Atomic-Scale Analysis of Plastic Deformation in Thin-Film Forms of Electronic Materials

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ATOMIC-SCALE ANALYSIS OF PLASTIC DEFORMATION IN THIN-FILM FORMS OF ELECTRONIC MATERIALS

A Dissertation Presented

by

KEDARNATH KOLLURI

Submitted to the Graduate School of the University of Massachusetts Amherst in partial fulfillment of the requirements for the degree of

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ATOMIC-SCALE ANALYSIS OF PLASTIC DEFORMATION IN THIN-FILM FORMS OF ELECTRONIC MATERIALS

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ACKNOWLEDGMENTS

But for the company and help of many people, the five and half years I spent at UMass-Amherst could not have been as interesting and as adventurous as they have been. I am very happy to acknowledge their contribution to the completion of this thesis.

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ABSTRACT

ATOMIC-SCALE ANALYSIS OF PLASTIC DEFORMATION IN THIN-FILM FORMS OF ELECTRONIC MATERIALS

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Nanometer-scale-thick films of metals and semiconductor heterostructures are used increasingly in modern technologies, from microelectronics to various areas of nanofabrication. Processing of such ultrathin-film materials generates structural defects, including voids and cracks, and may induce structural transformations. Furthermore, the mechanical behavior of these small-volume structures is very different from that of bulk materials. Improvement of the reliability, functionality, and performance of nano-scale devices requires a fundamental understanding of the atomistic mechanisms that govern the thin-film response to mechanical loading in order to establish links between the films’ structural evolution and their mechanical behavior.

Toward this end, a significant part of this study is focused on the analysis of atomic-scale mechanisms of plastic deformation in freestanding, ultrathin films of face-centered cubic (fcc) copper (Cu) that are subjected to biaxial tensile strain. The analysis is based on large-scale molecular-dynamics simulations. Elementary mechanisms of dislocation nucleation are studied and several problems involving the structural
evolution of the thin films due to the glide of and interactions between dislocations are addressed. These problems include void nucleation, martensitic transformation, and the role of stacking faults in facilitating dislocation depletion in ultrathin films and other small-volume structures of fcc metals.

Void nucleation is analyzed as a mechanism of strain relaxation in Cu thin films. The glide of multiple dislocations causes shearing of atomic planes and leads to formation of surface pits, while vacancies are generated due to the glide motion of jogged dislocations. Coalescence of vacancy clusters with surface pits leads to formation of voids. In addition, the phase transformation of fcc Cu films to hexagonal-close packed (hcp) ones is studied. The resulting martensite phase nucleates at the film’s free surface and grows into the bulk of the film due to dislocation glide. The role of surface orientation in the strain relaxation of these strained thin films under biaxial tension is discussed and the stability of the fcc crystalline phase is analyzed. Finally, the mechanical response during dynamic tensile straining of pre-treated fcc metallic thin films with varying propensities for formation of stacking faults is analyzed. Interactions between dislocations and stacking faults play a significant role in the cross-slip and eventual annihilation of dislocations in films of fcc metals with low-to-medium values of the stable-to-unstable stacking-fault energy ratio, $\gamma_s/\gamma_u$. Stacking-fault-mediated mechanisms of dislocation depletion in these ultrathin fcc metallic films are identified and analyzed.

Additionally, a theoretical analysis for the kinetics of strain relaxation in Si$_{1-x}$Ge$_x$ ($0 \leq x \leq 1$) thin films grown epitaxially on Si(001) substrates is conducted. The
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CHAPTER 1

INTRODUCTION

1.1. Motivation

A broad range of modern technologies relies increasingly on the use of nanometer-scale-thick films. Typically, over 500 processes are involved in the development of micro- and nano-scale devices. During these processes, the constituent materials are subjected to various thermo-mechanical environments, which introduce strain in the nano-scale-thick films, causing them to undergo plastic deformation. Such a response to strain has significant impact on the ductility, hardness, and strength of these ultrathin films and may lead eventually to their failure; the mechanical behavior of device interconnects in microelectronics offers typical examples of such materials response (Nix, 1989; Thompson & Lloyd, 1993; Gleixner et al., 1997; Hu & Harper, 1998).

The mechanical behavior of single-crystalline metallic and semiconductor thin films undergoing plastic deformation is a direct consequence of the nucleation and motion of crystalline line defects or dislocations, as well as of a variety of dislocation-dislocation interactions. For bulk materials, such mechanisms are nearly well understood and phenomenological models have been developed for analyzing their deformation. These phenomenological models, however, cannot be extended to the nanoscale and, more specifically, to describing the mechanical response of ultrathin films of metals and semiconductors. A fundamental understanding of the atomistic mechanisms of strain relaxation in ultrathin films, as well as the associated deformation
dynamics, is essential for accurate property predictions, for better reliability analysis, and for designing higher-quality devices. Such a fundamental understanding of the underlying mechanisms that govern mechanical behavior in small-volume structures of materials is also essential for the development of new phenomenological models of deformation or for extension of existing ones. In addition to nanometer-scale thick metallic and semiconductor films, material heterostructures such as heteroepitaxial films of metals and semiconductors are also increasingly used in various areas of micro- and nano-fabrication due to their enhanced material properties compared to those bulk materials structures (Mii et al., 1991). Such heterostructures pose serious reliability challenges from the film growth to the device service stages. A fundamental understanding of the interfacial response and stability in heterostructures, as well as quantitative predictions of defect nucleation and evolution are particularly significant for developing optimal strategies for fabrication of reliable electronic devices.

Nevertheless, experimental studies of strain relaxation mechanisms that are primarily due to dislocation dynamics are particularly difficult; for example, in single-crystalline face-centered cubic (fcc) metals such as Cu, the corresponding strain relaxation time scales under extremely high strain rates are in the sub-nanosecond range (Loveridge-Smith et al., 2001). Under these conditions, atomistic computational methods are most suitable for analyzing the strain relaxation mechanisms in thin films by direct monitoring of their dynamical response (Bulatov et al., 1998; Abraham, 2003). In this thesis, atomistic simulation techniques are used in order to analyze systematically the response to straining of thin-film forms of electronic materials. Specifically, the mechanical behavior is analyzed of ultrathin films of fcc metals,
primarily Cu, under applied biaxial tensile strains. The fundamental mechanisms of plastic deformation in metallic thin films are identified and links between these mechanisms and the films’ mechanical properties, such as their strength, are established. Additionally, a dislocation mean-field theory is developed to predict the defect evolution and strain relaxation kinetics in epitaxially grown Si$_{1-x}$Ge$_x$ films, and a proof-of-concept study is conducted to parameterize, based on atomic-scale computations, the dislocation mean-field theory for semiconductor heterostructures in order to enable computationally efficient quantitative predictions based on the theory.

1.2. Background

Fundamental understanding of deformation processes and determination of the associated material response to straining has been a subject of great importance over the last 50 years. Strain relaxation processes have significant impact on material properties like ductility, hardness and material strength. Beyond a critical strain, strain relaxation induces plasticity (i.e., plastic flow) in ductile materials due to the nucleation and glide motion of dislocations. The existence of dislocations was first proposed theoretically in 1934 in order to explain the low strength of crystals observed in experiments (Orowan, 1934; Taylor, 1934); the presence of dislocations in crystals was confirmed experimentally over two decades later (Hirsch et al., 1956). The properties of single crystalline materials undergoing plastic deformation are determined by the motion, multiplication, and interactions of these dislocations; a brief background information on defects in crystals and dislocations in fcc metals is given in Appendix A.
In bulk crystalline materials subjected to external stress, work hardening is caused by an increase in the dislocation density and by a sharp decrease in plastic flow. The dislocation density increases due to rapid dislocation multiplication through double cross-slip and Frank-Read-type mechanisms. The sharp decrease in plastic flow is caused by interactions between dislocations in multiple slip planes, which lead to the formation of sessile dislocations that act as barriers to dislocation motion, as well as by pile-ups of dislocations at obstacles such as grain boundaries and sessile dislocations (Hirth & Lothe, 1982).

Unlike bulk metals, metallic nanostructures and other small-volume structures exhibit, for example, ultra-high strength. Examples of such small-volume structures range from whiskers with small diameters that were reported (as early as almost 6 decades ago) to have much higher strength than that of bulk metals (Herring & Galt, 1952; Brenner, 1958) to nanometer-scale pillars of fcc metals that yielded under uniaxial compression flow strengths comparable to the materials’ ideal shear strengths (Uchic et al., 2004; Greer & Nix, 2005, 2006; Shan et al., 2008). Identifying and understanding the fundamental atomic-scale mechanisms of plastic deformation in small-scale materials, and specifically in ultrathin films of fcc metals, is one of the major goals of recent and current studies on the mechanical behavior of small-volume structures. As these fundamental mechanisms are difficult to probe in experiments, atomic-scale and other fine-scale dynamical simulation methods provide ideal means for analyzing deformation mechanisms in ductile thin films by direct monitoring of the dynamical response of the materials at the atomic/microscopic scale.
First-principles density-functional-theory (DFT) calculations, large-scale molecular-dynamics (MD) simulations, and large-scale dislocation-dynamics (DD) simulations have been used extensively to understand the fundamentals of plastic flow initiation and nucleation of dislocations (Li et al., 2002; Lilleodden et al., 2003), to explore and analyze dislocation intersections (Zhou et al., 1998; Bulatov et al., 2006), vacancy formation due to dislocation motion (Zhou et al., 1999), and pipe diffusion in mobile dislocations (Gungor & Maroudas, 2005a), and to investigate plastic deformation during nanoindentation of metal surfaces (Li et al., 2002; Lilleodden et al., 2003). Deformation processes at shock loading conditions have been reported to cause severe plastic deformation (Bringa et al., 2006), leading to formation of ultra-fine grain structures in bulk metals (Wu et al., 2002) and nano-domains in metallic thin-film materials (Gungor & Maroudas, 2005b). Similar to nanocrystallization, amorphization of fcc crystalline metallic nanowires due to strain-rate-limited shock loading at controlled temperatures has been reported in atomic-scale studies (Ikeda et al., 1999). Discrete DD simulations also have been used to study size effects in nanopillars of fcc metals; these simulations of response to mechanical loading of fcc nanopillars have shown that dislocation escape through the free surfaces and depletion or inactivation of dislocation sources play a significant role in the plastic deformation of small-volume structures (Balint et al., 2006; Tang et al., 2007, 2008). Furthermore, atomistic simulations have been used extensively to study polycrystalline metals; various deformation processes, including deformation twinning (Yamakov et al., 2002, 2003), grain boundary migration and material softening (Schiotz et al., 1998, 1999), the influence of grain size on the deformation mechanics (Zhu & Langdon, 2005), and the
dynamical complexity of work hardening in nanocrystalline materials (Buehler et al., 2005) have been studied using large-scale MD simulations for different grain sizes and at different strain rates.

Plasticity and deformation processes frequently lead to eventual failure of the material; in metals, the two most common precursors to failure are nucleation, growth, and coalescence of voids and crack propagation. Theoretically and computationally, void growth and void-void interactions and coalescence have been studied at continuum (McClintock, 1968; Koplik & Needleman, 1988) and atomistic (Gungor et al., 2000; Seppala et al., 2004a, 2004b, 2005; Gungor & Maroudas, 2005a) scales. Atomistic studies also have been used to understand the fracture dynamics of bulk materials (Zhou et al., 1997; Belak, 2002; Abraham, 2003). Atomistic simulations and first-principles studies also have been conducted to study dislocation nucleation and crack propagation, and to calculate the activation energy barriers of the underlying rate processes (Xu et al., 1997; Zhu et al., 2004b). Nevertheless, fundamental atomic-scale strain relaxation mechanisms that govern the mechanical behavior of ultrathin films remain largely unexplored.

In addition to metallic thin films that are vital ingredients of micro-electronic devices and other micro-electro-mechanical systems, strained semiconductor thin films grown epitaxially on semiconductor substrates of different composition, such as Si$_{1-x}$Ge$_x$ films grown on Si substrates, are becoming increasingly important due to their enhanced hole and electron mobility compared to unstrained structures of the same material (Mii et al., 1991). When an alloyed Si$_{1-x}$Ge$_x$ layer is grown on a Si substrate, biaxial strain develops in the epitaxial layer due to lattice mismatch between the
substrate and the film. Up to a critical film thickness, the state of strain in the film is elastic and the film/substrate interface is coherent with no dislocation defects introduced in the heterostructure. A substrate that can relieve strain due to lattice mismatch by elastic accommodation, i.e., by acting as if it were unconstrained at its base and relaxing parallel to the film/substrate interface is called a compliant substrate. Beyond the critical film thickness, mechanisms of strain relaxation in the epitaxial film include misfit dislocation generation at the film/substrate interface (van der Merwe, 1963), as well as surface morphological instabilities (Srolovitz, 1989; Spencer et al., 1991). In practice, a large number of threading dislocations are nucleated in the epitaxial film; after a short glide, these dislocations become immobilized, resulting in a high defect density in the film. However, device-quality materials require considerable strain relaxation, low threading dislocation densities, and smooth film surfaces. While various experimental techniques have been used to grow epitaxially thin-film heterostructures with significant strain relaxation and low dislocation density (Hull et al., 1990; Legoues et al., 1992; Fitzgerald et al., 1992; Watson et al., 1993; Mantl et al., 1999; Luysberg et al., 2002; Christiansen et al., 2002a; Cai et al., 2004), a priori estimation of the strain relaxation and dislocation density in such structures has not been achieved yet; such predictions are most important for the design of experimental procedures to manufacture device-quality materials.

1.3. Thesis Objectives and Outline

This thesis aims at a fundamental, atomic-scale understanding of the fundamental strain relaxation mechanisms in thin-film forms of electronic materials and at enabling the development of macroscopic models that can predict strain relaxation
due to plastic deformation and the resulting mechanical properties of the thin films. The studies in this thesis concentrate on two specific thin-film forms: (i) ultrathin films of fcc metals (primarily Cu) and (ii) strained-layer semiconductor heterostructures of Si_{1-x}Ge_{x} thin films grown on Si(001) substrates. The analysis of the mechanical behavior of such thin-film materials is based primarily on MD simulation and on other atomistic simulation methods.

A significant part of this thesis aims at a fundamental understanding of the deformation mechanisms in ultrathin fcc Cu films. The research focuses on understanding the strain relaxation of the metallic thin films under the application of biaxial strain, since the films are subjected to planar strain during their processing and service. In experiments of thin-film deposition, either by sputtering or by electroplating, the thin film surfaces are typically oriented normal to the \(<111>\), \(<110>\), and \(<100>\) crystallographic directions. In this thesis, therefore, I consider the effects of film surface orientation and study metallic films with \(<111>\) and \(<110>\) film planes, i.e., films whose free surfaces are parallel to high- and low-symmetry lattice planes with low Miller indices, respectively. In addition, I aim to understand the role of ductile void growth in strain relaxation and to identify and analyze the interplay of plastic deformation mechanisms mediated by dislocations nucleated at the film surface and at void surfaces. Furthermore, this thesis aims at a fundamental understanding of the mechanisms that govern the mechanical behavior during dynamic deformation of thin films of different fcc metals. Atomistic simulation methods used in modeling thin-film response to applied biaxial strain and the methods used to characterize the film nanostructures and to determine the mechanical properties of the thin films are detailed.
in Chapter 2; potential energy functions used to model the interatomic interactions also are discussed in Chapter 2.

In Chapter 3, a systematic computational analysis is presented of strain relaxation mechanisms and the associated defect dynamics in [111]-oriented, free-standing ultrathin Cu films subjected to a broad range of biaxial tensile strains. The analysis reveals five regimes in the thin film’s mechanical response with increasing strain level, $\varepsilon$. Within the considered strain range, after an elastic response at biaxial strain levels up to $\varepsilon = 5.5\%$, the strain in the metallic thin film is relaxed by plastic deformation. At low levels of the applied biaxial strain above the yield strain ($\varepsilon \sim 6\%$), threading dislocation nucleation at the surface of the thin film in conjunction with vacancy cluster formation in the film lead eventually to formation of voids that extend across the thickness of the film. For $6\% < \varepsilon < 8\%$, dislocations are emitted uniformly from the thin-film surface, inhibiting the nucleation of voids. For $\varepsilon \geq 8\%$, in addition to nucleation of dislocations from the film surface, dislocation loops are generated in the bulk of the film and grow to intersect the thin-film surface. For $\varepsilon \geq 10\%$, a high density of point defects in the film leads to nucleation of Frank partial dislocations that dissociate to form stacking-fault tetrahedra. In addition, dislocation-dislocation interactions due to the high dislocation density lead to formation of Lomer-Cottrell dislocation locks and complex stable dislocation junctions that act as obstacles to dislocation glide. A detailed analysis of the void formation mechanism has been published in *Applied Physics Letters* (Kolluri et al., 2007). The systematic analysis of
the films’ structural response and of the underlying atomic-scale mechanisms of strain relaxation has been published in Journal of Applied Physics (Kolluri et al., 2008a).

In Chapter 4, a systematic computational analysis is presented of strain relaxation mechanisms, based on isothermal-isostain MD simulations of the response of (110)-oriented ultrathin Cu films. The analysis in Chapter 4 includes strain relaxation due to growth of an already existing void. The analysis reveals that for applied biaxial strain levels, \( 2\% \leq \epsilon \leq 6\% \), biaxial strain relaxation is dominated by emission and propagation of dislocations (plastic flow) from the surface of the void accompanied by ductile void growth. For \( 6\% < \epsilon < 10\% \), the biaxial strain in the thin film is relaxed by both ductile void growth and emission of dislocations from the surface of the thin film. For \( \epsilon \geq 10\% \), strain relaxation is dominated by dislocation emission from the surface of the thin film, leading to a phase transformation from the fcc to a hexagonal close-packed (hcp) phase. In this martensitic transformation, martensites are found to nucleate at the film surface and grow into the bulk of the film due to dislocation glide; in this process, the magnitudes of the relative atomic slip displacements are identical to those proposed for Bain transformations. Mechanical stability analysis also shows that the onset of the phase transformation is consistent with the onset of a shearing instability of the thin film. A detailed analysis of the fcc \( \rightarrow \) hcp martensitic transformation has been published in Physical Review B (Kolluri et al., 2008b). The systematic analysis of the film’s structural response and of the underlying atomic-scale mechanisms of strain relaxation is in preparation to be submitted for publication in the Journal of Applied Physics.
In Chapter 5, a detailed analysis is presented based on large-scale MD simulations of dynamic deformation under biaxial tensile strain of different pre-strained, single-crystalline nanometer-scale-thick fcc metallic films. The results in this study indicate that the mechanical behavior of thin films of metals with moderate-to-high propensity for formation of stacking faults such as Cu and Ni is significantly different from those of metals with low propensity of formation of stacking faults such as Al ones. Plastic strain rate in Cu and Ni films is found to be greater than that in Al thin films, which leads to an extended easy-glide stage in Cu and Ni but not in Al films. These results reveal that stacking faults, which are abundantly present in some fcc metals (Ni and Cu), may play a significant role in the dissociation, cross-slip, and eventual annihilation of dislocations in small-volume structures of fcc metals. The underlying mechanisms are mediated by interactions within and between extended dislocations that lead to annihilation of Shockley partial dislocations or formation of perfect dislocations. While the mechanisms of dislocation annihilation in Ni and Cu thin films are due to stacking-fault-mediated cross-slip of dislocations, the analysis conducted in this study indicates that collinear interactions lead to dislocation annihilation in Al thin films. These results provide new insights into the mechanisms of dislocation depletion in fcc metals with ultra-thin film geometries, where dislocation depletion due to escape of dislocations at the surface is less likely. The findings in this study demonstrate dislocation starvation in small-volume structures of fcc metals with ultra-thin film geometry and underline the significant role of geometry (e.g., ultrathin films versus nanopillars) in determining the mechanical response of metallic small-volume structures. The analysis of the mechanical behavior of Cu thin films together
with a detailed characterization of the stacking-fault-mediated mechanisms of dislocation annihilation has been submitted for publication in the *Journal of Applied Physics*. A comparative study of the mechanical behavior under biaxial strain of ultrathin films of fcc metals with different propensities for formation of stacking faults has been submitted for publication in *Applied Physics Letters*.

In Chapter 6 of this thesis, strain relaxation is analyzed in epitaxial Si$_{1-x}$Ge$_x$ films on Si(001) substrates during growth or thermal annealing using a properly parameterized dislocation-mean-field theoretical model of plastic deformation dynamics. Furthermore, an integrated hierarchical computational approach, which employs various atomistic simulation methodologies in conjunction with continuum elasticity and dislocation theory, is presented toward the parameterization of predictive macroscopic models for the onset of dislocation generation in the heteroepitaxial systems and for the kinetics of strain relaxation. The development of the dislocation mean-field theoretical model, the comparison of its predictions with experimental measurements, and the hierarchical computational approach together with its implementation in prototypical systems have been published in *Materials Research Society (MRS) Symposia Proceedings* (Kolluri et al., 2005) and in *Applied Physics Letters* (Kolluri et al., 2006). Finally, future directions for research on deformation of thin films are discussed in a concise manner in Chapter 7.
CHAPTER 2

ATOMISTIC SIMULATION AND STRUCTURAL CHARACTERIZATION METHODS

In this Chapter, several computational methods, which are employed in this thesis for the atomic-scale simulations of thin metallic films and semiconductor heterostructures and for the analysis of the simulation results, are discussed. In Sec. 2.2, the interatomic potentials for metals and semiconductors that are used in the atomistic simulations for the description of interatomic interactions are discussed. The numerical details of the atomistic simulations are given in Sec. 2.3. The various techniques and computational tools for structural characterization and computation of materials properties from the atomistic simulations are presented in Sec. 2.4 and Sec. 2.5, respectively. The limitations of molecular-dynamics simulations, in the context of the studies within this thesis, are discussed in Sec. 2.6.

2.1. Molecular-Dynamics Simulations and Interatomic Potentials

Atomistic simulations are used to study the structure, energetics, and dynamics of a collection of interacting atoms following classical mechanics. For a system containing \(N\) atoms with positions \(\{\mathbf{r}_i\}_{i=1}^N\) and velocities \(\{\mathbf{v}_i = d\mathbf{r}_i / dt\}_{i=1}^N\), the total Hamiltonian is given by

\[
H(\{\mathbf{r}_i\}_{i=1}^N) = T + U = \sum_{i=1}^N \frac{1}{2} m_i \mathbf{v}_i^2 + U(\{\mathbf{r}_i\}_{i=1}^N) \tag{2.1}
\]
where $T$ and $U$ represent kinetic and potential energy, respectively, of the system and $m_i$ is the mass of atom $i$. A molecular dynamics (MD) simulation is based on the integration of Newton’s equations of motion for all $N$ interacting atoms and generates a trajectory of the atomic positions of all the atoms. Most of the thermodynamic properties of the material system can be calculated as time averages along the atomic trajectories and the corresponding transport properties can be calculated from computation of time correlation functions (Allen & Tildesley, 1990). MD simulations, however, are most useful in identifying the dynamical response of a material system to external forces; the structural evolution of the material can be monitored easily over the course of the simulation and atomic-scale information that is not easily accessible in experiments can be extracted from MD simulations. Hence, MD simulations are very useful for the fundamental understanding of structure-property relationships.

As can be seen from the expression for Hamiltonian, Eq. (2.1), the accuracy of the potential energy function, $U(\{r_i\}_{i=1}^N)$, in describing the interatomic interactions is vital to obtaining reasonably accurate predictions for the behavior of real materials. In the MD simulation studies of metallic systems conducted in this thesis, semi-empirical interatomic potentials that are based on the embedded-atom method (EAM) in order to describe the interatomic interactions in metals were employed (Foiles et al., 1986; Oh & Johnson, 1988; Angelo et al., 1995; Mishin et al., 1999, 2001). In the EAM formalism, the total potential energy $U$ of an elemental system is represented as

$$U = \frac{1}{2} \sum_q \mu(r_q) + \sum_i \phi(\rho_i)$$  \hspace{1cm} (2.2)
where \( u(\mathbf{r}_{ij}) \) is the pair potential as a function of the distance between atoms \( i \) and \( j \), \( \mathbf{r}_{ij} \), and \( \phi \) is the embedding energy that is a function of the host electron density, given by

\[
\rho_i = \sum_{i \neq j} \rho(\mathbf{r}_{ij}),
\]

induced at site \( i \) by all the other atoms in the system. While the functional form of the EAM potential is based on certain concepts and principles of solid-state physics, the comprising potential functions used in MD simulation practice are essentially empirical and their parameterization is based on fitting procedures to reproduce known (mainly from experimental measurements) material properties.

In almost all EAM parameterizations, and certainly all the EAM parameterizations used in the studies within this thesis, the total energy of the fcc lattice under uniform compression and dilation is required to obey the universal equation of state proposed by Rose and coworkers (Rose et al., 1984). As a result, the EAM potentials account properly for the lattice anharmonicity (Foiles & Daw, 1988) and provide a good modeling framework for simulating high-pressure and shock-loading experiments (Oh & Johnson, 1988; Johnson, 1988; Foiles & Daw, 1988). These potentials have been tested extensively regarding their predictions of structural and mechanical properties. Specifically, the potentials predict accurately point-defect properties and surface energies and favor energetically the fcc phase over the hexagonal close-packed (hcp) and body-centered cubic (bcc) phases. Furthermore, the computed unstable, generalized, and intrinsic stacking-fault energies are in good agreement with experimental results (Oh & Johnson, 1988; Angelo et al., 1995; Mishin et al., 1999, 2001).
Figure 2.1. Stress-strain curves for shearing deformation of copper in the [111] plane along the [11\(\bar{2}\)] direction. Shear strain is defined as the shear displacement divided by \(a/\sqrt{6}\). The black and red curves correspond to the potential parameterizations used in the present work. The blue and green curves correspond to \textit{ab initio} and EAM-Ackland calculations as reported by Zhu and co-workers (Zhu et al., 2004a).

In addition to their good predictions of single-value properties, the EAM potentials used in the studies within this thesis predict accurately the elastic deformation of Cu. Figure 2.1 compares the stress-strain response for a simple shearing deformation in the (111) plane along the [11\(\bar{2}\)] direction predicted by the Oh & Johnson potential (Oh & Johnson, 1988) and the EAM potential parameterization by Mishin and coworkers (Mishin et al., 2001) with those obtained from \textit{ab initio} density functional theory (DFT) calculations and from another EAM parameterization developed by Ackland and co-workers (Ackland et al., 1997) that was not used in the studies within this thesis. The stress-strain response from the \textit{ab initio}, EAM-Ackland, and EAM-Mishin potentials shown in Fig. 2.1 were calculated by Zhu and coworkers (Zhu et al., 2004a).
It is demonstrated clearly in Fig. 2.1 that the predictions of the Oh & Johnson potential and Mishin potentials are in good agreement with the *ab initio* DFT calculations. Zhu and coworkers verified further the ability of the EAM-Mishin potential to predict accurately the characteristics of the first nucleated dislocation in nanoindentation experiments; the EAM-Mishin potential accurately predicted the mixed shear mode of the Shockley partial in nanoindentation simulations (Zhu et al., 2004a).

In order to model silicon-germanium heterostructures (Si$_{1-x}$Ge$_x$, 0 ≤ x ≤ 1), an empirical many-body interaction potential proposed by Tersoff was used (Tersoff, 1989). In Tersoff’s formulation, the potential energy $U$ is expressed as

$$U = \frac{1}{2} \sum_{ij} f_c(r_{ij})[f_R(r_{ij}) + b_{ij}f_{\lambda}(r_{ij})],$$

where

$$f_R(r_{ij}) = A_{ij} \exp(-\lambda_{ij}r_{ij}), \quad f_{\lambda}(r_{ij}) = B_{ij} \exp(-\mu_{ij}r_{ij}),$$

$$f_c(r_{ij}) = \begin{cases} 
1, & r_{ij} < R_{ij} \\
\frac{1}{2} + \frac{1}{2} \cos[\pi(r_{ij} - R_{ij})/(S_{ij} - R_{ij})], & R_{ij} < r_{ij} < S_{ij} \\
0, & r_{ij} > S_{ij}
\end{cases}$$

$$b_{ij} = \chi_{ij}(1 + \beta^n_i \psi^n_{ij})^{-1/2n_i}$$

$$\xi_{ij} = \sum_{k \neq i} f_c(r_{ik})g(\theta_{ik}), \quad g(\theta_{ik}) = 1 + \frac{c_i^2}{d_i^2} - \frac{c_i^2}{d_i^2 + (h_i - \cos \theta_{ik})^2}$$

and

$$\lambda_{ij} = (\lambda_i + \lambda_j) / 2, \quad \mu_{ij} = (\mu_i + \mu_j) / 2,$$

$$A_{ij} = \sqrt{A_i A_j}, \quad B_{ij} = \sqrt{B_i B_j}, \quad R_{ij} = \sqrt{R_i R_j}, \quad \text{and} \quad S_{ij} = \sqrt{S_i S_j}$$
where the indices $i, j$, and $k$ are used to label the atoms in the system, $r_{ij}$ denotes the distance between atoms $i$ and $j$, and $\theta_{jk}$ is the bond formed between distance vectors $r_{ij}$ and $r_{ik}$ at vertex $i$. Single subscripted parameters, such as $\lambda$ and $n$, depend only the type of the atom. The Tersoff potential has been used quite successfully for modeling the solid-state mechanics and surface physics of semiconductor materials.

2.2. Atomistic Simulations of the Mechanical Behavior of Ultrathin Metallic Films

A significant part of this Ph.D. thesis research focuses on identification and analysis of fundamental mechanisms of strain relaxation and associated deformation processes in ultrathin metallic films. Toward this end, a computational strategy was developed to simulate strain relaxation in metallic thin films. Isothermal-isostrain MD simulations as well as dynamic deformation tests at constant applied strain rate and temperature were used to study the atomistic mechanisms of material response and the ensuing defect dynamics. In the MD simulations, the classical equations of motion were integrated using the velocity Verlet algorithm (Allen & Tildesley, 1990). The time-step used during the simulations conducted in this thesis is $8.12 \times 10^{-16}$ s and has been tested carefully for the accuracy and numerical stability of the algorithms. Since the focus of this study is not on thermal effects on the underlying deformation dynamics, the models of the films were maintained at a low constant temperature of 100 K by explicitly rescaling the velocities of all the atoms in the system at each time step. Since the strain imparted in the thin films under experimental conditions is planar, the focus of this thesis is on on biaxial straining, as well as the relaxation of biaxial strain in
freestanding thin fcc metallic thin films. In the MD simulations, slab supercells are employed with periodic boundary conditions applied in the $x$- and $y$-directions, while the boundaries in the $z$-direction in a Cartesian representation are free surfaces. Biaxial tensile strain is applied by expanding the edges of the simulation cell along the $x$- and $y$-directions. Large-scale MD simulations were performed with the number of atoms in the simulation cell varying from 350,000 to a few million. In the present thesis, ultrathin Cu films with film surface planes normal to the [111] and [1\bar{1}0] crystallographic directions are studied; the orientation of the film surface is motivated both for comparisons with available experimental observations and because the [111]-oriented surface has the highest symmetry while the [1\bar{1}0]-oriented one has the lowest symmetry in fcc metals among low-index crystallographic orientations. Additionally, the dynamic deformation of ultrathin metallic films of Ni and Al are studied in order to compare differences in the mechanical behavior of thin films of different fcc metals (Cu, Ni, and Al) and correlate these differences to the differences in their microstructure. In addition to developing in-house computer programs, public-domain software LAMMPS (Plimpton, 1995) was used for MD simulations of dynamic deformation at constant strain rate and temperature.

Finally, a dislocation mean-field theoretical model was implemented in order to study energetics and kinetics of strain relaxation in semiconductor heterostructures. In this context, preliminary simulation studies were conducted, as a proof-of-concept test, to show that parameterization based on atomistic simulations of the dislocation mean-field theoretical model can substantially improve the quantitative accuracy of the
model’s prediction. The computational strategy implemented in these studies is outlined systematically in Chapter 6.

2.3. Structural Analysis of MD-Generated Atomic Configurations

Common neighbor analysis (CNA) is employed in order to categorize the atoms according to their local environment (Honeycutt & Anderson, 1987). CNA can be used to identify atoms in locally perfect fcc and hcp environments. Two planes of atoms in a locally perfect hcp environment within an fcc lattice constitute an intrinsic stacking fault. Two planes of atoms in a locally perfect hcp environment separated by a plane of atoms in a locally perfect fcc environment constitute an extrinsic stacking fault. A single plane of atoms in a locally perfect hcp environment constitutes a twin boundary. Point and line defects in the films studied are identified by calculating the coordination number, \( Z \), of each atom; lines of atoms with \( Z = 12 \) that are neither in a locally perfect fcc nor in a locally perfect hcp environment, as well as atoms with coordination defects \( (Z \neq 12) \) other than isolated point defects or surface atoms are associated with dislocations.

Dislocations and dislocation-dislocation interactions were characterized with the aid of the Thompson tetrahedron (Hirth & Lothe, 1982; see also Appendix A). In order to analyze the Burgers vectors of glissile dislocations, slip vector analysis was used, where the slip vector is analogous to the Burgers vectors. The slip vector of an atom, \( \alpha \), in the thin film is given by
\[ s^\alpha = -(1 / n_s) \sum_{\beta \neq \alpha}^{n_s} (x^{\alpha \beta} - x^{\alpha \beta}) \]  

(2.9)

where \( x^{\alpha \beta} \) and \( X^{\alpha \beta} \) are the distance vectors between atoms \( \alpha \) and \( \beta \) (from \( \alpha \) to \( \beta \)) in their current and reference configurations, respectively, and \( n_s \) is the number of neighbors of atom \( \alpha \) that have undergone slip (Zimmerman et al., 2001); in the analysis of mechanical behavior of Cu thin films, atom \( \alpha \) is considered to have slipped with respect to atom \( \beta \) if \( |x^{\alpha \beta} - x^{\alpha \beta}| > 0.4 \ \text{Å} \). The slip vectors of atoms that have been swept by Shockley partial dislocations and gliding perfect dislocations are \((a/6) \langle 112 \rangle\) and \((a/2) \langle 110 \rangle\), respectively. Slip vector analysis cannot identify the Burgers vectors of dislocations that do not glide, such as stair-rod dislocations; sessile dislocations are often the products of interactions between glissile dislocations and can be identified easily once the Burgers vectors of the reactant and product dislocations are determined by slip vector analysis. Finally, public-domain software ATOMEYE (Li, 2003) was used in order to visualize MD-generated atomic configurations for analyzing the underlying plastic-deformation mechanisms.

2.4. Computing Material Properties from MD-Generated Atomic Configurations

Stress, dislocation density, stacking-fault surface area, and thin-film surface roughness are some of the important properties that are monitored in dynamic straining and strain relaxation of metallic thin films. The stresses in the thin films studied are computed using the virial formulation (Allen & Tildesley, 1990), according to which
\[
\sigma_i = \frac{1}{\Omega} \left( -m_i \mathbf{r}_i \otimes \dot{\mathbf{r}}_i + \frac{1}{2} \sum_{j \neq i} \mathbf{r}_{ij} \otimes \nabla_{ij} U(\mathbf{r}_{ij}) \right),
\]  
(2.10)

where \( \Omega \) and \( \sigma_i \) are the atomic volume of and the stress tensor acting on the atom labeled \( i \), respectively, and \( \otimes \) denotes a dyadic (tensor) product.

For the EAM formalism leading to Eq. (2.2), the stress on each atom is given by

\[
\sigma_i = \frac{1}{\Omega} \left( -m_i \dot{\mathbf{r}}_i \otimes \mathbf{r}_i + \sum_{j \neq i} \left[ \frac{1}{2} \frac{\partial U}{\partial \mathbf{r}_j} + \frac{\partial \phi}{\partial \rho(\mathbf{r}_{ij})} \frac{\partial \rho(\mathbf{r}_{ij})}{\partial \mathbf{r}_j} \right] \mathbf{r}_{ij} \otimes \mathbf{r}_{ij} \right),
\]  
(2.11)

Since the thin films are biaxially strained equally in each lateral dimension, multiple dislocation slip systems are activated. Under such circumstances, the von Mises stress, \( \sigma_{vM} \), which is the second invariant of the stress tensor, is a good measure of the mechanical state of the material; \( \sigma_{vM} \) is given by

\[
\sigma_{vM} = \frac{1}{\sqrt{6}} \sqrt{\left(\sigma_{xx} - \sigma_{yy}\right)^2 + \left(\sigma_{yy} - \sigma_{zz}\right)^2 + \left(\sigma_{zz} - \sigma_{xx}\right)^2 + 6\left(\sigma_{xy}^2 + \sigma_{yz}^2 + \sigma_{zx}^2\right)}
\]  
(2.12)

where \( \sigma_{ij} (\alpha, \beta = x, y, \text{or } z) \) are the elements of the stress tensor averaged for the entire thin film.

Two methods were implemented for the computation of the film’s volume, which is involved in computing the stress, as well as the dislocation density; computing the volume, \( V = \sum_i \Omega_i \), of a thin film that has free surfaces is not a trivial task. In the first method, a grid of multiple elements of known lateral dimensions were constructed on the thin-film surface; the thickness of each such element was computed as the distance, in \( z \), between the two atoms on the opposite surfaces of the film that were the farthest located from its center plane on each surface within the element. The total
volume of the film was then computed as the sum of the element volumes. In the
second method, thickness of the film was estimated as the distance, in \( z \), between the
two atoms on each of the opposite film surfaces that were the farthest located from the
center plane of the film. The first method underestimates the total film volume, while
the second method overestimates it. The stress values reported in the stress-strain curve
plots of this thesis (Chapter 5) were computed using the second method. Irrespective of
the method used for film volume computation, the corresponding qualitative features of
the stress evolution in dynamic deformation remained identical except at a stage close
to failure; at this stage, the volume computed according to the first method excludes
(incorrectly) the pits formed on the surface. The difference in the stress values
computed by the two methods were \(~100-200\) MPa; the first method yielded
consistently higher stress values.

Dislocation density is computed as \( \rho = l/V \) where \( l \) is the total length of the
dislocations in the film. Atoms with \( Z = 12\) that are neither in locally perfect fcc or
locally perfect hcp environments make up the dislocation “core.” Strictly speaking, the
dislocations including the stacking faults that they bind form the core of the dislocation.
Here, the term “core” is used loosely and corresponds to the atoms that are neither in
fcc nor hcp arrangements. The stacking fault area is defined as

\[
A_{sf} = \left( \frac{N_{hcp}}{2} \right) \times (1/3) \times \left( 3\sqrt{3}a^2 / 4 \right),
\]

where the first factor is the number of hcp atoms on one of the two planes of the
stacking fault, the second factor is the number of atoms per hexagonal unit of the
stacking fault lattice plane, and the third factor is the area of each such hexagonal unit,
where \( a \) is the lattice parameter of the fcc lattice of the materials studied.
While the dislocation density and stacking-fault area computations described above are rigorous, they are time consuming. The percentages of atoms that are in the dislocation cores and the percentages of hcp atoms provide equally good metrics for monitoring the evolution of dislocation densities and stacking-fault areas, respectively; in this thesis, both of these computational procedures are used to monitor the evolution of the thin films’ microstructure.

2.5. Limitations of MD Simulations of Dynamic Straining and Strain Relaxation

Although the simulations conducted in this thesis address relatively simple model systems, they provide insights into mechanisms that cannot be observed directly in experiments; as a result, the simulation predictions are valuable in designing experimental and computational protocols to understand further the role of dislocation interactions in the mechanical behavior of ultrathin metallic films. There are, however, serious limitations regarding length and time scales that can be accessed directly by MD simulation. Consequently, size effects (e.g., effects of film thickness) on the mechanical behavior were not analyzed systematically. Additionally, MD simulations are limited to very high strain rates that are typically greater by several orders of magnitude than those applied in dynamic deformation experiments. Hence, it is imperative that the characteristic stresses and time scales associated with the deformation-rate-limiting dynamical processes be computed and compared with the corresponding experimentally measured material parameters.
In the MD simulations conducted in this thesis, the highest shear stresses computed were far below the ideal shear strength of the material; the maximum shear stresses observed in the dynamic deformation simulations were up to 45% of the ideal shear stress. Moreover, the maximum dislocation velocities computed in the MD simulations conducted in this thesis were \(~1200\) m/s in isostrain simulations and \(~400\) m/s in dynamic deformation simulations on Cu films, which is well below the sound velocity in the material (\(~3700\) m/s for the Cu potentials used in this thesis and for unstrained samples). These comparisons suggest that the high strain rates applied in the MD simulations of dynamic deformation conducted in this thesis do not affect the underlying dislocation dynamics. The dislocation densities in the studies within this thesis are on the order of \(10^{17}\) m\(^{-2}\), which is considerably higher than the dislocation densities that are typically observed in experiments. High dislocation densities in MD simulations are, however, inevitable; for example, the resulting dislocation density with only one dislocation extended across the film thickness is \(~7.9 \times 10^{14}\) m\(^{-2}\) in the largest-area thin films used in the simulations conducted in this thesis.
CHAPTER 3

ATOMISTIC ANALYSIS OF BIAXially STRAINED ULTRATHIN COPPER FILMS: FILM SURFACE 
ORIENTED ALONG THE [111] CRYSTALLOGRAPHIC DIRECTION

3.1. Introduction

This study focuses on the identification and analysis of fundamental mechanisms of strain relaxation and the associated deformation processes in ultrathin metallic films. Toward this end, large-scale MD simulations of metallic thin-film response to the application of tensile biaxial strain were implemented. In the MD simulations conducted in this study, slab supercells were employed with periodic boundary conditions applied in the $x$- and $y$-directions in a Cartesian representation where the boundaries in the $z$-direction were traction-free surfaces; the Cartesian $x$, $y$, and $z$ axes were along the $[\bar{1}\bar{1}0]$, $[\bar{1}\bar{1}2]$, and [111] crystallographic directions, respectively. The choice of the film surface parallel to the (111) crystallographic plane was partly motivated by the common use of textured metallic thin-film interconnects with preferred $\langle 111 \rangle$ grain orientation (Thompson, 1993). For the results reported here, two simulation cells were considered: the smaller one contained 370,986 atoms and had edge sizes of 30.9, 32.3, and 4.4 nm in the $x$-, $y$-, and $z$-directions, respectively, and the larger one contained 1,467,690 atoms and had edge sizes of 61.6, 64.2, and 4.4 nm in
the x-, y-, and z-directions, respectively; in both cases, the cell dimensions given correspond to the unstrained state of the metallic thin film.

The simulations were performed at a constant temperature of 100 K at every given applied strain level. Initially, the supercell was unstrained, i.e., the cell-edge sizes corresponded to the equilibrium density at the simulation temperature. The metallic thin film was strained biaxially by expanding the supercell along the x- and y- directions up to the chosen strain level at a rate of approximately 3×10^{11} \text{s}^{-1}; specifically, a strain increment of 1\% (Δε = 0.01) was applied over a time period of 2.983×10^{-14} \text{s}, yielding a strain rate of 3.352×10^{11} \text{s}^{-1}. The effects of the strain rate were examined over a one-order-of-magnitude range; within this range, the atomistic mechanisms were not affected by the strain rate.

This chapter is structured as follows. The thin-film mechanical response to applied biaxial strain over the strain range studied are discussed in detail in Sec. 3.2. The elementary mechanisms of dislocation nucleation and propagation leading to strain relaxation are discussed in Sec. 3.3. The collective dislocation dynamics and its effects on thin film’s structural evolution are discussed in Sec. 3.4 and, finally, the main conclusions of this study are summarized in Sec. 3.5.

3.2. Thin-film Mechanical Response to Applied Biaxial Strain

In order to determine the fundamental mechanisms of strain relaxation and associated defect dynamics in biaxially strained ultrathin metallic films, mechanical
response was simulated of fcc metallic thin films over the range of tensile biaxial strain levels from 0% to 12%. The response of the thin film is found to be elastic up to a biaxial strain level $\varepsilon = 5.5\%$. Beyond this critical $\varepsilon$(yield strain), the biaxial strain in the metallic thin film is relaxed by plastic deformation mechanisms. This yield strain is rather large for the initially defect-free thin film; the yield strain of a Cu thin film with pre-existing voids modeled with same interaction potential and for the same mode of straining was $\varepsilon \approx 2\%$ (Gungor & Maroudas, 2005a). The presence of a pre-existing void in the thin film creates a high strain concentration in the vicinity of the void and causes dislocation emission from the void surface, thus lowering the yield strain.

The computed evolution of the potential energy per atom, $E(t)$, is plotted in Fig. 3.1(a) for various applied strain levels over the range $6\% \leq \varepsilon \leq 12\%$. Identical MD simulations were carried out for both the small and the large supercell described in Sec. 3.3; the corresponding $E(t)$ evolution curves were identical for both supercell models, i.e., for both smaller and larger film surface areas, indicating clearly that the mechanisms of strain relaxation observed are independent of the surface area of the slab model. The initial oscillations of the potential energy in Fig. 3.1(a) are characteristic of the elastic response of the thin film. Since the strain distribution is not perfectly uniform in the thin film during the course of a MD simulation, dislocations are emitted at the high strain concentration locations of the film’s surface at and beyond the onset of the elastic-to-plastic transition of the thin-film mechanical response in order to relax the strain. For strain levels above the yield strain, in Fig. 3.1(a), the elastic oscillations that are observed during the initial response of the thin film are attenuated subsequently by the generation of lattice defects. With increasing applied biaxial strain $\varepsilon \geq 10\%$, the
density of high-strain-concentration regions increases and such regions are distributed uniformly throughout the thin film; as a result, the elastic response time decreases and plastic deformation occurs at a faster rate, which reduces the total time required for biaxial strain relaxation.

Figure 3.1(b) shows the elastic response time (blue curve) and the total time to strain relaxation (black curve) of the thin films as a function of $\varepsilon$ for films subjected to different biaxial strains. The elastic response time decreases monotonically with $\varepsilon$ up to an applied biaxial strain level $\varepsilon = 14\%$, above which plastic response of the thin film occurs almost instantaneously; this suggests that the deformation caused by such high strain levels leads to the formation of a very high density of defects, thus suppressing the elastic response. Also, the total time for strain relaxation in the thin film decreases monotonically with $\varepsilon$ up to $\varepsilon = 10\%$ and remains almost constant for applied biaxial strain levels over the range $10\% < \varepsilon < 16\%$. This constant relaxation time suggests that the mechanisms of strain relaxation, which include dislocation nucleation mechanisms and dislocation-dislocation interaction processes, remain mostly the same over this strain range; this is demonstrated and discussed in Sec. 3.3. For $\varepsilon > 16\%$, regions with very high point-defect density develop in the thin film. Such dense point-defect clusters require longer time to anneal and, hence, the total time for relaxation of the applied strain increases.
Figure 3.1. (a) Evolution of potential energy (per atom), $E$, during biaxial strain relaxation; different evolution curves correspond to different applied biaxial strain levels. The response of the film is elastic up to a strain level of almost 5.5% and the response curves in the elastic deformation regime are not plotted. In the evolution curves shown, the observed oscillatory dynamics corresponds to the initial elastic response to the applied biaxial strain. (b) Dependence on the applied biaxial strain level, $\varepsilon$, of the strain-relaxation time of the thin film and the time for which the thin film exhibits an elastic response to the applied biaxial strain.

The surface morphologies of the thin films subjected to biaxial strains with $\varepsilon = 6\%, 7\%, \text{and } 8\%$ are shown in Fig. 3.2(a). In this depiction of surface morphology, the atoms in the thin films are colored according to their coordinate normal to the center plane of the film. Visual inspection of the strain-relaxed thin-film microstructure in Fig. 3.2(a) reveals nucleation of voids at low strain levels above the yield strain; specifically, pits were observed on the surface of the thin film that extend across the thickness of the film over the range $5.5\% < \varepsilon < 6.4\%$. As the applied strain increases, plastic
deformation becomes more uniform; the depth of the surface pits decreases with increasing $\varepsilon$ for $\varepsilon > 6.4\%$ and void nucleation is inhibited.

The evolution of surface roughness, $w(t)$, defined as the root-mean-squared fluctuation of the film thickness, is plotted in Fig. 3.2(b) for applied biaxial strain levels over the range $6\% \leq \varepsilon \leq 12\%$. At such strain levels, plastic deformation in the thin film is mediated by the nucleation, glide, and interactions of dislocations; the surface forms grooves, which are the primary cause of roughening of the thin film’s surface, due to the glide of dislocations and the migration of point defects to the surface. With increasing $\varepsilon$, the more uniform distribution of defects and plastic deformation in the film leads to a smoother film surface.

Figure 3.2(c) shows the final (steady-state) values of the surface roughness of the strain-relaxed thin films, $w_{ss}$, as a function of $\varepsilon$. At low $\varepsilon$ levels above the yield strain, point defects are generated randomly at the surface. Over the range $5.5\% < \varepsilon < 6.4\%$, multiple threading dislocations are emitted from discrete locations on the film surface. The surface step traces formed by gliding dislocations on intersecting and adjacent parallel glide planes lead to the formation and growth of surface pits and grooves that lead eventually to formation of voids extending across the film thickness. Formation of such surface pits and voids increases the surface roughness in the thin films, as can be seen clearly in Figs. 3.2(a) and 3.2(b) and from the abrupt increase of $w_{ss}$ in Fig. 3.2(c). For $\varepsilon > 6.4\%$, the population of threading dislocations emitted from the film surface increases and the dislocations are distributed more uniformly relative to their distribution at lower strain levels; such uniform distribution of defects inhibits the
formation of voids that extend across the thin film. The surface roughness, therefore, decreases as the deformation becomes more uniform across the thin film.

Figure 3.2. (a) Top view of the thin-film surface after strain relaxation for different applied biaxial strain levels, $\varepsilon$. The atoms are colored according to their height in the surface topography; greater color contrasts correspond to rougher film surfaces. (b) Evolution of the surface roughness, $w$, during biaxial strain relaxation; different curves correspond to different applied biaxial strain levels over the range $6\% \leq \varepsilon \leq 12\%$. (c) Dependence on $\varepsilon$ of the surface roughness, $w_{ss}$, of the strain-relaxed thin films; $w_{ss}$ corresponds to the steady-state value reached in the $w(t)$ evolution shown in (b).

For $\varepsilon \geq 10\%$, the surface roughness remains almost constant with increasing $\varepsilon$ due to the increased uniformity and saturation of defect nucleation in the thin film. Over this range of applied strain levels, $w_{ss}$ remains almost constant, $1.1$ Å $\leq w_{ss} \leq 1.7$ Å. However, under biaxial strains at levels higher than those that this study focuses on ($\varepsilon > 20\%$), thin-film deformation may lead to fracture eventually, resulting also in an abrupt change in the film surface roughness as a function of $\varepsilon$. The surface roughness
curves are found to be identical for both of the supercell models (smaller and larger) employed in this study. The mechanisms of dislocation nucleation and dislocation-dislocation interactions are discussed in sections 3.3 and 3.4.

**Figure 3.3.** (a) Evolution of defect atoms with \( Z = 12 \) during biaxial strain relaxation; different curves correspond to different applied biaxial strain levels over the range \( 6\% \leq \varepsilon \leq 12\% \). (b) Dependence on \( \varepsilon \) of the percentages of various kinds of atoms (fcc, hcp, and \( Z = 12 \) defect atoms) in strain-relaxed thin films. (c) Top view of the thin films after relaxation of the applied biaxial strain, showing the resulting dislocation density; dark and light blue atoms are in local perfect fcc and hcp arrangements, respectively. Atoms with other colors correspond to point defects, as well as atoms in the dislocation cores. Surface atoms are not shown for clarity.

In order to elucidate the mechanisms of strain relaxation, including nucleation and glide of dislocations and dislocation-dislocation interactions, this study was focused
on the strain range $5.5\% \leq \varepsilon \leq 12\%$. Five regimes were identified in the thin film’s mechanical response to the applied tensile biaxial strain:

(i) Elastic response for applied strain levels up to $\varepsilon \approx 5.5\%$;

(ii) Threading dislocation nucleation from the surface of the thin film, leading eventually to nucleation of voids for applied strain levels over the range $5.5\% < \varepsilon < 6.4\%$;

(iii) Nucleation of threading dislocations, distributed uniformly, at the surface of the thin film for applied strain levels over the range $6.4\% < \varepsilon < 8.0\%$;

(iv) Nucleation of dislocation loops from the bulk of the film in addition to the mechanism of regime (iii) for applied strain levels over the range $8\% < \varepsilon < 10\%$; and

(v) Nucleation of Frank partial dislocations by collapse of point defects in addition to the mechanisms of regimes (iii) and (iv), leading eventually to formation of nano-scale domains in the film for $\varepsilon \geq 10\%$.

The evolution of the percentage of atoms in the thin film with $Z = 12$ that are neither in an fcc nor in an hcp local environment is plotted in Fig. 3.3(a); different curves correspond to different levels of applied biaxial strain, $\varepsilon$. Such atoms surround point and line defects and provide a good measure of the defect density in the material. In the second regime of the film’s mechanical response, $5.5\% < \varepsilon < 6.4\%$, as the threading dislocation loops emitted from the surface expand, they either intersect with the opposite film surface and dissociate into two partial dislocations, or they interact with another threading dislocation that is nucleated inside the threading dislocation loop.
to form perfect screw dislocations (the corresponding mechanistic details are discussed in Sec. 3.3). The gliding dislocations create high strain concentrations at the surface of the thin film leading to the nucleation of more threading loops; eventually, the defect population reaches a steady-state value. In the third regime, $6.4% < \varepsilon < 8\%$, threading dislocations emitted uniformly across the surface of the thin film increase the rate of generation of the defect population. In the fourth regime, $8\% < \varepsilon < 10\%$, dislocation loops are emitted in the bulk of the thin film in addition to nucleation of dislocations at the film surface. The activation of this second mechanism of dislocation nucleation increases the rate of defect generation compared to that in the third regime, $6.4% < \varepsilon < 8\%$. After reaching a maximum, this defect population is reduced by annealing of point defects at the surface and in the bulk and by annihilation of dislocations as a consequence of various dislocation processes and dislocation-dislocation interactions; dislocation annihilation leads eventually to a steady value of the dislocation density. Finally, in the fifth regime, $\varepsilon \geq 10\%$, a very high density of point defects is generated, leading to an increased rate of generation of the defect population compared to that at lower applied strains. The supersaturated point defects collapse quickly to form Frank partials that dissociate subsequently, generating numerous stacking fault tetrahedra in the film.

The percentages of various types of atoms are plotted in Fig. 3.3(b) as a function of $\varepsilon$ after $t = 40.6$ ps, i.e., after steady state has been reached. Red, blue, and green curves correspond to the percentages of fcc atoms, hcp atoms (stacking faults), and $Z = 12$ structural defects, respectively. The different regimes in the film’s mechanical
response, (i)-(v), can be identified in the curves of Fig. 3.3(b). Figure 3.3(c) shows top views of the strain-relaxed thin films (at $t = 40.6$ ps) for films subjected to different biaxial strain levels. The dark and light blue atoms are in locally perfect fcc and hcp lattice arrangements, respectively; other atoms are in dislocation cores, while surface atoms are not shown for clarity. For applied strain levels up to $\varepsilon = 10\%$, the average stacking-fault length (area in 3D) were observed to decrease and the stacking-fault density were observed to increase with increasing $\varepsilon$; for $\varepsilon > 10\%$, the stacking-fault and dislocation densities are almost the same and visual inspection of the thin-film microstructure reveals no observable differences in the texture of the thin-film material.

At high strains ($\varepsilon \geq 10\%$), dislocation activity and dislocation-dislocation interactions lead to the formation of complex stable dislocation junctions, including Lomer-Cottrell locks. Such dislocation activity causes lattice rotations and leads to the formation of nano-scale domains in the thin films, which makes the film structure resemble that of a nanocrystalline film although the film remains single-crystalline. Two different views of a section of the surface of a strain-relaxed thin film subjected to $\varepsilon = 12\%$ are shown in Figs. 3.4(a) and 3.4(b). Maroon colored atoms are the surface atoms with $Z = 9$ and all the other atoms on the surface, colored orange and red, are traces of dislocation activity or locations where point defects are annealed. The atoms corresponding to the traces of dislocation activity act as boundaries between the nano-scale domains; detailed characterization yields an average domain size of approximately 1.5 nm with misorientation angles varying from $1.5^\circ$ to $4.5^\circ$ (Gungor & Maroudas, 2005b).
Figure 3.4. Top views of a section of a strain-relaxed thin film that has been subjected to a high strain (ε = 12%), showing the formed nanodomains and their misorientations. Maroon colored atoms are surface atoms with Z = 9; other atoms mark traces of dislocation activity or locations where point defects are annealed.

3.3. Mechanisms of Strain Relaxation

In this section, the dislocation nucleation and glide mechanisms observed during the strain relaxation of the Cu thin films are analyzed. The properties of the dislocations emitted and their corresponding glide planes are characterized using the Thompson tetrahedron (Hirth, 1982); a schematic of the (111) face of the Thompson tetrahedron is shown in Fig. 3.5(a). Dislocations with Burgers vectors in the [011] direction are labeled DB; perfect dislocations are labeled DB, AB, and AD, while partial dislocations are labeled Dγ, Bγ, and Aγ. This analysis of elementary mechanisms of dislocation generation and reactions is carried out primarily on the (111) glide plane; however, these mechanisms have been observed in all of the {111} glide planes in the fcc lattice and for all the dislocations with the corresponding Burgers vectors.
At low strain levels above the yield strain (5.5% < \( \varepsilon < 8\% \)), nucleation and glide of threading dislocation loops at the film surface are identified and characterized as the main mechanism of dislocation generation and propagation. Figure 3.5 also shows a three-dimensional (3D) close view of dislocation evolution. In Figs. 3.5(b), (c), and (d), atoms are colored according to both their atomic coordination and the corresponding local lattice arrangement, using CNA to aid in the identification of point and line defects. Dark blue atoms are in a locally perfect hcp lattice arrangement, maroon atoms are those at the film surface, light blue atoms have \( Z = 13 \), and light green atoms have \( Z = 11 \). For clarity, atoms in locally perfect fcc environments are not shown in Fig. 5.

Threading dislocations nucleate at locations of high strain concentration on the film surface, as shown in Fig. 3.5(b), following an initially elastic response. The threading dislocation half-loop shown in Fig. 3.5(b) consists of two Shockley partial threading arms, each \( \gamma_B \), with Burgers vectors \( \mathbf{b} = (a/6)[121] \), where \( a \) is the fcc lattice parameter. As this first threading dislocation extends into the bulk material of the film, a second threading dislocation half-loop is nucleated inside the stacking fault bounded by the first threading dislocation, and extends to unzip the stacking fault created by the first threading dislocation as shown in Fig. 3.5(c). This second threading dislocation half-loop also consists of two Shockley partial threading arms, each \( \mathbf{D\gamma} \), with Burgers vectors \( \mathbf{b} = (a/6)[112] \). As these threading dislocation loops glide, they leave step traces on the film surface due to the shearing of the lattice and generate point defects, as seen in Fig. 3.5(c). These point defects lead to nucleation of more threading dislocations at the same surface site. The inner threading loop of Fig. 3.5(c) extends and it interacts
eventually with the outer dislocation half-loop; as a result, two perfect screw dislocations, each characterized as \( \text{DB} \), are formed with Burgers vectors \( \mathbf{b} = (a/2)[011] \). Both of the perfect screw dislocations are observed to have jogs and one of them glides on the \( (\bar{1}11) \) plane, while the other one glides on the \( (\bar{1}1\bar{1}) \) plane, as shown in Fig. 3.5(d).

![Diagram](image)

**Figure 3.5.** (a) A \{111\} plane of the Thompson tetrahedron; (b)-(d) 3D close view of biaxially strained thin-film material (\( \varepsilon = 6\% \)) showing the evolution of a threading dislocation from one surface of the film, as it extends through the film and intersects with the other film surface. Dark blue atoms are in perfect hcp lattice arrangements. Light green atoms have \( Z = 11 \) and light blue atoms have \( Z = 13 \). Surface atoms are colored maroon and atoms in locally perfect fcc lattice positions are not shown for clarity. The corresponding time, \( t \), in the MD simulation is recorded, starting from the film in its unstrained state.
**Figure 3.6.** (a)-(d) A \{111\} view of biaxially strained thin-film material (\(\varepsilon = 8\%\)) showing the nucleation and evolution of a dislocation from the bulk of the film, as it grows and intersects with one of the film surfaces becoming a threading dislocation loop. Dark blue and light blue atoms are in perfect fcc and hcp lattice arrangements, respectively. Light green atoms have \(Z = 13\) and yellow atoms have \(Z = 11\). Surface atoms are colored maroon. (e)-(h) 3D close view of the same biaxially strained thin-film material for the same evolution sequence shown in the top row of configurations, (a)-(d), wherein atoms in perfect fcc lattice arrangements are not shown for clarity. The corresponding time, \(t\), in the MD simulation is recorded, starting from the film in its unstrained state.

At \(\varepsilon \geq 8\%\), in addition to the nucleation of threading dislocations at the surface of the thin film, the activation of a second mechanism was observed: nucleation of dislocation loops inside the bulk material of the film. The nucleation and evolution of such a dislocation loop is shown in Fig. 3.6. Figures 3.6(a)-(d) show the cross-sectional view (normal to the \([\bar{1}1\bar{1}]\) crystallographic direction) of the thin-film region, where the dislocation is nucleated. 3D views of the same region of the film from a different angle are shown in Figs. 3.6(e)-(h). The strain in the thin film causes atoms in the bulk to shear along \{111\} planes, generating a cluster of atoms, characterized by a high strain.
concentration, that are not in a locally perfect fcc environment as shown in Figs. 3.6(a) and (e). Upon further shearing, a dislocation loop is nucleated, as shown in Figs. 3.6(b) and (f); the loop grows to reach eventually one of the film surfaces, as shown in Figs. 3.6(c) and (g). This dislocation is characterized to be a Shockley partial, \( \mathbf{b}_y \), with Burgers vector \( \mathbf{b} = (a/6)[12\bar{1}] \). After reaching the surface, the threading dislocation dynamics is observed to be similar to the dynamics described in the previous regime, Fig. 3.5; specifically, after the dislocation loop reaches the surface of the film, a second threading dislocation is nucleated inside the first half loop, unzipping as it glides the stacking fault formed by the first dislocation loop.

At \( \varepsilon \geq 10\% \), a third mechanism of dislocation nucleation is activated. The application of biaxial strain higher than 10\% leads to a dramatic increase in the initial concentration of point defects in the thin film, which becomes evident upon careful film structural characterization. The pair correlation function, \( g(r) \), is plotted in Fig. 3.7(a) for a thin film strained to \( \varepsilon = 12\% \) over a time period of 0.358 ps. The red curve is the \( g(r) \) just before the defect concentration reaches a maximum in the thin-film material [see, e.g., Fig. 3.3(a)] at \( t = 1.624 \) ps and the green curve is the \( g(r) \) after the strain has been relaxed fully at \( t = 48.72 \) ps. The generation of a high concentration of point defects leads to loss of short-range order (focusing on \( r \leq 2.5a_0 \), where \( a_0 \) is the equilibrium lattice parameter of Cu at 0 K) in the initially perfect lattice structure of the thin film; this is evident by the absence of characteristic peaks in the \( g(r) \) at \( t = 1.624 \) ps that would correspond to specific coordination shells in the crystalline lattice structure and it is consistent with point-defect-induced amorphization. As the strain is relaxed,
however, the thin film recovers its crystalline order. The point defects generated by the application of high strains collapse quickly to form Frank partials (Hirth & Lothe, 1982), which bound stacking faults that are formed not as a result of any dislocation glide. The atomic shearing rearrangements in the process of formation of dislocations during the collapse of point defects is normal to \{111\} planes, unlike the earlier mechanisms in the previous two regimes where the atoms shear along \{111\} planes to form dislocation loops. Due to the high concentration of point defects and dislocations, these Frank partials dissociate quickly to form each a stair-rod dislocation and a glissile Shockley partial leading to the formation of stacking-fault tetrahedra (Hirth & Lothe, 1982).

Several stacking-fault tetrahedra are observed in the initial stages of strain relaxation. Most of these tetrahedra are only partially formed due to the very high defect density. Representative configurations of partial stacking-fault tetrahedra are shown in Figs. 3.7(b), (c), and (d), each of which is a top view of a section of the thin-film material during the initial stages of strain relaxation. In Figs. 3.7(b) and (c), surface atoms are not shown for clarity; in Fig. 3.7(d), for clarity, in addition to surface atoms, atoms in locally perfect fcc lattice arrangements are not shown. Figure 3.7(d) shows clearly the base of the tetrahedron formed by the stacking faults and the stair-rod dislocations at the intersection (marked by the arrow) of the stacking faults. Finally, it should be noted that the \( g(r) \) of Fig. 3.7(a) suggests that the relaxation of metallic thin films subjected to high biaxial strains is analogous to the quenching of metallic melts. Formation of stacking-fault tetrahedra during rapid quenching of metallic melts has been observed in numerous experiments (Hirth & Lothe, 1982).
**Figure 3.7.** (a) Pair correlation function, $g(r)$, of the thin-film material during application of biaxial tensile strain at a level $\varepsilon = 12\%$; red and green curves correspond to the $g(r)$ before and after strain relaxation. (b)-(c) Top views of the thin-film material during the initial stage of strain relaxation. Dark blue and green colored atoms are in perfect fcc and hcp lattice positions, respectively. Other colored atoms are associated with point defects and dislocations. Surface atoms are not shown for clarity. (d) Same top view as in (b) and (c), where neither surface atoms nor atoms in perfect fcc lattice arrangements are shown for clarity. Light blue atoms are in a local hcp lattice arrangement. (b)-(d) are snapshots at a time instant $t = 3.256$ ps in the MD simulation.

### 3.4. Dislocation-Dislocation Interactions and their Effects

With increasing applied strain level, the dislocation density increases and the gliding partial dislocations evolve subjected to various dislocation-dislocation interactions; these interactions are analyzed in detail in this section. For $\varepsilon = 6\%$, visual inspection of strain-relaxed thin films reveals nucleation of a single void for the thin-film model with smaller surface area and nucleation of multiple voids for that with
larger surface area, suggesting a strong correlation between the film surface area and the number of voids nucleated; the higher strain energy stored in the thin film with the larger surface area requires the nucleation of a larger number of voids for its relief. The slab supercell with smaller (30.9 × 32.3 nm²) surface area was selected for the detailed analysis of atomistic mechanisms that mediate void nucleation and growth, in order to focus on a single void in the supercell. Figure 3.8 shows in detail the structural evolution of the thin-film material for ε = 6%. The top views in Figs. 3.8(a)-3.8(d) show the surface area of the entire simulation cell, where atoms are colored according to their von Mises shear strain invariant (Li, 2000). Figures 3.8(e)-3.8(h) show cross-sectional views of the thin film on the xz-plane; these are extracted from the region of the film where the void nucleates, which is marked in Fig. 3.8(a). In Figs. 3.8(e)-3.8(h), atoms are colored according to both their local lattice environment using CNA and Z to identify various types of defects. Dark blue and light blue atoms are locally in perfect fcc and hcp environments, respectively, while other atoms are associated with point defects or dislocations, or they are surface atoms. A 3D close view of the thin film is shown in Figs. 3.8(i)-3.8(l), where atoms colored in maroon, light blue, and light green are surface atoms with Z = 9, atoms with Z = 10, and atoms with Z ≤ 8, respectively. Clusters of atoms with Z ≤ 10 correspond to vacancies and vacancy clusters. Atoms with Z = 11, Z = 12, and Z = 13 are not shown for clarity. Jogged threading dislocations are emitted from such high-strain-concentration locations at the surface. This mechanism of nucleation of threading dislocation is the same as the first nucleation mechanism described in Sec. 3.3. Subsequent dislocation glide and interactions between dislocations lead to relaxation of strain in the thin film, as shown in Figs.
3.8(b)-3.8(d). The threading dislocations emitted from each side of the film propagate into the film and grow to reach eventually the opposite film surface; then, the middle segment of the loop is annihilated and a stacking fault is formed on the slip plane extending across the film and bounded by the two Shockley partials. Shearing of the thin film along the slip planes by the gliding dislocations leads to formation of step traces on the film surface. Intersection of step traces formed by the glide of multiple dislocations in different \{111\} planes gives rise to a localized surface pit as shown in Fig. 3.8(f). Following the formation of a pit on the film surface, more threading dislocation half loops are emitted from the pit location.

Moreover, the half loops gliding along adjacent parallel slip planes shear the thin film further, causing deepening of the surface grooves and growth of the surface pit as shown in Fig 3.8(g). This process contributes to the formation of a void that extends across the film. The dynamical sequence of cross-sectional views in Figs. 3.8(e)-3.8(h) resembles the necking and ductile fracture of a metallic bar during a standard tensile test. In addition, glide of jogged dislocations and dislocation-dislocation interaction processes generate vacancies in the bulk, as shown in Fig. 3.8(j). Initially, vacancies either anneal in the bulk or migrate to the surface and deepen the surface pit further. As the concentration of vacancies in the film increases due to continued glide of jogged dislocations and dislocation intersections, the vacancies coalesce to form vacancy clusters in the film as shown in Fig. 3.8(k). The growing surface void (pit) and the vacancy clusters coalesce [Figs. 3.8(k) and 3.8(l)] to form a larger void that extends across the film, as seen in Figs. 3.8(d), 3.8(h), and 3.8(l). Upon further equilibration of the thin film at constant $\varepsilon$, negligible void growth is observed.
Figure 3.8. (a)–(d) Top views of the surface of the strained Cu thin film with an area of 30.9 \times 32.3 \text{ nm}^2 under applied biaxial strain $\varepsilon = 6\%$ during deformation and strain relaxation. Atoms are colored according to their von Mises strain. (e)–(h) Cross-sectional views of the strained Cu thin film over the same period. Atoms are colored according to their local environment. Dark blue and light blue atoms are in perfect fcc and hcp lattice arrangements, respectively. Other colors represent point defects, dislocations, and surface atoms. (i)–(l) 3D close view of the thin-film material over the same period; atoms with $Z = 11$, $Z = 12$, and $Z = 13$ are not shown for clarity. Atoms colored in maroon, light blue, and light green are surface atoms with $Z = 9$, atoms with $Z = 10$, and atoms with $Z \leq 8$, respectively. The corresponding total time $t$ in the MD simulation is recorded, starting from the film in its unstrained state.

As the applied strain level increases, stacking faults are observed to act as obstacles to dislocation glide, leading eventually to the annihilation of threading dislocations and other partial dislocations present in the film. The glide of partial dislocations in successive atomic planes causes the unzipping of existing stacking faults and leads to the formation of deformation twins. In addition, formation of Lomer-Cottrell junctions due to the interactions between Shockley partial dislocations, as well as formation of stable complex dislocation junctions that extend across the film.
thickness also as a result of dislocation-dislocation interactions were observed. These dislocation-dislocation interactions restrict dislocation mobility and plastic flow and they may increase the toughness of the thin-film material.

The MD simulations conducted in this study have revealed the formation of deformation twins during relaxation in thin films subjected to low biaxial strain levels above the yield strain. Deformation twins are formed due to the glide of Shockley partial dislocations in successive atomic planes. Twin boundaries are identified by a single plane of atoms in a locally perfect hcp environment. As defined earlier, a stacking fault consists of two layers of atoms in a locally perfect hcp environment and is formed by the glide of a Shockley partial dislocation, which displaces the atoms as it glides in the glide plane. When a Shockley partial dislocation glides on an atomic plane adjacent to an already existing stacking fault or when two Shockley partial dislocations glide past each other in adjacent atomic planes, the atoms in the region of overlap are unzipped back to a locally perfect fcc environment from a locally perfect hcp environment (Yamakov et al., 2002). However, because of the shearing, the atoms in the region of overlap undergo a displacement equal to the sum of the Burgers vectors of the two partial dislocations; the resulting atomic displacements in the region of overlap are such that these displaced atoms are mirror images of the atoms in the thin film that are not subjected to this shearing (Yamakov et al., 2002). The resulting atomic structure depends on the nature of the two Shockley partials gliding in successive atomic planes. If both of the participating stacking faults are intrinsic, the ensuing structure is an extrinsic stacking fault. On the other hand, if one of the participating stacking faults is intrinsic and the other one is extrinsic, a two-layer microtwin is
formed. Finally, if both of the initial stacking faults are extrinsic, a three-layer microtwin is formed (Yamakov et al., 2002).

![Image](image.png)

**Figure 3.9.** (a)-(c) Top view of a section of a biaxially strained thin film at a level $\varepsilon = 6\%$, showing in detail the different stages of dislocation activity in successive atomic planes leading to the formation of two- and three-layer microtwins. Dark blue and light blue atoms are in locally perfect fcc and hcp environments, respectively; red and orange atoms have $Z = 11$ and $Z = 13$, respectively; and green atoms are those surrounding point and line defects. Surface atoms are removed for clarity. The initial positions of the partial dislocations are labeled as A, B, and C in (a). The glide of dislocations B and C causes the formation of extrinsic stacking faults, while the glide of dislocation A leaves behind an intrinsic stacking fault. Dislocations A and B glide in opposite directions, which causes the unzipping of one layer of hcp atoms leading to the formation of a two-layer microtwin. The glide of dislocation C also leads to the unzipping of one layer of hcp atoms and leads to the formation of a three-layer microtwin. (d) A $[\bar{1}0\bar{1}]$ view of a single plane of atoms showing the twin and the twin boundary.

Figure 3.9 shows the top view of a section of the thin film subjected to an applied strain level $\varepsilon = 6\%$ and elucidates the different stages of dislocation activity in successive atomic planes that lead to the formation of two- and three-layer microtwins. The partial dislocations and the accompanying stacking faults prior to microtwin
formation are shown in Fig. 3.9(a); the partial dislocations are labeled as A, B, and C. The glide of dislocations B and C causes the formation of extrinsic stacking faults, while the glide of dislocation A leaves behind an intrinsic stacking fault. Dislocations A and B glide from their positions shown in Fig. 3.9(a) in opposite directions, which causes the unzipping of one layer of hcp atoms leading to the formation of a two-layer microtwin. The glide of dislocation C also leads to the unzipping of one layer of hcp atoms and leads to the formation of a three-layer microtwin. The two-and three-layer microtwins are shown in Fig. 3.9(b). The atomic packing in the twin, as can be seen in Fig. 3.9(b), is different from the surrounding \{111\} plane; careful inspection of this atomic packing confirms the formation and the structure of the twin. Specifically, these twins are oriented normal to the [322] crystallographic direction. Further glide of the dislocations leads to growth of the microtwins, as seen in Fig. 3.9(c). A (\{110\}) view of a single layer of atoms in the twin region, highlighting the structure of the microtwins, is shown in Fig. 3.9(d).

Dislocation-dislocation interactions often lead to the formation of immobile dislocation junctions that consist of various kinds of dislocations and associated stacking faults. In the MD simulations conducted in this study, Shockley partial dislocations, threading dislocation loops, and sessile stair-rod dislocations have been observed to contribute to the formation of dislocation junctions. A particularly important dislocation junction observed in these simulations is the Lomer-Cottrell lock (Hirth & Lothe, 1982). A Lomer-Cottrell junction is formed by two stacking faults in different \{111\} planes intersecting at 70.5° or 109.5° with Shockley partial dislocations bounding the non-intersecting ends of the stacking faults. The dislocation at the
intersection of the stacking faults is a sessile stair-rod dislocation, formed by the reaction of two Shockley partials gliding in different \{111\} glide planes.

The evolution of a representative Lomer-Cottrell junction captured in these MD simulations is shown in Fig. 3.10. Two stacking faults are shown in Fig. 3.10(a): one in the (\bar{1}1\bar{1}) plane and another one in the (\bar{1}11) plane. Two dislocations, \(D\alpha\) and \(\alpha D\), extend across the film thickness and bind the stacking fault in the (\bar{1}11) plane, while the threading dislocation, \(B\gamma\), binds the second stacking fault in the (\bar{1}1\bar{1}) plane. As the threading dislocation \(B\gamma\) extends, it reacts with dislocation \(D\alpha\) to form a stair-rod dislocation according to the reaction \(D\alpha + B\gamma \rightarrow DB/\alpha\gamma\); the stair-rod dislocation is represented in Fig. 3.10 by a straight line of atoms with \(Z \neq 12\) (usually atoms with \(Z = 13\) and \(Z = 11\)). The extension of the threading dislocation until it intersects with the top surface of the thin film facilitates the extension of the stair-rod dislocation. The evolution of the stair-rod dislocation, extending across the film thickness, can be seen in Figs. 3.10(b), (c), and (d). This dislocation junction, consisting of mobile Shockley partial dislocations together with the sessile strair-rod dislocation, forms a Lomer-Cottrell lock. Numerous such Lomer-Cottrell locks are observed in thin films subjected to high biaxial strains. These Lomer-Cottrell locks act as barriers to plastic flow by restricting dislocation glide.
Figure 3.10. (a-d) 3D close view of structural evolution in a biaxially strained thin film (ε = 9%) during the formation of a Lomer-Cottrell junction. Blue atoms are in perfect hcp lattice positions. Green and yellow atoms have Z = 13 and Z = 11, respectively, and these atoms represent the dislocation core. Other atoms correspond to lower coordination defects and surface atoms; atoms in locally perfect fcc lattice arrangements are not shown for clarity. The corresponding total time, t, in the MD simulation is recorded, starting from the film in its unstrained state. Two dislocations, \( \mathbf{D}_\alpha \) and \( \mathbf{a} \mathbf{D} \), extend across the film thickness and bind the stacking fault in the (\( \bar{1}11 \)) plane, while the threading dislocation, \( \mathbf{B}_\gamma \), binds the second stacking fault in the (\( \bar{1}1\bar{1} \)) plane. Dislocation \( \mathbf{B}_\gamma \) reacts with dislocation \( \mathbf{D}_\alpha \) to form a stair-rod dislocation according to the reaction \( \mathbf{D}_\alpha + \mathbf{B}_\gamma \rightarrow \mathbf{DB}/\alpha \gamma \), and is represented in (c) and (d) by a straight line of atoms with \( Z \neq 12 \). This dislocation junction, consisting of mobile Shockley partial dislocations together with the sessile stair-rod dislocation, represents a Lomer-Cottrell lock.

Formation of other complex dislocation junctions also is observed in the MD simulations. Figure 3.11 shows such a representative complex stable dislocation junction. Interacting dislocations become immobile as a result of their entanglement.
with one another to form these complex junctions. In Fig. 3.11, junction A is formed by the intersection of a threading dislocation and a Shockley partial extending across the thickness of the film. Junction B is formed by the entanglement of a threading dislocation, \(x\), and a stair-rod dislocation, \(y\). The junctions A and B together with the stacking faults and dislocation networks, which also include glissile dislocations, sessile dislocations, and associated stacking faults in adjacent planes [seen in Fig. 3.11(b)] form a complex stable junction. Such junctions of immobile dislocations and associated stacking faults also act as obstacles to dislocation motion and restrict the plastic flow in the thin-film material.

![Figure 3.11.](image.png)

**Figure 3.11.** (a) A \{111\} view and (b) a 3D close view of a complex immobile stable junction; dark blue and light blue atoms are in perfect local fcc and hcp environments, respectively, and other colored atoms have \(Z=12\), representing point defects and dislocations. Surface atoms and atoms in a locally perfect fcc lattice arrangement are not shown in (b) for clarity. The two junctions are denoted by A and B; \(x\) and \(y\) are a threading dislocation loop and a stair-rod dislocation, respectively, forming the junction B.

The number of such immobile dislocation junctions, including Lomer-Cottrell locks, increase with increasing \(\varepsilon\). For \(\varepsilon > 9\%\), a very high dislocation density followed by the formation of numerous dislocation junctions cause the rotation of nano-scale domains in the thin film material, leading to the structural transformation (from single-crystalline to nano-crystalline) of the thin film (Gungor & Maroudas, 2005b). The
increased density of immobile junctions, including Lomer-Cottrell locks, with increasing $\varepsilon$, suggests that the material toughness is closely related to the biaxial strain applied.

3.5. Conclusions

In conclusion, the strain relaxation mechanisms and the associated defect dynamics in [111] oriented ultrathin Cu films subjected to biaxial tensile strain over a broad range of strain levels, $\varepsilon$, were analyzed. Over the range $0 \leq \varepsilon \leq 12\%$, five regimes of thin-film mechanical response were identified. (i) Elastic response for $0 \leq \varepsilon < 5.5\%$; (ii) relaxation of the applied strain through nucleation of voids that grow to extend across the thin-film thickness mediated by emission of threading dislocation loops from certain locations at the film surface for $5.5 \leq \varepsilon < 6.4\%$; (iii) strain relaxation through emission of uniformly distributed threading dislocations from the film surface without void nucleation for $6.4 \leq \varepsilon < 8\%$; (iv) nucleation of dislocation loops in the bulk accompanied by threading dislocations emitted from the film surface for $8 \leq \varepsilon < 10\%$; and (v) nucleation of Frank partials and stacking-fault tetrahedra in addition to the dislocation mechanisms of regimes (iii) and (iv), that lead to formation of nano-scale domains in the film for $\varepsilon \geq 10\%$. The detailed analysis of these mechanisms also revealed deformation twinning at low $\varepsilon$ levels above the yield strain and formation of complex stable dislocation junctions for $\varepsilon > 7\%$, including Lomer-Cottrell locks, whose density increases with increasing $\varepsilon$. At high applied strain levels, very high defect densities cause local rotations of lattice regions and, eventually, the formation of nano-
scale domains through twinning mechanisms. The atomic-scale mechanistic information obtained by the MD simulations conducted in this study and their analysis is not accessible to experiments and has important implications for the mechanical behavior of metallic thin films. Furthermore, my atomistic analyses provide some of the required information for the development of first-principles-based constitutive theories, which are much needed for predictive modeling in the field of materials reliability.
CHAPTER 4

ATOMISTIC ANALYSIS OF BIAXIALLY STRAINED ULTRATHIN COPPER FILMS: FILM SURFACE ORIENTED ALONG A $\langle 110 \rangle$ CRYSTALLOGRAPHIC DIRECTION

4.1. Introduction

This study focuses on the identification and analysis of fundamental mechanisms of strain relaxation and the associated deformation processes in ultrathin films of Cu with and without pre-existing voids for thin-film free surfaces oriented along a $\langle 110 \rangle$ crystallographic direction. Toward this end, large-scale MD simulations of metallic thin-film response to the application of tensile biaxial strain were implemented. In these MD simulations, slab supercells were employed with periodic boundary conditions applied in the $x$- and $y$-directions in a Cartesian representation where the boundaries in the $z$-direction were free surfaces; the Cartesian $x$, $y$, and $z$ axes were along the $[111]$, $[\bar{1}1\bar{2}]$, and $[1\bar{1}0]$ crystallographic directions, respectively. For the results discussed here, a simulation cell that contained 1,272,000 atoms and had edge sizes of 56.35, 53.13, and 5.11 nm in the $x$-, $y$-, and $z$-directions was considered; these cell dimensions correspond to the unstrained state of the metallic thin film without any pre-existing voids. In ultrathin films with pre-existing voids, the initial configuration contained a cylindrical void at the center of the simulation cell that extended throughout
the film thickness with the axis of the cylindrical void was along [110] crystallographic direction; such voids are typical in metallic interconnects. The initial diameter of the void was 8.8 nm and the supercell contained 1,248,000 atoms and same edge sizes as the one without pre-existing voids. The supercell and the initial void size were chosen such that a realistic void morphology could be simulated during the void evolution, and void-void interactions between the periodic images were negligible. In order to examine further the deformation regimes where only the thin-film surface (as opposed to the void surface) played the dominant role in strain relaxation, a smaller simulation cell that contained 576,000 atoms and edge sizes of 37.6, 35.4, and 5.12 nm in the x-, y-, and z-directions was considered; this simulation cell did not contain any initial voids. In the absence of the void, the mechanical behavior of the film modeled by the larger simulation cell was found to be identical to that of the film modeled by the smaller simulation cell.

The MD simulations were performed at a constant temperature of 100 K at every given applied strain level. Initially, the supercell was unstrained, i.e., the cell edge sizes corresponded to the equilibrium density at the simulation temperature. The metallic thin film was strained biaxially by expanding the supercell along the x- and y-directions up to the chosen strain level, $\varepsilon$, at a rate of approximately $3 \times 10^{11}$ s$^{-1}$; specifically, a strain increment of 1% ($\Delta \varepsilon = 0.01$) was applied over a time period of $2.983 \times 10^{-14}$ s, yielding a strain rate of $3.352 \times 10^{11}$ s$^{-1}$. The effects of the strain rate were examined over a one-order-of-magnitude range; within this range, the atomistic mechanisms identified and analyzed were not affected by the strain rate.
4.2. Thin-Film Mechanical Response to Applied Biaxial Strain

To determine the mechanisms of strain relaxation and the associated defect dynamics in ⟨110⟩-oriented, biaxially strained ultrathin films of fcc Cu, films’ mechanical response over the range of biaxial strain levels from 0% to 18% were simulated. In addition to the initial elastic response for applied biaxial strains $\varepsilon < 2\%$, three deformation stages over the range $0 \leq \varepsilon \leq 18\%$ were identified. At low levels of applied biaxial strain, $2\% < \varepsilon < 6\%$ (stage I), strain relaxation is dominated by the nucleation of dislocations at the surface of the void and their propagation into the film as they are emitted from the voids surface. The change in the volume of the void after the thin film reaches a steady state is a good measure of the extent of plastic deformation originating from the void surface; steady state is reached over a time period after which the potential energy of the thin film remains practically constant. Figure 4.1 shows the change in the volume of the void, $(V-V_0)/V_0$, where $V_0$ and $V$ are the initial and steady-state volumes, respectively, as a function of applied biaxial strain level $\varepsilon$ at steady-state conditions ($t = 32.48$ ps as recorded starting from the film in its unstrained state) after the application of biaxial strain (blue curve). Figure 4.1 also shows the steady-state surface roughness $w_{ss}$, defined as the root-mean-squared fluctuation of the film thickness at steady-state conditions (red curve). During stage I, $2\% < \varepsilon < 6\%$, the strain relaxation is dominated by the emission of dislocations from the surface of the void; such a plastic deformation mechanism leads to void growth. As the dislocations glide away from the void surface, they create steps on the surface of the thin film, which
are the sources of the film surface roughness. As $\varepsilon$ is increased in stage I, the number of dislocations nucleating at the surface of the void increase and their glide leads to higher surface roughness.

![Graph showing dependence of $w_{ss}$ and $(V-V_0)/V_0$ on $\varepsilon$](image)

**Figure 4.1.** Dependence on $\varepsilon$ of the steady-state surface roughness, $w_{ss}$, and void volume change, $(V-V_0)/V_0$, of the strain-relaxed thin films; the steady-state values plotted correspond to $t = 32.48$ ps, i.e., after strain relaxation in the thin films. The solid lines are cubic-spline fits to the computed values as a function of $\varepsilon$.

At higher levels of applied strain, $6\% < \varepsilon < 10\%$ (stage II), the biaxial strain in the thin film is relaxed by both ductile void growth and emission of dislocations from the surface of the thin film. As more dislocations nucleate at the thin-film surface, the steps formed on the thin-film surface are more uniform than they were when formed due to glide of dislocations nucleating at the surface of the void; hence, the surface roughness of the thin film is reduced in stage II, as compared to the maximum values reached in stage I. As strain relaxation is increasingly dominated by the plastic deformation originating at the thin film surface with increasing applied biaxial strain, the change in the void volume also is reduced accordingly. For applied biaxial strains $\varepsilon$
\[ \geq 10\% \text{ (stage III), the strain relaxation is predominantly due to nucleation of dislocations at the surface of the thin film and their propagation into the film; negligible void growth is observed in stage III.} \]

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{Top views of the \([1\bar{1}0]\)-oriented thin films showing the thin-film microstructure immediately after the onset of plasticity for different applied biaxial strain levels, \(\varepsilon\). Green and orange colored atoms are in the dislocation cores and other atoms in the thin film are not shown for clarity.}
\end{figure}
The top views immediately after the onset of plasticity of the Cu thin films subjected to different applied biaxial strain levels are shown in Fig. 4.2. Green and orange colored atoms are in dislocation cores; atoms in locally perfect fcc and hcp environments, as well as surface atoms are not shown for clarity. The dislocation activity surrounding the void demonstrates clearly the different stages of plastic deformation identified. For $\varepsilon = 2\%$ [Fig. 4.2(a)], nucleation of Shockley partial dislocations at the surface of the void is observed. These Shockley partial dislocations have a pure edge character and extend across the film thickness. The glide of these Shockley partial dislocations in the $\{111\}$ planes causes strain relaxation in the thin films. In contrast in [111]-oriented Cu thin films, plastic deformation at similar strain levels is caused due to the nucleation at the void surface and subsequent glide of perfect screw dislocations (Gungor & Maroudas, 2005a).

For $\varepsilon = 4\%$, threading dislocation half loops emanate from the surface of the thin film in addition to the Shockley partial dislocations emitted from the void surface. These threading dislocation half loops nucleate very close to the surface of the void due to high stress concentration at the film surface near the void surface; such nucleation of threading dislocation loops at the film surface (i.e., close to the void surface) also was observed also for thin films oriented normal to the [111] crystallographic direction (Gungor & Maroudas, 2005a). These half loops of Shockley partial dislocations are seen clearly in Fig. 4.2(b). As it is evident in Fig. 4.2(c), for $\varepsilon = 6\%$, by comparison to Figs. 4.2(a) and (b), the density of the dislocations that are nucleated either at the void surface or at the film surface very close to the void increases with increasing applied biaxial strain. At $\varepsilon = 8\%$, nucleation of dislocations at the surface of the thin film
farther away from the void is observed in addition to nucleation of dislocations at the void surface and at the thin-film surface very close to the void. The partial dislocation loops emanating from multiple regions on the surface of the thin film lead to the reduction in the surface roughness of the thin film that was shown in Fig. 4.1.

Figure 4.3. Top views of the thin films after steady state is reached ($t = 32.48$ ps). Dark blue and light blue colored atoms are in locally perfect fcc and hcp environments, respectively, and other colored atoms are in the dislocation cores. Surface atoms are not shown for clarity.
Figure 4.3 shows the top views of the thin films at \( t = 32.48 \) ps, i.e., after steady state has been reached in terms of dislocation density and total energy of the thin film. Dark and light blue atoms correspond to atoms in locally perfect fcc and hcp environments, respectively. Other colored atoms are in the dislocation cores and surface atoms are not shown for clarity. As it is evident in Fig. 4.3, the void grows with increasing applied biaxial strain level, \( \varepsilon \). The void growth is accompanied by change in the void morphology from its initial perfect cylindrical shape (i.e., a circular top view) in accordance with the two-fold symmetry of the \( (1 \bar{1} 0) \) crystallographic plane, i.e., the film plane. The morphological evolution of the void surface is determined by the lattice symmetry of the free surface; for example, for a film surface oriented normal to the [111] crystallographic direction, a perfect circular void surface transformed to a hexagonal prism, i.e., to a six-fold symmetric hexagonal cross section on the \((111)\) plane (Gungor & Maroudas, 2005a). Increasing \( \varepsilon \) also leads to a higher density of stacking faults. For \( \varepsilon > 10\% \), dislocations nucleate primarily at the surface of the Cu thin film and void growth is suppressed; a similar response, i.e., suppression of void growth also was observed for [111]-oriented thin films. In addition to the similarities, however, there are marked differences in the structural response to biaxial strain between [111]- and \( \langle 110 \rangle \)-oriented Cu thin films; this is described and discussed in detail in Sections 4.3 and 4.4.

The percentage of various types of atoms in the thin film are plotted in Fig. 4.4 as a function of \( \varepsilon \) after \( t = 32.48 \) ps, i.e., after steady state has been reached at each strain level examined. Red, green, and blue curves correspond to the percentages of fcc
atoms, hcp atoms, and atoms in dislocation cores, respectively. The percentages of hcp atoms and atoms in dislocation cores increase with increasing applied biaxial strain level up to $\varepsilon \sim 10\%$. Such an increase is due to the increased plastic deformation with increasing applied strain. The increase in the percentage of atoms in stacking faults (hcp atoms) is unusually steep, however, for applied strain levels $\varepsilon > 10\%$, indicating a structural transition of the Cu thin film from a fcc to a hcp lattice; this transformation is discussed in detail in Sec. 4.4.

![Graph showing percentages of different types of atoms in the film as a function of applied strain.](image)

**Figure 4.4.** Percentages as a function of applied strain level, $\varepsilon$, of various types of atoms in the thin films at steady state ($t = 32.48$ ps). Red, green, and blue curves, which are cubic-spline fits to the computed values, denote to percentages of fcc atoms, hcp atoms, and atoms in dislocation cores, respectively.

### 4.3. Strain Relaxation Mechanisms and Defect Dynamics

In this section, dislocation nucleation mechanisms and dislocation dynamics observed during the strain relaxation of the (110)-oriented Cu thin films are analyzed. The properties of the emitted dislocations and their corresponding glide planes are characterized using the Thompson tetrahedron (Hirth & Lothe, 1982). At low applied biaxial strains ($2\% \leq \varepsilon < 4\%$), the primary mechanism of strain relaxation is the
nucleation and glide of pure edge Shockley partial dislocations from the void surface. For $\varepsilon \geq 4\%$, in addition to the nucleation of Shockley partial dislocations, nucleation of threading dislocations are observed at the surface of the film very close the void surface. Figure 4.5 shows a 3D close-view sequence of the evolution of the threading dislocation half loops; the 3D section of the thin film shown in Fig. 4.5 is marked in the accompanying image showing the top view of the entire thin film. The dark blue atoms are in a locally perfect hcp environment. Surface atoms are colored yellow and orange, and other atoms correspond to atoms in the dislocation cores; atoms in a locally perfect fcc environment are not shown for clarity. Additionally, a section of the void surface is seen clearly in the Fig. 4.5(a). Due to higher local stresses near the intersection of the void surface and the thin-film surface, the nucleation of dislocation half-loop pairs is seen to occur at the surface of the thin film and very close to the void surface. The sites of nucleation of these dislocation loops have two-fold symmetry, which is determined by the orientation of the film surface. $(\overline{1} \overline{1} \overline{1})$ and $(\overline{1}1\overline{1})$ slip planes [noted as (b) and (a); see Appendix A for a detailed description of the notation] are activated and the Burgers vectors of the dislocation half loops are $C\alpha(a)$, $C\beta(b)$, $D\beta(b)$, and $D\alpha(a)$; the Burgers vectors of the dislocations and their corresponding glide planes are marked in Fig. 4.5(a). As the dislocation half loops extend, each pair of partial dislocation loops, each of which lies in a different glide plane, grows for the loops to interact with each other and form one sessile stair-rod dislocation; the Burgers vectors of both of the resulting stair-rod dislocations are $\alpha\beta/DC$. 

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Figure 4.5. Three-dimensional view of the evolution of threading dislocation half loops that nucleate at the surface of the thin film, but very close the void surface and extend into the film. Dark blue colored atoms comprise the stacking faults; yellow and orange colored atoms are the atoms at the film and void surfaces and other colored atoms are in the dislocation cores. Atoms in locally perfect fcc environments are not shown for clarity.

Additionally, nucleation of a second partial dislocation is observed inside the initial threading dislocation half loop, as seen in Figs. 4.5(c) and 4.5(d) for the dislocation $\mathbf{D}_a(a)$, which emanates from the bottom surface of the Cu thin film. The Burgers vector of the secondary Shockley partial dislocation is $\mathbf{\alpha}_B(a)$, while that of the primary Shockley partial dislocation is $\mathbf{D}_a(a)$; consequently, the glide of the secondary partial dislocation unzips the stacking fault created by the primary partial and a perfect
dislocation \textbf{DB}(a) is formed. Such nucleation of a second Shockley partial dislocation inside a threading dislocation half loop has been observed in bulk Al metals (de Koning \textit{et al.}, 2003), as well as in the studies of [111]-oriented Cu thin films under biaxial strain (See Chapter 3).

An additional defect dynamical mechanism is observed at moderate applied biaxial strain levels \((4 \leq \epsilon \leq 8\%)\). Shockley partial dislocations, which nucleate at the void surface and at the film surface very close to the void surface, are observed to glide in successive atomic planes and lead to formation of twins. The structural evolution of a Cu thin film biaxially strained at \(\epsilon = 8\%\) to twin structure is shown in Fig. 4.6; a [100]-oriented cross section of the thin film is shown. The surface atoms, atoms in locally perfect fcc environments, and atoms in locally perfect hcp environments are colored dark red, dark blue, and light blue, respectively; other colored atoms are in the dislocation cores. The [100] orientation of the plane shown is evident in Fig. 4.6(a). In response to the application of biaxial strain, the partial dislocations that nucleate at the void surface or at the film surface close to the void are shown to glide away from the void. The low symmetry of the \([1\bar{1}0]\)-oriented film surface limits the availability of the \(\langle 110\rangle\{111\}\) and \(\langle 112\rangle\{111\}\) slip systems and leads to the nucleation of dislocations in successive \{111\} slip planes. The glide of such dislocations leads to formation of twin grain boundaries with \(\Sigma = 3\) and \(\theta = 54.47^\circ\); the grain boundary between the newly formed twin grain and rest of the thin film was found to be incoherent. The twins and the twin boundary are seen in Fig. 4.6(d). The \(\langle 111\rangle\) orientation of the plane, which had [100] orientation initially, is seen clearly.
Figure 4.6. [100]-oriented cross section of the thin film showing the evolution of twins forming a $\Sigma = 3$ twin as a consequence of glide of dislocations in successive atomic planes. The surface atoms, atoms in locally perfect fcc environments, and atoms in locally perfect hcp environments are colored dark red, dark blue, and light blue, respectively; other colored atoms are in the dislocation cores.

For applied biaxial strain levels $\varepsilon \geq 8\%$, in addition to nucleation of dislocations from the surface of the void and from the film surface close to the void, nucleation of
dislocations from the surface of the thin film far from the void is observed. These Shockley partial dislocations are nucleated in pairs and at different \{111\} glide planes. Figure 4.7 shows representative configurations of such dislocations. Figure 4.7(a) shows the top view ([1\overline{1}0] view) of the region of the thin film where these dislocations are nucleated, while Figs. 4.7(b) and 4.7(c) show the top ([1\overline{1}0]) and bottom ([\overline{1}10]) view of the interior of the dislocations. In Figs. 4.7(b) and 4.7(c), atoms in locally perfect fcc environments are not shown for clarity. The Shockley partial dislocations emanating from the surface of the film have Burgers vectors \( \mathbf{D\alpha} \) and \( \mathbf{D\beta} \), respectively, as indicated in Figure 4.7. The line vector shared by these two partial dislocations becomes effectively a sessile dislocation binding the two partial dislocations with a Burgers vector \( \mathbf{b} = (a/6)[\overline{1}\overline{1}0] \). Incipient plastic deformation caused by nucleation of dislocations at the surface of the thin film in the manner discussed above is different from that observed in the [111]-oriented films, where the partial dislocation loops glide in all the three available \{111\} planes (Gungor & Maroudas, 2005b; also see Chapter 3). Such differences between the mechanisms of incipient plastic deformation from the surface of the thin film are due to the differences between the crystallographic symmetries of the planes of the thin-film surfaces (two-fold and six-fold for [111]- and \( \langle 110 \rangle \)-oriented films, respectively).

At higher applied strain levels, plastic deformation in the thin films is dominated by the nucleation of dislocations at the surface of the thin film and their growth into the film; plastic deformation due to dislocation emission from the surface of the void (ductile void growth) does not contribute to strain relaxation at this stage of
deformation. While suppression of void growth was observed in both [111]- and ⟨110⟩-oriented thin films, the thin-film structural response was found to be very different for the two different film-surfaces orientations. Whereas the [111]-oriented thin films were found to undergo significant deformation leading to the formation of nano-domains, the ⟨110⟩-oriented Cu thin films were found to transform from a fcc crystalline phase to a hcp crystalline phase. This transformation is discussed in detail in the next section (Sec. 4.4).

![Figure 4.7](image.png)

**Figure 4.7.** Top views of a pair of dislocation half loops emanating from the surface of the thin film. The atoms are [(a)] and are not [(b), (c)] the atoms in locally perfect fcc environments

### 4.4. Martensitic fcc-to-hcp Phase Transformation and Stability Analysis

The steep increase in the percentage of hcp atoms for $\varepsilon \geq 10\%$ (seen in Fig. 4.4) is evidence for the structural transition of the Cu thin film from a fcc to a hcp lattice. A
systematic analysis is conducted of this structural transformation based on MD simulations employing simulation cells without pre-existing voids. The top views of the thin films subjected to applied biaxial strains $\varepsilon = 9\%$ and $12\%$ are shown in the insets to Figs. 4.8(a) and 4.8(b), respectively, and highlight the difference in the structural response to the different levels of $\varepsilon$; the top view of the thin film under $\varepsilon = 12\%$, which has undergone a fcc$\rightarrow$hcp phase transformation, resembles very closely that of experimentally observed martensites (Waitz & Karanthaler, 1997; Garcia et al., 1999; Lee et al., 2000; Hesemann et al., 2003; Sort et al., 2003; Liu & Huang, 2004; Espinosa et al., 2006b). Martensitic transformations induced by thermal or mechanical strains have been observed in experimental studies at moderate ($10^2$-$10^3$ s$^{-1}$) and high ($>10^6$ s$^{-1}$) strain-rate deformation of 304L stainless steel (Lee et al., 2000). Moreover, heterogeneous martensitic transformations induced by plastic deformation also have been reported in experimental studies where shock loading was applied through ball milling (Sort et al., 2003) or by conducting shock-recovery experiments (Hsiung, 2005). This fcc$\rightarrow$hcp transition is most evident from the evolution of the pair correlation function, $g(r)$, shown in Fig. 4.9 during the strain relaxation of a Cu thin film with a fcc lattice structure initially, after it has been strained biaxially to $\varepsilon = 12\%$. For example, an unambiguous indication of the fcc$\rightarrow$hcp transition is the appearance of the third and fifth peaks in the $g(r)$ curve shown in black, which is characteristic of a hcp lattice. The resulting hcp Cu has a lattice-parameter ratio $c/a = 1.612$, i.e., very close to that of the perfect hcp lattice, $(8/3)^{1/2}$. The structural relaxation after biaxial tensile straining to $\varepsilon \geq 10\%$, hence, results in a martensitic fcc$\rightarrow$hcp transformation.
Figure 4.8. Top views of the strain-relaxed thin films subjected to applied biaxial strain levels (a) \( \epsilon = 9\% \) and (b) \( \epsilon = 12\% \) biaxial strain; the surface plane is the \((\overline{1}0)\) plane. Dark and light blue atoms correspond to atoms in locally perfect fcc and hcp environments, respectively, and other colored atoms are in the dislocation cores. Surface atoms are not shown for clarity.

Figure 4.9. Evolution [shown by upward shifting] of the pair correlation function, \( g(r) \), during the strain relaxation of a Cu thin film with a fcc lattice structure initially strained biaxially to \( \epsilon=12\% \). Solid triangles and circles indicate peak positions in the \( g(r) \) that correspond to the sequence of coordination shells in perfect fcc and hcp crystals, respectively, and \( a_0 = 3.615 \) Å is the equilibrium lattice parameter of fcc Cu. The absence of sharp peaks in the early \( g(r) \) of the fcc film is due to a disordering of the fcc lattice as a result of straining at shock loading rates. The MD simulation time, \( t \), is recorded strating from the film at its unstrained state.
Figure 4.10. (a)-(d) A cross-sectional view of a biaxially strained (to $\varepsilon = 12\%$) Cu thin film showing the evolution of the film structure as it undergoes structural transition from an initial fcc lattice [(a)]; the plane shown is the (001) fcc lattice plane. Dark and light blue atoms are in perfect fcc and hcp lattice arrangements, respectively. Maroon and other colored atoms are on the surface and inside dislocation cores, respectively. The three subdomains shown in (d) embedded in the hcp-martensite phase are faulted hcp (middle) and fcc regions. The MD simulation time, $t$, is recorded strating from the film at its unstrained state.

Figure 4.10 shows a cross-sectional view of the structural evolution during strain relaxation of the thin film that has been strained biaxially to $\varepsilon = 12\%$; the plane shown corresponds to the (001) crystallographic plane of the original fcc phase, (001)$_{\text{fcc}}$. This
evolution sequence shows the film as it undergoes structural transition from its initial fcc lattice. After the biaxial straining of the film and during the subsequent structural relaxation at isostrain conditions ($\varepsilon = 12\%$), multiple partial dislocation loops are emitted simultaneously from the film surface and glide in successive $\{111\}$ slip planes along $\langle 112 \rangle$ directions; these loops have Burgers vectors $(a/6)\langle 112 \rangle$, where $a$ is the fcc lattice parameter. The glide of these loops in successive atomic planes is analogous to twinning mechanisms that are observed commonly in fcc metals (Yamakov et al., 2002) and leads to the nucleation of a shear-induced hcp phase near the film surface [Fig. 4.10(b)]. The martensite then propagates and the hcp phase grows as the dislocation loops extend into the bulk of the film [Figs. 4.10(c) and 4.10(d)]. The $(001)_{\text{fcc}}$ plane [Fig. 4.10(a)] now corresponds to the $(0001)_{\text{hcp}}$ plane [Fig. 4.10(d)], i.e., the basal plane of the hcp lattice structure. The habit plane of the heterogeneously nucleated $(001)_{\text{fcc}}\parallel(0001)_{\text{hcp}}$ structural transition, i.e., an invariant crystallographic plane common to both the parent lattice and the martensite, was found to be normal to the $[\overline{1}10]_{\text{fcc}}$ direction and parallel to the original film surface plane.

Martensitic transformations of the $(001)_{\text{fcc}}\parallel(0001)_{\text{hcp}}$ type also have been observed in large-scale MD simulations of shock loading of body-centered cubic (bcc) metals (Kadou et al., 2002), and analyzed in a recent first-principles study (Kim et al., 2006). In those studies, however, the martensite nucleated homogeneously. In addition, in the study of Kim and co-workers (Kim et al., 2006), the transformation mechanism involved slip of $(001)$ planes and an intermediate orthorhombic state, which is not observed in the MD simulations conducted in this thesis. The analysis conducted in this study reveals that the martensitic transformation in biaxially strained thin films is
mediated by plastic deformation. The structural evolution depicted in Fig. 4.10 shows that the transformation is facilitated by the dislocation glide motion and the martensite phase grows due to the propagation of the stacking faults bounded by these partial dislocations. Furthermore, in the MD simulations of strain relaxation in biaxially strained thin metallic films with [111]-oriented free surfaces, martensitic transformations were not observed (see Chapter 3). In this study, the low symmetry of the (110)_{fcc} surface plane limits the availability of the ⟨110⟩{111} and ⟨112⟩{111} slip systems and leads to nucleation of line defects in successive ⟨111⟩_{fcc} slip planes, which facilitates the martensitic transformation.

The magnitudes of the atomic displacements were analyzed for the atoms in the structurally transformed thin film that have undergone slip relative to their neighbors in the unstrained fcc state, which is taken as the reference configuration; the slip vector formulation has been presented in detail in Chapter 2. In Fig. 4.11, distributions are plotted of the slip vector magnitudes, \( s \equiv |\mathbf{s}| \), of all the atoms that have slipped in strain-relaxed thin films for various strain levels \( \varepsilon \). Specifically, the function \( \hat{f}(s) = f(s) / (f(s))_{\text{max}} \) is plotted, where \( \int_{s>0} f(s) ds = N_{\text{slipped}} \) and \( N_{\text{slipped}} \) is the total number of atoms in the thin film that have undergone slip displacements. For \( \varepsilon < 10\% \), the \( \hat{f}(s) \) distributions are centered at the Burgers vector magnitude, \( b \), of perfect and Shockley partial dislocations, \( b = a/\sqrt{2} \) and \( a/\sqrt{6} \), respectively. For \( \varepsilon > 10\% \), a shift is observed in these distributions, which become centered at \( a\sqrt{2}/6 \) and \( a/\sqrt{6} \).
Figure 4.11. Dependence on $\varepsilon$ of the distribution $\hat{f}(s)$ of the relative slip vector magnitudes in strain-relaxed thin films at $t = 32.48$ ps.

The relative slip vector magnitude, $a\sqrt{2}/6$, is consistent with the atomic displacements proposed in the theoretical study of Dmitriev et al. (Dmitriev et al., 1991), which provides a unified theoretical framework for the description of the Burgers and Bain mechanisms of bcc→hcp and bcc→fcc transformation, respectively. In both cases, the corresponding relative displacement magnitudes are equal to $a\sqrt{2}/6$. The fcc→hcp transformation can be considered as a combination of the Bain and the Burgers mechanisms (fcc→bcc + bcc→hcp) and, therefore, the corresponding relative displacement magnitude should also be $a\sqrt{2}/6$. Nevertheless, in the MD simulations of this study, these displacements are a result of dislocations gliding in successive atomic planes as opposed to concerted motion of adjacent atomic planes in opposite directions (as in the Burgers mechanism) or in the same direction (as in the Bain deformation).
The beginning of the shift in the distributions of Fig. 4.11 marks the onset of the martensitic transformation.

Figure 4.12. Eigenvalues of the symmetrized elastic stiffness moduli matrix, \( \mathbf{A} \), as a function of \( \varepsilon \), as calculated immediately after the application of the biaxial strain.

Mechanical stability analysis (Wang et al., 1995; Morris & Krenn, 2000) is carried out in order to predict the loss of the crystalline fcc lattice stability under the conditions of the simulations in this study. The analysis revealed that shear, as well as Born instabilities are triggered prior to the martensitic transformation in the metallic thin-film models considered in this study. Specifically, the modes of destabilization of the symmetrized version, \( \mathbf{A} = (1/2)(\mathbf{B} + \mathbf{B}^T) \), of the elastic stiffness moduli matrix, \( \mathbf{B} \), are calculated. Given that the habit plane is \((\bar{1}10)_{\text{fcs}}\), \( \mathbf{B} \) is transformed from the original coordinate system to one with coordinate axes \( x' \), \( y' \), and \( z' \) taken along the [110], [001], and [\bar{1}0] crystallographic directions, respectively. The form taken by matrix \( \mathbf{A} \) implies that the conditions for the stability of the fcc lattice are \( \det(\mathbf{A}_m) > 0 \), \( A_{44} > 0 \), \( A_{55} > 0 \), and \( A_{66} > 0 \), where the elements of matrix \( \mathbf{A} \) are expressed in contracted Voigt
notation and $A_m$ is the $3 \times 3$ matrix $[A_{ij}; i,j = 1,2,3]$. The real and positive eigenvalues of $A_m$ are denoted by $\lambda_1$, $\lambda_2$, and $\lambda_3$, with $\lambda_1 < \lambda_2 < \lambda_3$.

The eigenvalues of matrix $A$, $\lambda_1$, $\lambda_2$, $\lambda_3$, $\lambda_4 = A_{44}$, $\lambda_5 = A_{55}$, and $\lambda_6 = A_{66}$, are plotted in Fig. 4.12 as a function of $\varepsilon$. For each value of $\varepsilon$, the elements of $A$ were computed immediately after the application of the biaxial strain, i.e., at the end of the dynamic straining period to bring the film from its unstrained state to this strain state, $\varepsilon$. Figure 4.12 shows the onset of a shear instability in the (001) crystallographic plane associated with the vanishing of $A_{55}$ ($A_{55} = A_{x'x'z'} = 0$) occurring at a strain $\varepsilon \sim 10\%$. In the MD simulations, for $\varepsilon = 10\%$, the nucleation is observed at the surface of the Cu thin film of isolated hcp-martensite embryos, most of which do not extend into the bulk of the film. The MD simulations also reveal that martensite grains continue to nucleate and grow with increasing applied strain (e.g., $\varepsilon = 11\%$). The Born instability, associated with the vanishing of $\lambda_1$ ($\lambda_1 = 0$), occurs at $\varepsilon \sim 12\%$ and leads to breaking of the fcc lattice symmetry. This bifurcation, the onset of the Born instability, also causes the material to transform to a new stable lattice structure, the hcp phase. The bifurcation, in conjunction with the driving force provided by the applied strain lead to the growth of martensite embryos into the bulk of the film and completes the structural transition [Fig. 4.10(d)]; in this state, the hcp phase has grown throughout the film thickness and the film is predominantly hcp martensite. It can be concluded that the breaking of the fcc lattice symmetry in the thin film marks the onset of martensitic transformation, which is mediated by plastic deformation.
4.5. Conclusion

In conclusion, a systematic atomic-scale analysis was carried out of the strain relaxation mechanisms and the associated defect dynamics over a broad range of biaxial tensile strain levels, $\varepsilon$, in nanometer-scale-thick Cu films with [110] surface crystallographic orientation with and without pre-existing voids. At lower strain levels ($2\% \leq \varepsilon < 8\%$) strain relaxation occurs primarily due to ductile void growth by emission of dislocations from the void surface (plastic zone expansion). Dislocation arrays are emitted from the surface of the void and lead to formation of incoherent twins. At higher strain levels ($8\% \leq \varepsilon < 10\%$), dislocations emitted from the film surface contribute to strain relaxation and act as a competing mechanism to void growth. At even higher applied strain levels, void growth is inhibited completely and fcc$\rightarrow$hcp martensitic transformation is triggered. The martensite phase was found to nucleate heterogeneously at the thin-film surface and grow subsequently into the bulk of the film; the growth process is mediated by the propagation of stacking faults, which is caused by partial dislocation glide motion. The results also elucidate the role of stacking faults in facilitating such martensitic transformations in fcc metallic thin films. The analysis in this study supports recent theoretical studies that also suggest the emergence of lattice defects in reconstructive transformations, such as the fcc$\rightarrow$hcp martensitic transformations, as well as close-to-reconstructive transformations (Bhattacharya et al., 2004; Perez-Reche et al., 2007). Finally, mechanical stability analysis showed that the orientation relationship between the parent and the martensite phase and the onset of the phase transformation are consistent with the onset of a shear instability in the thin film.
CHAPTER 5

ATOMISTIC ANALYSIS OF DYNAMIC DEFORMATION
OF PRE-STRAINED ULTRATHIN FILMS OF FACE-CENTERED CUBIC METALS

5.1. Introduction

Dynamic deformation, i.e., constant strain-rate deformation at constant temperature, of materials is the most common method of testing an understanding of materials’ mechanical behavior in experiments. As already mentioned in Chapter 1, metallic nanopillars were found to exhibit an extended easy-glide regime, i.e., a deformation stage where the stress remained constant with application of strain. In addition to metallic nanopillars, which have been studied extensively in recent works on both fcc and bcc metals, ultrathin metallic films have a high surface-to-volume ration, therefore, potentially promising mechanical properties. Unlike nanopillars, however, dislocation depletion through escape of dislocations at the surface in metallic thin films is presumably less likely due to the differences in the geometry of these two categories of small-volume structures. In this chapter, the mechanical behavior is analyzed of fcc metallic ultrathin films (i.e., nanometer-scale-thick films) that contain a high initial dislocation density. First, the mechanical behavior and the corresponding dislocation dynamics for a given metal (Cu) is analyzed; then, the mechanical behavior is compared of ultrathin films three fcc metals with different propensity for formation of stacking faults. The chapter is structured as follows. Sample preparation methods and the criteria for selection of appropriate interatomic potentials are discussed in Sec. 5.2. The
mechanical response of Cu thin films during dynamic deformation is analyzed in Sec. 5.3 and the different underlying mechanisms of dislocation depletion in Cu thin films are analyzed and discussed in Sec. 5.4. A discussion, placing the findings of this study in the context of recent experimental studies on metallic nanopillars and computational studies of twin boundaries is presented in Sec. 5.5. A detailed analysis of dislocation-stacking fault interactions that lead to cross-slip of dislocations in fcc metals but do not necessarily cause dislocation annihilation is presented in Sec. 5.6. Finally, a comparative study of different fcc metals is presented in Sec 5.7 together with a discussion of the mechanisms of dislocation depletion in fcc metals with low propensity for formation of stacking faults.

5.2. Sample Preparation

In the MD simulations conducted in this study, slab supercells consisting of single-crystalline fcc metallic films are employed with periodic boundary conditions applied in the x- and y-directions in a Cartesian representation; the film planes were oriented normal to the z axis. Vacuum space above the top and below the bottom surfaces of the film was used in order to generate free surfaces and mimic free-standing thin films. The Cartesian x, y, and z axes were taken along the [110], [112], and [111] crystallographic directions, respectively, with the film plane corresponding to the (111) crystallographic plane. For the results reported in this Chapter, simulation cells are employed that contained 1,520,640 atoms with edge sizes of 240 \( d_x \), 432 \( d_y \), and 21 \( d_z \) in the x-, y-, and z-direction, respectively, in the films’ unstrained state; \( d_x \), \( d_y \), and \( d_z \), are the equilibrium interplanar spacings of the crystals in the [110], [112], and [111]
crystallographic directions, respectively. In addition, in the case of Cu, a simulation cell with edge sizes of 160 $d_x$, 264 $d_y$, and 54 $d_z$ in the $x$-, $y$-, and $z$-direction, respectively, is used to study Cu thin films that are about 2.5 times thicker than the original ones in order to confirm the results observed for thinner films.

In order to understand the role of stacking faults in the annihilation of dislocations, the response of three different fcc metals, Cu, Ni, and Al, is compared. These metals are characterized by different propensities for formation of stacking faults; this propensity is expressed by the ratio of stable to unstable stacking-fault energy, $\gamma_s/\gamma_u$, where $\gamma_s$ and $\gamma_u$ are the stable and unstable stacking-fault energies, respectively; high (low) values of $\gamma_s/\gamma_u$ imply small (large) total stacking-fault area in the sample. In this study, the interatomic interactions were described using EAM potentials; the parameterization developed by Mishin and co-workers was used for Al (Mishin et al., 1999) and Cu (Mishin et al., 2001), whereas that developed by Angelo and co-workers (Angelo et al., 1995) was used for Ni. The choice of the EAM potential for each material was based on its accuracy to predict $\gamma_s$, $\gamma_u$, and the ideal critical shear stress in comparison with the predictions of *ab initio* calculations (Ogata et al., 2004; Brandl et al., 2007) as shown in Table 5.1.

In the MD simulations of thin-film dynamic deformation under biaxial tensile strain, the single-crystalline films were pre-treated in order to generate the initial dislocation distributions before conducting the MD simulations of dynamic straining. Specifically, the thin films were strained in biaxial tension to a strain level of 8% at a constant true strain rate of $2.01 \times 10^{11}$ s$^{-1}$ (equally in each lateral direction). Under this
mode of loading, three \{111\} crystallographic planes are activated; the Schmidt factor for the possible slip directions in the (111) plane parallel to the plane of the film surface is negligibly small. After the desired strain level was reached, isothermal-isostrain MD simulations were performed until the complete relaxation of the thin-film structure. For pre-treatment under biaxial tensile strain at strain levels over the range from 6\% to 10\%, the resulting microstructure of the equilibrated thin films remained qualitatively identical.

Table 5.1 Stable and unstable stacking-fault energies, \(\gamma\) and \(\gamma_s\), respectively, and ideal simple shear strength for Al, Ni, and Cu according to different EAM potentials; the EAM values (in boldface) are compared with previously published values from first-principles calculations (in italics).\(^{a}\)Mishin et al., 1999;\(^{b}\)Angelo et al., 1995;\(^{c}\)Mishin et al., 2001;\(^{d}\)Brandl et al., 2007;\(^{e}\)Ogata et al., 2004.

<table>
<thead>
<tr>
<th>Metal</th>
<th>(\gamma_s) (mJ/m(^2))</th>
<th>(\gamma) (mJ/m(^2))</th>
<th>(\gamma_s/\gamma)</th>
<th>Ideal Shear Strength (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>195(^a)</td>
<td>158.9(^a)</td>
<td>0.82(^a)</td>
<td>3.81(^a)</td>
</tr>
<tr>
<td></td>
<td>178(^d)</td>
<td>146(^d)</td>
<td>0.83(^d)</td>
<td>3.73(^e)</td>
</tr>
<tr>
<td>Ni</td>
<td>263.8(^b)</td>
<td>124.7(^b)</td>
<td>0.47(^b)</td>
<td>5.88(^b)</td>
</tr>
<tr>
<td></td>
<td>278(^d)</td>
<td>137(^d)</td>
<td>0.49(^d)</td>
<td>6.29(^c)</td>
</tr>
<tr>
<td>Cu</td>
<td>181.1(^c)</td>
<td>44.4(^c)</td>
<td>0.25(^c)</td>
<td>3.97(^c)</td>
</tr>
<tr>
<td></td>
<td>164(^d)</td>
<td>38(^d)</td>
<td>0.23(^d)</td>
<td>3.45(^e)</td>
</tr>
</tbody>
</table>

The mechanical response of three Cu thin-film samples is analyzed; two of these, samples 1 and 2, had initial (unstrained-state) thicknesses of 4.38 nm and 11.27 nm, respectively. The third one, sample 3, was prepared by compressing sample 1 after its tensile straining back to its original lateral dimensions at the same constant high strain rate and then equilibrating the film’s structure by isothermal-isostrain MD simulation. In addition, the mechanical response is analyzed of Ni and Al films with
initial unstrained thickness of 4.26 nm and 4.9 nm, respectively. After pre-treatment, the final thicknesses of Cu samples 1, 2, and 3 were 3.84 nm, 9.76 nm, and 4.43 nm, respectively, and the final thicknesses of the Ni and Al samples were 3.78 nm and 4.1 nm, respectively.

The pre-treated thin film samples contained predominantly simple extended dislocations that either extended across the film thickness or were threading dislocation half loops emanating from the film surfaces. The initial dislocation density, \( \rho \), of these films ranged from \( 1.0 \times 10^{17} \text{ m}^{-2} \) to \( 3.2 \times 10^{17} \text{ m}^{-2} \); \( \rho = l/V \) where \( l \) is the total length of all the dislocations in the film and \( V \) is the film’s volume. In samples 1 and 3, as well as in the Ni and Al samples, many extended dislocations spanned across the film thickness, while only a few threading dislocation half loops were present. In the thicker Cu film (sample 2), however, the population of threading dislocation half loops was comparable with that of the extended dislocations that spanned across the film thickness. This initial film microstructure is similar to that of the Ni nanopillars used in mechanical annealing experiments (Shan et al., 2008); in those experiments, similar dislocation types were observed, i.e., most of them extended across the thickness of the nanopillar and a few of them were threading dislocation half loops at the surface of the nanopillar, and the dislocation density was \( \sim 10^{15} \text{ m}^{-2} \). The initial stacking-fault area in the Cu samples ranged from \( 12.5 \times 10^{-14} \text{ m}^{2} \) to \( 36 \times 10^{-14} \text{ m}^{2} \) and that in the Ni sample was \( 11.2 \times 10^{-14} \text{ m}^{2} \), whereas the initial stacking-fault area in Al sample was only \( 3.1 \times 10^{-14} \text{ m}^{2} \). After this pre-treatment stage, the final steady-state values of the von Mises shear stress, \( \sigma_{M} \), for Cu samples 1, 2, and 3 were 530, 460, and 105 MPa, respectively.
Although the stresses in the thin films were significant, most of the stress was due to elastic straining. In the thin films studied here, the shear strain, measured as the second invariant of the local atomic strain tensor (Li, 2000), varied between 0.7% and 1.59%; this indicates that in these pre-treated thin films the applied biaxial strain had relaxed significantly. Additionally, the pre-treatment equilibration stage was continued until ensemble averages of the stress and potential energy in the thin films reached constant (i.e., steady) values. Following pre-treatment, the samples were strained biaxially (again, equally in each lateral dimension) in tension at a constant strain rate of $7 \times 10^8 \text{s}^{-1}$ and the evolution of the film structure was monitored in detail.

5.3. Mechanical Behavior of Cu Thin Films

Figure 5.1(a) shows the evolution of the von Mises shear flow stress, $\sigma_{\text{VM}}$, dislocation density, $\rho$, and the stacking fault area, $A_{sf}$, in sample 1 (initial $\rho = 1.4 \times 10^{17} \text{m}^2$) during dynamic biaxial straining, where the applied strain level, $\varepsilon$, increases linearly with time. The decrease in $\rho$ and $A_{sf}$ until they each reach a minimum value indicates annihilation of dislocations. Subsequently, new dislocations nucleate, leading to an increase in $\rho$ and $A_{sf}$. From the shear stress-strain curve, it is evident that the thin film does not undergo stage-II hardening, which is typical of bulk metals. The maximum $\sigma_{\text{VM}}$ computed in the simulations conducted in this study varied between 1.56 and 1.74 GPa. These stress levels are 39.9%-44.5% of the ideal simple shear strength of the copper model used in this study [3.91 GPa according to the interatomic potential employed in the MD simulations conducted in this study, which compares well with first-principles calculations (Zhu et al., 2004a)]; these stress levels are consistent with
experimental measurements on Au nanopillars [indicating that the nanopillars reached 44% of the material’s theoretically predicted ideal pure shear strength (Brinckmann et al., 2008)].

The mechanical response of the thin film to the applied dynamic biaxial straining exhibits three different stages; these stages are indicated in Fig. 5.1(a) and are termed as Stage I, Stage II, and Stage III. Figure 5.1(b) shows three-dimensional views of the MD simulation cell at various states, which are also marked in Fig. 5.1(a). In Stage I, $\sigma_{xM}$ increases rapidly to about 1.1 GPa. The glide of most dislocations is activated at this stage. A few partial dislocations retract into the extended dislocations that they constitute and, in the process, they unzip the stacking faults that they bound. Such dislocation glide induces the interaction of the two Shockley partial dislocation components of a simple extended dislocation, resulting in a reduction of the stacking-fault area. The reduced stacking-fault area increases the mobility of the dislocations. In addition to the reduction in stacking-fault area, a few dislocations also annihilate in Stage I, primarily due to interactions between extended dislocations and, to a far lesser extent, due to interactions betweenpartials and anti-partialsthat bind a few stacking faults. Dislocation annihilation due to interactions between partial and anti-partial dislocations occurs possibly because such dislocations are unstable. In Stage II, for $1.5\% < \varepsilon < 4.5\%$ [Fig. 5.1(a)], $\rho$ and $A_{sf}$ decrease to less than one third of their initial values; $\sigma_{xM}$, however, remains nearly constant. Stage II is dominated by dislocation glide and, more importantly, by interactions within and between extended dislocations. The intersection of the gliding dislocations with the stacking faults of the extended dislocations causes cross-slip of the intersecting dislocations and break-up of the
obstacle stacking faults. At least one of the dislocations produced by the cross-slip reaction glides in the plane of the obstacle extended dislocation and interacts with its Shockley partials to either form a perfect dislocation or annihilate. Such stacking fault-aided cross-slip mechanisms cause further reduction in $\rho$ and $A_{sf}$ [Fig. 5.1(b), state (iii) at the end of Stage II]. In Stage III, the dislocation dynamics is similar to that observed in Stage II. By the end of Stage III, $\rho$ and $A_{sf}$ reach their minimum values; most dislocations that survive Stage III are perfect dislocations. Unlike Stage II, in Stage III, $\sigma_{\text{int}}$ increases with increasing $\varepsilon$ due to reduced plastic flow (dislocation starvation) in the thin film. At the end of Stage III, after a considerable build-up in shear stress, new dislocations start nucleating at the thin-film surface, leading to a sharp decrease in the thin-film stress. In sample 1 (Fig. 5.1), the propagation of the newly nucleated dislocations leads quickly to film failure; due to film failure, successive strain burst events similar to those observed in experiments on metallic nanopillars (Greer & Nix, 2006; Nix et al., 2007; Shan et al., 2008; Brinckmann et al., 2008) are not observed in sample 1.

Sample 3 also responds to applied dynamic biaxial straining in a similar fashion. The evolution of $\sigma_{\text{int}}$, $\rho$, and $A_{sf}$ in samples 2 and 3 is shown in Fig. 5.2(a) and Fig. 5.2(b), respectively. In samples 1 and 3, the stress increases rapidly and then remains constant until significant dislocation depletion and annihilation of stacking faults occur. Reduction in plastic flow due to dislocation depletion leads to build-up of stress in the thin films and, eventually, to nucleation of new dislocations. As dislocation density in sample 3 reaches a minimum faster (due to the much higher initial dislocation density in
sample 3 than in sample 1), the constant-stress stage (stage II) is shorter in sample 3 than in sample 1. Samples 1 and 3 are considerably thin; in these samples, the nucleation and propagation of new dislocations lead quickly to thin-film failure. In sample 2, however, the newly nucleated dislocations propagate without causing failure; instead, they lead to successive cycles of dislocation dissociation and annihilation followed by dislocation nucleation events. Consequently, the strain bursts that correspond to nucleation of new dislocations and the associated sharp decrease in the thin-film stress are identified clearly in Fig. 5.2(a); it is worth noting that these strain bursts are similar to those observed in the nanopillar experiments (Greer & Nix, 2006; Nix et al., 2007; Shan et al., 2008; Brinckmann et al., 2008). In Fig. 5.2(a), green shaded zones correspond to deformation stages associated with plateaus and local minima in the stress curve; these stages are characterized by a decrease in the stacking-fault area and dislocation density, indicating that these deformation stages of constant and decreasing stress are accompanied by dislocation dissociation and annihilation. On the other hand, red shaded regions in Fig. 5.2(a) are associated with local maxima in the stress curve and are characterized by an increase in the dislocation density and the stacking-fault area, indicating nucleation of new dislocations and their propagation in the thin film.
Figure 5.1. (a) Evolution (i.e., dependence on strain during dynamic deformation) of the shear stress (black), dislocation density (red), and stacking-fault area (blue) computed from molecular-dynamics simulation of biaxial tensile straining of a 3.84-nm-thick Cu film. (b) Three-dimensional view of the thin film at the various different states, (i), (ii), (iii), and (iv), marked in (a). Atoms in locally perfect hcp arrangements are colored blue; two consecutive layers of hcp atoms form a stacking fault. Atoms located inside dislocation cores also are colored. Atoms in locally perfect fcc arrangements and the surface atoms are not shown for clarity.
Figure 5.2. Evolution (i.e., dependence on strain during dynamic deformation) of the shear stress (black), dislocation density (red), and stacking-fault area (blue) computed from molecular-dynamics simulations of biaxial tensile straining of Cu films with initial thickness (a) 9.76 nm (sample 2) and (b) 4.43 nm (sample 3). In (a), deformation stages governed by dislocation nucleation and propagation and by dislocation dissociation and annihilation are depicted as red and green shaded zones, respectively.
5.4. Dislocation Depletion Mechanisms in Cu Thin Films

Through detailed analysis of the MD simulation results, various interactions are identified between the gliding dislocations and the stacking faults that lie in their path. These interactions can be classified as (I) reactions between the stacking fault of an obstacle extended dislocation and a mobile perfect dislocation: the Burgers vector, \( \mathbf{b} \), of the mobile perfect dislocation is such that the dislocation (a) can or (b) cannot cross-slip into the plane of the obstacle extended dislocation without dissociation; and (II) reactions between a partial dislocation and the stacking fault of an obstacle extended dislocation.

Figure 5.3(a) shows a sequence of atomic configurations along the pathway of a reaction under category I(a). An extended dislocation is shown to glide in the \((\overline{1}1\overline{1})\) plane, consisting of a leading and a trailing Shockley partial with \( \mathbf{b} = (a/6)[2\overline{1}\overline{1}] \) and \((a/6)[121] \), respectively, where \( a \) is the fcc lattice parameter. A second extended dislocation lies in the \((\overline{1}1\overline{1})\) plane and is bounded by Shockley partials with \( \mathbf{b} = (a/6)[\overline{1}12] \) and \((a/6)[211] \). The stacking fault of the second extended dislocation acts as an obstacle to the glide of the first one. When the leading partial of the mobile extended dislocation approaches the obstacle stacking fault, it “waits” for the trailing partial to “catch up.” This causes the combination of the two partials to form a perfect dislocation with \( \mathbf{b} = (a/2)[110] \). The resulting perfect dislocation glides to and dissociates on the obstacle stacking fault; from this dissociation, two Shockley partial dislocations are formed, with \( \mathbf{b} = (a/6)[1\overline{2}1] \) and \((a/6)[211] \), which glide on the \((\overline{1}1\overline{1})\)
crystallographic plane. The corresponding combination and dissociation reactions are given by

\[(a/6)[21\bar{1}](1\bar{1}1) + (a/6)[12\bar{1}](1\bar{1}1) \rightarrow (a/2)[110](\bar{1}1\bar{1})\]

and \[(a/2)[110](\bar{1}1\bar{1}) \rightarrow (a/6)[211](1\bar{1}1) + (a/6)[12\bar{1}](1\bar{1}1).\]

The glide of the new Shockley partials unzips the obstacle stacking fault and leads to the formation of two extended dislocations in the same plane; the partials of each of the two extended dislocations can combine to either form a perfect dislocation or annihilate if they have opposite Burgers vectors. In the simulations conducted in this study, combination of at least one pair of partials to form a perfect dislocation is observed frequently.

Reactions under category I(b) also were commonly observed in the MD simulations conducted in this study. Figure 5.3(b) shows a sequence of atomic configurations along the pathway for one such reaction. The stacking fault of the obstacle extended dislocation depicted in Fig. 5.3(b) lies on the (1\bar{1}1) plane. A perfect dislocation with \(b = (a/2)[011]\) is shown to glide on the (1\bar{1}1) crystallographic plane toward the obstacle stacking fault. Upon intersecting with the obstacle stacking fault, the perfect dislocation dissociates according to the dissociation reaction

\[(a/2)[011](1\bar{1}1) \rightarrow (a/6)[112](1\bar{1}1) + (a/6)[\bar{3}10] + (a/6)[211](\bar{1}1\bar{1})\]

to form two Shockley partials, one in the plane of the gliding perfect dislocation and another one in the plane of the obstacle stacking fault, and a sessile stair-rod dislocation with \(b = (a/6)[\bar{3}10]\). Subsequently, the produced stair-rod dislocation dissociates according to the reaction
\[(a/6)[\overline{3}10] \rightarrow (a/6)[\overline{1}21](11\overline{1}) + (a/6)[\overline{2}1\overline{1}](\overline{1}1\overline{1}).\]

Figure 5.3. Interactions between gliding dislocations and stacking faults in a Cu ultrathin film (sample 1) under biaxial tensile strain. Sequences of atomic configurations are shown along pathways of dislocation-stacking-fault reaction mechanisms; the structural evolution is from left to right. (a) A gliding extended dislocation effectively cross-slips into the plane of an obstacle stacking fault. The extended dislocation dissociates to form another extended dislocation on the obstacle stacking-fault plane. (b) A gliding perfect dislocation intersects with an obstacle stacking fault and dissociates effectively into two extended dislocations, one of which propagates on the obstacle stacking-fault plane, while the other one breaks through the obstacle stacking fault and glides on the same \{111\} plane as that of the initial perfect dislocation. (c) A partial dislocation dissociates upon intersecting with an obstacle stacking fault. The product dislocations then glide in the plane of the obstacle stacking fault and unzip the stacking fault. Atoms in locally perfect hcp arrangements are colored blue; two consecutive layers of hcp atoms form a stacking fault. Atoms located inside dislocation cores also are colored. Atoms in locally perfect fcc arrangements and the surface atoms are not shown for clarity.
The outcome of the two dissociation reactions is the formation of two extended dislocations in the final state. In effect, upon interacting with the obstacle extended dislocation, a perfect dislocation dissociates into its corresponding Shockley partials and splits the obstacle extended dislocations into two extended dislocations. Many reactions classified under category I(a) and I(b), which are similar to the Friedel-Escaig cross-slip mechanism (Escaig, 1968), are observed in the simulations conducted in this study.

Less frequently, the dissociation of a Shockley partial at the stacking fault of an obstacle extended dislocation is observed. Figure 5.3(c) shows atomic configurations along the pathway of one such reaction. The obstacle and the gliding extended dislocations lie on the (111) and (111) crystallographic planes, respectively. The leading and the trailing partials of the gliding extended dislocation have \( \mathbf{b} = (a / 6)[\overline{1}12] \) and \( (a / 6)[\overline{1}21] \), respectively. Initially, the trailing partial of the gliding extended dislocation is entangled with other dislocations and it is, therefore, unable to glide toward the leading partial, combine with it, and form a perfect dislocation. Upon contact with the obstacle stacking fault, the leading partial dissociates to generate a Shockley partial in the plane of the obstacle stacking fault and a sessile stair-rod dislocation according to the reaction

\[
(a / 6)[\overline{1}12](111) \rightarrow (a / 6)[\overline{1}2\overline{1}](1\overline{1}1) + (a / 3)[100].
\]

The trailing partial of the gliding extended dislocation, which is now separated from the entanglement, glides to react with the stair-rod dislocation according to the reaction

\[
(a / 6)[\overline{1}2\overline{1}](1\overline{1}1) + (a / 3)[100] \rightarrow (a / 6)[\overline{1}2\overline{1}](1\overline{1}1).
\]
Effectively, this reaction leads to annihilation of the gliding extended dislocation and break-up of the obstacle dislocation into two extended dislocations. This reaction mechanism is similar to the Fleischer cross-slip mechanism (Fleischer, 1959). As shown in Fig. 5.3(c), one of these two dislocations is annihilated.

In the thicker film (sample 2), dislocation nucleation mechanisms are observed at the intersection of the thin-film surface and the stacking fault of a threading dislocation. Such a newly nucleated threading dislocation half loop grows to unzip an already existing stacking fault and to meet, eventually, the Shockley partial of the initially present threading dislocation; this results either in formation of a perfect dislocation or in complete annihilation of the threading dislocation half loop. Such dislocation nucleation and interaction mechanisms have been observed in simulations of pristine gold nanopillars (Afanasyev & Sansoz, 2007) and of bulk aluminum (de Koning et al., 2003), as well as in the simulations of structural relaxation of copper thin films after shock loading discussed in Chapter 3.

5.5. Discussion on Dislocation Depletion Mechanisms in Metallic Small-Volume Structures

A stage of constant or decreasing stress during dynamic deformation, known as the “easy-glide” stage, also has been observed in compression experiments of Ni and Au nanopillars (Uchic et al., 2004; Greer et al, 2005; Greer & Nix, 2006; Nix et al., 2007; Shan et al., 2008; Brinckmann et al., 2008). In the nanopillar experiments, this stage was followed by a stage of increasing stress. The stress increase in these nanopillars has been attributed to reduced plastic flow due to a reduction in the
dislocation density. In the case of Ni nanopillars, which had a very high initial dislocation density ($\sim 10^{15}$ m$^{-2}$), the experiments showed that the dislocation density at the end of the “easy-glide” stage was always much lower than that at the beginning; in some cases, the nanopillars were dislocation free before a steep increase in the stress (Shan et al., 2008). Even though the dislocation annihilation mechanisms that are observed in the simulations conducted in this study are different from those discussed in the experiments of nanopillar compression (i.e., dislocation escape to the free surface), the mechanical response of the ultrathin films in these simulations is similar to that observed in the experiments on nanopillars of Ni and Au crystals; during the application of strain, the nanopillar strength reaches levels that are comparable to those of the ultrathin film strength in these simulations, i.e., a substantial fraction ($\sim 40\%$) of the bulk material’s ideal shear strength.

On the other hand, the mechanical behavior of nanopillars of molybdenum, a body-centered cubic (bcc) metal, was found to be similar to that of the bulk material and no size effects were reported (Brinckmann et al., 2008); bcc Mo pillars reached only 7% of bulk molybdenum’s ideal strength, while pillars of fcc Au reached 44% of the bulk crystal’s ideal strength (Brinckmann et al., 2008). Shockley partial dislocations and perfect dislocations are prevalent in fcc and bcc metals, respectively. The glide of perfect dislocations does not leave behind any stacking faults and these dislocations can cross-slip easily without dissociation; as a result, they are not constrained to glide in the same slip plane. Shockley partials, however, cannot cross-slip as easily as the perfect dislocations can without dissociation; therefore, they are constrained to their glide plane and leave stacking faults behind as they glide. These differences in the dislocation
dynamics between fcc and bcc metals are likely to cause the pronounced differences in their mechanical behavior. This finding that stacking faults play a significant role in the annihilation of dislocations in ultrathin films of fcc metals may provide a possible explanation for the experimentally observed differences in the mechanical behavior of fcc and bcc metallic nanopillars.

Friedel-Escaig- and Fleischer-type cross-slip mechanisms similar to those identified in this study also have been reported in recent studies of dislocation interactions with twin boundaries (Dewald & Curtin, 2007a, 2007b; Zhu et al., 2007). Particularly, twin-boundary-mediated dislocation dissociation reactions (also termed slip transfer reactions) were shown to be the rate controlling mechanisms of plastic flow in nano-twinned materials; the high ductility of these materials was attributed to the structural evolution caused by such mechanisms (Zhu et al., 2007). These twin-mediated mechanisms are similar to those in categories I and II in this analysis. For the twin-mediated mechanisms similar to those in categories I(a) and II, the activation barrier was reported to be \(\sim 0.49\) eV, whereas for the mechanisms similar to those in category I(b), it was reported to be \(\sim 0.67\) eV (Zhu et al., 2007); these quantities were computed for perfectly coherent twin-boundaries without any dislocations and for an applied stress that was 78% of the yield strength of the materials considered. When the first such dissociation occurs at the twin boundary, the product dislocations form networks on the obstacle twin boundary causing the twin boundary to become incoherent. Such dislocation networks on the twin boundary lead to higher activation barriers for subsequent dislocation absorption and transmission reactions, causing the hardening of the nano-twinned metal (Zhu et al., 2007); Zhu and co-workers (Zhu et al.,
2007) found that the activation barriers for absorption and transmission for a second dislocation interacting with the twin boundary increased to 1.01 eV and 1.3 eV, respectively.

In these simulations, with stacking faults being the obstacles to dislocation motion, the activation energies for similar mechanisms at similar high-stress conditions are expected to be the same as or lower than those reported in the twin-boundary-dislocation interaction studies. In the mechanisms that we have identified, the obstacle stacking fault unzips during the dissociation reaction and no dislocation networks similar to those observed on nano-twinned copper that lead to subsequent hardening are observed in the thin-film samples employed in this study. Additionally, other hardening mechanisms that are mediated by the dislocations are not observed. These observations and inferences suggest that the strength of small-volume structures of single-crystalline fcc metals is not strongly dependent on their initial dislocation density. These inferences are consistent with experimental observations in Ni nanopillars where the maximum strength of the material was found to be independent of the initial dislocation density (Shan et al., 2008). A rigorous statement on the dependence of the dislocation density on the strength of ultrathin metallic films, however, require further analysis similar to those conducted by Zhu and co-workers (Zhu et al., 2007).

5.6. Additional Mechanisms of Dislocation-Stacking Fault Interactions

The analysis presented in Sec. 5.4 is restricted to those mechanisms of stacking fault-mediated cross-slip of dislocations that lead eventually to dislocation annihilation.
There are, however, other possible outcomes of such stacking fault-dislocation interactions. One such possible outcome is when a partial dislocation passes through the obstacle stacking fault instead of either waiting for the trailing partial near the obstacle stacking fault to form a perfect dislocation (Category I) or cross-slipping on the obstacle stacking fault (Category II). Such a case is shown in Fig. 5.4; in the first sequence of images [top row Figs, 5.4(a)-(c)], only the dislocations and the stacking faults are shown while other atoms are not shown for clarity. In the second sequence of images in Fig. 5.4 [middle row Figs, 5.4(d)-(f)], only the atoms in dislocation cores are shown. In the third sequence of images [bottom row Figs, 5.4(g)-(i)], atoms are colored according to their slip vector. The reference state taken for computing the slip vectors is not shown in Fig. 5.4 (see Sec. 2.4 for details of slip-vector analysis). As shown in Fig. 5.4, an incident partial dislocation with Burgers vector $\mathbf{B}_\alpha$, passes through an obstacle stacking fault created by the glide of the Shockley partial with Burgers vector $\mathbf{D}_\gamma$; during this process, a sessile dislocation with Burgers vector $\alpha\gamma/\mathbf{B}_\mathbf{D}$ is created at the intersection between the obstacle stacking fault and the stacking fault of the extended dislocation with the incident Shockley partial as its leading partial. The sessile dislocation can be considered, effectively, as the product of the interaction of the two Shockley partial dislocations the glide of which leads to the formation of the stacking faults. As it is evident in Fig. 5.4, the leading and the trailing partials of the incident dislocation are far apart; the junction remains stable throughout the duration of the simulation, i.e., the sessile dislocation does not dissociate into Shockley partial dislocations. In this particular case, the incident Shockley partial, $\mathbf{B}_\alpha$, glides past the first obstacle stacking fault and passes successfully through a second obstacle stacking
fault generated due to the glide of another Shockley partial with Burgers vector $A\beta$ to form a second sessile dislocation with Burgers vector $A\alpha/B\beta$.

![Figure 5.4](image)

**Figure 5.4.** Formation of a sessile dislocation as a Shockley partial dislocation passes through the obstacle stacking fault. In the first sequence of images [top row (a)-(c)], dark blue atoms are in the stacking faults and other colored atoms form the dislocation cores; other atoms are not shown for clarity. In the second sequence of images [middle row (d)-(f)], only the atoms in the dislocation cores are shown while in the third sequence of images [bottom row (g)-(i)], atoms are colored according to their slip vector with respect to a reference state that is not shown. In each sequence of images, the evolution is from left to right.

Dislocations with such wide stacking-fault ribbons, however, are not common and the trailing partials can react with the newly formed sessile dislocation when finite-width dislocations interact; such interactions can lead to a variety of interesting dislocation-dislocation reactions. One such interaction identified in the simulations
conducted in this study is shown in Figs. 5.5 and 5.6. This interaction occurs in multiple stages. In the first stage (Fig. 5.5), the leading partial of the incident dislocation, \( \mathbf{B}\alpha \), passes through the obstacle stacking fault that is bound by the Shockley partials \( \mathbf{B}\alpha \) and \( \mathbf{D}\beta \); such an interaction is similar to that discussed above, and creates a sessile dislocation \( \alpha\mathbf{A}/\mathbf{B}\beta \).

**Figure 5.5.** Formation of a sessile dislocation due to the glide of a Shockley partial dislocation through an obstacle stacking fault; this mechanism is very similar to that highlighted in Fig. 5.4. The sequences of images, top row [(a)-(c)], middle row [(i)-(iii)], and bottom row [(1)-(3)], are analogous to those in Fig. 5.4. The color scheme is the same with that of Fig. 5.4. In each sequence of images, the evolution is from left to right.

In the second stage (Fig. 5.6), the trailing partial of the incident dislocation, unlike in the previous case (Fig. 5.4), quickly reaches the formed junction and interacts with the sessile dislocation; this interaction leads to the formation of a second sessile dislocation, \( \alpha\beta \), which has the same line vector as the reacting sessile dislocation \( \alpha\mathbf{A}/\mathbf{B}\beta \), and a Shockley partial dislocation, \( \mathbf{C}\beta \), that glides in the plane of the obstacle.
stacking fault (plane $b$). Interestingly, the trailing partial, $\mathbf{D}\beta$, of the obstacle extended dislocation then interacts with the sessile dislocation, $\mathbf{\alpha}\beta$, and, effectively, it cross-slips into the plane $a$, which is the glide plane of the incident dislocation. As a consequence of this multi-stage stacking-fault mediated interaction between the dislocations, the Burgers vectors of the incident and obstacle dislocations are altered. The overall effective reaction is given by $(B\alpha + \alpha C) (B\alpha + D\beta) \rightarrow (B\alpha + \alpha D) (B\alpha + C\beta)$. Although such interactions are observed rarely in the simulations conducted in this study, the underlying mechanisms shed new light into cross-slip processes in fcc metals with higher propensity for formation of stacking faults.

![Image](image-url)

**Figure 5.6.** Interactions between the sessile dislocation and the trailing Shockley partials of the participating dislocations. The sequences of images, top row [(d)-(f)], middle row [(iv)-(vi)], and bottom row [(4)-(6)], are analogous to those in Fig. 5.4. The color scheme is the same with that of Figs. 5.4 and 5.5. In each sequence of images, the evolution is from left to right.
5.7. Comparative Study of Ultrathin Films of Different Face-Centered Cubic Metals

The mechanical responses of the three different fcc metallic ultrathin films examined in this study are shown in Fig. 5.7. Figures 5.7(a) and 5.7(b) show the evolution of the von Mises shear stress as a fraction of the material’s ideal shear strength, \( \sigma_{vM}/\sigma_u \), and of the dislocation density as a fraction of the initial dislocation density, \( \rho/\rho_0 \), respectively, during the dynamic biaxial straining of the three (Al, Ni, and Cu) thin films; the applied strain rate is constant and the applied strain level, \( \varepsilon \), increases linearly with time until failure. In the simulations conducted in this study, the maximum \( \sigma_{vM} \) for the different metallic films varied between 35.2% and 44.5% of their ideal simple shear strength according to the EAM models employed in the simulations. These maximum \( \sigma_{vM} \) values are consistent with experimental measurements on Au nanopillars that reached 44% of their theoretically predicted ideal pure shear strength (Brinckmann et al., 2008). Figure 5.7(c) shows the evolution of the plastic strain rate, \( \dot{\gamma} = \rho b v \), where \( v \) is used to denote dislocation velocity. For the computation of \( \dot{\gamma} \), a discrete formulation has been implemented, where \( \dot{\gamma} \) is the sum of the contributions from all gliding dislocations: \( \dot{\gamma} = \sum_i \frac{b_i l_i \dot{x}_i}{V} = \sum_i \frac{b_i \dot{S}_i}{V} \), where \( V \) is the volume of the thin film and \( b_i, l_i, \) and \( \dot{x}_i \) are the Burgers vector of, length of, and distance swept by the \( i^{th} \) dislocation over a specified time interval, respectively; \( \dot{S}_i = l_i \dot{x}_i \) is the area swept by a dislocation of length \( l_i \) over the same time interval.
From the stress-strain curves of Fig. 5.7(a), it is evident that the mechanical response of the Al film is significantly different from that of the Ni and Cu films; note that the propensity for stacking-fault formation is higher in Ni and Cu compared to Al (Table 5.1). In the initial deformation stage (stage I), \( \sigma_{\text{eff}} \) increases in all three metallic films. Beyond this stage, in the Ni and Cu films, an extended easy-glide stage (stage II) is observed where the stress remains approximately constant; this easy-glide stage is then followed by a stage of stress build-up (stage III) in the films. In Al, however, \( \sigma_{\text{eff}} \) increases monotonically during stage II. As shown in Fig. 5.7(b), the dislocation density decreases for all three films until the stress build-up is high enough to cause nucleation of new dislocations; nevertheless, during stages II and III, the dislocation annihilation, as well as the rate of dislocation annihilation, is much lower in the Al film than in the Ni and Cu films. As shown in Fig. 5.7(c), for all three films, the plastic strain rate increases during stage I; however, the increase is \(~ 60\%\) greater for the Cu and Ni films compared to that in the Al film. Then, \( \dot{\gamma} \) decreases in the Cu and Ni films until new dislocations are nucleated, whereas it remains nearly constant in the Al film. With nucleation of new dislocations in stage III, \( \dot{\gamma} \) increases again for all three films. Interestingly, the decrease in \( \dot{\gamma} \) in the Cu and Ni films to levels comparable to those in the Al film marks the end of the easy-glide stage in the Cu and Ni films.

In Figs. 5.7(a)-5.7(c), the films’ mechanical response during stage I demonstrates that the increase in von Mises stress, \( \sigma_{\text{eff}} \), corresponds to an initial and early dislocation population that is highly mobile. In stage I, the plastic strain rate increases more in the Ni and Cu films than in the Al film, indicating higher dislocation
mobility in the metallic films with larger stacking-fault areas. In the Cu and Ni films, the high plastic flow rate leads to the dissipation of the stress imparted in the films due to the application of biaxial strain, causing $\sigma_{\text{in}}$ in these films to remain nearly constant during stage II. On the other hand, the higher dislocation mobility in the Ni and Cu films promotes interactions between dislocations resulting in greater dislocation annihilation rates in these films than that observed in the Al film. In the Cu and Ni films, this dislocation annihilation process causes the plastic flow rate to decrease with increasing applied biaxial strain during stage II, Fig. 5.7(c). In contrast, during stage II, the plastic flow rate in the Al thin films remains essentially constant and lower than those in the Ni and Cu films. Due to the fewer interactions between dislocations in the Al film, the dislocation annihilation rate in this film is nearly half as those in the Ni and Cu films. Additionally, the stress dissipation in the Al film is lower due to the lower plastic flow rate and $\sigma_{\text{in}}$ in the Al film increases monotonically in stage II, Fig. 5.7(a).

At the onset of stage III, the plastic flow rates in the Ni and Cu films are reduced below that of the Al thin film and the stress in the Ni and Cu films starts to increase. During stage III, Fig. 5.7(a), the increasing stress for the Ni and Cu films is accompanied by a very low dislocation density (up to about 20% of the initial dislocation density) in these films; the occurrence of such low dislocation densities can be characterized as “dislocation starvation.” In the Al film, however, the limited plastic flow does not promote interactions between dislocations and, consequently, the minimum dislocation density (as a fraction of initial dislocation density) in this film is more than twice as high as that in the Ni and Cu films, Fig. 5.7(b). These differences in the films’ mechanical behavior and dislocation population evolution suggest that
stacking faults play a significant role in the annihilation of dislocations in these ultrathin metallic films.

![Graphs showing stress-strain behavior](image)

**Figure 5.7.** Evolution (i.e., dependence on strain, $\varepsilon$, during dynamic deformation) of the (a) von Mises stress, $\sigma_{VM}$, as a fraction of the ideal shear strength, $\sigma_u$, (b) dislocation density, $\rho$, as a fraction of the initial dislocation density, $\rho_0$, and (c) plastic strain rate, $\dot{\gamma}$, from MD simulation of dynamic biaxial tensile straining of nm-scale-thick films of Al, Cu, and Ni (green, red, and blue lines, respectively).

In the Cu and Ni thin films, Shockley partial dislocations and stacking faults are more prevalent than perfect dislocations and the stacking faults act as sources for dislocation cross slip and eventual dislocation annihilation. These stacking fault-mediated mechanisms of dislocation dissociation and annihilation have been analyzed extensively Sec. 5.4. The mechanisms of dislocation annihilation in Al thin films, however, are primarily due to interactions between perfect dislocations in different glide planes and with opposite Burgers vectors. Figure 5.8 shows top views at different time
instants of the Al film that exhibited the mechanical response shown in Fig. 5.7. In Figs. 5.8(a) and 5.8(b), atoms in a locally perfect fcc environment and surface atoms are not shown for clarity; Fig. 5.8(a) shows the initial microstructure of the film. In Figs. 5.8(a) and 5.8(b), blue and other colored atoms denote atoms in a locally perfect hexagonal close-packed (hcp) environment and atoms in dislocation cores, respectively. In Fig. 5.8(c), atoms that have slipped due to the glide motion of perfect dislocations are shown; each color corresponds to a specific Burgers vector. For clarity, only three Burgers vectors, namely \( \mathbf{b} = (a/2)[101] \), \( (a/2)[\overline{1}00] \), and \( (a/2)[011] \) are included and other atoms are not shown; for example, the golden colored atoms correspond to those that slipped due to the glide of dislocations with \( \mathbf{b} = (a/2)[\overline{1}00] \). In Figs. 5.8(a)-5.8(c), multiple locations are marked to show clearly dislocations with opposite Burgers vectors in different glide planes that interact and annihilate one another.

**Figure 5.8.** Top view of the Al film under an applied biaxial strain of (a) \( \varepsilon = 0 \) and (b) \( \varepsilon = 3\% \). Light green and orange colored atoms form the dislocation cores, while atoms in stacking faults are colored blue; atoms in a locally perfect fcc environment and surface atoms are not shown for clarity. Dislocation pairs that are marked (in circles) in (a) have been annihilated in (b). (c) Top view of the Al film in (b) with atoms colored according to their slip vector (analogous to Burgers vectors). For clarity, only atoms that have slipped with Burgers vectors \( \mathbf{b} = (a/2)[101] \), \( (a/2)[\overline{1}00] \), and \( (a/2)[011] \) are shown and colored green, gold, and red, respectively.
A three-dimensional close view of a sequence of atomic configurations along a representative dislocation annihilation reaction pathway is shown in Fig. 5.9. It is seen that the interaction between two perfect dislocations extending across the film thickness and having Burgers vectors \( \mathbf{b} = (a/2)[\overline{1}0]11\) and \( \mathbf{b} = (a/2)[110](1\overline{1}) \) leads to the annihilation of both of the dislocations. The light green and golden colored atoms are in the dislocation cores, while the dark blue atoms are in the stacking faults; atoms in a locally perfect fcc environment and surface atoms are not shown for clarity. As expected for Al, the dislocation width is very narrow and the dislocation core is asymmetric as has been observed in previous studies of dislocations in fcc metals (Christiansen et al., 2002b). These dislocation annihilation mechanisms are completely analogous to collinear interactions that have been demonstrated in bulk fcc metals (Madec et al., 2003).

![Image](image_url)

**Figure 5.9.** Three-dimensional close view of a sequence of atomic configurations along a representative collinear interaction pathway; the structural evolution is from left to right. Two perfect dislocations gliding in different glide planes interact and annihilate one another completely.

### 5.8. Conclusion

The mechanical behavior was analyzed of nanometer-scale-thick fcc films of different metals based on large-scale MD simulations of dynamic deformation under
biaxial tensile strain. The analyses suggest that in small-volume structures with ultrathin film geometry, dislocation escape to free surfaces may not be the primary mechanism of dislocation starvation. These MD simulations indicate that there are significant differences in the evolution of the flow stress between thin films of metals that have high propensity for formation of stacking faults and those of metals that have low such propensity. In the simulations conducted in this study, the plastic flow in Ni and Cu films is far greater than Al films during the initial stages of the dynamic deformation, and it is likely responsible for the significant stress dissipation causing an extended easy-glide in Cu and Ni thin films.

This analysis identified the physical mechanisms of defect dynamics that govern this mechanical behavior. Specifically, the analysis revealed that stacking faults are likely to provide an important source of dissociation, cross-slip, and annihilation of dislocations in small-volume structures of fcc metals with moderate-to-high propensity for formation of stacking faults. The mechanisms of stacking-fault-mediated cross-slip events analyzed in this study lead to dislocation annihilation or formation of perfect dislocations. In geometries such as those in nanopillars, these perfect dislocations can readily escape to the free surfaces. The mechanism of dislocation annihilation in Al thin films is similar to collinear dislocation annihilation mechanisms identified in bulk fcc metals. The findings in this study lend support to the dislocation starvation phenomenon and expand the current understanding of dislocation depletion mechanisms in fcc metals. Furthermore, this analysis provides new insights into subtle differences in the mechanisms of dislocation depletion ultrathin films of fcc metals and the resulting mechanical behavior of such metallic small-volume structures.
CHAPTER 6
CONTINUUM- AND ATOMIC-SCALE MODELING OF STRAINED-LAYER SEMICONDUCTOR HETEROSTRUCTURES

6.1. Introduction

This Chapter focuses on a theoretical analysis of the deformation mechanics, interfacial stability, strain relaxation kinetics, and surface morphology of strained-layer Si$_{1-x}$Ge$_x$/Si heteroepitaxial systems. Toward this end, a hierarchical approach is developed combining continuum elasticity and dislocation theory with atomistic simulations of structural and compositional relaxation; emphasis is placed on the case of heteroepitaxial growth of Si$_{1-x}$Ge$_x$ on Si(100) compliant substrates of finite thickness. Starting with the implementation of a phenomenological continuum model and demonstrating its capabilities in Sec. 6.2, the analysis aims at a rigorous parameterization of such continuum theoretical models for predicting the mechanical response of strained-layer semiconductor heteroepitaxial systems in Sec. 6.3. A diagrammatic outline of the hierarchical computational approach that has been implemented toward this goal is given in Fig. 6.1.

In this hierarchical approach, a variety of atomic-scale simulation methods that are based on a classical interatomic potential are implemented. The interatomic interactions in the Si$_{1-x}$Ge$_x$/Si systems examined are described by Tersoff’s empirical potential (Tersoff, 1989). The potential has been tested extensively for accurate
description of Si-Ge interactions and has been presented in Chapter 2. Equilibrium Monte Carlo (MC) simulations are employed in order to obtain ensemble averaged equilibrium properties. Since equilibrium properties are of interest rather than dynamical response, MC methods are preferred in order to avoid time-scale limitations of MD simulations. Structural relaxation is obtained by molecular-statics methods for the geometric optimization of a system of interacting atoms, based on minimization of the system’s potential energy with respect to the atomic coordinates. Structural relaxation of atomic coordinates after MC equilibration is essential for computing the strain and the stored elastic energy in a heteroepitaxial film/substrate system. The elastic strain energy and the total strain in the Si$_{1-x}$Ge$_x$/Si film obtained from atomic-scale simulations are then used to parameterize the phenomenological continuum elastic and dislocation theory.

![Diagram](image)

**Figure 6.1.** Diagrammatic outline of the hierarchical computational approach implemented to study mechanical response of strained-layer heteroepitaxial systems.
6.2. Strain Relaxation in the Epitaxial Film During Growth or Thermal Annealing: Analysis Based on Dislocation Mean-Field Theoretical Model

Recently, it has been reported that He\textsuperscript{+} ion implantation and subsequent annealing can result in thin Si\textsubscript{1-x}Ge\textsubscript{x} layers possessing a high degree of strain relaxation, as well as relatively low densities of threading dislocations (Cai et al., 2004). In these experiments, a pseudomorphic Si\textsubscript{1-x}Ge\textsubscript{x} (0.15 < x < 0.30) layer was grown on a Si(100) substrate. He\textsuperscript{+} ions were then implanted into the Si\textsubscript{1-x}Ge\textsubscript{x}/Si heteroepitaxial system at room temperature, with an energy chosen so that the He atoms were distributed mostly below the Si\textsubscript{1-x}Ge\textsubscript{x}/Si interface. Subsequently, the structure was annealed at a temperature T over the range 1023 K ≤ T ≤ 1123 K. Motivated by these experimental findings, the kinetics of strain relaxation in such Si\textsubscript{1-x}Ge\textsubscript{x}/Si film/substrate systems is analyzed theoretically. A phenomenological model is employed in order to investigate strain relaxation as a result of thermal annealing, which is used for post-growth treatment of Si\textsubscript{0.8}Ge\textsubscript{0.2}/Si(100) systems. Both cases are examined, where the annealing may or may not be preceded by a post-growth ion implantation process.

To outline the basic elements of the phenomenological model, an epitaxial film is considered of thickness \( h_f \) grown on a substrate of finite thickness \( h_s \), such as a compliant substrate that is not mechanically constrained at its base (Freund & Nix, 1996; Zepeda-Ruiz et al., 1999). In addition to a model accounting for dislocation-mediated strain relaxation kinetics, the critical film thickness, \( h_{fc} \), for the introduction of the first misfit dislocation at the film/substrate interface is required; determining \( h_{fc} \) sets
an important initial condition for the kinetic model. Assuming the equilibrium lattice parameter of the film to be greater than that of the substrate, the lattice mismatch between the film and the substrate results in a compressive strain, $\varepsilon_f$, for the film and a tensile strain, $\varepsilon_s$, for the substrate. Assuming uniform biaxial deformation when the film/substrate system is coherently strained (i.e., for $h_f \leq h_{f,c}$), $\varepsilon_{xx,i} = \varepsilon_{yy,i} = \varepsilon_i$ and $\sigma_{xx,i} = \sigma_{yy,i} = M_i\varepsilon_i$, where $i = f$, $s$, and $M_f$ and $M_s$ are the corresponding biaxial moduli defined as $M \equiv 2\mu(1+\nu)/(1-\nu)$ with $\mu$ and $\nu$ being the shear modulus and Poisson’s ratio, respectively. Mechanical equilibrium for the biaxially strained film/substrate system requires that $\sigma_f h_f + \sigma_s h_s = 0$, which yields $M_s\varepsilon_s h_s + M_f\varepsilon_f h_f = 0$. In addition, the compatibility condition requires that $\varepsilon_f - \varepsilon_s = \varepsilon_m$, where $\varepsilon_m$ is the lattice-mismatch strain in the film in the limit $h_s \rightarrow \infty$ and is given by $\varepsilon_m = (a_s^0 - a_f^0)/a_f^0$, where $a_s^0$ and $a_f^0$ are the equilibrium (in-plane) lattice parameters for the substrate and film materials, respectively. The resulting strain in the film is $\varepsilon_f = \varepsilon_m/(1+\Lambda(h_f/h_s))$, where $\Lambda \equiv M_f/M_s$.

Considering a dislocation with Burgers vector $b = \{b_x, b_y, b_z\}$, a critical thickness condition can be derived by setting equal to zero the net work required for creating a single misfit dislocation at the interface of the strained heteroepitaxial system. A simple extension of the Freund & Nix theory for compliant-substrate heteroepitaxial systems (Freund & Nix, 1996) to account for different elastic moduli in the film and substrate materials yields the critical film thickness condition

$$\frac{\left[b_x^2 + b_y^2 + (1-v_s)b_z^2\right]}{8\pi(1+v_s)b_x} \left(h_s + \Lambda h_{f,c}\right) \ln \left(\frac{4h_f h_{f,c}}{b(h_s + h_{f,c})}\right) - \varepsilon_m = 0. \quad (6.1)$$
For Si_{1-x}Ge_{x}/Si systems, ε_{m} in Eq. (6.1) is calculated assuming that the dependence on x of the lattice parameter in the alloyed film is in accordance with Vegard’s law (Vegard, 1921).

After the onset of dislocation generation, the strain relaxation kinetics is studied by adopting a variant of the phenomenological model originally proposed by Alexander & Haasen to describe plastic deformation dynamics in semiconductor crystals (Alexander & Haasen, 1968; Alexander, 1986). In this formulation, the speed, v(t), of a gliding dislocation in the epitaxial film is given by

\[ v(t) = v_{0} \left[ \frac{\tau_{\text{eff}}(t)}{\mu_{f}} \right]^{m} \exp \left( -\frac{Q_{n}}{k_{B}T} \right), \]  

(6.2)

where \( v_{0} \) is a reference velocity in a pre-exponential mobility factor, \( \tau_{\text{eff}} \) is the effective stress on the dislocation, \( Q_{n} \) is the Peierls activation barrier (Hirth & Lothe, 1982), \( k_{B} \) is Boltzmann’s constant, \( T \) is temperature, and the exponent \( m \) is taken to be equal to 2 for Si_{1-x}Ge_{x}/Si (Houghton, 1991). The right-hand side of Eq. (6.2) can be viewed as the product of a stress-dependent mobility with a Peach-Koehler force (Hirth & Lothe, 1982) on the dislocation proportional to the effective stress. The rate of dislocation generation is given by

\[ \frac{dN}{dt} = B \left[ \frac{\tau_{\text{eff}}(t)}{\mu_{f}} \right]^{n} \exp \left( -\frac{Q_{n}}{k_{B}T} \right) N(t), \]  

(6.3)

where \( B \) is a material constant, \( n = 2.5 \), \( N \) is the mobile (threading) dislocation density, and \( Q_{n} \) is the activation barrier for dislocation nucleation (Houghton, 1991). The rate of strain relaxation in the film can be expressed by Orowan’s equation (Alexander & Haasen, 1968; Alexander, 1986),
\[
\frac{d\varepsilon}{dt} = b\nu(t)N(t)\cos\lambda,
\]  
\[(6.4)\]

where \( b \) is the magnitude of the Burgers vector and \( \lambda \) is the angle between the Burgers vector and the corresponding strain relaxation direction.

The effective stress, \( \tau_{\text{eff}} \), used in Eqs. (6.3) and (6.4) is defined as

\[
\tau_{\text{eff}} \equiv \tau_{\text{appl}} - \tau_{\text{int}} = M_f \varepsilon_f - \tau_{\text{int}},
\]  
\[(6.5)\]

where \( \tau_{\text{appl}} \) represents the resolved shear stress component of the full stress tensor on the dislocation glide plane and \( \tau_{\text{int}} \) denotes an internal stress arising due to the dislocation field. Equation (6.5) assumes that the deformation remains biaxial as strain relaxes through dislocation formation and propagation; the validity of this assumption depends on the surface orientation (Zepeda-Ruiz et al., 1999; Maroudas et al., 2002) and is guaranteed for \( \text{Si}_{1-x}\text{Ge}_x/\text{Si}(100) \). The internal resolved shear stress component can be expressed as

\[
\tau_{\text{int}} = C(h_f, h_s)M_f b^2 N + AM_f b\sqrt{N},
\]  
\[(6.6a)\]

where

\[
C(h_f, h_s) = \frac{\alpha}{h_f} \ln \left[ \frac{4h_fh_s}{b(h_f + h_s)} \right]
\]  
\[(6.6b)\]

and \( \alpha \) and \( A \) can be used as adjustable parameters. The first contribution to the internal stress, Eq. (6.6a), is due to the self-stress of the generated dislocations and, therefore, proportional to the dislocation density, \( N \). Note that the dependence of \( C \) on \( h_f \) and \( h_s \), Eq. (6.6b), accounts for the interaction of an individual dislocation with the surface of the film and the base of the substrate; this result is rigorous for a misfit dislocation and
is used here only as a scaling expression of a functional dependence generalized for mobile threading dislocations that may or may not be connected to a misfit dislocation. The second term on the right-hand side of Eq. (6.6a) arises due to dislocation-dislocation interactions, which scale (to leading order) with $1/l$, where $l = 1/\sqrt{N}$ is the mean dislocation separation. Finally, in simulating growth experiments, the constant growth rate of film, $v_G$, is taken as

$$\frac{dh_f}{dt} = v_G. \quad (6.7)$$

The results reported are according to the above model, Eqs. (6.1)-(6.7), for the kinetics of strain relaxation in Si$_{0.8}$Ge$_{0.2}$/Si (100) film/substrate systems. The various samples were first grown computationally at a constant growth rate by integrating Eqs. (6.2)-(6.4) and Eq. (6.7) using a Runge-Kutta algorithm. For quantitative predictions, the numerical values of the parameters $V_0$, $m$, $Q_g$, $B$, $n$, and $Q_h$ are taken from the work of Houghton (Houghton, 1991). During growth, the appropriate initial condition is provided by Eq. (6.1), for initiation of dislocation generation for infinitely thick substrates ($h_s \rightarrow \infty$); specifically, Eq. (6.4) is integrated after $h_f$ reached its critical level, $h_{fc}$, with the corresponding strain in the film given by $\varepsilon_f^0 = \varepsilon_f(h_{fc})$ for the coherently strained system under conditions of uniform biaxial deformation. A low dislocation density at the end of the growth process ($N < 10^3 \, \text{cm}^{-2}$) is obtained by adjusting the parameter $\lambda$. The samples were annealed subsequently at 850 °C. For modeling the annealing of the as-grown samples, the initial conditions were provided by the final strain and dislocation density values calculated by the kinetic modeling of the epitaxial growth process. During annealing, the growth velocity, $v_G$, was set to zero and the
governing equations were integrated to steady state. Ion implantation causes substantial dislocation nucleation; therefore, for modeling the post-implantation annealing of samples implanted by He\(^{+}\) ions, initial dislocation densities were used that were higher by orders of magnitude than those obtained from kinetic modeling of growth. In addition, ion implantation has direct effects on the deformation behavior of the substrate (Freund, 1997). Consequently, the implantation depth (defined as the difference \(R_p-h_f\), where \(R_p\) corresponds to the location of the maximum of the implanted atom distribution) was taken to be equal to an effective finite substrate thickness, \(h_s\), i.e., it was assumed that the implantation results practically in a thin substrate analogous to a silicon-on-insulator substrate arrangement (Powell \textit{et al.}, 1994). Therefore, for modeling the annealing of post-implanted samples, the substrate was not considered as infinitely thick. Furthermore, in modeling the thermal annealing process, \(\alpha\) and \(A\) were treated as adjustable parameters to maintain a final dislocation density of \(N \sim 10^7 \text{ cm}^{-2}\), consistent with the experiments (Cai \textit{et al.}, 2004).

The results of the modeling described above are shown in Fig. 6.2 and are compared with the experimental measurements of Cai and coworkers (Cai \textit{et al.}, 2004). The results demonstrate the dependence on the film thickness, \(h_f\), of the strain relaxation in the film, \(\delta = 1-\varepsilon_f/\varepsilon_m = (a_s^0-a_f)/(a_s^0-a_f^0)\), where \(a_f\) is the in-plane lattice parameter of the relaxed epitaxial film. Figure 6.2(a) shows results for thermal annealing of un-implanted samples. Different curves in Fig. 6.2(a) correspond to different values of the growth rate, \(v_G\), while the different symbols correspond to experimental measurements taken for samples grown in different reactors (Cai \textit{et al.}, 2004). Figure 6.2(b) shows
results for thermal annealing of samples after their implantation with He\(^+\) ions. Each curve corresponds to a given set of values for the adjustable parameters \(\alpha\) and \(A\), while different symbols correspond to different experimental conditions (Cai et al., 2004). It is evident from Figs. 6.2(a) and 6.2(b) that the phenomenological model has captured the strain relaxation behavior in the various Si\(_{0.3}\)Ge\(_{0.7}\)/Si(100) samples in accordance with the experimental measurements. In addition, the inset to Fig. 2(b) shows that \(\delta(h_f)\) reaches in all cases (i.e., independent of the experimental conditions) a common saturation level. This implies that strain relaxation of thick films upon annealing of implanted samples is insensitive to the implantation dose and depth, which is in excellent agreement with the experimental observations by Cai and coworkers (Cai et al., 2004). Furthermore, the model prediction for the \(\delta(h_f)\) saturation level for thick films is \(\sim 90\%\), in very good agreement with the experimental value of \(\sim 85\%\). Finally, the agreement of these phenomenological modeling results with the experimental data (Cai et al., 2004) is comparable to the agreement reported between the data and state-of-the-art discrete dislocation-dynamics simulations for the case of post-implantation annealing (Schwarz et al., 2004).
Figure 6.2. Strain relaxation, $\delta$, as a function of film thickness, $h_f$, for Si$_{0.8}$Ge$_{0.2}$/Si(100) samples annealed after epitaxial growth un-implanted (a) or after He$^+$ implantation (b). The solid curves correspond to the modeling results, while the discrete points correspond to experimental data; different symbols correspond to different experimental conditions (from Cai et al., 2004). The inset in (b) demonstrates that for very thick films, the $\delta(h_f)$ dependence reaches a common plateau value around 90% that is practically independent of the experimental conditions.
6.3. Parameterization of Dislocation-Mean-Field Models using Atomistic Studies

Further parameterization of simplified coarse-grained models, such as the ones described in Sec. 6.2 based on fundamental analysis of these systems using molecular-dynamics and dislocation-dynamics simulations will contribute significantly to efficient quantitative studies of strain relaxation in technologically important semiconductor heterostructures. As a first step toward such parameterizations of simplified coarse-grained models, the equations governing the deformation dynamics are parameterized before the onset of plasticity in the heteroepitaxial system. Mechanical equilibrium for the biaxially strained film/substrate system, as described in Sec. 6.2 requires that

\[ \varepsilon_x = -\varepsilon_m \frac{\Lambda(h_f / h_s)}{1 + \Lambda(h_f / h_s)} \]  
\[ \text{and} \]

\[ \varepsilon_f = \frac{\varepsilon_m}{1 + \Lambda(h_f / h_s)} \]  
\[ \text{where } \Lambda \equiv \frac{M_f}{M_s} \].

The elastic strain energy \( U_{el} = \int \frac{1}{2} tr(\sigma \varepsilon) dV \) stored in a coherently strained film/substrate system is given by

\[ U_{el} = \frac{1}{2} \sum_{i,f,x} (\sigma_{xx,i} \varepsilon_{xx,i} + \sigma_{yy,i} \varepsilon_{yy,i}) A_i h_i \]  
\[ \text{As a result, } \frac{U_{el}}{A_s} = \frac{M_f \varepsilon_m^2 h_f}{1 + \Lambda(h_f + h_s)}. \]
In Eq. (6.9), $U$ is the elastic strain energy stored in the heterostructure and $A_s$ is the surface area of the film/substrate interface.

Appropriate parameterization of the strain in the film, strain in the substrate, and the elastic strain energy as a function of overall film composition, grading in the film, and compliant substrate thickness will help parameterize Eq. (6.1) and improve the accuracy in the estimation of the critical film thickness beyond which onset of dislocation generation occurs. A variant of the MC simulation procedure, originally suggested by Foiles (Foiles, 1985; Kelires & Tersoff, 1989) is implemented, employing a three-step computational sequence. The first step in the sequence involves one compositional MC sweep over all Ge atoms in the film, where each MC step consists of a trial to exchange a Ge atom with a randomly chosen Si atom for “compositional” relaxation. If this trial is accepted, it is then followed by (second step) many (typically 50) MC sweeps over all atoms, where each step consists of a continuous-space atomic displacement trial of each atom for “structural” relaxation. This is then followed by (third step) one MC step for cell-size relaxation, where the MC step consists of a trial to adjust the cell dimensions in the principle directions normal to the film/substrate interface for “strain” relaxation. In all cases, the trial moves are accepted or rejected according to the Metropolis criterion (Allen & Tildesley, 1990). The resulting relaxed configurations are then treated using conjugate-gradient energy minimization to obtain the equilibrium energies for the final relaxed structural configurations that are then used to parameterize the continuum theory given by Eqs. (6.8) and (6.9).

First, a prototypical system is considered of a free-standing slab of an alloy with 50% Ge and 50% Si with free surfaces oriented normal to the [100] crystallographic
direction. Upon relaxing the slab using the hybrid compositional relaxation scheme described above, Ge segregates to the free surfaces of the slab. The equilibrium compositional distribution of Ge in the slab is shown in Fig. 6.3. The results of Fig. 6.3 for the Ge profile are in excellent agreement with results reported by Kelires & Tersoff using grand-canonical Monte Carlo simulations (Kelires & Tersoff, 1989).

![Graph showing Ge concentration profile](image)

**Figure 6.3.** Ge segregation at free surfaces after compositional relaxation of a free-standing slab with free surfaces oriented normal to the [001] crystallographic plane. The initial composition of the free standing film is a randomly distributed $\text{Si}_{0.5}\text{Ge}_{0.5}$ alloy, with a total of 28 atomic layers. The percentage of Ge atoms in each atomic layer is plotted. The inset plots the potential energy per atom (eV/atom) as a function of MC steps, indicating clearly convergence of the computational scheme and equilibration.

Next, a second prototypical system is considered wherein a pure Ge film is grown on a pure Si substrate. Those cases are considered where the substrate is infinitely thick compared to the film and where the substrate is compliant with finite thickness $h_s$. For an infinitely thick substrate, there is no biaxial strain relaxation in the epitaxial film parallel to the film/substrate interface. Upon relaxing the heterostructure using the above hybrid compositional relaxation scheme, the equilibrium energy and
atomic positions are obtained, which are used to calculate the biaxial strain in the film and the stored elastic energy in the film/substrate system. The resulting biaxial strain in the film, $\varepsilon_f$, as a function of film thickness, $h_f$, for a compliant substrate thickness, $h_s = 7$ nm, is shown in Fig. 6.4(a). The continuous curve is the best fit to the simulation results (solid symbols) according to the linear elastic theory, Eq. (6.8). The corresponding elastic strain energy, $U_{el}/A_s$, in the film as a function of film thickness for a compliant substrate thickness, $h_s = 7$ nm, and for an infinitely thick substrate are shown in Fig. 6.4(b). The continuous curves are the best fits to the simulation results (solid symbols) according to the linear elastic theory Eq. (6.9), for both substrate thicknesses. It is evident from Fig. 6.4 that the simulation results are in excellent agreement with the linear elastic theory. From fitting such simulation results according to Eqs. (6.8) and (6.9) and fits for various substrate thicknesses and film compositions, the elastic moduli of the film and the substrate ($M_f$, $M_s$, and $\Lambda$) can be parameterized as a function of film composition, substrate thickness, and compositional grading. The resulting elastic parameters can then be incorporated into Eq. (6.1) for a more accurate prediction of the critical film thickness, $h_{fc}$, for the onset of dislocation generation.
Figure 6.4. (a) Biaxial strain in a relaxed epitaxial film of pure Ge on a compliant substrate of pure Si of thickness $h_s = 7$ nm, as a function of film thickness. The continuous curve is the best fit to the simulation results (blue symbols) according to linear elastic theory, Eq. (6.8). (b) Elastic strain energy stored in the epitaxial film/substrate system as a function of film thickness for a thick Si substrate (green symbols) and for a compliant substrate of thickness $h_s = 7$ nm (blue symbols as a function of film thickness. The continuous curves are the best fits to the simulation results according to linear elastic theory, Eq. (6.9).
6.4. Conclusion

The kinetics of strain relaxation is analyzed in Si$_{1-x}$Ge$_x$ epitaxial films grown layer-by-layer on Si(001) substrates based on a phenomenological dislocation mean-field theoretical model. The model is successful in interpreting the experimentally observed strain relaxation behavior of Si$_{1-x}$Ge$_x$/Si(001) systems and yields results in excellent agreement with experimental data for Si$_{0.8}$Ge$_{0.2}$/Si(001) samples that are thermally annealed after epitaxial growth either unimplanted or after He$^+$ implantation. Furthermore, I have shown how atomic-scale simulation methods can be employed to provide proper parameterizations of such simplified coarse-grained models. Such parameterizations can contribute significantly to computationally efficient quantitative studies of strain relaxation in semiconductor heterostructures.
CHAPTER 7

FUTURE DIRECTIONS

7.1. Quantifying Interactions between Dislocations and Stacking Faults in Thin Films of Face-Centered Cubic Metals

Various possible interactions between stacking faults and dislocations, both Shockley partial dislocations and perfect dislocations, that lead eventually to dislocation annihilation during the dynamic deformation of single-crystalline ultrathin metallic films were discussed in detail in Sec. 5.4. Additionally, a few additional mechanisms of dislocation-stacking-fault interactions were analyzed in Sec. 5.6; these additional mechanisms do not lead necessarily to dislocation annihilation but provide some important fundamental insights into dislocation-stacking-fault interactions. Although many classes of such interactions during dynamic deformation simulations of biaxial straining of ultrathin films were identified in this thesis, a systematic study of the interactions between obstacle stacking faults and the incident (to the obstacle) dislocations is necessary in order to place these reactions and their relevance to materials deformation in a larger context. Limitations of the MD simulations, such as the time scales that can be captured, as well as the required high strain rates due to such time limitations, can be overcome by quantifying, as a function of the applied strains and stresses the activation barriers that govern the kinetics of these stacking-fault-mediated dislocation mechanisms (reactions).
Toward this end, systematic studies of various possible interactions can be conducted through the construction of optimal dislocation reaction pathways. The Climbing-Image Nudged Elastic Band method (CINEB) (Henkelman & Jonsson, 2000) is a chain-of-states optimization approach for accurate predictions of saddle points on reaction pathways and the associated thermal activation energy barriers. CINEB does not suffer from the severe strain-rate limitations associated with MD simulations and it can be used on MD-generated atomic configurations to determine the minimum-energy path (MEP) and the corresponding saddle point(s) and activation energy barrier(s) as the reaction (or kinetic process, in general) progresses. In a CINEB calculation, two end states are determined initially. A discretized elastic band consisting of a finite number (say N+1) of replicas of the system is constructed that are denoted by \([\mathbf{R}_0, \mathbf{R}_1, \mathbf{R}_2, \ldots, \mathbf{R}_N]\); these replicas are obtained from MD simulations. In this notation \(\mathbf{R}_0\) and \(\mathbf{R}_N\) are the end points of the elastic band. Each node \(i\) in the band is connected to node \(i+1\) by a spring with spring constant \(k_i\) and to node \(i-1\) by a spring with spring constant \(k_{i-1}\). For simplicity, the spring constants for the all the node connecting springs can be considered to be the same, \(k\). Then, the total force acting on each node is the sum of the spring force along the local tangent to the elastic band and the true force perpendicular to the local tangent, \(\mathbf{F}_i = \mathbf{F}_{i,i+1}^s - \nabla U(\mathbf{R}_i)_{\perp}\), where the true force is given by \(\nabla U(\mathbf{R}_i)_{\perp} = \nabla U(\mathbf{R}_i) - \nabla V(\mathbf{R}_i)\mathbf{\tau}_i\). Here, \(U\) is the energy of the system as provided by the interatomic potential and \(\mathbf{\tau}_i\) is the normalized local tangent to the elastic band at node \(i\). The spring force is given by \(\mathbf{F}_{i,i+1}^s = k(\mid\mathbf{R}_{i+1} - \mathbf{R}_i\mid - \mid\mathbf{R}_i - \mathbf{R}_{i-1}\mid)\mathbf{\tau}_i\). The force on the node with the highest energy, \(i_{\text{max}}\), along the MEP under construction, however, is taken as the full force due to the potential with the component along the elastic band inverted,
and is given by $\mathbf{F}_{\text{max}} = -\nabla U(R_{\text{max}}) + 2\nabla U(R_{\text{max}}) \cdot \mathbf{\tau}_{\text{max}} \cdot \mathbf{\tau}_{\text{max}}$. An optimization algorithm can be used to move the nodes according to the force on each node; the band then converges to the MEP. Such relaxations (constructions of optimal pathways) can be conducted for different shear stresses to determine the dependence on the applied stress of the corresponding energy barrier.

The physical attempt frequency, $\nu_0$, can also be determined using the CINEB method as $\nu_0 = \frac{1}{2\pi} \sqrt{\frac{C}{m}}$, where $C = \left( \frac{d^2 U(x_{\text{MEP}}(s))}{ds^2} \right)_{s=0}$ is the initial energy curvature along the MEP and $s = \int_{x_{\text{initial}}}^{x_{\text{final}}} \sqrt{dx_{\text{MEP}}^3 \cdot dx_{\text{MEP}}^3}$ is the arc used to parameterize the energy curvature curve in the proper hyperspace. With an appropriate, $\sigma(\dot{e})$, stress and strain rate relationship, harmonic approximation of the transition state theory can be used (Zhu et al., 2008) to obtain a strain rate and temperature dependence of the stacking-fault-mediated cross-slip events under construction. For example, the stress is related to the applied strain rate in the approximation of a perfectly elastic response by $\dot{\sigma} = E\dot{e}_{\text{ij}}$.

The survival rate of the initial state, in the probabilistic measure, can be related easily to the applied stress and applied strain rates (Zhu et al., 2008). The proper treatment, however, becomes more complicated in the case of dislocation-ridden thin films, where plastic strain $\gamma$ (defined by $\dot{\gamma} = \rho u b$ as shown in Sec. 5.7) contributes to the structural dynamics; such cases, which better represent experimental conditions, require careful formulation.
7.2. Dependence of Stacking-Fault Width on Characteristic Length of Small-Volume Structures

Another important factor in the interactions between incident dislocations and obstacle stacking faults is the width of the obstacle stacking fault. The probabilities of occurrence of such interactions are dependent on the width of the stacking faults and, hence, it is important to identify the dependence of the stacking-fault width on the simulation/experimental conditions. From the simulation studies in this thesis, it can be assumed that, in addition to the applied stress tensor, the stable width of the stacking faults of dislocations that extend across the film thickness depends strongly on the film thickness; this can be generalized to a strong dependence on the characteristic length of the small-volume structure under consideration. Systematic equilibrium studies with corresponding characteristic length as a parameter, using a wide variety of already available molecular-statics techniques, as well as Dislocation Dynamics methods (Madec et al., 2003; Bulatov et al., 2006), can provide quantitative relations for the dependence of the stacking-fault width on the characteristic length (the film thickness for ultrathin films). Such quantitative relations will enable good predictions of the stress and size ranges over which the stacking-fault-mediated interactions of dislocations in small-volume structures, e.g., those extending across the film thickness in ultrathin films, play an important role in determining the strength of such material structures.
7.3. Effects of Surface Orientation on Critical Strength of and Incipient Plastic Deformation Mechanisms in Ultrathin Films

The simulation studies in this thesis show that the surface orientation and, by extension, the stresses generated due to applied biaxial straining along crystallographic directions perpendicular to that of the film surface normal vector play an important role in determining the mechanisms of strain relaxation; for example, for high levels of applied biaxial strain, fcc-to-hcp martensitic transformation occurs in \((110)\)-oriented Cu thin films direction (Chapter 4) whereas nano-domains are formed for \((111)\)-oriented Cu thin films (Chapter 3). However, the role of film surface orientation in incipient plasticity is less clear. A systematic study (a) based on MD simulations of dynamic deformation to identify the differences in the mechanisms of incipient plastic deformation as a function surface orientation; and (b) based on molecular-statics calculations to determine the barriers for such mechanisms as a function of planar strain (or applied stress) would provide new and relevant information about the plastic response to planar strains of thin film orientations observed during sputtering and electro-plating processes.

7.4. Studies of Plastic Deformation in Capped Thin Films

Although the primary focus of this thesis was the plastic deformation mechanisms of free-standing thin films, capped thin films are more common in micro-electronic devices; for example, thin-film interconnects are capped by layers of \(\text{SiO}_x\text{N}_y\)
or by metals such as Ta or W. Misfit dislocations are formed at the interfaces of the
capped layers with the Cu films due to differences in the lattice structure of the capped
layer and the Cu interconnect that induce lattice-mismatch strains. Furthermore,
reactions can occur at the interface between the capped layer and the Cu thin film.
Under such circumstances, glide of dislocations that extend across the thin films or that
of the threading dislocations is controlled by the microstructure of the interface, while
in free-standing thin films the dislocations glide readily. Interactions between misfit
dislocations at the interface of the capped layer and the Cu thin film and those
extending across the Cu thin film (e.g., threading dislocations) are more likely to control
the mechanical properties of the nano-composite material structure rather than the
interactions between dislocations in the thin film. Understanding the underlying
dislocation dynamics in capped Cu thin films under planar strain (biaxial strain) is
essential for developing phenomenological models that can predict accurately the
behavior of micro-electronic devices.

Extending the simulation framework developed in this thesis to study Cu thin
films that are capped with metals such as Ni, Ta, W, or Co is a fruitful future research
direction. Interatomic potentials can be developed for such heterostructures by fitting
the EAM potentials to interfacial properties; these properties can be obtained by first-
principles DFT calculations. Alternately, ReaxFF (van Duin et al., 2001, 2003; Buehler
et al., 2006), a force field for reactive systems, can be used to describe interatomic
interactions at the Cu/SiO$_x$N$_y$ interface in order to model accurately the corresponding
dissociation of bonds. An EAM potential (Mishin et al., 2001) can be used to model Cu-
Cu interatomic interactions beyond a cut-off distance from the interface. Such a
simulation set up can be used to study the role of interfaces in the plastic deformation dynamics and, ultimately, the strength of Cu interconnects or other metallic layers in micro-mechanical devices.
APPENDIX A

DEFECTS IN CRYSTALS AND DISLOCATIONS IN
FACE-CENTERED CUBIC METALS

A.1. General Classification of Defects in Crystals

Defects in crystalline lattices are classified, in general, according to their dimensions. Point defects or atomic-size defects, which can be considered as 0-dimensional defects, include vacancies and self-interstitials, as well as substitutional and interstitial impurities. A vacancy refers to the absence of an atom from a lattice site. An interstitial, on the other hand, refers to the presence of an atom in a position not defined by the primitive lattice vectors (i.e., at an interstitial site of the lattice). Interstitials can be either self-interstitials, i.e., the interstitial atom and the atoms occupying the lattice sites are of the same element, or extrinsic interstitial or interstitial impurities, where the interstitial atom is of a different element from that of the atoms occupying the lattice sites. Atoms can be found to be knocked off from their ideal lattice positions and to occupy interstitial positions, leading to the existence of both vacancies and self-interstitials. Such vacancy-interstitial pairs are called Frenkel pairs. Figure A.1 shows different types of point defects in a simple lattice.
Figure A.1. Schematic of simple lattice structure showing different types of point defects (Foell, 2007).

One-dimensional defects or line defects are formed when atomic half planes are displaced or dislocated from their ideal lattice positions. The generation of such line defect or dislocation is shown in Fig. A.2. Let us consider a general perfect crystal shown in Fig. A.2(a) in which we wish to create a dislocation. We make a cut in the crystal from one edge to an arbitrary depth inside the crystal; the cut plane here is shown with red lines, and the unit vector along the dislocation line we intend to create is given by \( \mathbf{t} \). We can then move the two parts of the crystal away from each other; theoretically, we can move them in any direction and by any magnitude (these directions are often defined by the lattice structure, the material occupying the lattice sites, and the conditions of operation, i.e., external forces in the form of stresses). In this simple case, we move the two parts of the crystal away from each other as shown in Fig. A.2(b) by a vector \( \mathbf{b} \). We then fill the crystal with material in order to preserve the crystal structure. We have, thus, created a dislocation with unit line vector \( \mathbf{t} \) [parallel to the red line inside the crystal in Fig. A.2(c)] and Burgers vector \( \mathbf{b} \). A dislocation line cannot end arbitrarily in the interior of the material; it can end at the crystal surface, at
an interface, at a dislocation junction, or it can form a loop and, therefore, end on itself (in Fig. A.2, the dislocation line ends at the surface of the crystal).

Figure A.2. Construction of an edge dislocation, with Burgers vector $\mathbf{b}$ and unit line vector $\mathbf{t}$, using a “Volterra knife” (Foell, 2007).

Figure A.3. (a) Generation of an edge dislocation by a shear stress $\sigma$; as can be realized from the image, the Burgers vector of the dislocation is along the plane of the image whereas the dislocation line vector is perpendicular to the plane of the image. (b) Movement of the dislocation (glide motion) through the crystal (Foell, 2007).

If the unit line vector $\mathbf{t}$ and the Burgers vector $\mathbf{b}$ are perpendicular to each other, the dislocation is said to have pure-edge character; when $\mathbf{b}$ and $\mathbf{t}$ are parallel to each other, the dislocation has a pure-screw character. In reality, many dislocations are curved and they have a mixed character, i.e., the Burgers vector of the dislocation has an edge component, as well as a screw component. A dislocation can move on (glide) or perpendicular (climb) to its glide plane (the unit vector normal to the glide plane is
given by \( \mathbf{n} = \mathbf{b} \times \mathbf{t} \), grow, multiply, or annihilate; such a movement of and interaction between dislocations occurs due to forces (due to stresses) exerted on the dislocations, and leads to plastic deformation in crystals. A schematic of the dislocation glide motion is shown in Fig. A.3. The properties of dislocations in FCC metals are discussed in more detail in Sec. A.2.

In nature, crystalline materials usually consist of regions of crystals or grains with multiple orientations; if the different grains are made of the same material, the interfaces formed between them are called the grain boundaries. The grain boundary is a two-dimensional (i.e., areal) defect in the material. If the grains are made of different materials, the resulting boundary is called an interface and if the grains are of the same material but of different crystal phases (such as fcc or hcp), the boundary is called a phase boundary. Figure A.3 shows a schematic of a grain boundary. Understanding the role of such two-dimensional defects in determining materials properties is a topic of great interest to the materials science community.

![Schematic of a crystal with two grains (bicrystal) separated by a grain boundary](image)

**Figure A.4.** Schematic of a crystal with two grains (bicrystal) separated by a grain boundary (H. Foell, 2007).
Another kind of two-dimensional defects are called stacking faults, and they are created due to the glide of certain types of dislocations. Stacking faults, as the name suggests, are defects caused due to a local change in the stacking sequence of the atoms in a crystal. Stacking faults are observed commonly in fcc metals; when certain types of dislocations glide in the fcc crystal, they change the local stacking order from fcc to hcp, thus creating a stacking fault. Dislocations bind the stacking faults always or, the stacking faults end at the interfaces or free surfaces.

A.2. Dislocations in Face-Centered Cubic Metals

As discussed in the Sec. A.1, dislocations are formed due to the slip of atoms from their ideal lattice sites; such a slip and dislocation formation is the most preferred method of deformation in single crystals. A dislocation is characterized by its Burgers vector \( \mathbf{b} \), its unit line vector \( \mathbf{t} \), and unit vector normal to the glide plane \( \mathbf{n} \). Dislocation theory (Hirth & Lothe, 1982) predicts that only a few low-index glide planes and directions should be observed for a given crystal system and this prediction has been confirmed in numerous experimental observations (Hirth & Lothe, 1982). According to dislocation theory, the planes with the largest interplanar spacing, \( d \), are the closest-packed planes in a crystal, and in turn, these planes have the lowest \( \{hkl\} \) or Miller indices. In the case of fcc metals, \( \{111\} \) planes are the closest packed ones with \( d = a_0/\sqrt{3} \) where \( a_0 \) is the equilibrium lattice parameter. Therefore, dislocations in fcc metals glide most often on \( \{111\} \) planes.

Let us consider a dislocation produced by making a cut on a plane \( AB \), as shown in Fig. A.5(a), for a simple cubic lattice. If the displacement is a primitive lattice
vector, or a vector that is the sum of primitive lattice vectors, then we obtain a configuration such as that shown in Fig. A.5(b). This dislocation has a misfit energy, \( E(r) \), (potential energy of the dislocation field) mainly localized near the core (centered at \( r = 0 \)) and such a dislocation is called a perfect dislocation. In fcc metals, the stable perfect dislocation has Burgers vector \( \mathbf{b} = (a_0/2) \langle 110 \rangle \). If the displacement cannot be decomposed into a sum of primitive lattice vectors, a different type of configuration results, as shown in Fig. A.5(c); in such cases, in addition to the misfit energy, this configuration has a long-range, divergent misfit-energy contribution caused by the mismatch of atoms across the displaced surface \( AB \). For a restricted number of Burgers vectors in fcc metals, such a misfit energy is low compared to the misfit energy of the dislocation core and, hence, a fcc crystal can contain such imperfect or partial dislocations.

**Figure A.5.** (a) Cut crystal, which is displaced along \( AB \) to form either (b) a perfect dislocation or (b) an imperfect or partial dislocation (Hirth & Lothe, 1982).
Crystals with fcc lattice structures are generated by stacking close-packed layers on top of one another in a fashion illustrated in Fig. A.6. Given a layer \( A \), close packing can be extended by stacking the next layer so that its atoms occupy \( B \) or \( C \) sites. Here \( A, B, \) and \( C \) refer to the three possible layer positions in a projection normal to the close-packed layers. The stacking sequence corresponding to an fcc crystal is \( \ldots ABCABCABC \ldots \), while that for a hcp crystal is \( \ldots ABABABABA \ldots \). When a partial dislocation is created in a fcc crystal, the fcc stacking sequence changes to the hcp stacking sequence; the stacking sequence in the crystal with a stacking fault can be \( \ldots ABCABCAABCAB \ldots \).

**Figure A.6.** Projection normal to the (111) plane showing the three types of stacking layer positions \( A, B, \) and \( C \). Various vectors in an fcc lattice also are shown (Hirth & Lothe, 1982).

Let us suppose that the close-packed layer shown in Fig. A.7 corresponds to the bottom side of the glide plane of a dislocation, and that the atoms above the layer are originally in sites such as the one denoted by \( (a) \). The passage of the perfect dislocation with Burgers vector \( \mathbf{b} = (a_o / 2)[\overline{1}01] \) from the bottom of the figure to the top results in the atom originally at \( (a) \) being displaced to \( (c) \). Motion of the atom along the straight path from \( (a) \) to \( (c) \) involves a large dilatation (assuming a hard-sphere model) normal to the slip plane, and hence a larger misfit energy than does the motion along the path.
(a) to (b) to (c), so that the letter path should be favored. The intermediate position (b), however, is that of an intrinsic stacking fault and the displacement of \( (a_o / 6)[\overline{2}11] \) is required to form configuration (b). Although partial dislocations of many kinds have been observed in fcc crystals, the most commonly observed partial dislocations have Burgers vector, \( b = (a_o/6) \langle 112 \rangle \). These kinds of partial dislocations, called Shockley partial dislocations, have a low activation barrier to glide; consequently, these partial dislocations are mobile. As shown in this thought experiment, perfect dislocations can dissociate into partial dislocations and partial dislocations can combine to form perfect dislocations. More generally, dislocations can interact with each other, very often conserving the total Burgers vector, and lead to different types of dislocations or they can be annihilated completely.

![Diagram of crystal structure](image)

**Figure A.7.** Atoms on the bottom side of a (111) glide plane. A perfect dislocation and its component Shockley partial dislocations are also shown (Hirth & Lothe, 1982).

A convenient representation of the Burgers vectors (at least perfect dislocations and Shockley partial dislocations) was introduced by Thompson, who created a schematic, the so called *Thompson tetrahedron*, by joining the atoms in a one-eighth
conventional unit cell by straight lines (Fig. A.8). The inner and outer faces of the tetrahedron represent four (eight if positive and negative directions are distinguished) possible \{111\} glide planes and the edges of the tetrahedron correspond to six (twelve if positive and negative directions are distinguished) \langle 110 \rangle glide directions of the fcc structure. The atom at the origin is \(D\) and the others are labeled \(A, B, C\) in a clockwise sense. The midpoints of the faces opposite \(A, B, C, D\) are labeled \(\alpha, \beta, \gamma, \text{ and } \delta\), respectively (not shown in the figure), and the planes opposite \(A, B, C, D\) are labeled \(a, b, c, \text{ and } d\), respectively (not shown in the figure).

![Diagram of a one-eighth conventional unit cell in the fcc lattice with an enclosed tetrahedron, the faces of which mark the possible glide planes and the edges mark the possible glide directions (Hirth & Lothe, 1982).](image)

**Figure A.8.** A one-eighth conventional unit cell in the fcc lattice with an enclosed tetrahedron, the faces of which mark the possible glide planes and the edges mark the possible glide directions (Hirth & Lothe, 1982).

The opened up tetrahedron is shown in Fig. A.9: the \{111\} planes are represented by letters, for example \(111\) becomes \((d)\); the perfect dislocation \((a_0/2)[\bar{1}01]\) becomes \(CB\) and partial dislocation \((a_0/6)[\bar{2}11]\) becomes \(B\delta\). The dissociation of \((a_0/2)[\bar{1}01]\) to \((a_0/6)[\bar{2}11]\) and \((a_0/6)[\bar{1}2]\), for example is given by \(CB \rightarrow B\delta + \delta C\).
Figure A.9. A Thompson tetrahedron opened at the corner D. Both the Thompson notation and one possible set of indices for the same plane and directions are shown. The notation of the line vectors is such that the sense of the direction is also shown (Hirth & Lothe, 1982).

In addition to Shockley partial dislocations (which are mobile), there are other types of partial dislocations, often formed by interactions between Shockley partials; these other partials have Burgers vectors such that the activation barrier for them to glide is very high. Such partial dislocations that do not glide under most circumstances are called sessile dislocations. For detailed discussions on sessile dislocations, as well as the theory dislocations, the reader is referred to the textbook Theory of Dislocations (Hirth & Lothe, 1982).
BIBLIOGRAPHY


