Chapter 2

Size effects in single crystal thin films

An important source of stress in thin films arises from the thermal mismatch between the film and the substrate. Experiments that reveal this, typically involve cooling, heating or an alternating sequence of cooling and heating, with the average stress in the film recorded by wafer curvature measurements [1, 2] or by X-ray diffraction [1, 3, 4]. On cooling a film from an almost stress-free state at a relatively high temperature, the deformation is initially elastic, but as cooling proceeds plastic deformation eventually occurs. When the film is re-heated, the stress level in the film at first reduces (in absolute value) elastically, with reverse plastic deformation subsequently occurring for a sufficiently large temperature change. Reverse plastic deformation occurs earlier and stress levels increase more rapidly for thinner films. These effects have been observed for fine-grained as well as coarse-grained films [2] and also in passivated films [1, 3]. Unfortunately, experimental results on single crystalline films are not yet available, due to the difficulty in the sample preparation.

This size effect is not captured by conventional continuum plasticity theories because they lack an internal length scale. Nonlocal phenomenological continuum plasticity theories have been proposed, e.g. [5]–[9], that can capture size effects within a phenomenological theory of plasticity. There have also been studies aimed at explaining the thin film size effect based on considerations of dislocation nucleation and motion. In addition to arguments that make reference to well-known concepts such as the Hall-Petch relation [2], single dislocation models have

been proposed for thin films. As already mentioned in the preface, Freund [10] and Nix [11] have proposed a model based on the confined motion of a threading dislocation in a single crystal film, which suggests that the yield strength scales with the film thickness $h$ as $h^{-1}$. Similar single dislocation arguments have been used for polycrystalline films in [13, 14]. Hartmaier et al. [15] have discussed the role of thickness on the possibility of generation of new dislocations.

In this chapter we carry out a dislocation dynamics simulation of the evolution of plastic deformation in single crystal thin films subject to thermal loading.
2.1 Problem formulation

We consider a single-crystalline film, of thickness $h$, perfectly bonded to an elastic half-plane, as illustrated in Fig. 2.1. A two-dimensional, plane strain study ($\varepsilon_{33} = 0$) is carried out. The two-dimensional nature of the model is motivated by the consideration that it is the long straight edge part of threading dislocations that provides most of the plastic relaxation. Three dimensional effects such as line tension and the interaction between dislocation lines not parallel to existing misfit dislocation lines along the interface, as discussed in [11], are not accounted for. The substrate remains elastic, while the film can relax by dislocation activity on a set of discrete slip systems defined by the angle $\phi$ relative to the interface, see Fig. 2.1a. The dislocations are all of edge character with a Burgers vector in the $x_1$-$x_2$-plane of length $b$. Individual dislocations are modeled as singularities in an isotropic thermo-elastic continuum.

The boundary value problem is governed by the equilibrium and compatibility equations,

$$\sigma_{ij,j} = 0, \quad \varepsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i}) \quad (2.1)$$

where $\sigma_{ij}$ denotes the stresses, $\varepsilon_{ij}$ the strains and $u_i$ the displacements; $(,)_i$ denotes partial differentiation with respect to $x_i$.

The constitutive relation is specified by

$$\varepsilon_{ij} = \frac{1 + \nu}{E} \left( \sigma_{ij} - \frac{\nu}{1 + \nu} \delta_{ij} \sigma_{kk} \right) + \alpha \Delta T \delta_{ij}. \quad (2.2)$$

Here, $\Delta T$ is the temperature change from the undeformed state. The linear thermal expansion coefficient $\alpha$ for the film is denoted by $\alpha_f$ and that for the substrate by $\alpha_s$; Young’s modulus $E$ and Poisson’s ratio $\nu$ are taken to be identical for the film and the substrate. In [16] it was found that elastic property differences did not qualitatively affect the predictions and quantitatively even a factor of two difference in $E$ changed predictions by only a few percent. The values of $E = 70$ GPa and $\nu = 0.33$ used in the calculations are representative of aluminum.

To implement the boundary conditions, the problem is decomposed in two linearly additive parts as illustrated in Fig. 2.1b. One part treats the unconstrained thermal expansion of the film and substrate as if they have the same thermal expansion coefficient, $\alpha_t = \alpha_s$. The solution to this problem is

$$\varepsilon_{ij}^{th} = (1 + \nu)\alpha_s \Delta T \delta_{ij}, \quad \sigma_{ij}^{th} = 0 \quad (i, j = 1, 2) \quad (2.3)$$
for both film and substrate (the factor $1 + \nu$ is due to the plane strain constraint in the $x_3$-direction). Since the stresses vanish everywhere, this part of the solution does not interact with the dislocations.

The other part of the solution, which is denoted by $(\cdot)'$ and pertains to the problem sketched in Fig. 2.1b, describes the stress which builds up in the film due to the thermal mismatch between the film and the substrate, and accounts for the presence of the dislocations. This problem pertains to a film with a thermal expansion coefficient $\alpha = \alpha_f - \alpha_s$ on a substrate that does not undergo thermal expansion. The solution to the full problem is

$$u_i = u^{\text{th}}_i + u'_i, \quad \varepsilon_{ij} = \varepsilon^{\text{th}}_{ij} + \varepsilon'_{ij}, \quad \sigma_{ij} = \sigma^{\text{th}}_{ij} + \sigma'_{ij}. \quad (2.4)$$

With plasticity arising from the collective motion of discrete dislocations, the $(\cdot)'$ solution is not independent of $x_1$. As shown in Fig. 2.1a, a unit cell is introduced in order to reduce the computation to one over a finite region. The film-substrate system is taken to be periodic in the $x_1$-direction with period $w$. The boundary conditions on the unit cell consist of the stress-free surface conditions

$$\sigma'_{12}(x_1, h) = \sigma'_{22}(x_1, h) = 0 \quad (2.5)$$

and the periodicity conditions

$$u'_i(0, x_2) = u'_i(w, x_2), \quad (2.6)$$

while traction continuity implies continuity of $\sigma'_{12}$ and $\sigma'_{11}$ at the cell boundaries $x_2 = 0$ and $x_2 = w$. The $(\cdot)'$ fields are governed by (2.1)–(2.2) with the appropriate substitutions for $\alpha$ according to Fig. 2.1b. In the absence of dislocations, the solution is

$$\varepsilon'_{ij} = 0, \quad \sigma'_{11} = -\frac{(\alpha_f - \alpha_s)E \Delta T}{(1 - \nu)}, \quad \sigma'_{ij} = 0 \text{ otherwise } \quad (i, j = 1, 2) \quad (2.7)$$

for the film and $\varepsilon'_{ij} = \sigma'_{11} = 0$ everywhere in the substrate. The solution (2.7) can be interpreted as resulting from the film freely expanding by $(\varepsilon^{\text{th}}_{11})_i = (1 + \nu)(\alpha_f - \alpha_s)\Delta T$ and subsequently being compressed by a stress $\sigma'_{11}$ to remove the expansion so that the film fits on the undeformed substrate.

In the presence of dislocations, the governing equations, subject to (2.5)-(2.6), are solved by decomposing the $(\cdot)'$ field quantities into two additive parts, as described in [17], so that the stress, strain and displacement fields in the film are given by:

$$u'_i = \tilde{u}_i + \hat{u}_i, \quad \varepsilon'_{ij} = \tilde{\varepsilon}_{ij} + \hat{\varepsilon}_{ij}, \quad \sigma'_{ij} = \tilde{\sigma}_{ij} + \hat{\sigma}_{ij}. \quad (2.8)$$
Here, the ($\tilde{}$)-fields are the superpositions of the fields of individual dislocations in infinite space, e.g.,

$$\tilde{\sigma}_{ij} = \sum_I \sigma_{ij}^{(I)}$$

(the superscript $(I)$ denotes the $I$th dislocation), and are singular at the positions of the dislocations. The ($\tilde{}$)-fields in (2.8) are image fields that are superimposed on the individual dislocation fields so that the boundary conditions on the unit cell are satisfied. These fields are smooth and their solution is obtained by a finite element method. The infinite space ($\tilde{}$)-fields are constructed in such a way that they reflect the periodicity in the problem; i.e. the field corresponding to each dislocation in the cell is the field, with periodicity $w$, due to this dislocation and all its replicas in the other cells making up the film. The closed-form expressions for these fields are given in [18]. The use of periodic discrete dislocation fields avoids the possibility of artificial dislocation patterning that may be induced when using a cutoff distance [19].

Special attention is needed for dislocations that glide out of the film. They leave the film, but they cannot be removed from the set of dislocations in the calculation because they contribute to a slip displacement and to the resulting step at the free surface. This is accounted for by virtually extending the slip planes above the film and positioning a dislocation at a distance $h$ above the free surface, i.e. at $x_2 = 2h$, once it leaves the film. This virtual dislocation produces stresses $\tilde{\sigma}_{12}$ and $\tilde{\sigma}_{22}$ on the stress-free surface (even though it is outside the film). These stresses are corrected by the ($\hat{}$)-fields. The virtual dislocations at $x_2 = 2h$ are sufficiently far away from the surface that the finite element solution can accurately describe the necessary correction.

Initially, the film-substrate system is at a high temperature and stress free. At each step of the simulation a temperature increment $\Delta T = \dot{T} \Delta t$ is prescribed and the boundary value problem is solved for all field quantities in the cell. For each time step, the dislocation structure is updated and then the updated solution for all field quantities is obtained as described above. As suggested by Kubin et al. [20], the following dislocation mechanisms are accounted for through constitutive rules: (i) dislocation glide; (ii) dislocation generation and (iii) annihilation; (iv) pinning at obstacles. All of these are governed by the Peach-Koehler force, which is computed as

$$f^{(l)} = n_i^{(l)} \left( \sigma_{ij} + \sum_{J \neq I} \sigma_{ij}^{(J)} \right) b_j^{(l)},$$
Chapter 2  Size effects in single crystal thin films

Taking advantage of the fact that $\sigma_{ij}^{th} = 0$ according to (2.3).

Dislocation glide is taken to be drag controlled so that the velocity of dislocation $I$ is directly proportional to the Peach-Koehler force, $f^{(I)} = B v^{(I)}$, with $B$ the drag coefficient, which is taken to have the value $B = 10^{-4} \text{Pas}$. Annihilation of two dislocations with opposite Burgers vector occurs when they approach each other within an annihilation distance $L_c = 6b$. Generation of new dislocations is incorporated through a distribution of Frank-Read sources. In two dimensions, these are point sources which generate a dipole when the Peach-Koehler force on the source exceeds a critical value $\tau_{nuc} b$ during a time span $t_{nuc} = 10 \text{ ns}$. The sign of the dipole is determined by the direction of the force. The distance between the two dislocations, $L_{nuc}$, is set so that they will not immediately collapse and annihilate under an applied shear stress $\tau_{nuc}$, i.e.

$$L_{nuc} = \frac{\mu b}{2\pi(1-\nu)\tau_{nuc}}.$$  

(2.9)

with $\mu = E/(2(1+\nu))$ the shear modulus. A distribution of point obstacles, which are intended to mimic small precipitates or forest dislocations, is also introduced. Dislocations get pinned at such obstacles and are released once the Peach-Koehler force attains the obstacle strength $b\tau_{obs}$.

2.2 Results

The objective of the simulations is to gain insight into the thickness-dependent response of thin films. We consider representative values of the material parameters. The Burgers vector of the film material is taken to be $b = 0.25 \text{ nm}$. The linear coefficient of thermal expansion is taken to be representative of silicon for the substrate ($\alpha_s = 4.2 \times 10^{-6}/\text{K}$) and of aluminum for the film ($\alpha_f = 23.2 \times 10^{-6}/\text{K}$). Results are presented for values of the film thickness, $h$, ranging from 0.25 to 1 $\mu\text{m}$. In all calculations, the width of the periodic cell is taken to be $w = 2\mu\text{m}$. The potentially active slip planes for each slip system are spaced at $d = 100b$, so that there are $n = (w/d) \sin \phi$ slip planes with orientation $\phi$ inside the cell.

In all the simulations the density of Frank-Read sources randomly distributed on the slip planes is $\rho_{nuc} = 60/\mu\text{m}^2$. This implies that there are 120 sources per micrometer of film thickness in the cell, which means that not all slip planes are necessarily active. On the other hand, for the thickest films considered, $h = 1\mu\text{m}$, there are as many as 4 sources per slip plane. The strength of the sources is
taken randomly from a Gaussian distribution with mean strength $\tau_{\text{nuc}} = 25$ MPa and standard deviation of 5 MPa. With the chosen material properties, the mean nucleation distance from (2.9) is $L_{\text{nuc}} = 0.0625 \mu m$, which is $1/4$ of the smallest film thickness $h = 0.25 \mu m$, and, more importantly, only $1/8$ of the shortest slip plane length $h / \sin 60\degree$. However, since the strengths are taken from a Gaussian distribution, values of $L_{\text{nuc}}$ can deviate significantly from the average. In the distributions used in the calculations here, the smallest value of $\tau_{\text{nuc}}$ is 10 MPa, which corresponds to $L_{\text{nuc}} = 0.156 \mu m$. All sources are displaced by at least the distance $L_{\text{nuc}} \sin \phi$ from the top or bottom of the film, in order that both dislocations in a nucleated dipole are contained in the film. Whenever obstacles are considered, their density is taken to be the same as the source density and their strength is specified as $\tau_{\text{obs}} = 150$ MPa.

In order to limit the computational time, the cooling rate is specified as $\dot{T} = 40 \times 10^6 K/s$ and the total temperature decrease is 200K, which is smaller than usual in experiments. A small time step is required to accurately resolve the dislocation dynamics. Numerical experimentation showed that with the parameters used here, a time step $\Delta t$ no larger than 0.05 ns is needed mainly to capture the formation of dislocation junctions (dipoles) near the intersection of slip planes.

The finite element mesh used to solve for the $\mathbf{\hat{t}}$ fields depends on the thickness of the film. In all cases, four-node elements are used which are square in the film and which gradually elongate inside the substrate with increasing depth. For the thinnest films considered, $h = 0.25 \mu m$, we have used 10 elements through the film thickness. Numerical tests have shown that this gives sufficient resolution over the top surface to satisfy the stress-free condition with sufficient accuracy.

Each simulation starts with a dislocation-free film, so that when cooling begins, the response is initially elastic. As the temperature decreases, a uniform tensile stress builds up in the film. When the resolved shear stress on a slip system reaches the critical strength of the weakest point source, the source generates a dislocation dipole. One of the dipole dislocations glides in the direction of the free surface and the other glides toward the interface where it gets pinned. It is this movement which provides the mechanism of plastic relaxation of the thermal stress. As cooling proceeds, many other dislocations are nucleated. Because of the stress fields associated with the individual dislocations in the film, the Peach-Koehler force at a source can become large enough to induce a nucleation event, even if the average stress in the film is not high enough to activate the source.
2.2.1 Size effect

We first present results for three cases that differ in the film thickness only: \( h = 1 \mu m, 0.5 \mu m \) and \( 0.25 \mu m \). The film material contains three slip systems, with slip plane orientations: \( \phi^{(1)} = 0^\circ; \phi^{(2)} = 60^\circ; \phi^{(3)} = 120^\circ \). The three slip systems mimic in two dimensions the redundancy of the twelve available slip systems in FCC crystals.

Figure 2.2 shows the distribution of the dislocations and the in-plane stress \( \sigma_{11} \) at the end of the cooling process. The stress is normalized by the elastic stress
\[
\sigma_n = -\frac{(\alpha_f - \alpha_s) E \Delta T}{(1 - \nu)},
\]
which would be present in the film if plastic relaxation had not occurred, see (2.7). With the parameter values here, \( \sigma_n = 397 \text{MPa} \). For each film thickness, a single unit cell of the film as well as the top of the substrate is shown. For the chosen thermal expansion coefficients and with \( \Delta T < 0 \), the film is in a state of tension, \( \sigma_n > 0 \). The compressive stress in the substrate is very low on average, because of its large thickness, except in a thin layer directly below the interface which is affected by the dislocations in the film near the interface. Indeed, a relatively large number of dislocations are piled up in the film against the interface because the interface is modeled as being impenetrable. Due to this local high dislocation density, a boundary layer forms with a much higher in-plane stress than in the rest of the film.

A boundary layer is also seen in the dislocation density profiles \( \rho(x_2) \) across the film, shown in Fig. 2.3. The dislocation density plotted is the average dislocation density in a strip of height \( \lambda \), averaged in the \( x_i \)-direction. Making use of periodicity, this quantity is computed as
\[
\rho(x_2) = \frac{1}{b w \lambda} \sum_I b^{(I)}, \quad \forall I \text{ such that } x_2 - \lambda/2 < x_2^{(I)} < x_2 + \lambda/2.
\]

Using a strip height of \( \lambda = 0.025 \mu m \), the profiles in Fig. 2.3 show that there is distinct peak in the bottom strip which is roughly the same for all three thicknesses. The thickness of the highly stressed boundary layer is less than \( 0.025 \mu m = 100 \lambda \). The dislocation density in the rest of the film, where the tensile stress has been relaxed (Fig. 2.2), is at least a factor 4 lower and also appears to be about the same for the three cases.
2.2 Results

Figure 2.2  Distribution of $\sigma_{11}$, normalized by $\sigma_n$ defined in (2.10), and the dislocation distribution after cooling by 200K for three values of film thickness: (a) $h = 0.25\mu m$, (b) $h = 0.5\mu m$ and (c) $h = 1\mu m$. The films have three slip systems with slip plane orientations specified by $\phi^{(1)} = 0^\circ$, $\phi^{(2)} = 60^\circ$, and $\phi^{(3)} = 120^\circ$ (see Fig. 2.1).

Examining the near-interface dislocations shown in Fig. 2.2 reveals that they are either positive dislocations on the $\phi^{(2)} = 60^\circ$ slip planes or negative dislocations on the $\phi^{(3)} = 120^\circ$ slip planes. In both cases the horizontal component of the Burgers vector is in the positive $x_1$-direction. Neglecting the low density of dislocations in the rest of the film, the classical idealized picture emerges of a film that
Figure 2.3 Dislocation density profile across the film thickness $x_2$ for the films in Fig. 2.2. (a) $h = 0.25 \mu m$, (b) $h = 0.5 \mu m$ and (c) $h = 1 \mu m$.

is relaxed by a distribution along the interface of misfit dislocations with Burgers vector $b|\cos \phi|$ in the $x_1$-direction. Full relaxation of the film would require that the thermal strain $(\varepsilon_{11}^{th})_f = (1 + v)(\alpha_f - \alpha_s)\Delta T$ is entirely accommodated by such
misfit dislocations. The dislocation density in a strip of height \( \lambda \) needed for this is given by

\[
\rho = \frac{(1 + \nu)(\alpha_f - \alpha_s)\Delta T}{\lambda b \cos \phi}.
\]  

(2.12)

For \( \lambda = 0.025\mu m \), this expression gives a density \( \rho = 1.6 \times 10^3 \mu m^{-2} \). The dislocation density in the bottom strip \( \lambda \) of Fig. 2.3 is around 800\( \mu m^{-2} \). This is significantly less than the necessary dislocation density for a completely stress-free film. Thus we expect that there is a significant stress component \( \sigma_{11} \) left in the film.

Figure 2.2 gives insight into the nature of this stress state. An additional perspective is given by the \( x_1 \)-averaged \( \sigma_{11} \) profiles in Fig. 2.4a for the three film thicknesses considered at \( \Delta T = 200K \). The \( x_1 \)-averaged stresses, \( \langle \sigma_{11} \rangle(x_2) \), are computed in a strip-wise fashion as in (2.11), i.e.

\[
\langle \sigma_{11} \rangle(x_2) = \frac{1}{w\lambda} \int_{x_2-\lambda/2}^{x_2+\lambda/2} \int_0^w \sigma_{11}(x_1,z_2) dx_1 dz_2.
\]  

(2.13)

The integral is evaluated using \( 5 \times 5 \) trapezoidal quadrature in each strip with height \( \lambda \) of a finite element. A value of \( \lambda = 0.0167\mu m \) was found to give well-converged results of \( \langle \sigma_{11} \rangle(x_2) \). Also shown in Fig. 2.4a are the average stresses in the film: \( \langle \sigma_{11} \rangle_f = 50MPa, 70MPa \) and \( 130MPa \) for \( h = 1\mu m, 0.5\mu m \) and \( 0.25\mu m \), respectively (with \( \langle \rangle_f \) denoting the film average of a quantity). The profiles clearly show the presence of highly stressed boundary layers and also illustrate the variation in boundary layer thickness with film thickness. For the two thickest films, the boundary layer thicknesses are nearly the same, but the boundary layer in the thinnest film is significantly thinner. It is also of importance to note that the stress level in the core of the \( h = 0.25\mu m \) film is higher than for the other two films. It is primarily this lack of relaxation in the core that causes the \( h = 0.25\mu m \) film to have the highest average stress.

Average stress, \( \langle \sigma_{11} \rangle_f \), versus film thickness \( h \) is shown in Fig. 2.4b to illustrate the scaling with film thickness. The thicker two films suggest a Hall-Petch-like \( h^{-1/2} \) scaling. The data for the thinnest two films are consistent with the \( h^{-1} \) scaling found by Freund [10] and Nix [11] on the basis of energy considerations. Although data from three points is not conclusive, we believe that the dichotomy in scaling is due to a change in hardening mechanism for the thinnest film, as will be discussed in detail in Sec. 2.2.3. We note that the average film stresses are not sensitive to the source distribution; other realizations give results that only differ by a few percent.
Chapter 2. Size effects in single crystal thin films

Figure 2.4 (a) Profiles $\langle \sigma_{11} \rangle (x_2)$ of the in-plane stress in the films in Fig. 2.2 averaged in the $x_1$–direction. The vertical lines show the total film averages, $\langle \sigma_{11} \rangle_f$. (b) Average film stress versus film thickness $h$. The straight lines discribe a power law of the form $\langle \sigma_{11} \rangle_f \propto h^{-p}$, giving $p \approx 1$ for the thinnest two films and $p \approx 1/2$ for the thicker ones.

While we have focused until now on the stress states at the end of the cooling process, the full history is shown in Fig. 2.5. Rather than stress, however, Figure 2.5 shows the evolution of $\langle \varepsilon_{11}' \rangle_f$ as a function of the temperature reduction. The value of $\langle \varepsilon_{11}' \rangle_f$ represents the average lattice strain in the film due to the thermal
2.2 Results

Figure 2.5 Curves of $\langle \varepsilon'_{11} \rangle_f$ versus imposed temperature for three values of film thickness: $h = 0.25\mu m$, $h = 0.5\mu m$ and $h = 1\mu m$ (see Fig. 2.2 and Fig. 2.3).

Mismatch with the substrate. Adding $\varepsilon_{11}^h$ to it, eq. (2.3), gives the quantity which is usually measured experimentally by X-ray diffraction [3, 21]. From $\langle \varepsilon'_{11} \rangle_f$ and the corresponding $\langle \varepsilon'_{22} \rangle_f$, the average stress $\langle \sigma'_{11} \rangle_f$ can be directly computed from Hooke’s law, eq. (2.2), with $\alpha = 0$. Since $\sigma_{1j}^h = 0$, eq. (2.3), this immediately gives the total average film stress $\langle \sigma_{11} \rangle_f$, cf. (2.4). Thus, plots of the evolution of $\langle \varepsilon'_{11} \rangle_f$ give insight into the average stress development in the film.

The curves in Fig. 2.5 exhibit a distinct size effect on the hardening, with thinner films being harder. The yield point, which corresponds to a yield stress of about 30MPa, does not exhibit a size effect, as the initiation of plastic deformation is controlled by the statistical distribution of source strengths. Prior to the first nucleation event, the stress in the film is uniform, so that the first occurrence is determined by the weakest source. Since the source strengths are chosen randomly from a Gaussian distribution around a certain value, and since the specific values of source strength are different for different films, plastic deformation starts first in the film that contains the weakest source. For the cases shown in Fig. 2.5 this happened to be the thinnest film, where the minimum nucleation strength, out of the average of $\bar{\tau}_{\text{nuc}} = 25\text{MPa}$, is $\tau_{\text{nuc}} = 10\text{MPa}$.

Hardening, on the other hand, is a collective effect of the nucleation, glide and
annihilation of a large number of dislocations. Statistical effects are therefore smaller. The hardening rate averaged over a temperature drop of $\Delta T = 185K$ from the onset of yield, $\Delta\langle\sigma_{11}\rangle_{f}/\epsilon_{11}^{th}$ is 18 GPa, 39 GPa and 97 GPa for $h = 1\mu m$, $0.5\mu m$ and $0.25\mu m$, respectively. To check the sensitivity of the yield stress and hardening rate to the value of $\tau_{\text{nuc}}$, the calculations were repeated with the same source distributions but with the value of $\tau_{\text{nuc}}$ at each source multiplied by a factor of two. The values of the yield stress for each of the three films doubled, while
2.2 Results

Figure 2.7 Curves of $\langle \varepsilon'_1 \rangle_f$ versus imposed temperature for three values of film thickness: $h = 0.25 \mu m$, $h = 0.5 \mu m$ and $h = 1 \mu m$ for the calculation in Fig. 2.6 with a prescribed distribution of dislocation obstacles.

the values of the hardening rate remained essentially unchanged.

Calculations were repeated with all parameters fixed except that a uniform random distribution of point obstacles was added with a density of $60/\mu m^2$. Figure 2.6 shows that the dislocation density in the core region is higher with obstacles than without obstacles (especially for the thickest film), since the obstacles tend to prevent dislocations from leaving the film at the free surface. In fact, numerous dislocation dipoles form at slip plane intersections, leading to a harder core region than without obstacles which gives rise to the increased hardening rate seen in Fig. 2.7 compared with that in Fig. 2.5.

2.2.2 Effect of slip plane orientation

In order to investigate the influence of slip plane orientation, the simulations presented in the previous section have repeated with the crystal rotated by $\pm30^\circ$, so that $\phi^{(1)} = 30^\circ$; $\phi^{(2)} = 90^\circ$; $\phi^{(3)} = 150^\circ$ The source density is the same as before, but the source positions and strengths are different; there are no obstacles. Slip systems 2 and 3 are the most active ones, because the resolved shear stress
Chapter 2 Size effects in single crystal thin films

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{figure2.8.png}
\caption{Curves of \(\langle \varepsilon'_1 \rangle_f\) versus imposed temperature for three values of film thickness: \(h = 0.25\mu m\), \(h = 0.5\mu m\) and \(h = 1\mu m\). The films contain three slip systems with slip plane orientations \(\phi^{(1)} = 30^\circ\), \(\phi^{(2)} = 90^\circ\), and \(\phi^{(3)} = 150^\circ\).}
\end{figure}

\[\tau = -\sigma_{11}/2\sin 2\phi,\] caused by a nominal tensile stress \(\sigma_{11}\), is largest in absolute value. In fact, the Schmid factor \(|\sin 2\phi|\) is the same as for the \(\pm 60^\circ\)-slip systems in the original orientation. This explains that the onset of yield (Fig. 2.8) is roughly the same as for the original crystal orientation (Fig. 2.5).

The hardening in the film is reduced however, i.e. the stress is more relaxed in the rotated orientation for all film thicknesses, cf. Fig. 2.8 with Fig. 2.5. One explanation for this is that fewer dislocations are needed to relax the film in the rotated orientation: according to (2.12) with \(\phi = \phi^{(1)} = 30^\circ\), a dislocation density \(\rho = 900\mu m^{-2}\) is needed in the height \(\lambda = 0.025\mu m\) for complete relaxation. The dislocation density found near the interface is around \(600\mu m^{-2}\) for all three thicknesses. This is a higher percentage (67%) than for the previous orientation (50%), which is consistent with the film being more relaxed.

The dislocation distribution along with the boundary layer that forms in the thinnest film with orientation \(\phi^{(1)} = 30^\circ\) is seen in Fig. 2.9a. Profiles of \(x_1\)-averaged stresses for all three thicknesses reveal that the boundary layers have the same thickness, which is slightly smaller than that seen in Fig. 2.4a for the original orientation. A second difference is that the core region of the films is less stressed. The two effects explain the lower hardening in Fig. 2.8.

To investigate the orientation dependence further, simulations were carried out in single slip with slip plane orientations of \(\phi = 15^\circ\), \(30^\circ\), \(60^\circ\) and \(75^\circ\). In single slip, dislocations do not form junctions, so that the time step can be increased by an order of magnitude without losing accuracy. Figure 2.10 shows that the
2.2 Results

hardening rate increases with increasing \( \phi \). This is mainly due to a combination of the orientation dependence of the Schmid factor and of the slip plane length \( h / \sin \phi \). Also, we see that plastic flow occurs earlier for slip plane orientations of 30° and 60° than for those of 15° and 75°, because the resolved shear stress \( \tau \) is larger for the 30° and 60° orientations.

Figure 2.9 Internal stress states for the films in Fig. 2.8 after cooling by 200K. (a) Contours of \( \sigma_{11} \), normalized by \( \sigma_n \) defined in (2.10), and the dislocation distribution for \( h = 0.25 \mu m \). (b) Profiles \( \langle \sigma_{11} \rangle(x) \) of the in-plane stress in all films with this orientation. The vertical lines show the total film averages, \( \langle \sigma_{11} \rangle_f \).
Chapter 2. Size effects in single crystal thin films

Figure 2.10 Curves of $(\varepsilon_{11}')_i$ versus imposed temperature for films with a single slip system having the slip planes oriented at $\phi^{(1)} = 15^\circ$, $\phi^{(2)} = 30^\circ$, $\phi^{(3)} = 60^\circ$ and $\phi^{(1)} = 75^\circ$. All films have thickness $h = 0.25 \mu m$.

2.2.3 Origin of hardening

Examination of the average strain-temperature curves in Figs. 2.5 and 2.8 indicates that there is a more or less pronounced kink in the average slope after the onset of plasticity as exemplified in Fig. 2.5. These kinks reflect a change in the hardening rate in the plastic regime. For the smallest thickness, $h = 0.25 \mu m$, the kink is most clearly visible; also for the thickest film, $h = 1 \mu m$, a kink can be observed but occurs at a later stage. Moreover, we observe that the hardening in the second part of the curve increases with the slip plane angle, as seen by comparing Figs. 2.5 and 2.8. A significant increase in hardening has been noted experimentally by Leung et al. [1] in various types of films, and can also be seen in the experimental results in [3]. The strongest effect is always seen, as here, for very thin films. The effect is not found by Leung et al. [1] for passivated films, which has led them to suggest that the effect is due to additional relaxation by surface diffusion at higher temperature. However, the experimental results in [3] also show a two-stage hardening effect for very thin ($h = 0.3 \mu m$) passivated films. In our calculations there is no diffusion, and therefore it is interesting to explore
2.2 Results

Figure 2.11 Distribution of resolved shear stress on the slip plane $\phi^{(2)} = 60^\circ$, $\tau_{60}$ and the dislocation distribution at final temperature for the film in Fig. 2.2a. The point sources on the this slip system are shown as circles.

the origin of the kink in the simulations.

One possible cause is a sudden increase in dislocation density, leading to an increase in the number of dislocation junctions and therefore to an increase in hardening. However, since the kink is present in multiple slip as well as in single slip (no junctions), Fig. 2.10 and Fig. 2.5, this is excluded. Another possible cause is an abrupt reduction of the rate of dislocations nucleated. This is what happens in our calculations, with the reduction in nucleation rate arising from the back stress generated by the dislocations in the boundary layer adjacent to the interface. For each source, nucleation first occurs when the resolved shear stress reaches $\tau_{\text{nuc}}$. The stress field of the dipole generated by this source shields the source from further nucleation. The back stress at the source reduces as the dipole spreads, with the least effect occurring when one of the dislocations has left the film through the free surface and the other is blocked near the interface. In very thin films the back stress in this configuration is still high enough to have a significant effect at the source. During the first stage of the cooling process, other sources in the film will be activated before the back stress at previously activated sources has been overcome by the applied stress. This gives rise to the initial hardening rate. At some stage of the deformation history, all sources have been activated and collectively they have produced back stress throughout the film. Subsequently, the only way in which sources can be activated is by overcoming the back stress through further straining of the film. Thus, nucleation is delayed, which gives rise to additional hardening in the $\langle \varepsilon_1 \rangle_{\varepsilon - \Delta T}$ curves.

To support this explanation, Fig. 2.11 shows the distribution of the resolved shear stress $\tau$ for the film with $h = 0.25 \mu m$ on slip system $\phi^{(2)} = 60^\circ$ at the same time as in Fig. 2.2a. Also shown are all sources that are present on these slip
planes to demonstrate that they are all in regions with relatively low stress due to the back stresses caused by the dislocations piled up against the interface. As the thickness of the film increases, the back stress at a source caused by the dislocation pile-ups at the film-substrate interface will, on average, be lower because of the larger distance between the pile-up and the source. Hence, for thicker films, the kink in hardening is delayed and is less intense. This is confirmed by the results in Figs. 2.5 and 2.8.

The presence of the back stress is expected to give an important contribution to the response when the temperature change is reversed. This is verified for the thinnest film, by re-heating from the final temperature of $T = 400K$ reached previously. As seen in Fig. 2.12, reverse plasticity occurs almost immediately after temperature reversal for $h = 0.25 \mu m$. Without the presence of the long-range back stresses, elastic unloading would occur over a larger interval.

It should also be noted that the resolved shear stress distribution in Fig. 2.11 does not exhibit the same clear boundary layer as does the distribution of $\sigma_{11}$ in

---

Figure 2.12 Curves of $\langle \varepsilon_{11} \rangle_f$ versus imposed temperature for a thermal cycle between 600K and 400K for films of thickness $h = 0.25 \mu m$ and $h = 0.5 \mu m$. 
2.3 Thermal cycling

In this section are presented results of simulations in which the two thinnest films are at first cooled down from 600K to 400K, successively heated up to 600K and at last cooled again down to 400K.

Fig. 2.2a. This suggests a limitation to the classical picture of misfit dislocations with Burgers vector parallel to the interface. The piled-up dislocations on the inclined slip planes do not neatly combine to such misfit dislocations: on average they do, but not point-wise along the interface. In fact, closer examination of the dislocation structure shown in Fig. 2.5a shows the presence of pile-ups of two or three dislocations on the same slip plane. As there are no dislocations on a nearby inclined slip plane to cancel the resulting long-range back stress, the back stress remains effective for blocking nucleation on the same slip plane.

Figure 2.13 Dislocation distribution and in-plane stress, normalised by the elastic stress $\sigma_n$, in the film with thickness $h = 0.25\mu m$: (a) at 400K after the first cooling, (b) at 600K after heating and (c) at 400K after the second cooling.
Chapter 2. Size effects in single crystal thin films

Figure 2.14 Average in-plane stress in the film versus imposed temperature for film thicknesses: (a) $h = 0.25 \mu m$ and (b) $h = 0.5 \mu m$.

The dislocation distribution for the 0.25\( \mu m \) film at the end of the first cooling stage ($T = 400K$) is shown in Fig. 2.13a, together with the distribution of $\sigma_{11}$, the normal stress parallel to the interface. If the temperature is kept constant at 400K, no significant evolution of the dislocation structure is observed, indicating that the dislocation structure is close to equilibrium.

During heating, when the film straining changes sign, the direction of dislocation motion is reversed. The high back stress built up during cooling acts to enhance dislocation motion, so that reverse plastic deformation occurs. The dislocations that were forming pile-ups at the interface progressively reach the free surface and leave the film. At the end of the heating process (see Fig. 2.13b) only a few dislocations are left in the film. A few of those dislocations have signs opposite to the signs of the dislocations nucleated during cooling; the opposite-signed dislocations were nucleated during heating when the mean stress state became sufficiently compressive. The average $\sigma_{11}$ stress (in absolute value) reached after heating is much lower than that after cooling (note that the stress range in Fig. 2.13b is smaller than in Figs. 2.13a and 2.13c) but is not zero as it is prior to the first cooling. Thus, the response is not reversible, as is indeed seen in experiments. After the second cooling (Fig. 2.13c) the stress state in the film is very similar to the one obtained after the first cooling, with small differences in the dislocation structure.

The evolution of the average in-plane stress in the two films during the imposed thermal history is shown in Fig. 2.14. Comparison between Fig. 2.14a and b for
2.3 Thermal cycling

Figure 2.15 Dislocation distribution and in-plane stress normalised by the elastic stress $\sigma_n$ in the film with thickness $h = 0.5\mu m$ after cooling to 400K and re-heating to 600K.

$h = 0.25\mu m$ and $0.5\mu m$ gives evidence of a quite pronounced size effect. During the first cooling cycle, the $0.25\mu m$ film hardens much more than the film with $h = 0.5\mu m$, as discussed in more detail in the previous section. In the $0.25\mu m$ film, the high stress level pushes the dislocations in the pile-ups close together. This results in large back stresses which cause reverse plasticity in the early stages of the subsequent heating process.

In the $h = 0.5\mu m$ film the back stress associated with the pile-ups is lower. As a consequence, this thicker film unloads elastically almost until the stress changes sign. A compressive stress builds up in the film at around 430K, leading to the dislocation distribution at 600K shown in Fig. 2.15. The dislocation density after unloading is greater in the $0.5\mu m$ film than in the $0.25\mu m$ film. (Fig. 2.15 versus Fig. 2.13b).

The dislocation structure at 600K strongly influences the material response during the last cooling sequence. The dislocation density in the thin $h = 0.25\mu m$ film is very low, Fig. 2.13b, and most of the dislocations have been generated during the first cooling cycle. This situation is very similar to the initial condition, when the film was dislocation free. Therefore the response of the $0.25\mu m$ film during first and second cooling differs only in the initial stages (until 550K), see Fig. 2.14a.

During re-cooling of the $0.5\mu m$ film, Fig. 2.14b, the dislocations that were nucleated during heating are already available to move and relax the stress, giving rise to a difference in initial plasticity compared to the first cooling. Subsequently, around 500K, relaxation becomes mainly nucleation controlled, and the stress level reaches and overtakes the level in the first cooling.
2.4 Conclusions

Discrete dislocation analyses have been carried out of the stress evolution in single crystal films arising from the thermal mismatch between the film and its substrate. Attention was confined to plane strain, with the substrate remaining elastic and any effect of elastic mismatch between the film and substrate neglected. The film is initially dislocation free and the dislocations in the film, which are all of edge character, nucleate from Frank-Read sources on a specified set of slip planes. At the start of a calculation, the film-substrate system is stress free and the deformation arises from a prescribed temperature history. The results exhibit the follow trends:

- The stress evolution and the hardening show a clear dependence on film thickness for the thicknesses analyzed which range from 0.25 $\mu$m to 1 $\mu$m.

- The effect of film thickness is mainly due to the formation of a hard boundary layer at the film-substrate interface. The width of the boundary layer, which arises from dislocation pile-ups at the interface, does not scale with the film thickness.

- The boundary layer width depends on the orientation of the slip systems in the film.

- Below a certain film thickness, an additional contribution to hardening arises from a reduction in dislocation nucleation caused by the back stress associated with the dislocation pile-ups at the film-substrate interface. This reduction in the rate of dislocation nucleation can occur abruptly and lead to a two-stage hardening behavior as seen experimentally.

- In very thin films all the available dislocation sources are affected by the back stress early in the stress relaxation process. Further nucleation is suppressed until the back stress at the sources is overcome by additional straining of the film. The absence of dislocations that can eliminate the long-range back stress is related to the limited availability of sources.

Thermal cycling of the two thinnest film between 600K and 400K shows that:

- For both values of the film thickness, the high long-range back stress accumulated inside the films during the first cooling process induces early reversed plasticity during subsequent heating.
• When heating starts, de-stressing is elastic over a range that is almost equal to two times the initial elastic range, with an average yield stress of 40MPa in both films, predicting that hardening in thin films is essentially of kinematic character.

• Plastic relaxation during heating takes place mainly by the movement of dislocations already present in the films. In the 0.5µm film, there is a small contribution to relaxation by nucleation of new dipoles at the end of the cooling process, when the mean stress becomes sufficiently compressive to activate the sources.

• Upon re-cooling, the dislocation density in the 0.25µm film is so low, that the plastic behavior during the first and second cooling cycles are very similar. However, the dislocations nucleated during heating in the thicker $h = 0.5\mu$m film influence the hardening of that film during the second cooling.

References


