Discrete dislocation dynamics simulations to interpret plasticity size and surface effects in freestanding FCC thin films

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Abstract

Strong size effects have been experimentally observed when microstructural features approach the geometric dimensions of the sample. In this work experimental investigations and discrete dislocation analyses of plastic deformation in metallic thin films have been performed. Columnar grains representative of the film microstructure are here considered. Simulations are based on the assumptions that sources are scarcely available in geometrically confined systems and nucleation sites are mainly located at grain boundaries. Especially, we investigated the influence on the mesoscopic constitutive response of the two characteristic length scales, i.e., film thickness and grain size. The simulated plastic response qualitatively reproduces the experimentally observed size effects while the main deformation mechanisms appear to be in agreement with TEM analyses of tested samples. A new interpretation of size scale plasticity is here proposed based on the probability of activating grain boundary dislocation sources. Moreover, the key role of a parameter such as the grain aspect ratio is highlighted. Finally, the unloading behavior has been investigated and a strong size dependent Bauschinger effect has been found. An interpretation of these phenomena is proposed based on the analysis of the back stress distribution within the samples.

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1. Introduction

Thin film plasticity is currently an active field of research essentially for two reasons: (i) metallic thin films are widely used in electronic components; (ii) the plastic properties of thin films differ significantly from the properties of the corresponding bulk materials due to the reduced length scales of the microstructure, which become comparable to the geometric structural dimensions. Metallic thin films on substrates usually exhibit a flow stress which is an order of magnitude higher than the same material in bulk form and this flow stress increases with the decrease of the film thickness (Arzt, 1998; Spaepen, 2000; Arzt et al., 2001).

So far, thin film plasticity has mostly dealt with films (essentially Al and Cu) deposited on Si substrates. Following the pioneering work of Nix (1989) for single crystalline films, several authors analyzed the response of polycrystalline thin films; they focused on the effect of film thickness, grain size and orientation, on the stress/strain response and other important features like the level of thermal stresses as a function of the thickness and the effect of passivation layers (Venkatraman and Bravman, 1992; Yu et al., 1997; Keller et al., 1998; Baker et al., 2001; Hommel and Kraft, 2001). Recently, Baker et al. (2003) found an “anomalous” large Bauschinger effect for thin Cu films on substrates when the films were exposed to air.

For polycrystalline free-standing thin films, the experimental data and setups are more limited (Keller et al., 1996; Huang and Spaepen, 2000; Espinosa et al., 2003; Haque and Saif, 2004). Espinosa and co-workers (Espinosa and Prorok, 2001; Espinosa et al., 2003, 2004), using the membrane deflection experiment (MDE), identified major size effects in the mechanical properties of free-standing submicron FCC thin films (electron beam evaporated Au, Cu, and Al). These films were polycrystalline in nature with thicknesses ranging from 200 nm to 1 μm and were tested by applying macroscopic pure homogeneous uniaxial tension, i.e., in the absence of any macroscopic strain gradient. The average grain size (about 200 nm) was independent of the film thickness. This feature is quite important to eliminate the so-called Hall–Petch effect. The reader should note that for the case of electron-beam evaporated Cu, similar results concerning the deviation from linear elasticity and onset of plastic flow were obtained using a micro-tensile device with in situ TEM by Keller et al. (1996). These authors did observe dislocation nucleation and emission from grain boundaries consistent with current understanding of plastic flow in metals.

Haque and Saif (2004) also explored size effects in FCC metal films such as sputtered pure Al and Au, by measuring the tensile stress–strain response of submicronic (down to a thickness of 0.1 μm) free-standing films subjected to loading and unloading. However, in their experiments, the grain size varied with film thickness, which makes the interpretation more complex. The average grain size in the investigation was 100 nm or smaller. Upon performing in situ TEM observations, they did not notice dislocation activity for an average grain size lower than 50 nm, and they concluded that a grain boundary-based mechanism was the dominant contribution to deformation. These observations are in agreement with the results obtained for “bulk” nanocrystalline materials, either through in situ TEM, or by means of large scale molecular dynamics simulations, which are usually performed as a “guide” to experiments (Kumar et al., 2003).
Balk et al. (2001) used in situ TEM to study the thermomechanical behavior of polycrystalline Cu thin films with a grain size on the order of 200 nm. They observed in the cooling phase emission of dislocations from a grain boundary triple junction and subsequent glide on a (111) plane parallel to the film surface.

These experimental results tend to prove the idea that dislocation sources are scarcely available in geometrically confined systems. In this paper, we discuss new results from both the experimental and the modeling point of view. A new interpretation of size scale plasticity is proposed based on the reduced number of available dislocation sources when size is decreased. When the film thickness and particularly the number of grains through the thickness are reduced, the mechanical behavior of thin films becomes more and more affected by the probability of finding dislocation sources.

Efforts to understand and to model plasticity size effects have emerged very rapidly in the last decade. Two different approaches have been chosen to interpret and capture size effects.

The first approach based on continuum mechanics has been initiated by phenomenological strain gradient plasticity (Aifantis, 1992; Fleck and Hutchinson, 1993; Fleck and Hutchinson, 1997) and later on by “mechanism-based” strain gradient plasticity (Nix and Gao, 1998; Gao et al., 1999; Huang et al., 2000). Both theories have been applied successfully to non-uniform macroscopic straining experiments (micro-indentation, torsion or bending) for systems with characteristic dimensions from 0.1 to 100 μm. However, further insight is still needed to use strain gradient plasticity for describing the size dependent mechanical behavior of thin films in the submicronic range in the absence of macroscopic strain gradient (Hutchinson, 2000). Recently, Bažant et al. (2004) have also proposed a continuum-based model which suggests the existence of a boundary layer of constant thickness located at the film surfaces and originated by the deposition process on the substrate. The “boundary layer model” is a phenomenological model based on a non uniform initial yield stress distribution in the film due to a gradient of dislocation density in the boundary layer. It has been shown that this analytical model based on simple assumptions captures the experimental results of Espinosa et al. (2004) in pure tension reasonably well. However, the experimental identification of such boundary layer remains elusive.

The second approach to plasticity size effects is to perform large scale simulations based on molecular dynamics or on dislocation dynamics. This is the method adopted in this paper for identifying the relevant mechanisms through which the microstructure and the film characteristic dimensions lead to the observed size effects in pure tension. Following the conclusions of Espinosa et al. (2004), the misfit dislocation strengthening mechanism (Nix, 1989) is not present in polycrystalline free-standing unpassivated FCC thin films. This proves the existence of another mechanism, as previously observed by Venkatraman and Bravman (1992). Emission and motion of dislocations are key features to explain the observed experimental results as discussed in Espinosa et al. (2005). Thus, from the modeling point of view, there is a need to account for the spatio-temporal occurrence and evolution of discrete events such as the ones given by dislocations in confined samples. The self-organization of dislocations within a small structure is well captured using discrete dislocation dynamics (DDD) (e.g. Liu et al., 2000; Madec et al., 2003). Likewise, the influence of image stresses on this self-organization was studied by Khraishi and Zbib (2002a) and Ohashi (2004). Experimental TEM observations can be conducted to identify the mechanisms and validate the
DDD calculations. The numerical technique is computationally intensive, but we believe it may allow determining the relevant mechanisms responsible for size and grain boundary effects. Among the different codes recently developed (Kubin et al., 1992; Van der Giessen and Needleman, 1995; Verdier et al., 1998; Zbib et al., 1998; Schwarz, 1999; Ghoniem et al., 2000; Weygand et al., 2002; Bulatov et al., 2004; Deshpande et al., 2005; Nicola et al., 2003), the PARANOID code (Schwarz, 1999) is used in this paper. The code has been modified to account for the presence of grain boundaries, to compute the plastic strain, and to define different initial dislocation source configurations in order to interpret grain boundary and size effects. Note that boundary effects in DDD implementations were also explored by Fivel et al. (1996), Fivel and Canova (1998), Ghoniem and Sun (1999), Khraishi et al. (2001), Yasin et al. (2001), Khraishi and Zbib (2002b), Yan et al. (2004), and Nicola et al. (2005).

This paper is based on the grain boundary source model first proposed by Espinosa et al. (2005). Here we provide additional computational details and examine the model in the context of grain size and Bauschinger effects. The paper is organized as follows: Section 2 discusses the new original experimental investigations performed to characterize the mechanical behavior and plasticity size effects observed in free-standing thin films. In Section 3 the DDD method is presented in the framework of the fully parallelized code PARANOID. The numerical procedure, the problem definition, and the assumptions used are presented and discussed. Section 4 concerns numerical results obtained by DDD. In particular, we examine the effect on the material mesoscopic response of the system characteristic dimensions, i.e. film thickness, $h$, and grain size, $d$. This section also illustrates the presence of size scale effects during unloading. Finally, conclusions of the present study are drawn in Section 5.

2. Experimental results and TEM observations

2.1. MDE experiments and TEM/SEM observations

As mentioned before, the MDE experiment is a recently developed technique able to characterize the mechanical behavior of free-standing thin films. Using this experimental procedure Espinosa et al. (2003, 2004) showed that for electron beam evaporated Au the onset of plasticity is a strong function of film thickness, with thinner films exhibiting a much higher yield stress. Furthermore, a major transition in failure mode was observed when the thickness decreased from 1.0 to 0.5 μm, which implies different strengthening mechanisms. For electron beam evaporated Cu and Al membranes, engineering stress/strain curves obtained for specimen thicknesses of 0.2 and 1.0 μm also showed strong thickness dependence, like for Au. In all these experiments, the dimensional constraint appears clearly to be the thickness because the grain size is kept approximately the same for all the investigated film thicknesses. As previously stated, Keller et al. (1996) obtained similar results regarding the flow stress and the rate of hardening for Cu specimens with thicknesses of 0.2 and 1.7 μm.

Postmortem transmission electron microscopy (TEM) and scanning electron microscopy (SEM) observations on MDE tested films were performed to gain insight into the origin of the size scale experimental observations. Samples for plan view TEM observations were prepared by placing of the membranes on TEM grids followed by ion milling of the film from both top and bottom sides. Cross-sectional TEM samples were prepared by
focused ion beam (FIB) processing. For this purpose, the membrane specimen was first glued to an approximately 50-µm-wide Si block to provide a handling base, and then placed into a dual SEM/FIB chamber. Platinum (Pt) was deposited on top of the film surface to protect it from excessive FIB etching. The area to be observed was thinned with FIB on both sides to an electron-transparent thickness following the method of Ishida and Sato (2003). Both top and cross-sectional samples were observed in a JOEL2010 TEM at room temperature.

SEM observation of the 0.3-µm-thick Au films revealed no major plastic activity as evident from the smoothness of the surface of the specimen near the fracture plane (Fig. 1A).

Fig. 1. Study of the plastic activity of 0.3-µm-thick Au films (A) (SEM picture showing the fracture surface) and 1-µm-thick Au films, (B) (SEM picture showing large deformation slip bands close to the fracture edge), (C) Cross-sectional TEM view of fracture region, (D) high aspect ratio grain exhibiting dislocation network.
The TEM cross-sectional view displays some residual dislocation patterns within a few isolated grains, with a decrease in dislocation activity as we depart from the fracture surface. TEM images before testing show the initial grain structure of the film, with grains containing annealing twins and basically free of dislocations. For 1-µm-thick Au films, a significant plastic activity is evident in the SEM images of the specimen surface (Fig. 1B). Several deformation bands are also noted. This is consistent with the observed softening in the stress–strain behavior Espinosa et al. (2004). The TEM cross-sectional views of the same specimen (Figs. 1C and D) show residual dislocation patterns only within grains with high aspect ratio $h/d$. This suggests that dislocations have moved mainly to the free surfaces or grain boundaries and that they have formed networks only when the probability of forming dislocation junctions was increased. We will come back to this feature later in the discussion of the DDD simulation results. Again, our TEM observations are in agreement with the ones reported by Keller et al. (1996). In their in situ TEM studies, the films exhibited limited plastic activity with dislocation nucleation at grain boundaries. Dislocations were found to sweep the grains with very short glide distances, which did not allow significant dislocation interaction.

2.2. Self-similar experiments

To further investigate the plasticity size scale effect, e.g., in Cu films, we tested self-similar specimens until failure in tension occurred. The experiments were conducted using a miniature tensile stage (Fullam, Inc., Latham, NY) equipped with a 2000 lb load cell. The specimens were machined out of a copper foil and processed to have all the characteristic dimensions (thickness, grain size) 1000 times larger than the specimens used in the MDE. As a result, the number of grains through the thickness was about the same as in the thin films but the area of grain boundaries was increased 1 million times! The setup was placed under an optical microscope and pictures were taken throughout the stretching process to keep track of deformations at the grain level. The specimen surface roughness was used to determine local strains by means of digital image correlation. Fig. 2

![Fig. 2. Self-similar experiments in pure tension for Cu films: comparison between the mechanical response of submicron thin films (MDE) and millimeter thick coarse-grained films (Fullam tensile stage).]
compares the stress–strain responses for *coarse-grained films* with thicknesses of 0.3 and 1 mm (using the Fullam tensile stage) with the ones for *thin films* with thicknesses of 0.3 and 1 μm (using the MDE). No size effect is observed in tension for the *coarse-grained films*, with an average grain size of 200 μm, whereas a dramatic size effect is observed in the submicronic *thin films* as previously stated. In Fig. 3 some snapshots of the deformation process for the *coarse-grained films* are displayed. Fig. 3A shows the initial grain structure, while Fig. 3B is taken at a deformation of approximately 10%. The features are similar to plastic deformation within *bulk polycrystalline metals* where dislocations nucleate and propagate at relatively low levels of deviatoric stress, forming multiple slip lines across the grains. Figs. 3C and D show the measured local strain field, by means of digital image correlation, at an applied external strain of 10% and at the end of the test, respectively. One may notice the large values of local strain and some degree of grain rotation, which are typical for bulk polycrystalline plasticity. In the case of millimeter-thick films, due to the increased grain boundary area and the enlarged probability to find intra-granular dislocation sources, defect nucleation does not control the plastic activity any more. Furthermore, dislocation multiplication leading to deformation bands within the grains is clearly observed. For this reason the macroscopic bulk response is achieved and no significant statistical variations in the sample behavior occur.

Fig. 3. Deformation of coarse-grained films: (A) initial grain structure, (B) grain configuration for an external applied strain of approximately 10%, (C) local strain field, \( e_{xx} \), corresponding to an external applied strain of approximately 10%, (D) local strain field, \( e_{xx} \), at failure.
3. Discrete dislocation dynamics

3.1. General description of the code

In this work we have employed the PARANOID code to perform discrete dislocation dynamics (DDD) simulations. PARANOID is a 3D code that follows the motion of dislocation lines modeled as smooth curves passing through a sequence of tracking points (Schwarz, 1999). There are two main limitations in our calculation of dislocation motion: (i) we consider only the glide of perfect dislocations in a linear, isotropic medium; (ii) the problems of dislocation self-interaction and interaction with free surfaces are treated in an approximate way which introduces errors on the order of 10–20% (Schwarz, 1999). To this limit of accuracy, the code has previously provided quantitatively accurate predictions of dislocation configurations in small epitaxial islands (Liu et al., 2000), and of the residual misfit dislocation structure in plastically relaxed single-crystal layers (Schwarz et al., 2004). The polycrystal line films considered have not yet been as cleanly characterized in terms of a well-defined computational model, and our present aim is the more qualitative one of understanding the main deformation mechanisms rather than achieving an accurate comparison with experimental results.

Like the other mainstream DDD codes mentioned earlier, PARANOID is able to capture 3D effects like line tension effects, and to include the full 3D interactions between dislocation lines. Moreover, the code is able to run in a highly resolved manner, in which the point spacing can be reduced down to atomic dimensions. Recently, Schwarz (2003) introduced approximation rules for strong interactions such as at junctions, line–line crossing regions, and dipoles. The results obtained through these approximations have been found in good agreement with fully resolved calculations.

PARANOID is a parallel, load-balanced program that runs on SP-type or Linux-cluster-type distributed memory machines. It is written in Fortran-77 and uses MPI calls for communication between processors.

3.2. Dislocation dynamics

The evolution of the dislocation configuration is obtained by tracking the motion of the discrete meshing nodes. An explicit numerical scheme is used. The local stress tensor is given by the stress contribution due to the applied strain, stresses arising from the dislocation themselves, and corrections due to boundary effects. Once the stress tensor has been computed at each node, the Peach–Koehler equation is used to determine the force per unit length acting locally on the dislocation at that position. The Peach–Koehler force, $f_g$, to move a dislocation element $dl$ in the glide plane is given by:

$$f_g = (b \sigma \cdot n) n \times dl,$$

where $b$ is the Burgers vector of the dislocation, $n$ is the unit normal to the glide plane and $\sigma$ is the local stress tensor. The singularity in the self-interaction stress is regularized by means of the Brown splitting procedure (Brown, 1964), which has been modified in order to avoid the unstable behavior pointed out by Schwarz (1999). Each node is then moved in the direction perpendicular to the local line tangent following the simplest response model, i.e., with a velocity proportional to the local force component given by (1):
\[ v = \frac{f_g}{d/B}. \]  

(2)

\( B \) is a phonon viscous drag coefficient and a commonly used value for copper at room temperature \( B = 1.5 \times 10^{-5} \) Pa s is here employed following Verdier et al. (1998). Only a viscous drag type equation of motion is considered through the constant \( B \). The inertial forces, sound velocities and internal forces arising from interactions of dislocations with obstacles like the Peierls barriers are neglected. A complete thermodynamic description can be found in Hirth et al. (1998) and Zbib et al. (2002).

3.3. Problem definition and boundary conditions

In contrast with plastic deformation in bulk materials, where interactions between many (forest) dislocations may lead to self-organization such as cell structures, the present simulations involve a much smaller number of dislocations.

For each simulation, a single FCC crystal representing a grain within a polycrystalline film of copper is considered. In the simulations the following material parameters for copper have been employed: \( \mu = 42.3 \) GPa (shear modulus), \( \nu = 0.3 \) (Poisson ratio), \( a = 0.36148 \) nm (lattice parameter), \( b = 0.2556 \) nm (magnitude of Burgers vector). Since the films tested by MDE have a pronounced \( \{111\} \) fiber texture, the orientation of the single crystal is chosen so that the crystallographic \( \langle 111 \rangle \)-direction is parallel to the global Z-axis (axis normal to the film plane) as described in Fig. 4B. The two other crystallographic axes respectively parallel to the global X- and Y-axes are chosen arbitrarily. The grain is either represented by a cubic box with edges equal to the average grain size \( d = 0.2 \) \( \mu \)m or by a rectangular box (corresponding to a columnar grain, i.e., only one grain through the thickness) with a square base (0.2 \( \mu \)m \( \times \) 0.2 \( \mu \)m) and a varying height (i.e., the thickness of the film \( h \)) in the \( \langle 111 \rangle \)-direction (Fig. 4A).

In our simulations grain boundaries are modeled as impenetrable surfaces, which is a first order approximation since dislocation transmission from one grain to another may

Fig. 4. Problem definition for DDD calculations. (A) Columnar grains simulated by rectangular boxes with various heights. (B) Present study: single crystal (grain) considered as a rectangular box (thickness: \( h \), grain size: \( d \)) with a crystallographic direction \( \langle 111 \rangle \) perpendicular to the film plane (consistent with the strong \( \{111\} \) texture of the film) and with an applied tensile loading parallel to the \( \langle 112 \rangle \) direction (arbitrary).
occur (De Koning et al., 2002). Top and bottom faces of the box are attractive in order to represent the effect of free surfaces and image stresses on dislocations.

The box is subjected to a pure tensile loading in the $Y$-direction at a prescribed computational strain rate $\dot{\varepsilon}$. In order to limit the strain rate effects inherent to the dislocation dynamics scheme and to run calculations with a reasonable CPU time, we chose a strain rate $\dot{\varepsilon} = 6.66 \times 10^5$ s$^{-1}$. The effect of the strain rate on the material response has been parametrically investigated as we will report later.

The computation of the local stress tensor is subjected to corrections when free surfaces and other interfaces are present. For sufficiently thin free standing films, two kind of corrections must normally be accounted for. The first one concerns the interactions of dislocations with the free surfaces and the second takes into account the interactions between the two free surfaces (top and bottom). This problem has been presented and solved by Hartmaier et al. (1999) using the so-called Boussinesq operator and the principle of superposition for boundary conditions (Van der Giessen and Needleman, 1995). Different approaches to the more general problem of finding image corrections have been discussed by several authors (Fivel et al., 1996; Fivel and Canova, 1998; Ghoniem and Sun, 1999; Liu et al., 2000; Khrashi et al., 2001; Yasin et al., 2001; Khrashi and Zbib, 2002a,b; Yan et al., 2004; Liu and Schwarz, 2005). In all cases, an accurate accounting for image effects, in particular when dislocations can terminate on some of the surfaces, requires a massive increase in the computational load. However, a recent detailed study by Liu and Schwarz (2005) has demonstrated that for systems like that considered here, in which the dislocations are curved on the scale of the geometry, it is possible to obtain results accurate to the 10–20% level using the following simple approximations: (a) surfaces that repel dislocations are treated as impenetrable, while dislocations are allowed to reconnect to surfaces that attract dislocations, such as a free surface; (b) wherever a dislocation intersects the free surface, it locally feels the force induced by a free surface on a straight dislocation line (Lothe et al., 1982). The latter assures that the dislocation enters the free surface at the correct angle. DDD simulations in general approximate the detailed structure of the dislocation core in terms of some simplified continuum model, and are thus themselves accurate only to the 10–20% level. There is little point, therefore, in going beyond these simple approximations, unless one is treating nearly 2D systems where the dislocations have significant curvatures, systems which develop buildups of large-scale correlated dislocation arrays leading to large mean fields, like the misfit array in a single-crystal strained layer. A good illustration of how well this seemingly oversimplified approach can reproduce observed dislocation behavior in a small structure similar to a grain is given in Liu et al. (2000).

3.4. Plastic strain tracking and dislocation densities

The formula to compute the plastic strain tensor due to a dislocation at any point $\mathbf{x}$ is given by Mura (1987, p. 46):

$$
\varepsilon^p_{ij}(\mathbf{x}) = -\frac{1}{2} (b_i n_j + b_j n_i) \delta(\mathbf{S} - \mathbf{x}),
$$

where $\mathbf{S}$ is the dislocation slip plane and $\delta(\mathbf{S} - \mathbf{x})$ is the one-dimensional Dirac delta function in the normal direction of $\mathbf{S}$, which is unbounded when $\mathbf{x}$ is on $\mathbf{S}$ and zero elsewhere. The sign “$-$” is due to the use of the right-handed screw convention for the normal direction.
of S. Using (3) and the same sign convention, the total macroscopic plastic strain increment denoted by $d\varepsilon_{ij}^p$ yields:

$$d\varepsilon_{ij}^p = -\frac{1}{2V} \sum_{k=1}^{N} (b_i^{(k)} dS_{j}^{(k)} + b_j^{(k)} dS_{i}^{(k)}),$$

(4)

where $V$ is the volume of the crystal, and $dS^{(k)}$ is the algebraic increment of area swept out by the segment of dislocation between mesh points $k$ and $k + 1$. Initially and at each step, the total dislocation density is also computed. Its expression is:

$$\bar{\rho}_{\text{tot}} = \frac{\sum_{I} l^{(I)}}{V},$$

(5)

where $l^{(I)}$ is the length of the dislocation segment ($I$). To identify which are the most active slip systems for the different initial configurations, the dislocation density per slip system is also computed for the 12 primary slip systems of the FCC structure for which the Schmid and Boas notation is used in the following.

4. Numerical results and discussion

4.1. Activation stress of a Frank–Read source

As a preliminary analysis we examine the stress needed to activate a Frank–Read source within a grain (volume source). Such activation stress is a function of two internal length scale parameters, defined in Fig. 5A, i.e., $s$ and $l$, the Frank–Read source size and its distance from the grain boundary towards which it is propagating, respectively. For this purpose a cubic grain with $d = h = 0.2 \mu m$ is considered and both parameters, $l$ and $s$, are normalized to the film thickness. We define the activation stress as the stress needed...
by the source to overcome a critical configuration from which dislocation multiplication becomes possible. In order to compute the activation stresses, we have applied a strain rate to the sample. The activation stress corresponds to a plateau in the stress vs. plastic strain response of the material. Since this stress value may be affected by the applied strain rate, we investigated the effect of different strain rates on the computed activation stress. In this study a Frank–Read source of size $0.16 \mu m$ has been positioned in the center of the grain with all impenetrable surfaces. The source is located on the $(111)$ plane with a Burgers vector in the $(011)$ direction. For this initial configuration the stress vs. plastic strain is simulated for several values of the applied strain rate (Fig. 5B). This curve is characterized by three different parts which correspond to different phases in the evolution of the source. In the first stage of the simulation, the source is increasing the total swept area with increasing applied stress; this corresponds to the first part of the curve in which significant plastic strain is produced. Then, the dislocation encounters the impenetrable surfaces and the stress is rising without any significant increase in the plastic strain (second part of the stress–plastic strain curve). This status is preserved until the so called activation stress is reached and allows the dislocation to overcome a critical configuration beyond which dislocation multiplication becomes possible. This discrete event corresponds to a plateau in the stress–plastic strain curve, since a sudden large increase in the plastic strain occurs. Fig. 5B shows that the plastic behavior of the crystal is converging for decreasing strain rates. This feature was also identified by Rhee et al. (1998). Therefore, in the calculations reported in the following, a strain rate of $6.66 \times 10^5 \text{s}^{-1}$ has been applied in order to reduce the CPU time and at the same time minimize the effect arising from viscosity.

The activation stress has been parametrically investigated by considering three values of the ratio $s/h$ (0.1, 0.3 and 0.8) and several values of the distance $l$. In Fig. 6A, we plot a cross-section of the grain in which the left and right surfaces are modeled as impenetrable (grain boundaries) and the top and bottom surfaces are modeled as attractive (free surfaces). In Fig. 6B the activation stress to reach the final configuration displayed in Fig. 6A (where only half loop is generated) is plotted. Fig. 6B indicates that for large $l/h$, the stress needed to activate the dislocation is source-size dependent and, as in Von Blanckenhagen et al. (2001a,b), the smaller the source, the higher the activation stress.

![Fig. 6](image.png)
Fig. 6A shows that forming dislocation arms, close to free surfaces, are attracted by the surfaces further assisting their glide; this was also shown by Khraishi et al. (2001). On the contrary, when the source is very close to an impenetrable grain boundary, the latter acts as a strong obstacle and a high stress, independent of $s/h$, is needed to activate the source. This result shows that in geometrically confined systems, the yield stress and the hardening rate will be affected by the statistical distribution of sources in terms of both length scale parameters $l/h$ and $s/h$. When the microstructural characteristic dimensions, e.g. source size, are comparable to the geometrical features, such as film thickness and grain size, confinement exerts a strong constraint and only favorably located sources may be activated. Therefore, the scarce availability of dislocation sources and the reduced probability of nucleating defects are likely to dominate the plastic process. This may also explain the limited ductility exhibited by these systems in the aforementioned experiments.

While the picture emerging from the analysis of volume sources is somewhat consistent with the experimentally observed plasticity size effects, there is no TEM evidence of volume sources being active in ultrafine-grained FCC materials, Cheng et al. (2003). It is worth mentioning that in the context of 2D discrete dislocation calculations, Biner and Morris (2003) considered grain boundary sources as the only sources for nucleation of the dislocations. In the following we use a statistical model based solely on grain boundary sources as proposed by Espinosa et al. (2005). The reader should not confuse grain boundary sources with Frank–Read sources in the sense that the latter are typically volume sources capable of developing a whole glide loop around them and consequently multiply at relative constant stress. This feature is not present in grain boundary sources as highlighted by molecular dynamic simulations.

4.2. Effect of film thickness

In this section the focus is to examine the mesoscale constitutive response of a single crystal with multiple interacting grain boundary sources. Specifically, we analyze the effect of a change in the film thickness at constant grain size. As previously mentioned, the films tested experimentally exhibited a few grains through the thickness, as illustrated in Fig. 4A. However, in order to overcome the current limitations of DDD, only columnar grains are here considered. Therefore, the computational domain is made of a rectangular box with a square base of area $d^2$ (with $d$ being the grain size: 0.2 μm in the simulations of this section) and a varying height $h$, representing the thickness of the polycrystalline film (Fig. 4B). This columnar geometry is actually exhibited by some grains of the tested films (Fig. 1C), which will be referred to as “high aspect ratio grains”. Samples with four thicknesses have been considered: 0.2, 0.4, 0.6 and 1 μm, while the grain size was constant and equal to 0.2 μm. Thus, the four samples had aspect ratios, respectively, equal to 1, 2, 3 and 5.

In the present DDD simulations of size scale plasticity in thin films, dislocation sources have been randomly distributed only at grain boundaries. Therefore, the sources are not of the Frank–Read type in the classical sense, because they are not able to generate circular or elliptical pile ups. They are rather characterized by a one-time activation process leading to the formation of one dislocation line at the opposite grain boundary. The simulations clearly show that when grain boundary dislocation sources are activated, they shear the slip plane and are either absorbed by free surfaces or stored at grain boundaries. This appears to be consistent with a number of TEM observations and atomistic simulations.
reported in the literature. For a grain size of 200 nm, like the one here considered, grain boundary dislocation sources are expected to control the plastic process. As a matter of fact, Cheng et al. (2003) proposed the existence of four ranges of grain size in FCC metals for which distinct plastic deformation mechanisms exist. In the so-called “ultrafine” regime, which is the case of the tested freestanding thin films, grain boundary dislocation sources appear to be predominant. They also suggested that the upper limit of this material behavior may be up to 1 \( \mu \)m. Only for larger average grain sizes are intragranular dislocation sources (volume sources) expected to contribute significantly to the plastic activity.

A constant initial dislocation density \( 2.4 \times 10^4 \text{ cm}^{-2} \) has been used to characterize the grain boundary, the concept of a constant density of boundary sources being consistent with atomistic simulations of nanostructured metals (Farkas and Curtin, 2004). We want to highlight that for a change in the film thickness, a constant area density on the grain boundary also implies a constant volume density, which allows us to compare the average plastic strain over the volume. For each sample three different runs with different initial configurations of defects have been performed. The stress–strain curves for the three calculations have been averaged in order to reduce the scatter arising from the statistical nature of the sample. The physical meaning of this averaging procedure consist of taking into account three different grains within the polycrystalline film. The source lengths have been generated by means of a normal distribution with a mean value of 0.08 \( \mu \)m and a standard deviation of 0.02 \( \mu \)m. The parameters of this distribution have been kept constant for all the thicknesses.

4.2.1. Strain rate analysis

As already mentioned, the computational samples are loaded in pure tensile mode in the \( Y \)-direction by prescribing a strain rate \( \dot{\varepsilon} \). Therefore, before analyzing and comparing the results for the samples with different thicknesses, the effect of the adopted computational strain rate is investigated. In this analysis the 0.2-\( \mu \)m-thick sample has been considered and tested for several values of the applied strain rate. Fig. 7A shows the stress–strain curves for these computational tests; a high strain rate produces an overestimation of the activation stresses at which boundary sources are activated (discrete events which correspond to the sudden drops in the stress–strain behavior). It may be noticed that the two results at \( \dot{\varepsilon} = 6.66 \times 10^5 \text{ s}^{-1} \) and \( 5 \times 10^4 \text{ s}^{-1} \) are not very different from both a qualitative and quantitative point of view. This suggests that the material response is converging for diminishing values of the strain rate as discussed by Rhee et al. (1998). In order to validate this hypothesis, another simulation was performed in a “quasi-static” fashion, which was obtained by applying a stepped strain to the sample (step strain of amplitude 0.1) and relaxing it at constant applied strain for sufficiently long time intervals. In this fashion, the quasi-static stress corresponding to the value of the applied strain was determined. This result is also reported in Fig. 7A in the form of dots. These dots are in full agreement with the continuous stress–strain curve obtained for \( \dot{\varepsilon} = 5 \times 10^4 \text{ s}^{-1} \). However, simulations performed at this strain rate require very large computational time while the objective of the present study is to identify the main deformation mechanisms underlying size scale plasticity in small volumes. Therefore, in the following simulations a strain rate of \( 6.66 \times 10^5 \text{ s}^{-1} \) has been adopted in order to compromise between reduction of strain rate artifacts and reasonable computational time.
4.2.2. Stress–strain response

Fig. 7B displays the averaged stress–strain curves for the samples with varying thickness and aspect ratio. The error bars represent the statistical scatter of the three runs at the end of the simulation for each sample. The stress drops in the stress–strain curves correspond to dislocation emissions from boundary sources. The results in Fig. 7B, which were obtained for an initial source density of $2.4 \times 10^4$ per cm, reproduce the experimentally-observed size effects for the thicknesses $h = 0.2 \mu m$, $h = 0.4 \mu m$ and $h = 0.6 \mu m$. For these thicknesses “thinner is stronger” and the simulations clearly predict differences in the yield
stress and hardening rate. This feature is not manifested for the 1-μm-thick sample, which, in the early stages of the simulation, presents a response very similar to the 0.6-μm-thick sample and exhibits a larger hardening rate after a deformation of approximately 0.6%.

The following interpretation of the aforementioned results is proposed. Thinner films present less grain boundary area and, consequently, a lower probability to activate boundary sources at a certain stress level. This is consistent with our self-similar experiments, which do not exhibit any size effect as a consequence of the large grain boundary area available for defect nucleation. Moreover, for smaller thicknesses, there is a higher probability for dislocations to encounter free surfaces and to be partially adsorbed; thus, the available free path for dislocations is reduced by the presence of free surfaces and only a limited amount of plastic strain may be produced. However, the effect of these two features is diminished when the film thickness and the grain aspect ratio are increasing. In fact, for larger aspect ratios \((h = 1 \, \mu m, \, h/d = 5)\), the probability for dislocations to pile up at grain boundaries and intersect each other by forming networks and junctions is higher. We refer to this characteristic behavior as the “aspect ratio effect”, and it reflects an increase of the internal stress (back stress) during the deformation process, which causes an obstacle to further dislocation nucleation and leads to an increase in hardening rate. Fig. 8 shows snapshots of the dislocation structure for an applied strain of 0.8% in three different samples: 0.2, 0.6, and 1 μm. Only active dislocation sources are plotted. These snapshots show the formation of junctions in the 0.6-μm-thick and the 1-μm-thick samples. This finding is consistent with our TEM observations (Fig. 1D), which present evidence of networks formation only in the grains with highest aspect ratio.

In order to confirm our interpretation, similar simulations were performed for a smaller initial dislocation density of about \(8 \times 10^6 \, \text{cm}^{-1}\). The results are illustrated in Fig. 9. As expected, a smaller initial density decreases the possibility of junctions and network formation; therefore, in this case, the saturation of the size effect does not occur. It is evident from this analysis that the initial source density is the main parameter governing the critical aspect ratio for which a reversal in the hardening rate is manifested.

In the following two sections a more detailed analysis of the thickness effect for the case with density \(2.4 \times 10^4 \, \text{cm}^{-1}\) will be performed. To achieve this we will consider the evolution of the dislocation density on the 12 slip systems of the FCC structure and the history of the internal stresses during the straining process. In this regard we have selected for each sample the most representative run, i.e., the simulation that was closest to the average response of the sample.

4.2.3. Analysis of dislocation density evolution

The dislocation density may be computed at any time during the simulation by the expression in (5). The same definition may be applied to the 12 slip systems by defining local densities which evolve independently in the simulation. In order to identify the 12 slip systems we have adopted the Schmid and Boas notation which is recalled in Table 1. In the same table the Schmid factors have also been computed with regard to the loading direction prescribed in our calculations. In Fig. 10 the dislocation densities on each slip system are plotted as functions of the applied total strain, for varying thickness. In all four cases, the most active slip systems are the A3 and D1, as expected, having the highest Schmid factor. Two main features may be noticed: (i) thicker samples exhibit a higher degree of activation of the primary slip systems (A3,D1); (ii) in thicker samples, secondary slip systems (A6,B4,D6), with Schmid factor 2/3, participate earlier in the plastic process. This is
Fig. 8. Final dislocation configurations for an applied strain of 0.8% (left image: top view, right image: 3D view). (A) 0.2 \( \mu \)m sample; (B) 0.6 \( \mu \)m sample; (C) 1 \( \mu \)m sample.
caused by the higher probability of nucleating defects in larger samples. However, as already noticed, this effect is saturated for the 1-μm-thick sample. This sample exhibits less plastic activity than the 0.6-μm-thick one, due to the significant raise in internal stresses manifested in grains with high aspect ratios. This feature is analyzed quantitatively in the next section.

4.2.4. Back stress analysis

The objective in this section is to analyze the stress field within our samples and especially to differentiate the contribution coming from the applied external strain and the dislocation themselves (stress that we refer to as “back stress”). For this purpose, we compute the local stress field at any fixed point inside the computational box. In particular, we analyze the contribution of the back stress to the Peach–Kohler force acting per unit length on a dislocation on the A3 slip system (Schmid factor 1) in proximity of one of the grain

![Figure 9](image)

Fig. 9. Stress–strain curves for samples with different thicknesses and aspect ratios. Initial grain boundary dislocation density: 0.8 × 10⁴ per cm. The error bars represent the statistical scatter of different runs at the end of the simulation for each sample.

Table 1

<table>
<thead>
<tr>
<th>Schmid and Boas notation</th>
<th>Slip plane: (hkl)</th>
<th>Slip direction: ⟨uvw⟩</th>
<th>Schmid factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>A2</td>
<td>(111)</td>
<td>(011)</td>
<td>1/3</td>
</tr>
<tr>
<td>A3</td>
<td>(111)</td>
<td>(101)</td>
<td>1</td>
</tr>
<tr>
<td>A6</td>
<td>(111)</td>
<td>(110)</td>
<td>2/3</td>
</tr>
<tr>
<td>B2</td>
<td>(111)</td>
<td>(011)</td>
<td>2/3</td>
</tr>
<tr>
<td>B4</td>
<td>(111)</td>
<td>(101)</td>
<td>2/3</td>
</tr>
<tr>
<td>B5</td>
<td>(111)</td>
<td>(110)</td>
<td>0</td>
</tr>
<tr>
<td>C1</td>
<td>(111)</td>
<td>(011)</td>
<td>0</td>
</tr>
<tr>
<td>C3</td>
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</tr>
<tr>
<td>D4</td>
<td>(111)</td>
<td>(101)</td>
<td>1/3</td>
</tr>
<tr>
<td>D6</td>
<td>(111)</td>
<td>(110)</td>
<td>2/3</td>
</tr>
</tbody>
</table>
boundaries. The evolution during straining of the average profile of this force along the thickness of the film for the four samples is reported in Fig. 11. As expected, the force arising from dislocations is negative, in the sense that it opposes the external stress field. Moreover, the average absolute value of this back stress force is growing with the applied strain as a consequence of the increased dislocation density. This result is also summarized in Fig. 12 where the average absolute value of the force profile is plotted as a function of the applied total strain for the four samples. This plot represents the average effect of the back stress on the grain boundary sources for different thicknesses at different applied strains.

The average magnitude of the back force per unit length is very similar for all the thicknesses in the early stages of the simulation. However, increasing the deformation produces an obvious rise in back stress which is more marked in the case of thick samples. As mentioned before, this is a consequence of the high aspect ratio of thicker samples, which results in a higher probability for dislocations to encounter grain boundaries rather than free surfaces, and also to intersect each other and form junctions. These events produce the observed significant increase of the back stress, which is an obstacle to subsequent dislocation nucleation. This explains the fact that the 1-μm-thick sample...
possesses a larger hardening rate than the 0.6-μm-thick one at the assumed dislocation density of $2.4 \times 10^4$ cm$^{-1}$ (Fig. 7B).

4.3. Effect of grain size

In this section, we investigate the influence of the grain size $d$ on the mesoscale constitutive response of the columnar grain. For this purpose, a grain with $h = 0.2$ μm and $d = 0.4$ μm is simulated and the computed stress–strain response is compared with the one of the cubic sample, i.e., $h = d = 0.2$ μm. In these calculations, it is not possible to keep constant both the initial density of sources per unit of grain boundary area, and the initial volume density of sources. Since plastic strain is an average volume quantity, we have chosen to preserve the initial volume source density of the samples ($4 \times 10^{14}$ per m$^2$) which is the only way to compare the plastic activity in the two tests. Again, three simulations with different initial configurations have been performed. The average stress–strain results are compared in Fig. 13, which also shows the scatter of the results for an applied strain of 0.8%. The response of the crystal with larger grain size is significantly softer, and, particularly, the yield stress and hardening rate are much smaller.

Fig. 11. Evolution of the back stress force profile along the thickness during straining: (A) 0.2 μm sample; (B) 0.4 μm sample; (C) 0.6 μm sample; (D) 1 μm sample.
than in the sample with $d = 0.2 \mu m$. The significant increase in plastic activity may have two explanations: (i) as in the case of a change in the thickness a larger grain boundary area produces a higher probability of dislocation nucleation; (ii) the aforementioned effect is more marked than in the case of an equivalent change in the thickness, because when the grain size is increased, opposite grain boundaries are moved farther apart. This implies a less significant internal stress generated during straining, because the dislocation walls are more distant from each other. Moreover, for larger grain sizes there is a bigger surface area available for dislocation adsorption; therefore, dislocations are

![Figure 12](image1.png)  
Fig. 12. Average magnitude of the back force per unit length as a function of the applied strain for the four samples.

![Figure 13](image2.png)  
Fig. 13. Stress–strain curves for samples with different grain sizes. Initial volume dislocation density: $4 \times 10^{14}$ per m$^3$. The error bars represent the statistical scatter of different runs at the end of the simulation for each sample.
more likely to encounter free surfaces. This causes a release of the internal stresses within the grain.

4.4. Bauschinger effect

As we mentioned before, there is experimental evidence in the literature of a strong “anomalous” Bauschinger effect for thin Cu films on substrates (Baker et al., 2003; Xiang and Vlassak, 2005). Recently, Xiang and Vlassak (2005) proposed an interpretation for this material behavior. The authors suggested that in the loading phase of the tensile test, a large number of dislocation pile-ups are formed at the interface between the film and the substrate. This produces a strong back stress in the film, which, upon unloading, leads dislocations to glide in the opposite direction. The early sweeping of dislocations in the opposite direction may cause an unloading nonlinear behavior.

In order to investigate this phenomenon in free-standing thin films, we have tested the unloading behavior of the samples with $h = 0.2 \mu m$ and $h = 0.4 \mu m$. We have chosen to perform the unloading tests quasi-statically, i.e., by applying a stepped strain and relaxing the sample to reach a stable configuration. This allows computing the equilibrium relaxation stress corresponding to the applied strain. The results are illustrated in Fig. 14 in the form of points connected by straight segments. Two main features may be noticed. First, a strong Bauschinger effect is indeed manifested in the simulations. Reverse plastic strain begins very early, when the overall stress in the sample is still in tension. Second, this effect appears to be size-dependent, since, for the sample with $h = 0.4 \mu m$, the deviation from linearity starts much earlier in the unloading phase. Again, an explanation may be advanced based on back stress arguments as shown by Leger et al. (2004). As we underlined in previous sections, even in free-standing films, a strong back stress at grain boundaries arises from piling up of dislocations and formation of junctions within the sample. This is more marked in the case of high aspect ratio grains, see Section 4.2.4, which causes

Fig. 14. Unloading behavior of the samples with different thicknesses. Significant size dependent Bauschinger effect is manifested.
a significant nonlinearity during unloading (Fig. 14). Simulations clearly show that reverse gliding of dislocations starts earlier in the case of the grain with \( h = 0.4 \mu m \), i.e., with higher aspect ratio. In order to illustrate this process, Fig. 15 compares the two dislocation configurations for the thicker sample at an applied strain of 0.7% and 0.6% upon unloading. The arrows show the reverse motion of some of the dislocations segments. It is also observed the motion of junctions along sessile segments. All these features provide an indication of the mechanism for reversed plasticity during unloading.

5. Concluding remarks and future challenges

In this work experimental investigations and DDD simulations have been performed in order to study thin film plasticity and the relevant deformation mechanisms. An interpretation of size scale plasticity has been proposed based on the idea of a limited initial number of defects with most probable sites for dislocation nucleation at grain boundaries. This assumption is well supported by a large number of in situ TEM observations as well as our postmortem TEM observations, which show that the grains are basically defect-free in the initial state and with few residual dislocation networks in isolated grains after straining. Especially, residual networks were observed in the coarsest grains and in the ones with highest aspect ratio.

In order to validate the idea of a dislocation-source-dependent plasticity, self-similar experiments were performed by comparing the results for submicron Cu thin films and coarse-grained millimeter-thick films. As expected, coarse-grained films did not exhibit any size effect and their deformation behavior appeared to be very similar to the one of bulk materials.

This statistical concept was used to implement a computational model in DDD in order to simulate the activation and the evolution of grain boundary dislocation sources in a columnar grain of a polycrystalline thin film. By changing the thickness of the columnar...
grain and, hence, the available surface for dislocation nucleation, we were able to capture qualitatively the same trend than the experimentally observed size effects. However, these features gradually disappeared for a critical value of the aspect ratio of the columnar grain, due to the increase in internal stresses generated by junction formation and pile up at the impenetrable boundaries. This critical value of the aspect ratio has been found to be strongly dependent on the initial density of sources.

An even stronger size effect has been obtained through DDD simulations when the grain size of the crystal was varied. This has been attributed to the larger available area of free surfaces, which attract dislocations partially adsorbing them and releasing the internal stresses. In this situation, a strong increase in the number of activated grain boundary sources was observed.

Finally, the statistical grain boundary source model captured a strong size-dependent Bauschinger effect, which can also be interpreted based on the significant back stress generated during the loading stage.

We have shown that the approach adopted here is able to capture the essential features of the discrete mechanisms involved. However, strong assumptions were required in order to simplify the computational problem. For instance, we have assumed a density of sources distributed at grain boundaries with sizes randomly generated by means of a normal distribution. This starting configuration can only qualitatively represent the real nature and complexity of a grain boundary. This structure, at least for low angle grain boundaries, can be seen as arrays of dislocations, which under an appropriate stress condition may be pulled out and propagate into the interior of the grain. On the other hand, grain boundaries have also manifested, to a certain extent, the ability to adsorb dislocations approaching the boundary by modifying its structure. This emission-adsorption process requires a deeper modeling effort where atomistic simulations are likely to improve our knowledge of modifications in the grain boundary structure and to formulate an energetic activation criterion to be incorporated into our DDD model.

In this contribution, local discrete phenomena have been described efficiently in a qualitative way by an adequate numerical approach, underscoring the important roles of (i) the characteristic internal length parameters which are grain size and film thickness and (ii) the boundary conditions prescribed (here free surfaces, impenetrable boundaries representing grain boundaries). However, the description does not allow us to directly simulate the tensile response of the polycrystalline thin films, since the sample used for DDD simulations is not representative enough of the peculiarities of the microstructure. These are: a given grain size distribution (here a bimodal distribution) with a given crystallographic orientation distribution function of the grains which can significantly modify internal stresses (i.e. the back stress profile) within the grains. Moreover, grain shape and the presence of triple junctions may also modify the spatial dislocation source distribution so that the probability density function of source size is different from a grain to another when considering a polycrystalline sample. We have already found through this study that the aspect ratio of the grain is important. Lastly, mechanisms like intergranular dislocation slip transmission (see e.g. De Koning et al., 2002) instead of pile-up may also affect the profile of back stresses and the propagation of mobile dislocations at the local scale. While this complexity can be incorporated in future DDD models, we expect the statistical grain boundary source and other features of the model advanced in this work will be the dominant reasons for the experimentally observed size scale effects.
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