Microplasticity and yielding in crystals with heterogeneous dislocation distribution

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Abstract

In this study, we use discrete dislocation dynamics simulation to investigate the effect of heterogeneous dislocation density on the transition between quasi-elastic deformation and plastic flow in face-centered cubic single crystals. By analyzing the stress–strain curves of samples with an initial, axial dislocation density gradient, we arrive at the following conclusions: (i) in the regime of quasi-elastic deformation before the onset of plastic flow, the effective elastic modulus of the simulated samples falls significantly below the value for a dislocation-free crystal. This modulus reduction increases with decreasing dislocation density gradient: crystals with homogeneous dislocation distribution are thus weakest in the quasi-elastic regime; (ii) the transition towards plastic flow occurs first in regions of reduced dislocation density. Therefore, the overall yield stress decreases with increasing dislocation density gradient; (iii) crystals with dislocation density gradient exhibit a more pronounced hardening stage during which stress is re-distributed onto stronger regions with higher dislocation density until the sample flows at a constant flow stress that is approximately independent of dislocation density gradient. We interpret these findings in terms of a continuum dislocation dynamics inspired model of dislocation density evolution that accounts for geometrically reversible ('inversive') dislocation motions. The transition between quasi-elastic and plastic deformation is interpreted as a transition from inversive to non-inversive dislocation motion, and the initial differences in elastic modulus are related to a density dependent polarizability of the dislocation system. The

subsequent plastic flow behavior is analyzed in terms of a modified version of Mughrabi's composite model.

Keywords: dislocation dynamics, microplasticity, yielding, dislocation density gradient

(Some figures may appear in colour only in the online journal)

1. Introduction

Spatially graded microstructures can be observed in many biological materials, such as bones, bamboo, shells, etc, where the microstructure change gradually. These graded structures are the result of natural selection and natural evolution, and almost all of them have some special excellent properties [1]. Therefore, inspired by the gradient biological material, metallic materials with graded microstructures were also fabricated and have shown excellent mechanical properties, fatigue properties, friction and wear behavior [2, 3]. Gradients of microstructural properties may exist on the levels of grain size distribution, twin density, dislocation density, solute or precipitate density, or combinations thereof [4]. There are many studies of the effects of grain size gradients on plasticity [5–7], but there are few studies of the effects of dislocation density gradients despite the fact that such gradients are ubiquitous. Because of the loss of dislocations at surfaces, the existence of a dislocation density gradient in the near-surface region and a concomitant dependency of hardening on distance-to-surface can be demonstrated even in uni-axial deformation of macroscopic samples [8, 9]. Even more pronounced are gradient effects in samples where an intrinsically heterogeneous deformation mode imposes a gradient of strain and hence of strain hardening, see e.g. an investigation of dislocation density gradients of pure nickel under torsion, where deformation and dislocation density gradients were found to be accompanied by a decrease in Vickers hardness from surface to center [10, 11].

The purpose of our study is to understand the effect of dislocation density gradients on the small-strain deformation of face-centered cubic (fcc) metals by using the discrete dislocation dynamics (DDD) simulation method to investigate the deformation behavior in the microplastic regime and during yielding. The paper is organized as follows: we first give a concise presentation of the simulation method and present the results obtained by DDD simulation, which we discuss in terms of characteristic differences of dislocation behavior in the quasi-elastic/microplastic and the plastic deformation regime of microcrystals with and without dislocation density gradient. We then move to a more theoretical description, where we formulate a dislocation density based framework to interpret the characteristic differences between dislocation behavior in graded and non-graded samples, and between quasi-elastic and plastic deformation regimes. We conclude with a brief discussion, that puts our results into the general context of recent interpretations of the elastic-plastic transition.

2. Simulation set-up

We use DDD simulation to describe plastic deformation in terms of dislocation motion and dislocation interactions (see e.g. [12–15]) and apply this method to investigate the effect of dislocation density gradients. Specifically, we use the parallel dislocation simulator (ParaDis) code which uses an adaptive nodal description of the dislocation lines to trace the evolution of the dislocation system [16]. For a detailed description of the code, the reader is referred to the

Table 1. Material and model parameters used in the DDD simulation and density based model.

Material parameters	Symbol	Value
Shear modulus (GPa)	μ	54.6
Poisson ratio	ν	0.324
Burgers vector (nm)	b	0.256
Drag coefficient (Pa s)	B	10^{-4}
Model parameters	Symbol	Value
Mean dislocation density (m^{-2})	$\bar{\rho}$	7.6×10^{13}
Mean dislocation source length (nm)	l_{FR}	400
Fraction of dislocations on active slip systems	η_{E}	1/3
Taylor constant	α	0.4
Polarizability for mean dislocation density	P_0	0.883
Density derivative of polarizability	P_1	0.0425
Stress re-distribution factor	η_{E}	0.1

literature [12]. Originally developed by Lawrence Livermore National Laboratory for large-scale hardening simulations [12], ParaDis has become a popular tool for DDD simulation that has been adapted to a wide range of problems including dislocation processes at surfaces [17], size dependent plasticity of small samples [18], and interactions of dislocations with twin boundaries [19].

2.1. Sample geometry and loading mode

In our investigation we consider cuboidal samples of an fcc metal. We use material parameters indicative of Cu as summarized in table 1. All samples have dimensions $4000b \times 4000b \times 12000b$, which, with a Burgers vector length $b = 0.256$ nm, corresponds to sample dimensions of approximately $1 \mu\text{m} \times 1 \mu\text{m} \times 3 \mu\text{m}$. The samples are loaded uni-axially, the load axis is aligned with the $[1, -1, 0]$ axis of a Cartesian coordinate system and the load-perpendicular sample surfaces have $[1, 1, 1]$ and $[-1, -1, 2]$ orientation. This implies that four slip systems on the $[1, -1, 1]$ and $[-1, 1, 1]$ slip planes are loaded symmetrically. These slip systems have a common Schmidt factor of $M = 1/\sqrt{6} \approx 0.408$. The initial configuration and sample geometry are shown in figure 1. In addition to uni-directional tests conducted at an imposed strain rate of 5000 s^{-1} , loading-unloading tests were simulated. In these tests, the system was unloaded by inverting the straining direction after total strains of $\varepsilon = 0.05\%$, $\varepsilon = 0.1\%$, and $\varepsilon = 0.2\%$ had been reached. The strain rates during forward loading and unloading were the same, and unloading was conducted until the axial stress had reached zero.

2.2. Initial and boundary conditions

Our initial dislocation configuration consists of randomly located Frank-Read Sources, i.e. initially straight segments with pinned endpoints and a source length $l_{\text{FR}} = 400$ nm. We create a total of $n_{\text{FR}} = 600$ sources in the volume V of each simulated sample, which amounts a total dislocation density of

$$\bar{\rho} = \frac{n_{\text{FR}} l_{\text{FR}}}{V} = 76.5 \times 10^{12} \text{ m}^{-2}. \quad (1)$$

Figure 1. Initial dislocation configurations, blue dots represent F–R dislocation source pinning points, the red lines represent dislocation lines; the coordinate system indicates the crystal orientation of the three samples; (a) sample without dislocation density gradient, (b) low dislocation density gradient, (c) high dislocation density gradient. Graph (d) shows the axial dislocation density profile of sample (c) (density values averaged over 10 slices perpendicular to the sample axis).

The sources are equi-distributed over the 12 possible slip systems. The orientations of the pinned segments within the respective slip planes are equi-distributed over the unit circle, and the segment centerpoints are located at random positions, under the provision that the segments do not intersect the sample surface.

Sources with pinned endpoints represent artificial configurations and it is necessary to ensure that the artificial pinning points do not control dislocation motion and dislocation multiplication. To avoid artefacts caused by the initial pinning points, it is necessary to ensure that the dislocation source length is significantly larger than the dislocation spacing: In that case, operation of a source is controlled by interactions with other dislocations and by the requirement that the emerging loop can move through the dislocation forest. As discussed in [20], this can be ensured by taking the source length to be at least 2.5–3 times the dislocation spacing, i.e. $\rho l_{FR}^2 > 10$. This condition is in our simulations always fulfilled. It ensures that the flow stress τ_f is not mainly controlled by the Orowan stress for source activation, but by

the standard Taylor stress representative of forest hardening

$$\tau_f \propto \alpha \mu b \sqrt{\rho}, \quad (2)$$

where $\alpha \sim 0.2$ – 0.5 is the Taylor factor which depends on the distribution of dislocations over the different slip systems and μ is the shear modulus of the material.

For the present study of the effect of dislocation density gradients on the initial deformation behavior, the samples are divided along their long axis into three layers of width $1 \mu\text{m}$ and volume $V/3$. The centerpoints of a given number n_i of dislocation sources are located randomly within each layer, hence, the characteristic dislocation density per layer is

$$\rho_i = \frac{3n_i l_{\text{FR}}}{V}. \quad (3)$$

Characteristic dislocation densities and dislocation spacings in each layer are summarized in table 1, together with the overall dislocation density gradients of the three sample types. Realizations of initial dislocation configurations for the three sample types are illustrated in figure 1. The dislocation sources have finite extension and may extend across layer boundaries. Hence, despite the step-wise change of the source centerpoint densities during the construction process, the overall dislocation density changes smoothly in form of an approximately constant axial dislocation density gradient, see figure 1(d) for illustration. (Note that a slight decrease of the dislocation density in the layers at the top and bottom stems from the fact that dislocation sources are not allowed to intersect the surface.)

Regarding boundary conditions, we assume that dislocations can freely leave through the sample surfaces. This boundary condition as implemented in standard ParaDis (i.e. without surface corrections to the dislocation stress fields) poses certain problems when compared with simulation schemes that fully account for the elastic boundary value problem [21]. While the stress state for uni-axial deformation is correctly represented on average, the same is not true for the stress fluctuations caused by the intrinsic heterogeneity of dislocation slip. In particular, image forces acting on near-surface dislocations are not accounted for. This implies that, in the near-surface region where image forces prevail over the mutual interactions of dislocations, our simulations do not accurately represent the actual dynamics. This region can be envisaged as a layer with a width of about $\bar{\rho}^{-1/2} \approx 0.1 \mu\text{m}$ from the specimen surfaces. As a consequence the results of our simulations, while representing a qualitatively correct picture of the effects caused by surfaces and dislocation density gradients on plastic deformation behavior, cannot be considered fully accurate in quantitative terms. However, for the case of a homogeneous dislocation distribution, the quantitative differences in comparison with comparable simulations that account for the correct surface boundary conditions [22] are minor. For the purposes of gaining a qualitative understanding of the phenomena, which is what we are aiming for, the present simplified approach is therefore fully adequate.

3. Simulation results

In DDD, the initial configuration has a significant influence on the simulation results. Due to the different initial configurations, the stress–strain curves and the dislocation density evolution are different in the presence of a dislocation density gradient. Figure 2 shows stress–strain curves for samples without dislocation density gradient and for samples with small and large dislocation density gradient as specified in table 2.

Each stress–strain curve is averaged over multiple realizations of the initial conditions. From the figure, we can see that the stress–strain curves exhibit two distinct regimes. In the regime of small strains, we observe an approximately linear increase of stress with total strain.

Figure 2. Stress strain curves of samples with different dislocation density gradients. The inset shows part of the quasi-elastic regimes together with the effective elastic moduli of the simulated samples.

Table 2. Initial dislocation density distribution for the different sample configurations.

Cases	No gradient	Low gradient			High gradient		
Gradient (10^{18} m^{-3})	0	18.714			37.428		
Layers	1–3	1	2	3	1	2	3
Number of sources	200	150	200	250	100	200	300
Density (10^{12} m^{-2})	76.5	57.4	76.5	96	38	76.5	114
Dislocation spacing (nm)	114	132	114	102	162	114	93

However, the behavior in this stage is not simply elastic. This can be seen from the fact that the slope of the stress strain curves (shown in the inset of figure 2) falls significantly below the theoretical value for an isotropic material with $\nu = 0.324$ and $\mu = 54.6$ GPa, for which we expect an elastic slope of $E = 2\mu(1 + \nu) = 144.4$ GPa. The actual values of the quasi-elastic slope (henceforth: effective elastic modulus E_{eff}), however, are close to 110 GPa. Moreover, the effective elastic modulus is found to depend systematically on the dislocation microstructure. From the initial slope of the stress–strain curve, we find an effective elastic modulus, which for the homogeneous dislocation arrangement amounts to 107.8 GPa, for the sample with weak dislocation density gradient to 110.9 GPa, and for the sample with high dislocation density gradient to 112.6 GPa. We can thus conclude that, in the initial quasi-elastic regime of the stress–strain curve, the presence of a dislocation density gradient leads to a stiffer response of the material.

The opposite behavior is observed during the initial stage of plastic flow. The transition from quasi-elastic to plastic behavior occurs earliest in the material with high dislocation density gradient, later in the samples with small density gradient, and even later in the samples with homogeneous dislocation density distribution. Accordingly, during the initial stage of plastic flow, the presence of a dislocation density gradient makes the material softer. On the other hand, the initial hardening slope increases with increasing dislocation gradient. As a consequence, the flow stress differences between samples with graded and with homogeneous dislocation microstructure decrease with increasing plastic strain, and above a

Figure 3. Curves of dislocation-related strain and average dislocation density versus total strain for the three types of samples; full lines: dislocation-related (inverse plus plastic) strain, dashed lines: total dislocation density; inset: relative change of local dislocation density in the three layers, for sample with high dislocation density gradient.

total strain of about 0.35% (plastic strain of about 0.25%) the flow stress of all samples saturates at a level of about 170 MPa that does not depend on the initial microstructure.

The onset of plastic flow is not associated with any marked increase in the total dislocation density, which, as shown in figure 3, changes by less than 10% during the entire simulation. The same is true if we look at the three layers individually (inset in figure 3): even for the largest dislocation density gradient, the dislocation densities in the three layers remain close to their initial values, and there is no systematic dependency of the dislocation density change on the initial dislocation density. Indeed, as seen in figure 4(a), the initial dislocation density distribution remains practically unchanged as the system makes the transition from quasi-elastic deformation to plastic flow.

This finding is remarkable: the simplest explanation for the observed deformation behavior would be that, in line with equation (2), deformation starts in the layer where dislocation density and flow stress are lowest. The ensuing local plastic flow then would lead to an increase in dislocation density that, through isotropic hardening, compensates for the initial flow stress differences such that, ultimately, the sample deforms homogeneously and the flow stress reached no longer depends on initial conditions. This explanation is wrong, as the simulations neither show a significant increase of dislocation density with plastic strain, nor a reduction of the dislocation density gradient. We discuss, in section 4, the reasons for this behavior and propose a better explanation for the observed flow characteristics and dislocation density evolution.

Before doing so, it is worthwhile to look at the dislocation activity, defined as the fraction $\eta = \dot{\epsilon}_{pl} / \dot{\epsilon}$ of the local strain rate that is due to dislocation motions, and investigate how this quantity correlates with the dislocation density. To this end, we divide all samples with high dislocation density gradient into 10 slices and determine, for each slice, the dislocation density (see figures 1(d) and 4(a)) as well as the dislocation activity, averaged over short strain intervals. In addition, we determine the average dislocation activity, i.e. the plastic contribution to the imposed strain rate. Figure 4(b) shows the average dislocation activity as

Figure 4. (a) Evolution of dislocation density profile for a sample with high strain gradient, axial dislocation density profiles as in figure 1(d) for different total strains. (b) Correlation between dislocation density ρ and dislocation activity η as expressed by Pearson correlation coefficient $r_{\rho,\eta}$, as function of total strain (red data points); density and activity were determined for 10 layers along the specimen axis and averaged over all specimen with high dislocation density gradient; the average dislocation activity is shown in comparison (blue data points).

well as the Pearson correlation coefficient

$$r_{\rho\eta} = \frac{\sum_{i,s} \rho_{i,s} \eta_{i,s} - \bar{\rho} \bar{\eta}}{\sigma_{\rho} \sigma_{\eta}}, \quad (4)$$

where $\rho_{i,s}$ and $\eta_{i,s}$ are the dislocation density and activity found, at a given strain, in slice i of sample s , and $\bar{\rho}$, $\bar{\eta}$, σ_{ρ} and σ_{η} are the respective mean values and standard deviations.

Initially, during the quasi-elastic regime, the dislocation activity is low as most of the strain increase is of elastic nature. In this regime, we observe a clear positive correlation between dislocation activity and dislocation density—in other words, regions of high dislocation density produce a higher dislocation strain (they are softer) whereas regions of low dislocation density tend to produce less dislocation strain (they are harder). This picture is reversed once the samples make, at a strain of about 0.12%, the transition towards plastic flow as indicated by a sharp increase of the overall dislocation activity towards values close to $\eta = 1$ —from this point onwards, most of the imposed strain is accommodated by dislocation motions. At the same time, we now see a strong negative correlation between dislocation density and dislocation activity as the transition towards plastic flow occurs first in the regions with the lowest dislocation density: these regions are now soft, whereas the dislocation dense regions are hard. Finally, at strains above 0.22% we see another change in sign of r as the dislocation dense regions catch up until, finally, dislocation activity homogenizes and further deformation proceeds in a compatible manner.

Loading–unloading tests reveal that, at all strains, a certain fraction of the dislocation related strain reverts upon unloading. Figure 5 shows curves of dislocation-related strain versus stress and of dislocation density versus stress during loading followed by unloading from different end strains.

Figure 5. Left: stress versus strain and dislocation density versus strain curves during loading- unloading cycles (strain rate reversal at strains of 0.05%, 0.1% and 0.2%); right: evolution of dislocation related strain versus stress during loading and unloading, inset: recoverable (inversive) strain versus stress during unloading.

It can be seen from the figure that there is a significant dislocation related strain even during the initial, quasi-elastic regime of the stress-strain curve. To a large extent, this strain reverts upon unloading. A recoverable fraction of the dislocation-related strain is even present in the plastic regime, and indeed the slopes of the dislocation strain versus stress curves during unloading do not depend on the degree of pre-strain (inset in figure 5, right). Remarkably, also the dislocation density increase that occurs during straining is to a large extent reversible (figure 5, left), indicating that such increase is due to the geometrically reversible bowing out of dislocations: the recoverable strain is associated with a recoverable change in dislocation density. At the same time, certain dislocation motions do not revert. Initially, these are associated with dislocations bowing out from near-surface sources. Such dislocations annihilate at the surfaces of the simulated micropillars, which leads to a slight reduction of the dislocation density after a loading-unloading cycle and to a small amount of non-recoverable strain even at low stresses. This non-recoverable strain and concomitant dislocation density reduction increase with increasing overall strain as more dislocations interact with the surface (figure 5, red and blue curves). In the plastic regime, finally, the dislocation related strain is mostly non-recoverable as most moving dislocations leave the surface. Owing to the surface dominated nature of the dislocation dynamics in the simulated micropillars, this strain is not accompanied by any significant increase in dislocation density. In the next section we transfer these observations into a quantitative dislocation based model.

4. Analysis and dislocation density-based modeling

4.1. Quasi-elastic deformation regime

To understand the observed behavior, we first consider the initial regime of quasi-elastic deformation. In this regime, the total strain consists of an elastic contribution, plus a strain that is due to displacement of dislocations out of their configurations of minimum energy (i.e. the bowing out of source segments). Since this displacement reverts upon unloading, we cannot call this a plastic strain. On the other hand, because the corresponding motions are associated with energy dissipation, we can also not call them reversible in the thermodynamic sense. We therefore adopt a proposal of Zaiser and Seeger [23] and use the neologism ‘inversive’ for this type of geometrically reversible, but thermodynamically irreversible

dislocation motion. Accordingly, the strain caused by inversive dislocation motions is denoted as the inversive strain ε_{inv} , and the transition from the initial quasi-elastic to the subsequent plastic deformation regime is understood as a transition from pre-dominantly inversive to pre-dominantly non-inversive dislocation behavior.

To model the behavior of the dislocation system in the inversive regime, we first consider the paradigmatic case of widely spaced Frank–Read sources that operate independently. We use a line tension approximation where we envisage the pinned source segments as elastic lines of line tension $T = Kb^2 \ln(l_{\text{FR}}/b)/(4\pi)$ where K depends on line direction. For Cu, DeWit and Koehler [24] give an approximately sinusoidal dependency of K on the orientation angle θ with average value $K = 59.3$ GPa. The shape of the bowed out segment is then an elliptical segment (oblate for screw and prolate for edge orientations). We average over all orientations to obtain an average line tension $T = \beta\mu b^2$ where $\beta \approx 0.5$ depends logarithmically on segment length. The average segment shape is circular. The radius of curvature of such a segment is $R = \beta\mu b/\tau$, and in the process of bending the segment sweeps the area

$$\Phi(\tau) = \frac{l_{\text{FR}}^2}{4u^2} [\arcsin(u) - u\sqrt{1-u^2}], \quad u = \frac{l_{\text{FR}}}{2R} = \frac{\tau}{\tau_c}. \quad (5)$$

Here $\tau_c = T/(bR_{\text{FR},c}) = 2\beta\mu b/l_{\text{FR}}$ is the critical stress for source activation when the radius of curvature reaches the critical value $l_{\text{FR}}/2$. For stresses up to 60% of τ_c , equation (5) can be well approximated by a linear stress dependency

$$\Phi(\tau) \approx \frac{2}{3} R^2 u^3 = \frac{1}{12} \frac{l_{\text{FR}}^3}{\beta\mu b} \tau. \quad (6)$$

The corresponding strain is inversive as long as the source does not pass its critical configuration. Assuming a system of non-interacting sources of volume density n_{FR} , of which a fraction f_A is located on active slip systems with non-zero Schmid factor M , the inversive strain is in the low-stress regime given by

$$\varepsilon_{\text{inv}}(\tau) = M\Phi(\tau)f_A b n_{\text{FR}} = \frac{M}{12} \frac{f_A n_{\text{FR}} l_{\text{FR}}^3}{\beta\mu} \tau. \quad (7)$$

It is useful to re-write this expression in terms of the axial stress and Young's modulus. We introduce the parameter $f_{\text{FR}} = l_{\text{FR}}^2 \rho$, which is proportional to the average number of dislocations threading the activation area of a FR source (the area which a dislocation must sweep before it reaches the saddle point configuration of an isolated source). We also note that for our initial configuration $\rho = n_{\text{FR}} l_{\text{FR}}$. With these relations, we find

$$\varepsilon_{\text{inv}}(\sigma) = \frac{f_A f_{\text{FR}} (1 + \nu) \sigma}{36\beta E}, \quad (8)$$

where we used that $M^2 = 1/6$. Equation (8) predicts a linear stress dependency of the inversive strain, consistent with the findings in our unloading tests (figure 5). The total strain in the quasi-elastic regime then follows as the sum of the inversive and elastic strains

$$\varepsilon = \varepsilon_{\text{el}} + \varepsilon_{\text{inv}} = \frac{\sigma}{E} \left[1 + \frac{f_A f_{\text{FR}} (1 + \nu)}{36\beta} \right], \quad (9)$$

From which the effective elastic modulus is deduced as $E_{\text{eff}} = E/[1 + f_A f_{\text{FR}} (1 + \nu)/36\beta]$. We thus expect the effective elastic modulus to be reduced by a factor that depends on dislocation density only weakly, through the dislocation density dependence of the coefficient $\beta \propto \ln[f_{\text{FR}}/(b\sqrt{\rho})]$.

While the above argument has been formulated for a system of independently operating FR sources, it can be also stated in much more general terms, exploiting the generic scale invariance of dislocation systems [25]. Consider a dislocation system of total dislocation density ρ and with additional geometrical parameters f_i , that is in static equilibrium under a shear stress τ , and let us assume that motions of dislocations during loading have caused an inversive strain $\varepsilon_{\text{inv}}(\tau, f_i)$ which reverts to zero upon unloading. Next, we consider a second system, which is identical with the first, but where all lengths have been changed by a factor λ . This system will be in equilibrium at stress τ/λ . The distances traveled by the dislocations will have changed by λ , and the dislocation density has changed to ρ/λ^2 . Hence, the inversive strain in the second system has the value $\varepsilon_{\text{inv}}(\tau, f_i)/\lambda$, provided the geometrical parameters are formulated in such a manner that they are invariant upon the described spatial rescaling. It follows that the ratio between stress and inversive strain does not depend on λ , and is therefore independent on dislocation density. In a situation where all strain is inversive, the relationship between stress and strain will therefore have the structure of equation (8) for *any* quasi-statically loaded dislocation arrangement. Of course, the above argument does not say anything regarding the question whether there actually exists a regime of linear proportionality between stress and inversive strain, or what is its extent—it only tells us that, if such a regime exists, it is dislocation density independent. The question of its existence needs to be demonstrated by looking at the simulations.

In quantitative terms, with $\beta = 0.5$, $f_A = 1/3$ and $f_{\text{FR}} = 12.3$, we get an effective modulus $E_{\text{eff}} = 0.77 E = 110 \text{ GPa}$ which is close to the values found in our simulation. For comparison, we refer to simulations reported in [26]. In those simulations, which use a different DDD code developed by D. Weygand, the dislocation density was as in our homogeneous case, but the source lengths were shorter ($f_{\text{FR}} = 3.8$) and the crystal orientation of the simulated Al samples was [100], hence $f_A = 2/3$. Accordingly, we find from equation (8) an effective modulus $E_{\text{eff}} = 0.845 E$, which compares well with the initial slope of the stress strain curves in those simulations, which amounts to $0.825 E$.

However, there are certain issues when applying the same argument to situations with a dislocation density gradient. If we calculate the average inelastic strain from equation (7), the resulting effective elastic modulus is independent of the presence or absence of a dislocation density gradient, in disagreement with the simulation results. Our systems are constructed in such a manner that the source length is constant, independent on dislocation density. Accordingly, $f_{\text{FR}} = l_{\text{FR}}^2 \rho$ is linearly proportional to density, and according to equation (8) one would expect a linear dependency between dislocation density and dislocation-induced inversive strain. Thus, there should be a Pearson correlation of 1 between dislocation density and dislocation-induced strain rate (dislocation activity) in the early stage of the stress strain curve. However, the actual correlation coefficients shown in figure 4, while positive, are significantly smaller. Thus, the assumption of independently polarized FR sources needs to be modified.

We can arrive at a better description by noting that we do NOT have isolated FR sources as assumed in the derivation of equation (8). In fact, a higher value of $f_{\text{FR}} = l_{\text{FR}}^2 \rho$ implies that more dislocations are threading the activation area of a FR source and thereby reduce its polarizability in comparison with an isolated source. We thus replace equation (7) by the expression

$$\varepsilon_{\text{inv}}(\sigma) = M \Phi_{\text{eff}}(\tau) f_A b n_{\text{FR}} = \frac{M f_A f_{\text{FR}}}{12 \mu} \tau P(f_{\text{FR}}), \quad (10)$$

and accordingly $E_{\text{eff}} = E/[1 + P(f_{\text{FR}}^2) f_A^2 f_{\text{FR}}^2 (1 + \nu)/36\beta]$. For treating the case of inhomogeneous dislocation arrangements, we expand the polarizability around the mean

Table 3. Effect of dislocation density gradient on effective elastic modulus.

	Value from simulation	Calculated value from equation (11)
No gradient	107.7 GPa	108.2 GPa
Low gradient	110.8 GPa	110.6 GPa
High gradient	112.4 GPa	112.9 GPa

value $\bar{f}_{\text{FR}} = \langle \rho \rangle l_{\text{FR}}^2$ to obtain $P(f_{\text{FR}}) \approx P_0 - P_1(f_{\text{FR}} - \bar{f}_{\text{FR}})$. Inserting this relation into the expression for the effective modulus, we find for an inhomogeneous dislocation arrangement:

$$\varepsilon_{\text{inv}}(\sigma) = \frac{f_A \bar{f}_{\text{FR}} (1 + \nu)}{36\beta} \frac{\sigma}{E} \left(1 - P_1 \bar{f}_{\text{FR}} \left(\frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 \right) \right), \quad (11)$$

where the angular brackets denote spatial averages. It is easy to see that, for a heterogeneous dislocation arrangement of given average density, the mean square density increases with increasing degree of heterogeneity in comparison with a homogeneous system. Accordingly, the inversive strain is reduced and the effective modulus increases. In quantitative terms, we can evaluate the effective elastic modulus of a graded dislocation system as

$$E_{\text{eff}} = E \left[1 + \frac{f_A \bar{f}_{\text{FR}} (1 + \nu)}{36\beta} \left(1 - P_1 \bar{f}_{\text{FR}} \left(\frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 \right) \right) \right]^{-1}. \quad (12)$$

With $P_1 = 0.0425$ we obtain the effective modulus values in table 3, which are in good agreement with the simulation data. We may thus conclude that the observation of stiffer behavior in samples with dislocation density gradient results from the fact that the polarizability of the dislocation system decreases when the ratio between the dislocation spacing and the spacing of pinning points is reduced.

4.2. Plastic deformation regime

The transition towards plastic flow occurs when the externally applied stress ‘tilts’ the elastic energy landscape to such an extent that dislocations cross barriers in the initial energy landscape, such that upon stress removal they relax into new configurations. In our terminology, this corresponds to a transition from inversive to non-inversive dislocation motion, and from a situation with conserved dislocation density to a situation where both dislocation multiplication and annihilation at the sample surface are present. In order to understand the observed deformation behavior, we thus need to formulate a model describing these processes. To this end, we use the recently formulated approach of continuum dislocation dynamics (CDD) in a version which accounts for dislocation loop generation by FR sources, dislocation line generation by loop expansion, and dislocation losses at the sample surface [27]. We use the most simple possible CDD version which considers the dislocations on a given slip system in terms of their scalar density ρ and loop density q and note that, because of symmetry, we can treat all four active slip systems in our simulations as symmetry equivalent. Balance equations for ρ and q which account for FR sources have been formulated in [27]. Generalization to include dislocation losses at surfaces is straightforward: Let A denote the cross-sectional area of the specimen volume with an active slip plane. The characteristic rate for a loop to leave the specimen through its surface is given by $v_L = v/\sqrt{A}$ leading to a loop loss rate $\partial_t q^{(-)} = -qv/\sqrt{A}$ and a corresponding dislocation density loss $\partial_t \rho^{(-)} = -\rho v/\sqrt{A}$. In our simulations, $A = 1.22$ and a simple estimate shows that for these

values and our dislocation densities, dislocation loss at surfaces prevails over losses due to mutual annihilation. Combining these relations with the generation rates due to loop emission and loop expansion as formulated in [27] leads to the simple evolution equations

$$\begin{aligned}\frac{\partial \rho}{\partial t} &= qv - \rho \frac{v}{\sqrt{A}}, \\ \frac{\partial q}{\partial t} &= \frac{4\pi v_{\text{FR}} \rho_{\text{FR}}}{l_{\text{FR}}^2} - q \frac{v}{\sqrt{A}}.\end{aligned}\quad (13)$$

The loop emission rate is controlled by the characteristic velocity v_{FR} of dislocations that are crossing the saddle point configuration of a FR source that is interacting with surrounding dislocations. In a bulk material, as discussed in [27], the velocity at the source is indirectly controlled by the velocity v at which dislocations are convected away from the source as these dislocations exert a back stress, which limits source operation. The same is not true in small samples as studied here, since in these samples the emitted dislocations together with their back stress readily disappear at the surface.

The dislocation velocity v is controlled by collective dislocation interactions, which we express in terms of a dislocation density dependent ‘Taylor stress’ $\tau_{\text{T}}(\rho) = \alpha \mu b \sqrt{\rho}$. The resulting velocity is given by

$$v = \frac{b}{B} \tau_{\text{eff}} = \frac{b}{B} (\tau - \alpha \mu b \sqrt{\rho}), \quad (14)$$

whereas the source velocity is additionally influenced by the Orowan stress arising from the need to bend the segment between the source pinning points. Assuming that both effects are additive, we have

$$v_{\text{FR}} = \frac{b}{B} \tau_{\text{eff,FR}} = \frac{b}{B} \left(\tau - \alpha \mu b \sqrt{\rho} - \frac{T}{bl_{\text{FR}}} \right). \quad (15)$$

We can now use this framework to look at the steady state which is reached after some straining when dislocation emission and surface losses mutually balance. In this case we can look at steady state solutions of equation (13). This gives us the following relations connecting, in the quasi-steady state, the plastic strain rate with the initial FR density, the density of gliding dislocation loops, and the velocities of FR source activation and subsequent glide:

$$\dot{\epsilon}^{\text{pl}} = M \eta \rho b v = M \eta \rho_{\text{FR}} b v_{\text{FR}} \frac{2\pi A}{l_{\text{FR}}^2}. \quad (16)$$

From this, we can infer the FR source velocity in our simulations. We obtain for the parameters in our simulations (homogeneous case) the value $v_{\text{FR}} = 0.048 \text{ m s}^{-1}$. The corresponding effective stress in the saddle-point configuration is negligibly small as compared to the Orowan stress and the Taylor stress. This allows us to set, to a good approximation, $\tau_{\text{eff,FR}} = 0$, $\tau_{\text{eff}} \approx T/(bl_{\text{FR}})$, from which the velocity and density of moving dislocation loops follow as $v = T/(Bl_{\text{FR}}) = 88.5 \text{ m s}^{-1}$ and $\rho = 5.4 \times 10^{11} \text{ m}^{-2}$ which is much less than the dislocation density associated with the source segments.

We can thus conclude, in agreement with the simulation results, that the increase of dislocation density due to the emission of loops is expected to be small, as only a small number of sources are active at any moment and the emitted loops travel only a short distance before disappearing at the surface. Accordingly, the dislocation density in the surface-dominated samples remains essentially at its initial level even in the plastic regime. This raises

the question why the initial flow stress differences due to a dislocation density gradient disappear in the course of deformation.

To answer this question we note that the flow stress is controlled by the stress needed to activate sources, whereas the rate-dependent contribution to the flow stress is small. We can therefore, for a region that is plastically activated, express the local flow stress by the approximate rate-independent relation

$$\tau_f = \alpha\mu b\sqrt{\rho} + \frac{T}{bl_{FR}} \approx \alpha\mu b\sqrt{\rho} + \frac{\beta\mu b}{l_{FR}}, \quad (17)$$

which for a homogeneous dislocation distribution gives us, with the parameters in table 1, an axial flow stress of $\sigma = \tau_f/M = 175$ MPa, in good agreement with the simulation data shown in figure 2.

To describe the situation of a gradient dependent dislocation distribution with average gradient $\rho' = \partial\rho/\partial x$, we note that in this case the local flow stress is given by

$$\tau_f(x) = \alpha\mu b\sqrt{\rho(x)} + \frac{T}{bl_{FR}} \approx \alpha\mu b\sqrt{\rho + \rho'(x - \bar{x})} + \frac{\beta\mu b}{l_{FR}}, \quad (18)$$

where $\bar{x}$ is the midpoint along the specimen axis. To evaluate the corresponding global flow stress, we resort to a variant of Mughrabi's composite model of heterogeneous dislocation density distributions [28]. We assume that the stress is composed of an external stress, which for the present geometry is constant throughout the sample and given by $\sigma_{\text{ext}} = E(\varepsilon - \varepsilon^{\text{pl}})$, where the angular brackets denote averages over the sample length, and an internal back stress

$$\sigma_{\text{int}}(x) = -\varphi_E E(\varepsilon^{\text{pl}}(x) - \langle \varepsilon^{\text{pl}} \rangle), \quad (19)$$

which counter-balances plastic strain heterogeneities. In the original composite model, the Eshelby-like factor φ_E is set to unity. This is equivalent to making an iso-strain assumption, according to which the total strain ε is constant throughout the sample. Such an assumption is warranted in bulk deformation, but clearly not correct in axial deformation of micropillar samples as considered here. We therefore relax this assumption by allowing for incomplete elastic compensation of plastic strain heterogeneities, setting $\varphi_E < 1$ to correctly reproduce the observed post-yield stress transients. In the plastic regime, we then require the following inequalities to hold:

$$\begin{aligned} \sigma_{\text{ext}} + \sigma_{\text{int}}(x) &= \frac{\tau_f(x)}{M}, & x \in L_p \\ \sigma_{\text{ext}} + \sigma_{\text{int}}(x) &= \frac{\hat{\tau}_f}{M}, & x \in L_e, \end{aligned} \quad (20)$$

where L_p denotes the plastically deforming part of the sample, L_e is the part of the sample that undergoes only elastic (or inversive) deformation, and the stress within the plastically inactive region is defined by setting $\sigma_{\text{ext}} + \sigma_{\text{int}}(\varepsilon^{\text{pl}} = 0) = \frac{\hat{\tau}_f}{M}$. These relations allow us to evaluate the mean (external) stress as

$$\sigma^{\text{ext}} = \frac{1}{MV} \left[\int_{V^{\text{pl}}} \tau_f(x) dx + \hat{\tau}_f V^{\text{el}} \right], \quad (21)$$

the local plastic strain within the active region as $\eta\varepsilon^{\text{pl}}(x) = \varepsilon - \tau_f(x)/(ME)$, the mean plastic strain as $\varepsilon^{\text{pl}}(x) = (\hat{\tau}_f/M - \sigma^{\text{ext}})/(\eta E)$, and the overall strain as $\varepsilon = \sigma^{\text{ext}}/E + \varepsilon^{\text{pl}}$. This leads to the stress-strain curves shown in figure 6 (left) which follow the qualitative characteristics seen in the simulations: with increasing dislocation density gradient we

Figure 6. Left: stress–strain curves computed with modified composite model without accounting for inersive strain contributions, center: stress–strain curves including inersive dislocation strain, right: simulated stress–strain curves; the error bars show the scatter between different simulations in the plastic regime (strain > 0.15 , standard deviation of stresses averaged over a moving window of strain $b\bar{\rho}^{-1/2}$).

observe an earlier transition from elastic to plastic behavior, with plasticity first initiating in the low-density regions. Stress re-distribution from regions of low dislocation density (low local flow stress) to regions of high density (high flow stress) then leads to a build-up of internal stresses and kinematic hardening that is the more pronounced the bigger the heterogeneity of the dislocation structure.

4.3. Elastic-plastic transition

Until now we have disregarded the inersive strain contribution in our discussion of the plastic deformation behavior. In case of a spatially uniform dislocation density this leads to the unrealistic prediction of a sharp transition between quasi-elastic and plastic regimes. This shortfall can be rectified by noting that the plastic strain corresponds to the motion of dislocations that have passed the critical saddle point configuration for source activation and then move to the surface, and therefore disregards the strain that has been produced by the motion of the same dislocations from their initial equilibrium to the saddle point configuration. For a general inhomogeneous dislocation arrangement, this strain can be evaluated as (see equations (5) and (7))

$$\epsilon_{\text{inv}}(\tau, \rho) = M\Phi_{\text{eff}}(\tau, \rho)b\frac{\eta_A\rho_{\text{FR}}}{l_{\text{FR}}}, \quad (22)$$

where now $\tau = M(\sigma_{\text{ext}} + \sigma_{\text{int}})$ and the swept area is evaluated according to

$$\Phi_{\text{eff}}(\tau, \rho) = P(\rho l_{\text{FR}}^2)\frac{l_{\text{FR}}^2}{4u^2}[\arcsin(u) - u\sqrt{1 - u^2}], \quad (23)$$

where now $u = \tau/\tau_{\text{f}}(\rho)$ if we are in the quasi-elastic regime, and $u = 1$ in the plastic regime. Evaluating the ensuing local inersive strains, integrating them over the specimen length, and adding the result to the elastic and plastic strain contributions gives the stress–strain curves shown in figure 6 (right). The curves are now in good agreement with the simulation findings. In particular, we observe even for a homogeneous dislocation arrangement a gradual transition from elastic to plastic behavior, which is associated with a divergence of the inersive polarizability: the stress derivative of the inersive strain, equations (22) and (23), diverges at the critical stress where the system passes its critical saddle point configuration. In

the stress–strain curves this implies that the stress approaches the flow stress with a horizontal tangent, but not with a discontinuity of slope.

5. Discussion and conclusions

We have studied the transition from quasi-elastic behavior to plastic flow in small samples containing either a homogeneous distribution of dislocations or a heterogeneous distribution with a dislocation density gradient. Dislocation motions and dislocation-associated strain are already present in the very first stage of deformation, where they are associated with geometrically reversible (‘inversive’) motions of dislocation segments. In this regime, samples with heterogeneous dislocation distribution exhibit a higher slope of the stress–strain curve. We could interpret this phenomenon in terms of an effective polarizability of the dislocation system that is a decreasing function of dislocation density. Also, we observe that in the inversive regime there exists a positive correlation between dislocation density and dislocation strain, i.e. the regions of highest dislocation density are most deformable.

At a critical stress (the flow stress), the polarizability of a homogeneous dislocation system diverges as the system passes over a saddle point and reaches a state of sustained plastic flow. Similar transitions from inversive to irreversibly flowing behavior have been discussed in a wide range of physical contexts, ranging from low-strain deformation of amorphous solids [29] over colloidal suspensions [30], and motion of dislocations under oscillatory stress [31] to vortices in superconductors under oscillating fields [32]. Here we have given for the first time expressions that relate the inversive strain and its diverging susceptibility to basic parameters of the dislocation system in a deforming crystal such as source length and dislocation density. The transition to plastic flow is associated with a significant increase in dislocation activity, which occurs first in those regions where the dislocation density is smallest (lowest local flow stress). Accordingly, the correlation between dislocation density and dislocation activity changes sign: now the regions of lowest dislocation density are most deformable, whereas regions of high dislocation density show comparatively low dislocation activity.

In the plastically flowing regime, we have found that the behavior of the dislocation system in a small sample with open boundaries is governed by the interplay of three factors: (i) activated dislocations (loops emitted from FR sources) rapidly reach the sample surfaces; as a consequence, the dislocation density does not significantly increase and the dislocation microstructure is dominated by the initial, pinned configurations. Accordingly, initially present dislocation density gradients do not decrease during plastic flow; (ii) in heterogeneous dislocation arrangements (density gradients) plastic flow initiates preferentially in the regions of lowest dislocation density, corresponding to lowest local flow stress. Accordingly, the samples with highest dislocation density gradient pass first into the plastic regime; (iii) during heterogeneous plastic deformation, internal stresses re-distribute stress from deformed to less deformed regions (‘back stress’ effect). This leads, in samples with dislocation density gradients, to a kinematic hardening stage which is more pronounced if dislocation density and hence flow stress gradients are larger. At the end of this stage, the sample deforms compatibly at a constant stress level, which equals the mean flow stress, in line with the prediction of Mughrabi’s composite model.

Of course, the distinction between a purely inversive and a plastic regime made in our model implies a strong idealization. In our simulations, dislocation motions occurring during the initial quasi-elastic regime of the stress–strain curve are pre-dominantly inversive, whereas the behavior during the later stages is pre-dominantly plastic. However, some non-

recoverable dislocation strain is present even at low stresses as near-dislocation sources interact with the surface, leading to dislocation losses. At the same time, even after prolonged deformation when the emission of dislocation loops from sources and the annihilation of these loops at the surface has produced a large non-recoverable strain, an inversive strain contribution is present.

In conclusion, we note that the simulation and model predictions compiled in this paper can be easily checked experimentally. Dislocation density gradients are a ubiquitous feature of the near-surface region of deformed samples [8, 9]. Near the surface of a tensile sample, the dislocation density gradient is oriented perpendicular to the loading axis whereas here, we have a parallel orientation, however, this difference is of minor importance as shown in [33] where both gradient-parallel and gradient-perpendicular orientations of the tensile axis were studied. Owing to general scaling relations, dislocation density gradients are bound to grow as the overall dislocation density increases in the course of strain hardening. It is therefore possible to prepare surfaces with different dislocation density gradients and then use FIB to produce micropillar samples for controlled testing. It will be an interesting task for future studies to use this technique to gain a clearer understanding of the deformation properties of samples with heterogeneous and graded dislocation structures. This is particularly desirable in view of the technological importance of processes such as shot peening, which induce large dislocation density gradients in the near-surface region in order to enhance fatigue and wear resistance.

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