^{(\alpha)}] / S_z^{(\alpha)}$ a minimum, where $S_z^{(\alpha)} = (n_z \cdot s_z)^{(\alpha)}$ is the Schmid factor for tension along the z-axis. Pre-compression tends to spread out the $\tau_{c\text{eff}}$ distribution, so that soft region $B$ and hard region $C$ in the original distribution (Fig. 4.3b) shift respectively to even softer $B'$ and harder $C'$ (Fig. 4.3c). This is consistent with residual tension ($\sigma_z > 0$) for region $B'$ and residual compression for $C'$. The grains in region $B'$ readily yield as the tensile test commences because $\tau_{c\text{eff}}$ is so small. The analytic model (ref. Section 3.2.3) demonstrates that the resulting “rounded” $\sigma$-$\epsilon$ behavior stems from the abrupt rise in the distribution at $B'$ (Eq. 3.5). Alternately, pre-tension produces a more symmetric $\tau_{c\text{eff}}$ distribution (Fig. 4.3d) that is associated with a less rounded $\sigma$-$\epsilon$ response. In particular, regions $B$ and $C$ in the original distribution (Fig. 4.3b) respectively shift to relatively less soft regions $B''$ and less hard $C''$ (Fig. 4.3d). Thus, pre-compression and pre-tension produce opposite shifts in regions $B$ and $C$, and generate very different $\tau_{c\text{eff}}$ distributions (asymmetric vs. symmetric).
Figure 4.3: (a) The tensile stress-plastic strain response of an untextured polycrystal with $\tau_c$ = asymmetric distribution, $\varepsilon^p_{\text{pre}} = -3\%$, 0, and 3%, and $\tau_{\text{bias}} = 0$. Square symbols denote the experimental data for electrodeposited NC Ni with $d_{\text{mean}} = 50$ nm [110]; (b) the asymmetric $\tau_c$ distribution; (c) $\tau_c^{\text{eff}}$ distribution after plastic pre-deformation $\varepsilon^p_{\text{pre}} = -3\%$ and (d) $\varepsilon^p_{\text{pre}} = 3\%$. 
4.3.2 Non-uniqueness of Monotonic Response

Similar monotonic $\sigma$-$\varepsilon$ responses can be achieved with more than one combination of $(\tau_c, \varepsilon_{\text{pre}}^p)$. Figure 4.4a shows that the asymmetric $\tau_c$ distribution (Fig. 4.3b) with $\varepsilon_{\text{pre}}^p = 0$ (Case $A$: solid curve) and a symmetric $\tau_c$ distribution (Fig. 4.4b) with $\varepsilon_{\text{pre}}^p = -3\%$ (Case $S$: dashed curve) produce similar $\sigma$-$\varepsilon$ responses. This observation, coupled with the discussion in Section 4.3.1, suggests that both combinations of $(\tau_c, \varepsilon_{\text{pre}}^p)$ should produce similar $\tau_c^{\text{eff}}$ distributions. A comparison of Figs. 4.3b (Case $A$) and 4c (Case $S$) confirms similar $\tau_c^{\text{eff}}$ distributions. Thus, a unique combination $(\tau_c, \varepsilon_{\text{pre}}^p)$ cannot be deduced from monotonic $\sigma$-$\varepsilon$ data alone.
Figure 4.4: (a) The tensile stress-plastic strain response of an untexured polycrystal for Case A: $(\tau_c, \varepsilon_{p_{\text{pre}}}) = (\text{asym}, 0)$ and Case S: $(\tau_c, \varepsilon_{p_{\text{pre}}}) = (\text{sym}, -3\%)$. $\tau_{\text{bias}} = 0$; (b) symmetric $\tau_c$ distribution; and (c) $\tau_c^{\text{eff}}$ distribution after plastic pre-deformation $\varepsilon_{p_{\text{pre}}} = -3\%$. 
4.3.3 Uniqueness from Cyclic Response

Cyclic data at both large and small strain enables unique combinations of ($\tau_c$, $\varepsilon_{\text{pre}}^p$) to be deduced, as well as estimates of $\tau_{\text{bias}}$. Figure 4.5a shows the predicted $\sigma$-\(\varepsilon\) response when strained well into the plateau region ($>2.5\%$), then cycled. The hysteretic $\sigma$-\(\varepsilon^p\) response is relatively independent of $\varepsilon_{\text{global}}^p$ and $\varepsilon_{\text{pre}}^p$. However, it is very dependent on the $\tau_c$ distribution and $\tau_{\text{bias}}$. In particular, Case A ($\tau_c =$ asymmetric, $\varepsilon_{\text{pre}}^p = 0$, $\tau_{\text{bias}} = 0$) has a very pronounced hysteretic width ($\Delta\varepsilon^p = 0.9\%$ at $\sigma = 600\text{MPa}$) while Case S ($\tau_c =$ symmetric, $\varepsilon_{\text{pre}}^p = -3\%$, $\tau_{\text{bias}} = 0$) has negligible width ($\Delta\varepsilon^p \sim 0$).

Some insight is gained from the $\tau_c$ and $\tau_c^{\text{eff}}$ distributions. During monotonic straining along the plateau, $\tau$ is expected to reach $\tau_c$ in numerous grains, so that the $\tau$ distribution mimics the asymmetric $\tau_c$ distribution (Fig. 4.3b) for Case A and symmetric $\tau_c$ distribution (Fig. 4.4b) for Case S. Thus, the polycrystalline system reaches criticality along the plateau [119]. Upon unloading to points $A'$ and $S'$ in Fig. 4.5a, the asymmetric distribution offers a large fraction of weak (small $\tau_c$) sites at the onset of reverse slip and accordingly, there is a large amount of reverse slip (region $A''$, Fig. 4.5a). In contrast, the symmetric distribution has a smaller fraction of weak sites and consequently less reverse slip (Region $S''$, Fig. 4.5a). Upon reloading, the $\tau_c^{\text{eff}}$ distributions (Figs. 4.5b,c) apply. The distribution for Case A has a larger number of sites at smaller $\tau_c^{\text{eff}}$, so that forward slip is more pronounced compared to Case S.
A general characteristic of the asymmetric distribution (Fig. 4.3b) is that a relatively large fraction of weak (small $\tau_c$) sites are balanced by a relatively small fraction of strong sites. These weak sites supply large amounts of forward and backward slip events during cycling, while strong sites provide heterogeneous internal stresses as the driving force. In principle, symmetric (normal) distributions other than Fig. 4.4b can be considered. However, the distributions cannot be shifted or widened arbitrarily, since the mean controls the plateau stress (Eq. 4.3, Table 4.2) and the minimum controls the onset of yield and also must satisfy the positive driving force/propagation condition (Section 4.2.3).
Figure 4.5: (a) The cyclic stress-plastic strain response of an untextured polycrystal at large strain, for Case A \((\tau_C, \varepsilon_{p,\text{pre}}) = (\text{asym}, 0)\) and Case S \((\tau_C, \varepsilon_{p,\text{pre}}) = (\text{sym}, -3\%)\), \(\tau_{\text{bias}} = 0\); (b) \(\tau_c^{\text{eff}}\) distribution for Case A, after unloading to pt. A'; (c) \(\tau_c^{\text{eff}}\) distribution for Case S, after unloading to pt. S'.
Figure 4.6 shows the corresponding outcomes when a smaller plastic strain ($\varepsilon_{\text{global}} \sim 0.3\%$) is imposed, followed by cycling. As before, Case $A$ ($\tau_c = \text{asymmetric}, \varepsilon_{\text{pre}}^p = 0, \tau_{\text{bias}} = 0$) has a larger hysteretic width ($\Delta\varepsilon^p = 0.02\%$ at $\sigma = 425\text{MPa}$) than Case $S$ ($\tau_c = \text{symmetric}, \varepsilon_{\text{pre}}^p = -3\%, \tau_{\text{bias}} = 0$). Compared to Fig. 4.5a, the hysteretic widths at this smaller strain are an order of magnitude or even smaller.

![Figure 4.6](image)

Figure 4.6: (a) The cyclic stress-plastic strain response of an untextured polycrystal at small strain, for (a) Case $A$: ($\tau_c, \varepsilon_{\text{pre}}^p$) = (asym, 0); and (b) Case $S$: ($\tau_c, \varepsilon_{\text{pre}}^p$) = (sym, $-3\%$). $\tau_{\text{bias}} = 0$. 

93
4.3.4 Application to ED NC Ni (d_{mean} \sim 30 \text{ nm})

The results in Sections 4.3.1-4.3.3 produce a formal, multiple-step procedure to calibrate the QCP simulations to monotonic and cyclic data. These steps are summarized in Table 4.2 and applied to NC Ni with a mean grain size of 30 nm [38]. The first four steps originate from the work in Chapter 3 [103]. First, the maximum stress jump $\Delta \tau$ (Fig. 4.2) associated with a slip event is estimated from the mean grain size $d_{\text{mean}}$, Burgers vector magnitude $b$, elastic shear modulus $\mu$, and Poisson’s ratio $\nu$ (Eq. 4.2, Table 4.2). Second, the mean of the critical strength distribution $\tau_{c,\text{mean}}$ is estimated from the plateau stress $\sigma_{\text{plateau}}$ and the polycrystalline average of the largest Schmid factor $\bar{S}_{z,\text{max}} (= 0.45$ for an untextured FCC polycrystal) in each grain (Eq. 4.3, Table 4.2). In addition, the minimum in the critical strength distribution $\tau_{c,\text{min}}$ is estimated from the initial polycrystalline yield strength $\sigma_0$ and the global maximum Schmid factor $S_{z,\text{max}} (= 0.5$) (Eq. 4.4, Table 4.2). Third, $\tau_{c,\text{mean}}$ and $\tau_{c,\text{min}}$ are used to generate two candidate $\tau_c$ distributions: an asymmetric Gamma distribution and a normal distribution (Eqs. 4.5a and 4.5b, Table 4.2). Fourth, the quantized jump $\gamma_{\text{target}}$ for each grain is specified as discussed in Section 4.2.3 and summarized in Table 4.2, Eq. 4.6.

The remaining steps 5 and 6 identify the best distribution (asymmetric or normal) and optimal values of $\tau_{\text{bias}}$ and $\epsilon_{\text{pre}}^p$. Step 5 determines the best fit to cyclic $\sigma$-$\epsilon^p$ response. Figure 4.7 shows the QCP results (solid curve) that best fit the experimental data (dotted curve). The asymmetric $\tau_c$ distribution successfully captures the hysteretic widths and also the magnitude of reverse strain at unloading points 1 to 4 (Fig. 4.7). However, three
values of $\tau_{\text{bias}}$ are required: 90 MPa for $\varepsilon_{\text{p,global}} < 0.5\%$, 30 MPa for $0.5\% < \varepsilon_{\text{p,global}} < 1.5\%$, and 0 MPa for $\varepsilon_{\text{p,global}} > 1.5\%$. The normal distribution is unable to produce a sufficient hysteretic width and reverse slip, even for large $\tau_{\text{bias}}$. Finally, step 6 determines $\varepsilon_{\text{p,pre}} = -0.4\%$ as the best match to the initial yield and small strain monotonic $\sigma$-$\varepsilon$ behavior. This is generically consistent with experimental measurements of very inhomogeneous stress in ED NC Ni [114], and pre-existing compressive stress state of ED NC Ni [120].

Overall, the best fitting parameters are summarized in Table 4.3 and include modest adjustments to the analytic estimates from steps 1-4, obtained by iteration based on repeated QCP simulations.
Figure 4.7: Cyclic stress-plastic strain results for the best fitting QCP simulation (solid curve) vs. experimental data (dotted curve) for electrodeposited NC Ni with mean grain size \( d_{\text{mean}} = 30 \text{ nm} \). The simulation uses \( \tau_c = \text{asym.} \), \( \varepsilon_{\text{pre}}^p = -0.4\% \). \( \tau_{\text{bias}} = 0, 30, \) and \( 90 \text{ MPa} \) depending on the imposed global plastic strain. See Table 4.3 for parameters.
Table 4. 2: Multi-step Process to Calibrate QCP Simulations to Experimental Data

<table>
<thead>
<tr>
<th>Step</th>
<th>Quantity</th>
<th>Method to Calculate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\Delta \tau$</td>
<td>$\Delta \tau = -c_1 M b/d$, $c_1 = 1.44$, $M = \mu (7 - 5 \nu) / (15(1 - \nu))$ (4.2)</td>
</tr>
<tr>
<td>2a</td>
<td>$\tau_{c,\text{mean}}$</td>
<td>$(\tau_c - 0.5</td>
</tr>
<tr>
<td>2b</td>
<td>$\tau_{c,\text{min}}$</td>
<td>$\tau_{c,\text{min}} = \sigma_0 \bar{S}<em>{z,\text{max}}, \bar{S}</em>{z,\text{max}} = 0.5$ (4)</td>
</tr>
<tr>
<td>3</td>
<td>$\tau_c$ distrib. candidates</td>
<td>$P_d(\tau, k, \theta) = (\tau - \tau_{c,\text{min}})^{-1} (\Gamma(k) \theta)^{-1} \exp(- (\tau - \tau_{c,\text{min}})/\theta)$ (4.5a) $k = 1$, $\theta = \tau_{c,\text{mean}} - \tau_{c,\text{min}}$ $P_s(\tau, \tau_{c,\text{mean}}, \sigma) = (\sqrt{2 \pi} \sigma)^{-1} \exp(- (\tau - \tau_{c,\text{mean}})^2 / 2 \sigma^2), \sigma = \theta / 3$ (4.5b)</td>
</tr>
<tr>
<td>4</td>
<td>$\gamma_{\text{target}}$</td>
<td>$\gamma_{\text{target}} = \min(\tau_c / c_2 M, c_2 b/d), c_2 = 1.2, M = \text{(see 4.2)}$ (4.6)</td>
</tr>
<tr>
<td>5a</td>
<td>$\tau_c$ distrib.</td>
<td>best fit to cyclic data at large strain</td>
</tr>
<tr>
<td>5b</td>
<td>$\tau_{\text{bias}}$</td>
<td>best fit to cyclic data at large and small strain</td>
</tr>
<tr>
<td>6</td>
<td>$\varepsilon_{\text{pre}}$</td>
<td>best fit to tensile data at small strain</td>
</tr>
</tbody>
</table>

Table 4. 3: Best Fit of QCP Parameters to NC Ni ($d_{\text{mean}} = 30$ nm)

<table>
<thead>
<tr>
<th>$d$</th>
<th>30 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\tau_c$</td>
<td>asymmetric distribution (Eq. 4.5a)</td>
</tr>
<tr>
<td></td>
<td>$\tau_{c,\text{mean}} = 1050$ MPa, $\tau_{c,\text{min}} = 210$ MPa</td>
</tr>
</tbody>
</table>

| $\tau_{\text{bias}}$ | 90 MPa, for $\varepsilon_{\text{global}}^{\text{p}} < 0.5\%$; |
| $\tau_{\text{bias}}$ | 30 MPa, for $0.5\% < \varepsilon_{\text{global}}^{\text{p}} < 1.5\%$; |
| $\tau_{\text{bias}}$ | 0 MPa, for $\varepsilon_{\text{global}}^{\text{p}} > 1.5\%$ |
| $\gamma_{\text{target}}$ | $3.5 \cdot 10^{-3}$ to $1.0 \cdot 10^{-2}$ (Eq. 4.6) |
4.4 Prediction of the Calibrated QCP/FE Model

4.4.1 Fraction of the Grains Undergoing Forward and Reverse Slip

The QCP simulations are capable of providing statistics on the nature of forward vs. backward slip. Table 4.4 shows the QCP simulation results on plastic recovery, as calibrated to the ED NC (30 nm) Ni data in Fig. 4.7. The results for reverse plastic strain $\varepsilon_{rev}^p$ and the ratio $\varepsilon_{rev}^p / \varepsilon^p$ of reverse plastic strain to imposed plastic strain are within 10% of experimental values. A qualification is that the $\varepsilon_{rev}^p$ values adopted from experimental data include the time-dependent relaxation strain obtained after unloading to 10 MPa and holding for 20 min [38]. In contrast, the $\varepsilon_{rev}^p$ values from simulations are time-independent and are simply the instantaneous values upon unloading to 0 MPa. The experimental data (and simulations) show that $\varepsilon_{rev}^p$ increases to a peak of 0.28% and $\varepsilon_{rev}^p / \varepsilon^p$ decreases to a minimum of 13% as the stress $\sigma_u$ at unload increases to $\sigma_{plateau}$. The simulations predict the fraction $f_f$ of forward slipping grains to reach 87% at $\sigma_{plateau}$. Thus, there is a subset (13%) of grains that remain elastic even at large strain ($\varepsilon^p = 2\%$). After unloading, the fraction $f_b/f_f$ of backward-slipped to forward-slipped grains decreases from $>60\%$ at small $\varepsilon^p$ to $\sim37\%$ at large $\varepsilon^p$. 
**Table 4.4: Plastic Recovery in NC Ni ($d_{\text{mean}} = 30 \text{ nm}$)**

<table>
<thead>
<tr>
<th>$\sigma_u$ (MPa)</th>
<th>$\varepsilon_{\text{rev}}^p$</th>
<th>$\varepsilon_{\text{rev}}^p/\varepsilon^p$</th>
<th>$f_f^*$</th>
<th>$f_b^{**}$</th>
<th>$f_b/f_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 800</td>
<td>0.0007</td>
<td>40%</td>
<td>33%</td>
<td>20%</td>
<td>62%</td>
</tr>
<tr>
<td>2 1100</td>
<td>0.0013</td>
<td>33%</td>
<td>45%</td>
<td>27%</td>
<td>60%</td>
</tr>
<tr>
<td>3 1400</td>
<td>0.0020</td>
<td>20%</td>
<td>70%</td>
<td>32%</td>
<td>45%</td>
</tr>
<tr>
<td>4 1600</td>
<td>0.0028</td>
<td>13%</td>
<td>87%</td>
<td>32%</td>
<td>37%</td>
</tr>
</tbody>
</table>

* $f_f$: fraction of slipped grains at $\sigma = \sigma_u$

** $f_b$: fraction of backward slipped grains upon unloading

### 4.4.2 Characteristic of Grains with Backward Slip

Grains with backward slip tend to have a relatively small $\tau_c$ and a relatively large number of forward slip events compared to their neighbors. **Figure 4.8a** shows the number of forward slip events $q_f$ (Eq. 4.1) in each grain at $\sigma_{\text{global}} = 800 \text{ MPa}$ (pt. 1', Fig. 4.7). The plastically deformed grains are mainly isolated and surrounded by elastically deformed grains. **Figure 4.8b** shows the result upon unloading (pt. 1, Fig. 4.7). During unloading, elastic-only grains attempt to return to their original dimension, driving 62% of plastically deformed grains to slip backward (Table 4.4). This leaves a small subset of grains with a non-zero $q_f$ after unloading.

The QCP simulations reveal the nature of grains that slip backward. **Figure 4.8c** displays the number of backward slip events $q_b$ in grain $i$ vs. $\tau_c$ for grain $i$, for $i = 1$ to 1000 grains in the polycrystal, during unloading from $\sim 800 \text{ MPa}$. The results show that backward
slip tends to occur in grains with $\tau_c < 500$ MPa, and the number of backward slip events increases with decreasing $\tau_c$. Figure 4.8d displays $q_b$ vs. the ratio $(q^n_f/q_f)_{800 \text{ MPa}}$ of forward slip events per neighboring grain (on average) to forward slip events in grain $i$. The ratio is evaluated at $\sigma_{\text{global}} = 800$ MPa, just prior to unloading. The results show that number of backward slip events tends to be larger in grains with a relatively large forward slip (i.e., $q^n_f/q_f < 1$).
Figure 4. 8: Grain-to-grain distribution of the number of forward slip events $q_f$ obtained from QCP simulations for electrodeposited nanocrystalline Ni with mean grain size $d_{\text{mean}} = 30$ nm (see parameters in Table 3) at (a) $\sigma_{\text{global}} = 800$ MPa (pt. 1', Fig. 4.7); and (b) after unloading to $\sigma_{\text{global}} = 0$ MPa (pt. 1, Fig. 4.7); (c) number of backward slip events $q_{b,i}$ in grain $i$ during unloading from pts. 1’ to 1, vs. the critical resolved shear stress $\tau_{c,i}$ in grain $i$; (d) $q_{b,i}$ vs. the ratio $(q_{f,i}^n/q_{f,i})_{800\text{MPa}}$ of the average number of forward slip events in grains that border grain $i$ to the number of forward events in grain $i$. The ratio is evaluated at $\sigma_{\text{global}} = 800$ MPa (pt. 1’, Fig. 4.7).
4.4.3 Plastic Deformation and Residual Stress

Plastic deformation is capable of either reducing or enhancing the magnitude of inhomogeneity of residual stress. Figure 4.9 shows the predicted distribution of residual stress $\sigma_{z,\text{res}}$ at three unloaded states: just prior to tension testing, after unloading from $\sigma_{\text{global}} = 800$ MPa, and after unloading from $\sigma_{\text{global}} = 1400$ MPa. These correspond to states 0, 1, and 3 in Fig. 4.7. The state 0 distribution (Fig. 4.9a) is due to pre-deformation ($\varepsilon_{\text{pre}} = -0.4\%$). The state 1 distribution is very sharp, indicating that the residual stress is dramatically reduced if the sample is stressed to 800 MPa and unloaded. This is achieved on a local grain scale by reversing slip events produced by pre-deformation. The state 3 distributions is very broad, indicating that the residual stress is dramatically increased if the sample is further stressed (to 1400 MPa) and unloaded. Such a large stress induces more slip events, beyond that needed to reverse the effects of pre-deformation.
Figure 4. 9: The distribution of residual stress $\sigma_{z,\text{res}}$ at different states in Fig. 4.7: (a) initial state 0, prior to loading; (b) state 1, after unloading from $\sigma_{\text{global}} = 800$ MPa; and (c) state 3, after unloading from $\sigma_{\text{global}} = 1400$ MPa.
4.5 Concluding Remarks

In this chapter, the QCP model is further developed to incorporate reverse slip, and is implemented in FE simulations to study the monotonic as well as cyclic stress-strain responses of NC metals. Two new key parameters are included: backward slip ($\tau_{c,b}$), and the amount of plastic pre-deformation ($\varepsilon^p_{\text{pre}}$); in addition to quantized shear increment $\gamma_{\text{target}}$, and the grain-to-grain distribution of critical strength ($\tau_c$) to activate slip events. A principal outcome is a 6-step process (Table 4.2) by which to extract these quantities from experimental measurements of monotonic and cyclic $\sigma$-$\varepsilon$ response. This process is applied to data for ED Ni with a 30 nm mean grain size. Several observations are made:

- Plastic pre-deformation ($\varepsilon^p_{\text{pre}}$) induces a residual stress state that enhances or suppresses slip events during subsequent loading. This creates an effective distribution of critical strengths ($\tau^\text{eff}_c$) that can increase or decrease the macro yielding stress and make the elastic-plastic transition abrupt or extended.

- Similar $\tau^\text{eff}_c$ distributions and thus similar monotonic responses can be achieved by more than one combination of ($\tau_c$, $\varepsilon^p_{\text{pre}}$). Thus, $\tau_c$ and $\varepsilon^p_{\text{pre}}$ cannot be uniquely determined from monotonic $\sigma$-$\varepsilon$ data alone.

- A less redundant determination of $\tau_c$, $\varepsilon^p_{\text{pre}}$, and $\tau_{\text{bias}}$ can be made by fitting the QCP predictions to monotonic and cyclic $\sigma$-$\varepsilon$ at small and large strain.
- The best fit for ED Ni \((d_{\text{mean}} = 30 \text{ nm})\) has a truncated (asymmetric) \(\tau_c\) distribution, \(\varepsilon_{\text{pre}}^p = -0.4\%\), and \(\tau_{\text{bias}} = 90 \text{ MPa} \) at small strain and 0 MPa at large strain. The asymmetric distribution is consistent with an abrupt onset of slip in a large fraction of grains at a critical resolved shear stress \(\sim 1/\text{grain size}\). The bias is consistent with backward slip requiring a smaller mechanical driving force (stress) than forward slip.

- The simulations capture the large reversible deformation observed in experiments, and predict reverse slip to occur in relatively soft grains having smaller \(\tau_c\) and more slip events than their neighbors.

- The QCP simulations predict that quantized slip often induces very inhomogeneous stress states. These inhomogeneous states store large elastic energy that can drive reversible deformation.

- The QCP simulations also show that inhomogeneous stress induced by prior deformation can be reduced, in principle, via subsequent deformation.

Despite the capacity of the QCP simulations to capture monotonic and cyclic \(\sigma-\varepsilon\) data, there are important qualifications. First, the distribution of critical strengths \((\tau_c)\) is quite simple. It does not evolve with deformation and all slip systems within a grain are assumed to have the same \(\tau_c\). In reality, the nature of grain boundaries (and thus \(\tau_c\)) is expected to evolve with deformation. This is reflected, in part, by a fitted value of \(\tau_{\text{bias}}\) that evolves from 90 MPa at small strain to 0 MPa at large strain. The increasing difficulty for reverse slip consists with the increasing density of dislocation pinning site at GBs with deformation. Second, one cubic element is employed to represent each grain so that specific grain geometries, constraints from neighboring grains, and stress...
concentrations from localized slip are captured in a grain-average sense. Third, only the largest slip events are modeled in these simulations, leaving out numerous smaller events associated with slip across corners or smaller cross sections of grains. Fourth, there is no explicit time or temperature dependence to the slip processes. Finally, there is no explicit modeling of grain boundary sliding or grain growth during deformation. Despite these deficiencies, the QCP simulations capture a unique characteristic of plastic deformation observed in MD simulations, allowing NC deformation to be studied at length and time scales not accessible to MD simulations. In Section 4.3, QCP simulations reveal the importance of highly heterogeneous internal stress on unique NC mechanical behaviors, e.g. plastic recovery of NC metals. The details of internal stress state will be explored in Chapter 5: QCP Model on lattice strain evolution.
CHAPTER 5: QCP ON LATTICE STRAIN EVOLUTION OF NC Ni

5.1 Background on Lattice Strain Evolution

5.1.1 Lattice Strain Evolution of CG FCC Metals

5.1.1.1 Internal Stresses in Polycrystals

The internal stresses induced by inhomogeneous deformation of polycrystal results in the change of lattice spacing, i.e. lattice strain $e_{<hkl>}$. $e_{<hkl>}$ is a direct measurable quantity from diffraction experiment, and imparts the stress distribution within polycrystal.

Figure 5.1 illustrates the three-type internal stresses based on their averaged distance with respect to grain size [121]. Type I stress, $\sigma_i$, is homogeneous over the entire polycrystal (macro-stress). Type II stress, $\sigma_{ii}$, entails grain-to-grain variation but is the homogeneous stress within one grain, termed inter-granular stress. Type III stress reflects the inhomogeneous stress within a grain, termed intragranular stress. Type II and III are also called micro-stresses. $\sigma_i$ and $\sigma_{ii}$ influence the measured X-ray diffraction peak position, and $\sigma_{ii}$ and $\sigma_{iii}$ influence the peak broadening.
Figure 5.1: Schematic illustration of three type stresses in a randomly orientated polycrystalline assembly. Type I ($\sigma_I$) is macro-stress, Type II ($\sigma_{II}$) is intergranular stress, and Type III($\sigma_{III}$) is intra-granular stress [121].

5.1.1.2 Canonical Evolution of $\bar{e}_{<hkl>}$ in CG FCC Metals

The development of intergranular stress (Type II) strongly depends on the elastic and plastic anisotropy intrinsic to the materials. Experiments performed on elastic anisotropic FCC metals suggest the following canonical behaviors [121-123]: (1) tensile intergranular microstrain established in {200} diffraction planes, i.e. $\bar{e}_{<200>}>0$; (2)
compressive microstrain in \{220\} group, \(\varepsilon_{<220>} < 0\); (3) \(\varepsilon_{<311>} \approx 0\), and \(\varepsilon_{<111>} \approx 0\).

These trends hold for both axial and transverse directions, where the lattice strain is either parallel or transverse to the loading axis. This canonical way of stress redistribution in CG FCC metal is induced by deformation anisotropy from grain orientation, and is inferred to be a characteristic of dislocation slip based crystal plasticity.

### 5.1.2 Lattice Strain Evolution of NC FCC Metals

#### 5.1.2.1 Distinct Observations of \(\varepsilon_{<hkl>}\) of NC Metals

Recent in-situ diffraction measurements on lattice strain evolutions of NC metals and alloys observe different behaviors compared to its CG counterparts. A measurement performed on NC Ni-15 wt % Fe shows tensile deflection developed in lattice strains for all the measured reflections [64]. This contra-equilibrium observation has been attributed to the existence of a ‘soft’ GB phase that accommodates majority of the deformation. This in-situ measurement seems to provide an evidence for GB mediated plasticity. In contrast, the residual lattice strain measured on pure ED NC Ni (\(d_{\text{ave}} \sim 30\text{nm}\)) in the transverse direction (see Figure 5.6a) exhibits a tensile and compressive shift in \{200\} and \{220\} reflections respectively when plastic strain exceeds 0.6%. This trend consists with CG Ni (see Section 5.1.1.2). But distinctively for NC Ni there exists a region (~ 0.6%) where \(\varepsilon_{<hkl>}\) remains constant for all the reflections [65], yet no such “constant” region is observed for UFG and CG Ni [65]. Further, texture development, which is considered to be a signature of dislocation-based plasticity, is observed in NC Ni upon large deformation [124, 125]. The reported texture maintains similarity to CG Ni,
though exhibits somewhat weaker [124]. These observations suggest that dislocations continue to play significant roles in NC deformation, but probably in a different manner from CG metals, e.g. interacting more actively with GBs.

5.1.2.2 Distinct Observations of Peak Width Evolution of NC Metals

Figure 5.7a shows that the evolution of full width-half maximum (FWHM) of ED NC Ni with $d_{ave} \sim 30\text{nm}$ at several subsequent unloaded states. The most distinctive feature is that FWHM reduces in magnitude for all the reflections over the initial region. Coincidently, this is also the region where $\bar{\sigma}_{\text{chi}}$ remains constant in Fig. 5.6a. In stark contrast, FWHM of CG metals keeps increasing till saturation [124]. FWHM is a direct measurement of the inhomogeneous stresses, i.e. Type II and III, in the polycrystal. This reduced magnitude suggests that there is a substantial recovery of pre-existing inter-granular and intra-granular inhomogeneity. The removal of those pre-existing strains could be achieved by reverse dislocation slip or GB relaxation. One distinction between these two mechanisms is large activation volume for dislocation slip vs. small for GB relaxation. The activation volume determines rate dependence of a process. Further assessment is required to address which one is dominant.
5.1.3 Existing Models on Micro-stresses

5.1.3.1 Self-consistent Model

Conventional polycrystal models have encountered some difficulties in capturing the unique lattice strain evolution of NC metals [122, 123]. Among the existing polycrystal models, self-consistent model, which inherently includes elastic anisotropy and crystallographic plasticity, has been succeeded in predicting the lattice strain evolutions and texture development of CG metals [123, 125, 126]. In the self-consistent scheme, grains are regarded as spherical inclusion in an infinite homogenous crystal matrix with effective overall polycrystal moduli. The elastic-plastic interaction between grains and matrix is incorporated, but not direct grain-to-grain interaction[127]. Two factors prohibit self-consistent model from predicting NC responses. First, the homogenization of matrix grain properties contradicts with highly heterogeneous deformation nature of NC metals, particularly in the micro-plasticity region. Second, a uniform distribution of critical shear stress results in grain orientation the only key factor for plastic anisotropy. As a result, any CG-unlike feature cannot be captured. Yang has modified self-consistent by reducing the number of active FCC slip system from 12 to 8, and improved the texture prediction of NC Ni under cold rolling. The reduction of active slip systems is motivated by enhancing particular texture development, but is lack of physical basis.

5.1.3.2 GB and Two-Phase Models

GB [84] and two-phase NC models [75, 77], in which GB behaves as only or key deformation contributor, are unlike to capture the NC micro-strain evolution properly,
provided that the large strain trend of NC metals is similar to the CG counterparts, where crystallographic slip is significant. The two-phase argument is recently promoted on explaining NC lattice strain based on the *contra*-equilibrium observation that all the lattice strains deviate to the same direction as plastic yielding starts (see Section 5.1.2.1). But this trend has not been observed by other research groups, where a pure NC Ni is investigated instead of NC Ni-Fe alloy. Further discussion on GB model is presented in Section 5.4.4 with comparison to our QCP model.

### 5.1.4 Controversial and Unsolved Issues

Experimentally, the *in-situ* diffraction testing sheds a light on probing deformation mechanisms of NC metals. However, the variation in the material microstructure, such as growth twins [65], solid solution [64] and initial texture [65], results in a set of large scattered data and thus weakens the connection between experimental observations and proposed deformation mechanisms. For instance, the evolution of $\bar{e}_{\text{chkl}}$ entails a ‘constant region’. But the reported value of such a region varies from 0.5% [64] to 2% [65] macro-imposed plastic strain. Then what produces the ‘constant’ region? How does it relate to the pre-existing stress state? These questions are worth further investigating through mechanistic models.

The existing models encounter some difficulty in explaining the aforementioned NC lattice strain evolution features. In this Chapter, a new view from *quantized crystal plasticity* is provided to explain the observed NC macro as well as micro stress-strain features. Derived from the cyclic stress-strain response in Chapter 4, three important
caveats of QCP simulations remain essential here. They are that: (1) single slip events induce large (~1%) jumps in grain-averaged plastic strain; (2) there is a large, asymmetric grain-to-grain distribution in the critical stress to activate such events, with numerous relative easier-to-slip grains balanced by a minority of hard-to-slip grains; and (3) prior deformation history induces large residual stress states that are removed by subsequent deformation. In this chapter, the Method section 5.2 describes the procedure of post-processing QCP simulation results into lattice strain. The orientation dependence of material properties in the model is emphasized. The Result section 5.3 reports on the QCP predictions for macro stress-strain response and inelastic recovery, as well as the evolution of residual lattice strain and peak width with small and large amounts of plastic deformation. The Discussion section 5.4 underscores the essential physics that enables the QCP approach to capture the experimental observations and assesses other existing hypotheses.

5.2 QCP/FE Simulations on Lattice Strains

5.2.1 In-situ X-ray Diffraction Geometry

The experimental data of NC lattice strain presented in this chapter is obtained by in-situ X-ray diffraction performed at Paul Scherrer Institute (PSI), Switzerland. The in-situ technique allows the measurement of diffraction profiles continuously with mechanical testing. Figure 5.2a is a schematic drawing of the diffraction setup [128]. A monochromatic focused synchrotron beam is illuminating a sample, which is mounted in a tensile machine. A CCD camera is employed to observe the deformation of sample
during testing. The diffracted x-ray beam is recorded using a unique micro-strip detector. Two components are essential for the \textit{in-situ} testing: (1) micro-strip detector which covers an angular range of 60° with an intrinsic angular resolution of 0.004°; (2) high intensity of synchrotron beam from Swiss light source. Those two allows a complete diffraction spectrum to be measured in a few seconds.

\textbf{Figure 5.2b} shows the diffraction geometry [128]. The tensile axis of the sample is perpendicular to the view plane along Z direction. The X-ray beam comes along the X direction, and is diffracted by the sample at its lower surface. The micro-strip detector acquires only the component of the diffracted beam cone, which lies in the transverse (X-Y plane). Therefore, the diffraction vector, and the further calculated lattice strain is perpendicular to the tensile axis.
Figure 5. 2: (a) A schematic drawing of the experimental diffraction setup at PSI. (1) the incoming x-ray beam; (2) the tensile machine; (3) the CCD camera; (4) the diffracted beam, and (5) the micro-strip detector. (b) Diffraction geometry. The tensile axis is perpendicular to the viewing plane [128].
5.2.2 **QCP/FE Simulations**

The QCP model employs a crystallographic description of dislocation-mediated plasticity, and is appropriate for NC and UFG materials. Here, individual slip events impart large, quantized jumps in grain-averaged plastic shear strain that scale as $\Delta \gamma_p \sim 1/d$. Thereby $\Delta \gamma_p$ is on the order of 1% for NC, and 0.1% for UFG materials. A large *grain-to-grain* variation in $\tau_c$, slip strength, is required to capture $\sigma$-$\epsilon$ response of ED NC Ni (Fig. 5.5a). Further, $\tau_{c(\text{min})} \sim 1/d$, consistent with a minimum condition to overcome dislocation pinning [103]. This motivates the *asymmetric* probability distribution $\rho(\tau_c)$ for NC Ni (see Table 5.1, Eq. 5.2). The *asymmetric* feature of $\tau_c$ distribution reduces as grain size $d$ increase, particularly in the UFG regime. A uniform $\tau_c (= 380\text{MPa, see Table 5.1, Eq. 5.3})$ is employed to simulate UFG response. The single value of $\tau_c$ is chosen such that the resultant macro flow stress reproduces experimental value [65]. Even for UFG Ni, a non-uniform distribution of $\tau_c$ would produce a better fitting to stress-strain response (see Chapter 3). But a uniform one follows the trend that asymmetric feature in $\tau_c$ reduces as grain size increases, and also provides more pronounced contrast between NC and UFG results.

As before, QCP physics is implemented in a $10 \times 10 \times 10$ array of 3D cube finite elements (C3D8) using the FE code ABAQUS. Each element represents one grain with a particular random texture orientation, $\tau_c$ assigned from $\rho(\tau_c)$, and $\Delta \gamma_p$. Uniaxial loading and unloading is imposed along Z direction with a magnitude of strain rate $10^{-3}$. 
The diffraction group \(<hkl>\) consists of grains with their \(<hkl>\) plane normal perpendicular to loading axis Z as the experimental diffraction geometry (see Fig. 5.2b). Therefore the computed \(e_{T<hkl>}\) is in the transverse direction, and \(2^\circ\) tolerance is allowed. The choice of transverse direction provides better grain statistics than axial (Z) direction, and also allows a direct comparison with the experimental results. A typical analysis with 1000 grains and 100 time steps requires ~4.5 CPU hr. on the Glenn System [118]. Table 5.1 provides the model and computational parameters.
Table 5. 1: QCP Simulation Parameters for NC and UFG Ni

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elastic constants (GPa) [106]</td>
<td>( C_{11}: 246.5; C_{12}: 147.3; C_{44}: 124.7 )</td>
</tr>
<tr>
<td>Burgers vector (nm)</td>
<td>( b: 0.25 )</td>
</tr>
<tr>
<td>Grain size (nm)</td>
<td>( d: 30 ) (NC), 150 (UFG)</td>
</tr>
<tr>
<td>Texture</td>
<td>Random</td>
</tr>
<tr>
<td>Schmid factors (avg. of max. in ea. grain)</td>
<td>( S_{\text{poly}}: 0.45 )</td>
</tr>
<tr>
<td></td>
<td>( S_{z,\text{mx,&lt;200&gt;:}}: 0.47 )</td>
</tr>
<tr>
<td></td>
<td>( S_{z,\text{mx,&lt;311&gt;:}}: 0.46 )</td>
</tr>
<tr>
<td></td>
<td>( S_{z,\text{mx,&lt;111&gt;:}}: 0.45 )</td>
</tr>
<tr>
<td></td>
<td>( S_{z,\text{mx,&lt;220&gt;:}}: 0.41 )</td>
</tr>
<tr>
<td>Quantized pl. strain</td>
<td>( \Delta \gamma_p = \min(1.2 \frac{b}{d}, \frac{\tau_c}{60 \text{ GPa}}) ) (6.1)</td>
</tr>
<tr>
<td>Crit. stress distrib.</td>
<td>NC: ( \rho(\tau_c) = \frac{\tau_c^{k-1} e^{-\frac{\tau_c}{\theta}}}{\Gamma(k)\theta^k} ) (6.2)</td>
</tr>
<tr>
<td></td>
<td>( k = 2, \theta = 525 )</td>
</tr>
<tr>
<td></td>
<td>UFG: ( \rho(\tau_c) = \delta(\tau_c - 380) ) (6.3)</td>
</tr>
<tr>
<td>Axial plastic prestrain</td>
<td>( \varepsilon_{\text{p(pre)}} = -0.5% )</td>
</tr>
<tr>
<td>Macro strain rate (1/s)</td>
<td>( \dot{\varepsilon} = 10^{-3} )</td>
</tr>
<tr>
<td>Local QCP strain rate (1/s)</td>
<td>( \dot{\gamma}_0 = 2 \cdot 10^{-2} )</td>
</tr>
<tr>
<td>Local slip events (integer)</td>
<td>( q )</td>
</tr>
</tbody>
</table>
### 5.2.2.1 Diffraction-Group-Dependent $S_{z,mx}$

A random orientation is assigned randomly to the 1000 grains. Figure 5.3 shows the probability density of maximum Schmid factor $\rho(S_{z,mx})$ for the entire polycrystal as well as various $<\text{hkl}>$ subgroups. $\rho(S_{z,mx})$ shows orientation dependent. $\rho(S_{z,mx,<200>})$ is higher in the large value range with $\bar{S}_{z,mx,<200>}/\sigma = 0.47 > \bar{S}_{z,mx,poly} = 0.45$. In contrast, $\rho(S_{z,mx,<220>})$ peaks at small value with the smallest $\bar{S}_{z,mx,<220>}/\sigma = 0.41$ among the selected diffraction groups. $\rho(S_{z,mx,<311>})$ is the most similar to $\rho(S_{z,mx,poly})$. Due to the small size of $<111>$ diffraction group, $\rho(S_{z,mx,<111>})$ is different from $\rho(S_{z,mx,poly})$ with a higher density around its mean value. But as sample size increases, $\rho(S_{z,mx,<111>}) \approx \rho(S_{z,mx,<311>}) \approx \rho(S_{z,mx,poly})$. Here $\bar{S}_{z,mx,<111>}$ and $\bar{S}_{z,mx,<311>}$ are close to $\bar{S}_{z,mx,poly}$. $\bar{S}_{z,mx,<\text{hkl}>}$ is essential to dictate $\bar{\varepsilon}_{<\text{hkl}>}$ trend, especially for CG metals.
Figure 5. 3: The probability density $\rho(S_{z, mx})$ for the polycrystal and various $<hkl>$ sub-groups. (See Table 5.1 for $S_{z, mx}$, $<hkl>$).

5.2.2.2 Diffraction-Group-Independent $\tau_c$

Figure 5.4 shows asymmetric probability density $\rho(\tau_c)$ for the entire polycrystal and various $<hkl>$ sub-groups. This distribution must be (1) uncorrelated with grain orientation and (2) overwhelm the grain-to-grain variation in Schmid factor. The combined effect of these two features is that stress redistribution is orientation independent in $\tau_c$ dominant regime.
Figure 5.4: Asymmetric probability density $\rho(\tau_c)$ for the entire polycrystal and various $<hkl>$ sub-groups (See Eq. 5.2, Table 5.1).

### 5.2.3 Model Calculation of Lattice Strain

The lattice strain $e_{<hkl>}$ at a material point is calculated based on stress state $\sigma$ computed by FEM, the crystal orientation, and the single crystal stiffness matrix.

First, $\sigma^c$ with respect to crystal coordinate system is calculated based on

$$\sigma^c = Q^T \sigma^g Q$$  \hspace{1cm} (5.4)

where $\sigma^g$ is $\sigma$ with respect to fixed global coordinate system, and $Q$ is the rotation matrix transforming crystal basis to global basis.

Second, $e^c$ is calculated from

$$e^c = C^{-1} \sigma^c$$  \hspace{1cm} (5.5)
where \( C \) is the single crystal Ni stiffness matrix (see Table 5.1 for values).

Third, lattice strain of \( \{hkl\} \) plane \( e_{<hkl>} \) is

\[
e_{<hkl>} = l_{<hkl>}^c e^l_{<hkl>}
\]

(5.6)

where \( l_{<hkl>}^c \) is the lattice strain direction.

Fourth, the average value of \( e_{<hkl>}, e^l_{<hkl>} \) and standard deviation \( s_{<hkl>} \) are constructed over the subgroup of grains satisfying diffraction condition: \( l^g_{<hkl>}, Z = 0 \), where \( Z \) is the global loading direction. This diffraction condition gives \( e^T_{<hkl>} \), and \( s^T_{<hkl>} \).

5.3 QCP vs. In-situ XRD Results

5.3.1 Macro Stress-Strain Response of NC Ni

The QCP prediction (solid curve) in Figure 5.5b captures three key experimental features for NC Ni: (I) an extended plastic transition strain \( \varepsilon_{p(\text{trans})} \sim 1\% \) at which 90\% of the plateau \( \sigma \) is reached; (II) reversible plastic strain \( \varepsilon_{p(\text{rev})} \sim 0.2\% \) during unloading and (III) a reloading strain gap \( \varepsilon_{p(\text{gap})} \). The QCP simulations imposed a plastic prestrain, \( \varepsilon_{p(\text{pre})} = -0.5\% \), which enhances \( \varepsilon_{p(\text{trans})} \). For comparison, the interrupted dashed line is the QCP prediction with a spatially uniform \( \tau_c (= 900 \text{ MPa}) \) distribution. This case does not capture any of these features. The model suggests the following physics of deformation during a loading/unloading/reloading sequence. Upon loading, plastic yielding gradually percolates from soft grains to hard grains. This process produces a much extended \( \varepsilon_{p(\text{trans})} \).
\( \sim 0.85\% \) (in Fig. 5.5a) in NC Ni, which is more than 4 times of \( \varepsilon_{p(\text{trans})} \) (~0.2\%) of CG Ni. Upon unloading, those soft yield grains slip back and result in recovered plastic strain \( \varepsilon_{p(\text{rev})} \). The re-slip occurs in these back slip grains upon reloading, and delays the reloading curve to join the previous loading one, and thus produces a strain gap \( \varepsilon_{p(\text{gap})} \). Pre-compression induces a residual stress state that enhances the gradual percolation of slip and thus \( \varepsilon_{p(\text{trans})} \) (see Section 4.3.1).

![Graph showing σ-εp response of electrodeposited (ED) NC Ni (d_{avg} = 30 nm) from (a) experiments [38] and (b) QCP simulations with asymmetric \( \tau_c \) distribution (solid) vs. uniform \( \tau_c \) (900 MPa) distribution (dashed). See Table 5.1 for other properties.](image-url)

Figure 5.5: \( \sigma-\varepsilon_p \) response of electrodeposited (ED) NC Ni (\( d_{avg} = 30 \) nm) from (a) experiments [38] and (b) QCP simulations with asymmetric \( \tau_c \) distribution (solid) vs. uniform \( \tau_c \) (900 MPa) distribution (dashed). See Table 5.1 for other properties.
5.3.2 Mean Transverse Residual Lattice Strain, $\bar{\varepsilon}_{T<hkl>}$, for NC Ni

The QCP predictions in Figure 5.6b capture two additional features for NC Ni: (IV) an initial region $\varepsilon_p(const)$ for which $\varepsilon_{T<hkl>}$ $\approx$ constant; and (V) a large strain region with CG-like trends ($\varepsilon_{T<200>} > 0$, $\varepsilon_{T<220>}$ $< 0$, and $\varepsilon_{T<111>}$ and $\varepsilon_{T<311>}$ $\sim 0$). Experimental results for UFG or CG Ni have no initial region with constant $\varepsilon_{T<hkl>}$ [123]. Likewise, the QCP results for UFG Ni ($d = 150$ nm)—simulated using single $\tau_c$ ($= 380$ MPa) and reduced $\Delta\gamma_p$ ($= 0.2\%$)—lack such a region and predict $\varepsilon_{T<200>}$ to be consistently larger for UFG vs. NC material. Like the NC experiment, the NC simulations show a clear transition between initial and large strain regions. The initial region $\varepsilon_p(const)$ results from removing residual stress state created by pre-compression. Thus, pre-compression is predicted to extend $\varepsilon_p(const)$. The large strain region $\varepsilon_{T<hkl>}$ is assumed to be mainly dictated by grain orientation, e.g. $S_{z, mx}$ (see Table 5.2), similarly to CG metals. The fluctuation within $\varepsilon_p(const)$ is affected by the number of grains in FEM sample and spatial distribution of $\sigma_c$ ($= \tau_c / S_{z, mx}$). Thus to address small sample statistics inherent in 1000 grain simulations, the NC predictions in Figs. 5.5b, 5.6b are an average of 5 instantiations of the same $\rho(\tau_c)$. 

124
Figure 5.6: (a) Change in transverse residual lattice strain $\Delta \bar{e}_{T<hkl>}$ vs. $\varepsilon_p$ for ED NC Ni ($d_{avg} = 30$ nm) [117] and UFG and CG Ni [65] from experiments. (b) $\Delta \bar{e}_{T<hkl>}$ from QCP simulations of NC Ni (asymmetric $\tau_c$ distribution and $\Delta \gamma_{p(max)} = 1\%$) and UFG Ni (uniform $\tau_c = 380$ MPa and $\Delta \gamma_p = 0.2\%$). See Table 5.1 for other properties.

Table 5.2 QCP Simulation Parameters and Results for Diffraction Groups of NC Ni

<table>
<thead>
<tr>
<th>plane</th>
<th>$S_{z, mx&lt;hkl&gt;}$</th>
<th>$\tau_c&lt;hkl&gt;$ (MPa)</th>
<th>$\sigma_c&lt;hkl&gt;$ (MPa) $^{(1)}$</th>
<th>$\bar{e}_{T&lt;hkl&gt;}$ (MPa)</th>
<th>$\varepsilon_p$ $@\varepsilon_p = 2%$</th>
</tr>
</thead>
<tbody>
<tr>
<td>polycrystal</td>
<td>0.45</td>
<td>1050</td>
<td>2340</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>200</td>
<td>0.47</td>
<td>1046</td>
<td>2250</td>
<td>230</td>
<td></td>
</tr>
<tr>
<td>311</td>
<td>0.46</td>
<td>1060</td>
<td>2310</td>
<td>-65</td>
<td></td>
</tr>
<tr>
<td>111</td>
<td>0.45</td>
<td>1060</td>
<td>2370</td>
<td>-313</td>
<td></td>
</tr>
<tr>
<td>220</td>
<td>0.41</td>
<td>1066</td>
<td>2630</td>
<td>-65</td>
<td></td>
</tr>
</tbody>
</table>

$^{(1)} \sigma_c = \tau_c / S_{z, mx}$ for each grain.
5.3.3 Deviation in Residual Lattice Strain, $s_{T<hkl>}$, for NC Ni

The QCP predictions in Figure 5.7b capture two additional features for NC Ni: (VI) a dip $h_{<hkl>}$ in all reflections at small strain and (VII) a monotonic increase in $s_{T<hkl>}$ at larger strain. The initial region contrasts with CG Ni, for which $s_{T<hkl>}$ gradually increases and ultimately saturates [63]. The initial dips are predicted if a negative plastic pre-strain ($\varepsilon_{p(pre)}$) is imposed in the QCP simulations, prior to the uniaxial test. In contrast, the predicted dip for UFG Ni is significantly reduced. Pre-compression is essential for $h_{<hkl>}$ in simulations. The resultant pre-compressive grains are returning back to their original shapes upon tension. Consequently, the grain-to-grain heterogeneity in residual strain, indicated by $s_{T<hkl>}$, reduces until reaching minimum at 0.5% plastic strain (marked by * in Fig. 5.7b), which is the initial stress free state. Such heterogeneity again increases at large strain in the course of continuous tension.
Figure 5.7: (a) Experimental *residual* full width half maximum (FWHM) vs. $\varepsilon_p$ for ED NC Ni ($d_{\text{avg}} = 30$ nm) [117]. (b) Standard deviation in residual transverse lattice strain $\sigma_{T<hkl>}$ from QCP simulations of NC and UFG Ni. See Fig.5.6 caption for simulation parameters.

The experimentally measured peak width (Fig. 5.7a) does not evolve the same way for all the measured diffractions, i.e. $\{200\}$ reaches minimum and remains; while $\{220\}$ and $\{311\}$ increase after touching the bottom. The relative positions and dips in the predicted $\sigma_{T<hkl>}$ are complex functions of the multi-axial pre-strain history. Figure 5.8 displays $\sigma_{T<hkl>}$ for NC Ni obtained by pretension in the transverse direction, which produces the
same axial residual strain $\epsilon_{p(\text{pre})} = -0.5\%$. As before, minimum of $s_{T<hkl>}$ appears in all reflections (Fig. 5.8b). But $s_{T<200>}$ increases much slower after the minimum. Other reflections $s_{T<111>}, s_{T<200>}$, and $s_{T<311>}$ are not symmetric any more with respect to the minimum strain in the small strain region showing dip. Therefore the discrepancies between the experimental and simulated results can be addressed, at least in part, by considering multi-axial pre-deformation histories.

Figure 5. 8: QCP simulations of standard deviation in residual transverse lattice strain $s_{T<hkl>}$ vs. $\epsilon_p$ for ED NC Ni ($d_{\text{avg}} = 30$ nm) with properties in Table 5.1, except that the pre-strain $\epsilon_{p(\text{pre})} = -0.5\%$ is achieved by transverse tension rather than axial compression.
5.3.4 QCP Simulations of XRD Peaks

Figure 5.9 shows the distribution of $e_{T^<220>}$ at (a) $\varepsilon_p = 0\%$, (b) 0.5\%, (c) 1.0\%, and (d) 2.5\%. The evolution of $e_{T^<220>}$ distribution illustrates the stress transfer in the course of tensile test. The initial broadening in Fig 5.9(a) is induced by residual stress state after pre-compression ($\varepsilon_{p\text{(pre)}} = -0.5\%)$. Mechanically soft ($\tau_c < \bar{\tau}_c$) grains tend to deform more in compression, and thus $\sigma_Z > 0$ and $e_T < 0$. By stress balance, the hard $<220>$ grains ($\tau_c > \bar{\tau}_c$) have $e_T > 0$. During the initial stages of a tension test (Fig. 5.9a to Fig. 5.9b), stress transfers between hard and soft grains within $<220>$ groups, producing a sharp but nearly stationary peak. Ideally, this within-group stress transfer could maintain up to Fig. 5.9c ($\varepsilon_p = 1\%)$, in which individual grain has an opposite stress state of that in Fig. 5.9a. Further tensile straining generates the motion of peak position (Fig. 5.9d), which indicates that the stress start to transfer among different reflection groups. $<220>$ group have the smallest $\tilde{S}_{z,\text{ext}}$, therefore, the peak position moves towards hard grain side. In addition, the peak starts to become asymmetric, which reflects the feature of $\tau_c$ distribution as deformation approaching criticality.
Figure 5.9: The distribution of $e_{T<220>}$ at $\epsilon_p = 0\%$ (a), 0.5\% (b), 1.0\% (c), and 2.5\%(d) from QCP simulations of NC Ni (asymmetric $\tau_c$ distribution and $\Delta\gamma_p(\text{max}) = 1\%$).
5.4 Insight into NC Deformation from QCP

5.4.1 A Physical View Based on Stress Redistribution

Figure 5.10a shows that the constant $\bar{\varepsilon}_{T<hkl>}$ (e.g., <200>) at small $\varepsilon_p$ is due to a balance in the trends for plastically soft ($\tau_c < \bar{\tau}_c$) vs. plastically hard ($\tau_c > \bar{\tau}_c$) <hkl> grains. Pre-compression ($\varepsilon_{p(pre)} = -0.5\%$) along the z-axis induces “negative” slip events ($q < 0$), particularly in soft grains. Therefore at very beginning state, the more deformed soft grains undergo axial tension ($\sigma_Z > 0$, and $e_z > 0$) exerted by hard and less deformed grains. The Poisson effect gives the residual $e_T < 0$ in soft grains. As such the resulting residual state satisfies $\sigma_z < 0$, $e_z < 0$, $e_T > 0$ in hard grains (Fig. 5.10, left inset). During the initial stages of a tension test, these slip events are reversed, particularly in soft grains. Along the way, $q = 0$ is achieved, leading to a minimum in $s_{T<hkl>}$ at $\varepsilon_p = \ast$ (Fig. 5.10, center inset). Further tensile straining generates positive slip events ($q > 0$), particularly in soft grains, thereby reversing the residual stress state (Fig. 5.10, right inset). A similar trend occurs for other <hkl> reflections since the same $\rho(\tau_c)$ is assumed for each <hkl> diffraction group.
Figure 5.10: QCP simulations of residual transverse lattice strain $\bar{\varepsilon}_{T<200>}$ vs. $\varepsilon_p$ for soft, hard, and all <200> grains. Colored insets show the axial stress $\sigma_z$ in <200> diffracting grains at different $\varepsilon_p$. $q = \#$ of slip events in a grain. Simulation parameters are in Table 5.1.
5.4.2 The Connection Between NC Properties and QCP Parameters

Table 5.3 summarizes key NC properties and the QCP parameters that control them. An asymmetric distribution \( \rho(\tau_c) \), Eq. 5.2 in Table 5.1) is required to capture most of the NC properties. For comparison, uniform \( \tau_c \) distributions produce an abrupt elastic-plastic transition and negligible reverse slip, hysteresis, and loading gap (Fig. 5.5b), as well as non-uniform \( \bar{\tau}_{T<hkl>} \) at small \( \epsilon_p \) (UFG, Fig. 5.6b). Quantized jumps in plastic strain (\( \Delta \gamma_p \), Eq. 5.1) are needed for the pronounced dips \( (h_{<hkl>}) \) and large lattice strain deviation \( (s_{T<hkl>} \) shown in Fig. 5.7, and larger values of \( \Delta \gamma_p \) enhance \( \epsilon_p(\text{trans}) \), \( \epsilon_p(\text{rev}) \), and \( \epsilon_p(\text{gap}) \).

Pre-compression is needed to obtain significant \( h_{<hkl>} \) and it increases \( \epsilon_p(\text{trans}) \) and \( \epsilon_p(\text{const}) \) [116]. Finally, all \( <hkl> \) sub-groups must have the same \( \tau_c \) distribution in order to capture the CG-like trends at large strain—namely \( \bar{\tau}_{T<200>} > \bar{\tau}_{T<111}> \approx \bar{\tau}_{T<311}> \bar{\tau}_{T<220}> \).

This ordering is controlled by the average Schmid factor \( S_{<hkl>} \) for each sub-group (Table 5.2), within which there is a large variety of hard and soft grains.

A systematic investigation of Table 5.3 is provided below by comparing QCP simulations for NC Ni (denoted as CASE B) vs. three supplemental cases (CASE C, C’ and C”). Those cases are obtained by turning off or reducing one or two essential QCP properties in CASE B. Table 5.4 summarizes the QCP material parameters for CASE C, C’, and C”.

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133
Table 5.3: QCP Parameters vs NC Properties

<table>
<thead>
<tr>
<th>NC Properties</th>
<th>QCP Parameters</th>
<th>asymmetric ρ(τc) (Eq. 5.2)</th>
<th>QCP Δγp (Eq. 5.1)</th>
<th>pre-compression εp(pre) &lt; 0</th>
</tr>
</thead>
<tbody>
<tr>
<td>I: εp(trans) (Fig. 5.5)</td>
<td>√</td>
<td>↑</td>
<td>↑</td>
<td></td>
</tr>
<tr>
<td>II: εp(rev) (Fig. 5.5)</td>
<td>√</td>
<td>↑</td>
<td></td>
<td></td>
</tr>
<tr>
<td>III: εp(gap) (Fig. 5.5)</td>
<td>√</td>
<td>↑</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IV: εp(const) (Fig. 5.6)</td>
<td>√</td>
<td>↑</td>
<td>↑</td>
<td></td>
</tr>
<tr>
<td>V: CG-like τ_{t&lt;\delta&gt;} (large εp) (Fig. 5.6)</td>
<td>*</td>
<td>x</td>
<td>x</td>
<td></td>
</tr>
<tr>
<td>VI: dip h_{&lt;\delta&gt;} (εp(const)) (Fig. 5.7)</td>
<td>x</td>
<td>√</td>
<td>√</td>
<td></td>
</tr>
<tr>
<td>VII: large s_{T&lt;\delta&gt;} (large εp) (Fig. 5.7)</td>
<td>↑</td>
<td>√</td>
<td>x</td>
<td></td>
</tr>
</tbody>
</table>

∨: necessary; ↑: enhances; x: not important;
*: same ρ(τc) required for each sub-group.

Table 5.4: QCP Simulation Parameters for Supporting CASE C, C’ and C”

<table>
<thead>
<tr>
<th></th>
<th>τc distribution</th>
<th>quantized slip jump Δγp</th>
<th>pre-compression εp(pre)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CASE B</td>
<td>asymmetric</td>
<td>1%</td>
<td>0.5%</td>
</tr>
<tr>
<td>CASE C</td>
<td>single</td>
<td>1%</td>
<td>0.5%</td>
</tr>
<tr>
<td>CASE C’</td>
<td>asymmetric</td>
<td>0.2%</td>
<td>0.5%</td>
</tr>
<tr>
<td>CASE C”</td>
<td>single</td>
<td>0.2%</td>
<td>0.5%</td>
</tr>
</tbody>
</table>
First, the asymmetric $\tau_c$ distribution is necessary for $\varepsilon_{p(\text{trans})}$ and $\varepsilon_{p(\text{rev})}$ from Fig. 5.5b vs. CASE C & C’ in Fig. 11. $\varepsilon_{p(\text{trans})}$ are promoted by heterogeneous yielding from soft to hard grains. Pre-compression can further spread $\tau_c$ distribution by inducing a heterogeneous internal stress state and results in an even pronounced $\varepsilon_{p(\text{trans})}$ [116].

Moreover, the internal stresses are capable of driving soft grains to slip back and produce $\varepsilon_{p(\text{rev})}$ upon unloading. Quantized jump $\Delta \gamma_p$ can enhance $\varepsilon_{p(\text{rev})}$ (see Fig. 5.5b vs CASE C’ in Fig. 5.11c). A larger $\gamma_{\text{target}}$ for a slip event results in relatively fewer highly deformed grains surrounded by elastic hard neighbors. This configuration promotes highly inhomogeneous internal stresses and produces larger $\varepsilon_{p(\text{rev})}$. Furthermore, the pronounced inelastic recovery $\varepsilon_{p(\text{rev})}$ relaxes the large and highly inhomogeneous internal stress state, and helps reducing the overall magnitude of $\bar{\varepsilon}_{\tau_{\text{chd}}}$ in NC sample (e.g. V in Fig. 5.5b vs. V in Fig. 5.11), especially at small strain.
Second, the asymmetric as well as orientation independent features of $\rho(\tau_c)$ (Fig. 5.4) are essential for NC residual $\overline{\epsilon}_{T<\text{hkl}>}$ behavior. In single $\tau_c$ model (CASE C in Fig.5.12a), grain orientation is the key factor to determine load redistribution among reflections. Such model fails to predict $\epsilon_{p(\text{const})}$, though it works for CG metals properly. On the other hands, the asymmetric $\tau_c$ model is more proper to dictate the NC load redistribution among reflections. The augmented density of $\tau_c$ at small stresses enables grains, which are less favored by orientations, e.g. small $S_{s_{\text{max}}}$, deform first or earlier. $\overline{\epsilon}_{T<\text{hkl}>}$ is mainly
dictated by $\tau_c$. Thereby the difference among reflections in $\bar{e}_{T<\text{hkl}>}$ reduces, provided that $\tau_c$ distribution is identical to all the reflections. As deformation further proceeds to criticality, all the reflections take barely the same $\tau_{c<\text{hkl}>\text{~mean}}$, and grain orientations start to play dominant roles. This gives rise to $\bar{e}_{T<\text{hkl}>}$ evolving similarly to its CG counterpart.

Physically $\tau_c$ is the critical strength to complete a slip event, and combines the strength requirement for dislocation nucleation as well as propagation at grain average level. In CG metals, it is reasonable to assume the same $\tau_c$ throughout the polycrystal assembly considering that intra-granular dislocation source strength is the mainly slip barrier. The source strength is proportional to $1$/source length; and the average source length, which is much less than grain size, should not entail much grain-to-grain variation. As grain size decreasing into nano scale, the intragranular source is anticipated to shut down due to the extremely large activation strength. Instead, MD simulations indicates that GB sources and depinning strengths are important, by observing that dislocations nucleate at GB, depin from GB ledges and propagate through the entire grain [17]. In addition, our FEM simulations require a propagation condition to be satisfied, which is during propagation stress drop $|\Delta \tau| < \tau_c$. In both scenarios, either depinning or propagation, there exits a minimum stress required on $\tau_c$ (scaling as $1$/grain size), and sets a threshold on critical slip strength distribution. This gives the asymmetric feature in $\tau_c$. Such a grain-to-grain variation in $\tau_c$ is a key quantity in the QCP model to probe GB properties [103], yet GB is not dealt explicitly as other multi-phase models for NC metals [75, 77].

Third, pre-compressive $\varepsilon_{p(\tau_c)}$ is necessary to capture the reduced deviation $h$ (Fig.5.7b, $h_C$ and $h_{C''}$ in Fig.5.12b). The magnitude of $h$ is determined by $\Delta \gamma_p$. Reducing $\Delta \gamma_p$ will
decrease \( h \) under the same \( \varepsilon_{p\text{(pre)}} \). This is attributed to a large fluctuation in the local stress induced by \( \Delta \gamma_p \). In addition, the large \( s_{T<}\text{hkl}> \) can be ascribed to both the quantized jump \( (s_{T<200>>} \text{ of } C \text{ vs. } C'' \text{ in Fig.5.12b}) \) and asymmetric \( \tau_c \) \( (s_{T<200>>} \text{ of } B \text{ in Fig.5.7b vs. } C \text{ in Fig.5.12b}) \). Experimentally, NC diffraction peak is much broader than its corresponding CG one. In addition to the effect from grain refinement, the higher heterogeneity in deformation of NC metals, which produces large deviation, is also a key source of broadening [63].
Figure 5.12: (a) $\bar{\varepsilon}_{T<hkl>}$ vs. $\varepsilon_p$ from QCP simulations of CASE C (uniform $\tau_c$ distribution and $\Delta\gamma_{p(max)} = 1\%$). (b) $s_{T<hkl>}$ vs. $\varepsilon_p$ from QCP simulations of CASE C and C". See Table 5.4 for QCP parameters.

5.4.3 NC vs. UFG Lattice Strain Evolution

Two differences between NC and UFG lattice strain evolution have been identified experimentally. They are (1) NC $\varepsilon_{p(const)} \sim 0.6\%$ vs. UFG $\varepsilon_{p(const)} \sim 0$, and (2) UFG $\bar{\varepsilon}_{T<200>}$
QCP simulations are able to capture these two features by assigning an asymmetric $\rho(\tau_c)$ to NC Ni vs. a single $\tau_c$ to UFG Ni. The trend that $\tau_c$ becomes more uniform with increasing grain size has been validated by our previous work [103] on macroscopic stress-strain response. At small strain, the NC $\bar{\tau}_{<hkl>}$ trends are dictated mainly by $\rho(\tau_c)$. This causes stress redistribution between soft and hard grains at small strain, leading to a substantial $\epsilon_{p(\text{const})}$ (Section 5.4.1). In contrast, the UFG $\bar{\tau}_{<hkl>}$ trends are determined by grain orientation, so that they have a deterministic trend from the very beginning. At large strain, NC Ni reaches criticality whereby grain orientation becomes the key factor, similar to UFG Ni. But the asymmetric $\tau_c$ distribution scatters the determinacy and thus reduces the magnitude of $\epsilon_{<hkl>}$, particularly $\epsilon_{<200>}$. 

5.4.4 Quantized Slip vs. Grain Boundary Deformation

Earlier modeling of polycrystalline thin films [84] predicts large inelastic response if a bimodal distribution $\rho(D)$ in GB diffusivity and sliding velocity is assumed. Such disparate GB diffusivities create stress redistribution between hard and soft regions, similar to the role of $\rho(\tau_c)$ in the present model. Thus, two approaches appear to be successful: a QCP approach with a continuous $\rho(\tau_c)$ and quantized slip $\Delta \gamma_p$ vs. a GB approach with a bimodal $\rho(D)$ and smooth (non-quantized) GB deformation.

However, there are important distinctions between these two approaches. The bi-modal $\rho(D)$ case is an artificial means to maximize inhomogeneous deformation. A more realistic continuous (non bimodal) $\rho(D)$ is less likely to sufficiently localize deformation,
so that experimental magnitudes of $\varepsilon_{p(\text{rev})}$ and other unique NC features are not captured. This is compounded by a lack of quantized deformation in the grain boundary model. Further, at first sight stress transfer between soft and hard GB region would render grain interior deforming only elastically, which could produce $\varepsilon_{p(\text{const})}$. But it is hard to image that soft GB region only sheds load to ‘hard’ GB rather than ‘hard’ grain interiors which surround it. Moreover, GB model does not involve the physical basis for CG-like trend at large strain. In addition, the presence of a dip in FWHM (Fig 5.7a) for NC Ni suggests a significant amount of inhomogeneous stress in pre-tested samples. GB sliding would relax stress over long pre-test periods, thereby removing a dip in FWHM.

5.5 Concluding Remarks

The quantized crystal plasticity (QCP) model is applied to explore footprints in NC lattice strain evolution. Combining with NC macroscopic stress-strain responses, seven NC properties are identified (see Table 5.3). A connection is established between these seven properties and three key QCP parameters: (1) discrete slip events imparting large plastic increment $\Delta \gamma_p (\sim 1/\text{grain size})$; (2) an asymmetric distribution of critical stress for slip activation and (3) an initial, residual stress state that is removed during the early stages of uniaxial tension. Some main conclusions include:

- The initial constant region of NC lattice strain evolution ($\varepsilon_{p(\text{const})}$) can be explained by stress transfer between soft and hard grains within the same $<hkl>$ diffraction group.
A grain-to-grain distribution of $\tau_c$ is the key factor to dictate soft and hard grains, particular at micro-plastic region. Essentially, $\rho(\tau_c)$ is required to be the same for all the $<hkl>$ diffraction group, and overwhelm the effect of grain orientation. As a result, the stress transfer does not depend on diffraction group.

The amount of residual stress state $\epsilon_p^{(pre)}$ can vary $\epsilon_p^{(const)}$ by providing the residual state that is able to be removed during post tensile loading. The upper limit of $\epsilon_p^{(const)}$ is controlled by micro-plastic region which is enhanced by asymmetric $\rho(\tau_c)$. Once the fully plasticity is established after reaching criticality, material tends to ‘forget’ its initial state, and thus residual state becomes non-removable.

After deformation reaching criticality at large strain, grain orientation becomes the key factor to dictate the stress transfer within polycrystal. Each diffraction group has the same $\bar{\tau}_{c<hkl>}$ but different $\bar{S}_{c,mr,<hkl>}$ (see Table 5.1). As a result, NC behavior follows CG trend.

Pre-compressive $\epsilon_p^{(pre)}$ is necessary to capture the reduced deviation $h$ in $e_{<hkl>}$, which mimics the dip in FWHM. The magnitude of $h$ is determined by $\Delta\gamma_p$.

QCP simulation with a reduced $\Delta\gamma_p$ and a uniform $\tau_c$ distribution captures lattice strain evolution of UFG Ni. $\bar{\tau}_{T<hkl>}$, in particular $\bar{\tau}_{T<200>}$, is observed to be larger than that in NC Ni[65]. The asymmetric $\tau_c$ distribution for NC simulations scatters the determinacy and thus reduces the magnitude of $e_T$.$<hkl>$. 

An in-depth comparison between QCP and a heterogeneous GB sliding model infer that the GB model cannot capture all the seven NC features identified here. Therefore QCP model provides an alternate view on deformation in NC metals.
Diffraction experiments demonstrate that footprints of inter-granular stress are different for ED NC Ni vs. CG Ni. This behavior is captured by QCP model, which comprises most features from conventional EPSC models, but including discrete dislocation slip, and an asymmetric distribution of critical stress for slip activation together with an initial residual stress state. The QCP simulations predict violent stress redistributions between soft and hard grains that allow for the removal and creation of residual intra-granular stress. This first time establishes the connection between unique NC footprints, whereby at small deformation, the FWHM dips while the average $<hkl>$ lattice strain stays constant, and at large deformation, CG trends are recovered. One essential requirement is that critical stress distribution must be uncorrelated with grain orientation and overwhelm the grain-to-grain variation in Schmid factor. This dominance of $\tau_c$ only exists in small deformation landscape, *e.g.* in elastic-plastic transition regime; whereas grain orientation takes over in large landscape. This short-range aspect of $\tau_c$ consists with the picture of dislocation interacting with GBs.
CHAPTER 6: CONCLUSIONS

A quantized crystal plasticity (QCP) model is developed within a large strain formulation to investigate unique mechanical responses of NC metals. The QCP model employs a crystallographic description of dislocation-mediated plasticity, and features with a discrete/quantized shear increment for each individual dislocation slip event. Such a quantized feature is motivated, in part, by MD simulations, which illustrates an abrupt jump in grain-averaged shear strain when a dislocation spontaneously transverses a nano grain after depinning from GB ledges. With this discrete characteristic, the developed QCP model is employed to explore the unique macro as well as micro stress-strain responses of NC metals. Key model features and outcomes are summarized below.

First, with respect to model development and contribution to literature,

- The QCP model incorporates size dependence in a unique way by virtue of discrete plasticity, which is quite lacking in the literature.
- The developed QCP relation is implemented through a rate independent crystal plasticity flow law under an implicit time integration scheme. This flow type implementation avoids instability that can be caused by a dramatic change of desired shear strain, and is capable of achieving a quasi-instantaneous shear increment.
There are two key material parameters, which control QCP response: quantized shear increment $\Delta \gamma_p$ for one dislocation slip event; and the grain-to-grain distribution in critical slip stress $\tau_c$ for completing a slip event. $\Delta \gamma_p \sim b/d$, which introduces a geometric scaling; and $\tau_c$ is spatially non-uniform in general and depends on numerous microstructural factors. The relation between them is established through a dislocation propagation condition, which imposes a physical requirement: a positive driving force on dislocations during completing a slip event. In this implementation, the resolved shear stress on the active slip system is not permitted to change sign during a slip event.

**Second**, the model is applied to study the monotonic tensile responses of NC and UFG Ni. The developed QCP model is first employed to investigate monotonic stress-strain characteristics, e.g. flow stress and elastic-plastic transition strain of NC and UFG Ni:

- An enhanced flow stress at small grain size is required to satisfy propagation condition. In particular, an Eshelby type analysis indicates that the minimum stress for activating an slip event, $\tau_{c(min)}$, scales as $\sim 1/d$ (Eq. 2.15). This provides an alternate view of size-dependent strengthening in addition to the hypothesis of dislocation nucleation at GBs in nano grains.
- An asymmetric distribution of $\tau_c$ is identified to reproduce the unique extended elastic-plastic transition strain of NC Ni. This asymmetric $\tau_c$ distribution is characterized by a large fraction of easier-to-slip grains balanced by a minority of hard-to-slip grains. This asymmetric feature reduces as grain size is increased to the UFG regime.
QCP simulations predict that NC metals are more prone to localized plastic strain (Fig. 3.9). This stems, in part, from the more violent stress redistributions associated with quantized slip events. QCP simulations predict that NC metals have a smaller fraction of plastically deformed grains for a given global plastic strain, compared to larger grain counterparts. For example, for Ni with $d = 50$ nm, only 40% of grains are predicted to have slipped at a macro plastic strain of 0.2% (Fig. 3.9).

Third, the model is applied to cyclic stress-strain response of NC metals. The QCP model incorporates reverse slip to address two additional issues of NC metals: (1) effect of residual stress state; and (2) source of pronounced plastic recovery. Two new model parameters are included: (1) critical stress for backward slip $\tau_{c,b}$; and (2) residual plastic strain $\varepsilon_{p(pre)}$. The latter factor is usually not included in models of NC mechanical properties, so that a stress-free initial state is often assumed. Yet diffraction studies show that NC metals have large internal stress.

- The residual stress state $\varepsilon_{p(pre)}$ can enhance or suppress slip events during subsequent loading, and thus make the elastic-plastic transition abrupt or extended.
- The residual stress state challenges the model requirement of asymmetric $\tau_c$ distribution by providing other possibility of reproducing monotonic stress-strain responses. A further investigation of cyclic responses of NC Ni validates the heterogeneous nature of slips associated with an asymmetric $\tau_c$ distribution.
- The QCP simulations capture the large reversible plastic deformation observed in experiments by reversing slips in relatively soft grains (with smaller $\tau_c$). This is
driven by highly heterogeneous internal stresses. The quantized nature of slip further enhances this heterogeneity by inducing large local stress fluctuation.

- An evolution of backward slip strength $\tau_{c,b}$ is predicted. This reflects, in part, the nature of grain boundaries that evolve with deformation.

**Fourth**, the QCP model provides an alternate insight into underlying deformation phenomena in NC metals. This is achieved by interpreting recent lattice strain measurement by synchrotron diffraction. Combining with *in-situ* macroscopic stress-strain characteristics, seven unique NC mechanical properties are identified (see Table 5.3). A connection is established between these seven NC properties and three key QCP parameters: (1) discrete slip events imparting large plastic increment $\Delta \gamma_p (\sim 1/$grain size$)$; (2) an asymmetric $\tau_c$ distribution (3) an initial, residual stress state $\varepsilon_{p(pre)}$ that is removable during the early stages of uniaxial tension.

- An asymmetric and orientation independent $\tau_c$ distribution is essential to produce the initial constant region $\varepsilon_{p(const)}$ in NC lattice strain $\bar{\varepsilon}_{T<hkl>}$ evolution. $\varepsilon_{p(const)}$ can result from stress transfer between easier-to-slip grains and harder-to-slip grains within the same $<hkl>$ diffraction group. In the initial micro-plastic regime, $\tau_c$ is the key factor to dictate easy and hard grains. Its orientation independent feature ensures that the stress transfer does not depend on diffraction groups.

- $\varepsilon_{p(pre)}$ can vary $\varepsilon_{p(const)}$ by providing a residual state that can be removed during post tensile loading. The upper limit of $\varepsilon_{p(const)}$ is controlled by the micro-plastic region $\varepsilon_{p(trans)}$ which can be enhanced by an asymmetric $\tau_c$ distribution. Once the fully
plastic state is established after reaching criticality, material tends to ‘forget’ the initial state, and thus residual state will depend on subsequent deformation.

- After deformation reaches criticality at large strain, the grain orientation becomes the key factor to dictate the stress transfer within polycrystal. All \(<hkl>\) diffraction group have the same \(\bar{\tau}_{c<hkl>}\) but different \(\bar{S}_{z,mc,<hkl>}\) (see Table 5.1). As a result, NC behavior recovers a CG trend.

- Precompressive \(\varepsilon_{\text{p(pre)}}\) is necessary to capture the reduced deviation \(h\) in \(e_{<hkl>}\), which mimics the dip in experimental FWHM. The magnitude of \(h\) increases with \(\Delta \gamma_p\).

- QCP simulations with a reduced \(\Delta \gamma_p\) and a uniform \(\tau_c\) distribution capture the lattice strain evolution of UFG Ni. \(\bar{e}_{T<hkl>}\), in particular \(\bar{e}_{T<200>}\), is observed to be three times larger than \(\bar{e}_{T<200>}\) of NC Ni [65]. It is believed that the statistical distribution of \(\tau_c\) for NC simulations scatters the determinacy and thus reduces the magnitude of \(e_{T<hkl>}\).

**Finally**, an outlook for future work includes coupling quantized crystal plasticity with other inelastic deformation mechanisms to explore multi-structural or multi-functional material systems. First system comes into mind is bimodal-grain-size polycrystalline structure. As mentioned in Section 1.2.2, this engineering structure is capability of achieving reasonable high ductility and at the same time maintaining relatively high strength, but an optimal spatial distribution of two set of grains are desired. Coupled with conventional crystal plasticity, which simulates coarse grain dislocation behavior, \(e.g.\) strain hardening, the QCP model can investigate the evolution of stress redistribution.
among small harder grains and large softer but hardenable grains. This study will help optimize spatial grain distribution from mechanical point of view. A second application is sub-micron or nano scale shape memory alloy (SMA) microstructures. SMA features a smart memory on shape and generally involves coupling of phase transformations and dislocation plasticity. One of its appealing applications is an actuator in MEMs, a system working in micro- or submicron scales. A hypothesis is that phase transformation does not depend on size, whereas dislocation does. For instance, from a QCP point of view, dislocation plasticity becomes more discrete and dramatic just like transformation as the activity size reduces. It will be interesting to investigate how these two discrete processes compete with each other, i.e. enhance or suppress one another in a small system.

In addition to quantized/discrete slip, a grain-to-grain distribution in critical slip strength is also essential when probing NC behaviors. Such a distribution, however, in the current QCP simulations, is quite simple: it neither evolves with deformation, nor depends on time or temperature. The short landscape nature of $\tau_c$ distribution (see Section 5.4.2) indicates that $\tau_c$ originates, in part, from some fairly rate-sensitive processes, e.g. dislocations depin from GB ledges. One possible way to incorporate evolution and rate dependence of $\tau_c$ is to implement a kinetic Monte Carlo algorithm, where an activation rate for a dislocation slip event could be determined from an activation energy barrier with various activation volumes (see Section 1.3.2.3). The kinetics of dislocation slip (forward and backward) will provide a physical insight into the unique rate dependent plastic recovery phenomenon in NC thin films (see Section 1.2.4).
REFERENCES


