Grain Size Strengthening in Microcrystalline Copper: A Three-Dimensional Dislocation Dynamics Simulation

C. de Sansal[a,1], B. Devincre[b] and L. Kubin[c]

LEM, CNRS-ONERA, 29 Av. de la Division Leclerc, BP 72, 92322 Châtillon Cedex, France

[a]christophe.de-sansal@areva.com, [b]benoit.devincre@onera.fr, [c]ladislas.kubin@onera.fr

Keywords: Microcrystalline materials; Plasticity; Size effects; Hall-Petch relation; dislocation dynamics simulations.

Abstract. This article reports on a study of the microstructure and mechanical response of copper polycrystals with grain sizes in the micrometer range. Three-dimensional dislocation dynamics simulations are used for the first time to investigate grain boundary strengthening and the Hall-Petch law. The methodology, which involves constructing a microcrystalline representative volume element with periodic boundary conditions, is briefly presented. Simulation results show that the initial density of dislocation sources and the cross-slip mechanism are two key factors controlling the heterogeneity of plastic deformation within the grains. At yield, the smaller the grains size, the more plastic deformation is heterogeneously distributed between grains and homogeneously distributed inside the grains. A size effect is reproduced and it is shown that the Hall-Petch exponent decreases from the very beginning of plastic flow and may reach a stable value at strains larger than the conventional proof stress.

Introduction

The Hall-Petch Law. This law [1, 2] states that the yield stress of polycrystals depends on grain size through a relation of the form $\sigma = \sigma_o + k_{HP} d^{-1/2}$, where d is the grain size, σ_o is assumed to be a constant, although this may not be true for grain sizes in the micrometer range and below, and k_{HP} is a material constant. This relation is well verified experimentally for very large grains sizes and seems to hold down to nanometric grain sizes of the order of 10-20 nm [3, 4], with a large scatter and provided that the samples are carefully prepared. For even smaller grain sizes, molecular dynamics simulations (see e. g. [5]) show that Hall-Petch strengthening breaks down because plastic deformation is carried out by complex grain boundary sliding events. Experimental studies on coarse-grained samples [6], as well as microcrystalline ones [7], show that the Hall-Petch law is also strain-dependent. It follows that the constant k_{HP} should depend on strain.

Micrometric Grains. For grain sizes at or below the micrometer range, the situation is a bit confused. Experimental observations and theoretical predictions suggest that some dislocation mechanisms may change. In particular, heterogeneous dislocation generation by grain boundary sources is clearly observed in nanocrystalline materials, whereas the classical Frank-Read source mechanism is observed for grain sizes larger than a few μm. This led to the belief that the domain of micrometric grain sizes is a critical one [8]. Whether these changes entail modifications in the Hall-Petch law is also not very clear.

Modeling. An historical perspective on the Hall-Petch law and its modeling can be found in the review article [9]. The earliest model is the well-known pile-up model [10, 11], which yields a size effect of the Hall-Petch type but does not account for its strain dependence. As was discussed by Friedel [12], the deformation of a polycrystal is rather heterogeneous in the early deformation stage that follows the yield stress, so that the far field of dislocation pile-ups should induce a progressive homogenization of plastic flow. However, no extended dislocation pile-ups were found by

---

[1] Now at: AREVA NP SAS/NEER-F, Tour AREVA BAL-1851B, 92084 Paris La Défense, France.

transmission electron microscopy in polycrystals of pure metals with coarse grain sizes. For this reason, alternative models were developed that do not involve dislocation pile-ups. In Ashby's model [13] the strain dependence of the size effect arises from a strain-dependent density of geometrically necessary dislocations that accommodates the strain incompatibilities between grains. An equivalent but simpler formulation was proposed by several authors [14-16], in which dislocations loops are stored at grain boundaries after having sheared a grain. This storage induces an increase of the flow stress through the Taylor relation between stress and dislocation density. This type of model was further incorporated into the storage-recovery framework developed by Kock and Mecking [17].

Mesoscale Simulations. Dislocation dynamics (DD) simulations are adequate tools for investigating the relation between microstructure and the mechanical response of polycrystals. In the two-dimensional (2D) simulations carried out by Biner and Morris [18], the dislocation sources emit dislocation pile-ups, whereas in the 2.5D simulations carried out by Lefevbre, Devincre and Hoc [19] on micrometric and sub-micrometric grains, a transition is observed between dislocation pile-ups and a rather uniform storage of dislocation loops at grain boundaries. In parallel, the Hall-Petch relation does not seem to be modified. Balint et al. [20] further confirmed the occurrence of this transition in another 2D simulation. In the present work, 3D DD simulations are employed for the first time with the objective of obtaining a more realistic insight into dislocation mechanisms and the size effect in this critical domain of micrometric grains. In what follows, the methodology for constructing and deforming a model microcrystalline copper sample is first outlined. The main results are then presented and discussed with emphasis on the mechanisms by which plastic flow is homogenized, specifically multiplication and cross-slip.

Simulation Method

The Model Polycrystal. In a preliminary step, it is necessary to attribute to the original grains an adequate shape that paves the three-dimensional space. For this purpose, the grain shape selected is a tetrakaidecahedron, that is, a truncated octahedron (Figs. 1–*a* and 1–*b*). Each grain has then 14 nearest-neighbors, which is consistent with experimental observations. The simulated polycrystal contains 16 grains with randomly selected orientations, such that the Taylor factor is $M = 3.05$ as in face-centered cubic polycrystals with a random texture. The simulation cell is duplicated by periodic boundary conditions to allow simulating specimens without free boundaries. Five grain sizes are selected, d = 0.935, 1.25, 1.56 and 1.875 μm. Polycrystalline copper is assumed to be elastically isotropic with a shear modulus $\mu = 42$ GPa and a Poisson's ratio $\nu = 0.33$.

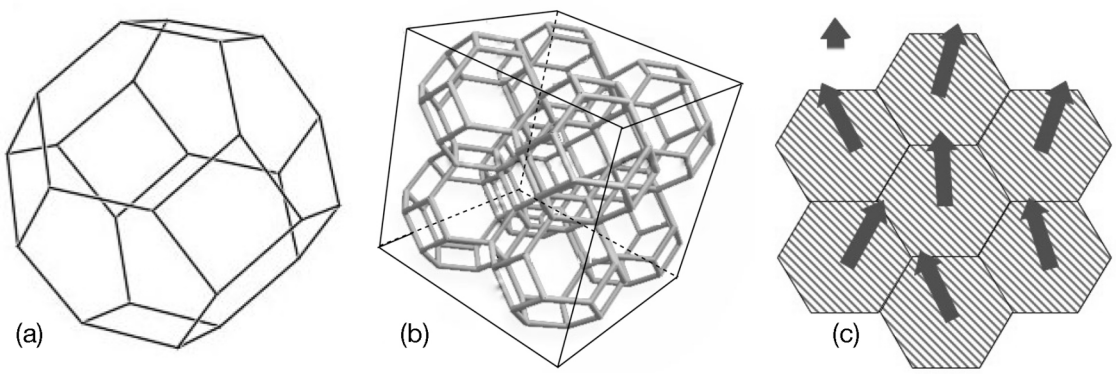

Figure 1. The simulated polycrystal and the applied stress. (a) – A tetrakaidecahedron with 14 facets representing a single grain. (b) – A cubic simulation volume containing 16 grains. Nine grains are entirely included in this volume. In addition, the periodic boundaries split three grains in two parts, three grains in four parts and one grain in eight parts. (c) – The externally applied stress (short vertical arrow) applied to the misoriented grains of the real polycrystal is replaced by an equivalent misoriented stress on grains with same crystallographic orientation.

Treating the deformation of misoriented grains under the effect of an externally applied stress would entail substantial modifications to the code used in the present work. For this reason an alternative procedure is used. It consists of simulating an equivalent situation in which all grains have same orientation and are submitted to suitably chosen misoriented stresses (Fig. 1–*c*). A numerical estimate of the deformations and lattice rotations shows that they are identical in the two types of configurations as a consequence of the small strain range investigated here.

A last question to solve is that of the compatibility of deformations at grains boundaries. Actually, no specific procedure has to be devised for this purpose. On the one hand, dislocations are by definition compatible defects and their fields ensure the compatibility of plastic strains across grain boundaries. On the other hand, there are no incompatibilities of elastic origin in elastically isotropic polycrystals.

The DD Simulation. The model polycrystal shown in Fig. 1–*c* is treated using a 3D DD simulation for the single crystal, of which the basic principles are not recalled here (see [22] and references therein). Three modifications have then to be implemented, which are detailed in [21].

i)- In agreement with experimental observations, the grain boundaries are considered as impenetrable obstacles for the dislocations.

ii) – The initial configurations, which usually consist of Frank-Read sources, are reconsidered. The critical stress for the operation of Frank-Read sources is proportional to $d^{-1/2}$. With decreasing sizes it increases faster than Hall-Petch strengthening, which implies the presence of a crossover typically around 0.2 μm. This cross-over is not experimentally observed, most probably because dislocations no longer multiply by usual source mechanisms for small grain sizes, as mentioned above. However, not enough is known about dislocation generation at grain boundaries to allow implementing this mechanism in DD simulations. The initial dislocation microstructure is then made up of randomly distributed sources, with one anchoring point inside the grain, within a prescribed distance from the grain center, and the other one at a grain boundary. This allows obtaining yield stresses in the range of the observed ones for micrometric grains. The effective density of sources is taken constant per unit area of grain boundary. Tests performed on single grains show that this density must be at least of 25 μm^{-2} in order to obtain a mechanical response that is not too much sensitive to the initial microstructure (cf. Fig. 3–*b*, below).

iii) – The implementation of the cross-slip mechanism was checked in order to ensure that it allows screw dislocations to escape the primary slip plane whenever they are involved in dislocation pile-ups. Figure 2 shows two examples of cross-slip processes in an isolated grain. In Fig. 2–*a*, primary dislocations pile-up at grain boundaries and double cross-slip of the screw dislocations occurs at the pile-up tips, inducing slip activity in the cross-slip plane. For a smaller grain size, double-cross slip produces new dislocation sources in the primary slip system. Thus, the cross-slip mechanism is effective in producing additional dislocation sources in the course of plastic deformation.

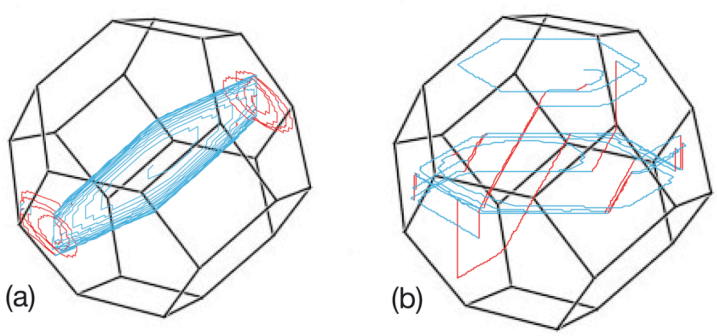

Figure 2. Cross-slip of screw dislocations produced by a source in a single grain (color online). (a) – d = 2.5 μm. The primary dislocations (blue lines) pile-up on grain boundaries and the screw portions cross-slip (red lines). (b) – d = 0.935 μm, the double cross-slip of screw segments piled-up at grain boundaries induces the formation of new primary sources.

Finally, the simulations are conducted with loading axes on each grain that are equivalent to an uniaxial loading of the polycrystal with misoriented grains. The imposed strain rate depends on grain size and is either 35 s^{-1} or 70 s^{-1}.

Results

Microstructure and Grain Size Effect. In what follows, the discussion is mostly restricted to the behavior at the maximum simulated plastic strain, $\varepsilon_{max} = 10^{-3}$. Figure 3–*a* shows a typical 3D microstructure obtained at this maximum strain. All the grains start deforming in single slip and additional slip systems, up to five of them, are activated as deformation increases. In average, the more a grain initially deforms, the larger is the number of active slip systems at ε_{max}. At this last strain value, the deformation of the grains is rather heterogeneous and this heterogeneity increases with decreasing grain size. For the smallest grain size and at ε_{max}, the ratio of the largest grain deformation to the smaller one is about eight. In contrast, the plastic deformation inside the grains becomes more and more uniform as the grain size decreases. This last effect is due to the increasing number of sources that are activated under an increasing stress and to the production of additional sources by a more active cross-slip mechanism. At the largest grain sizes, a few pile-ups relaxed by cross-slip and containing 3-4 dislocation loops are observed. No pile ups are, however, observed for the smallest grain sizes.

The deformation of the microcrystalline samples is sufficiently homogeneous at ε_{max} to allow fitting the stress vs. strain curves with a deformation curve of the Voce-type [17], which exhibits a strain hardening rate that linearly decreases with increasing stress (Fig. 3–*b*). This fit allows determining the Hall-Petch exponent with better accuracy by smoothening out fluctuations on the simulated curves.

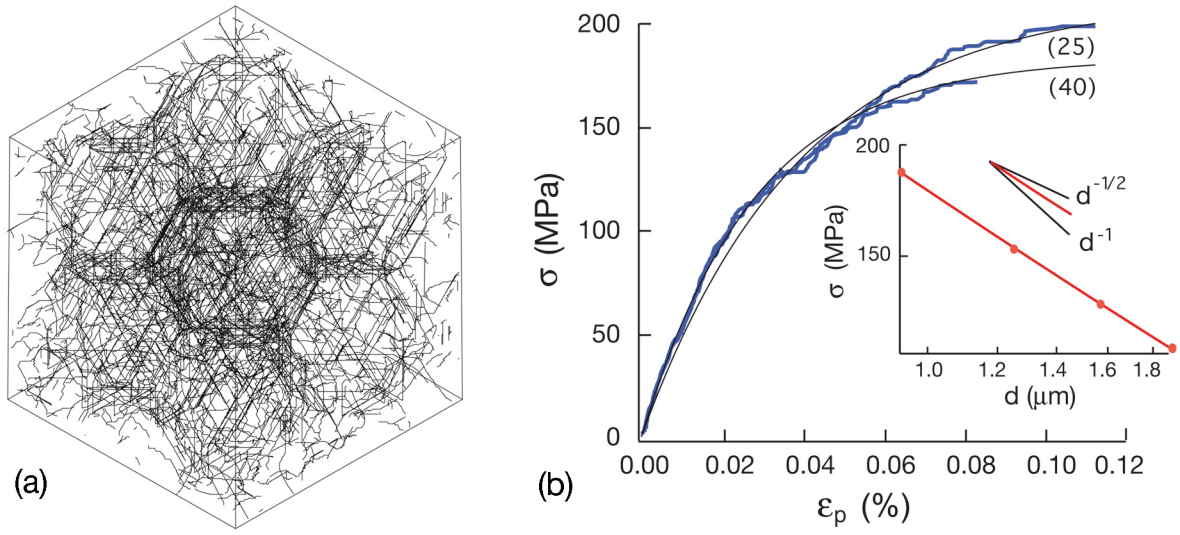

Figure 3. (a) – Three-dimensional dislocation microstructure in microcrystalline copper deformed up to a plastic strain of $\varepsilon_{max} = 0.1\%$. The grain size is 0.935 μm and the total dislocation density 1.5 10^{14} m^{-2}. Most of the dislocation density is stored at the grain boundaries. (b) – Stress vs. plastic strain curves for a periodic polycrystal with a grain size of 0.935 μm and two densities of sources, 25 and 40 per μm^{-2} (color online). The full lines are obtained by fitting these results to a stress-strain behavior of the Voce type. The insert shows a double logarithmic plot of the flow stress for the different grain sizes, at $\varepsilon_p = \varepsilon_{max} = 0.1\%$ and with 40 sources per μm^{-2}. The Hall-Petch exponent is intermediate between -1/2 and -1. With a lower density of sources or in the absence of cross-slip, it shifts to a value closer to -1.

The Hall-Petch exponent is -1 at yield and it continuously decreases with increasing strain. It also decreases with an increased number of initial sources and with increased cross-slip activity. As illustrated by Fig. 3–*b*, the expected value -1/2 is not yet reached at ε_{max}. This arises because the inter- and intragranular deformation is not fully homogenized. These simulations also show that the strain needed for obtaining a uniform deformation should increase with decreasing grain sizes.

Modeling. As was done for 2D simulations [19], the rate of storage of dislocations loops at the grain boundaries can be deduced from a simple argument and checked with respect to the simulations output. We consider a stored dislocation loop in a cubic grain of side *d*. It has a perimeter 4*d*, which corresponds to a stored density $\delta\rho = 4d/V$ in a polycrystal of volume *V*. The area swept by this loop when expanding through the whole grain is d^2, which corresponds to a shear strain $\delta\gamma = bd^2/V$. Assuming that the total dislocation density is essentially made up of identical stored loops, the total storage rate is of the form:

$$\delta\rho/\delta\gamma = k_s/bd, \tag{1}$$

with $k_s \approx 4$. The storage rate is inversely proportional to the grain size, which, in the present case, determines the mean free path of the mobile dislocations. Such a scaling was predicted by several authors [14, 16, 17]. Figure 4 shows that the simulated evolution of the total density with strain in the deformation follows this prediction with $k_s = 4.6$.

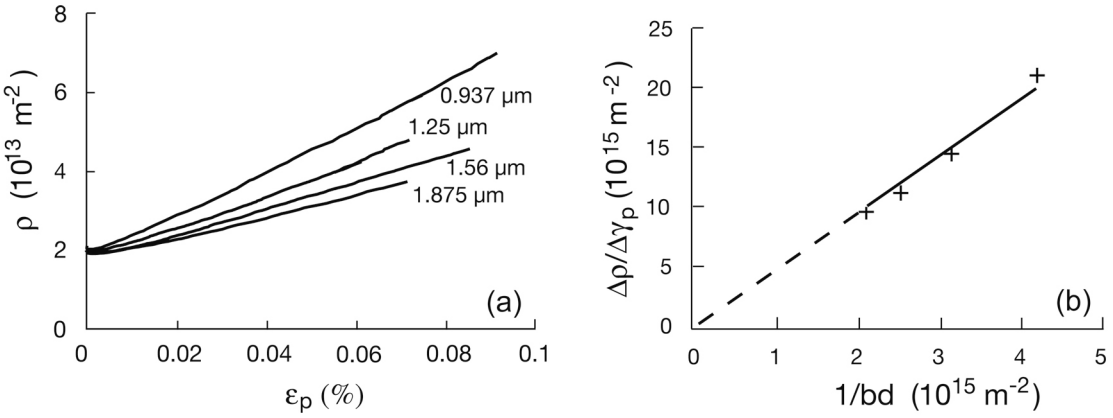

Figure 4. The dislocation storage rate as a function of grain size as derived from the DD simulations. (a) – Total density vs. axial strain in the simulated polycrystals for the different grain sizes. (b) – Total storage rate as a function of the inverse of the grain size times the Burgers vector.

By combining the integrated stored density $\rho = k_s\gamma/bd$ with the Taylor relation, $\tau = \alpha\mu b\rho^{1/2}$, one easily obtains a strain dependent Hall-Petch relation of the form:

$$\tau = \tau_o + \mu b(k_s\gamma/bd)^{1/2}, \tag{2}$$

where τ_o and ρ_o are the initial stress and density, respectively. This expression can be rewritten in terms of axial stress, σ, and strain, ε, with the help of the Taylor factor *M*. One eventually obtains:

$$\sigma = \sigma_o + k_{HP}d^{-1/2}, \tag{3}$$

with:

$$k_{HP} = M\alpha\mu(4.6bM\varepsilon). \tag{4}$$

Equations (1) to (4) are obtained within the framework of a storage model [17]. Only one type of obstacle is accounted for through a characteristic mean free path, the grain boundaries. The domain of application of such models can be extended by adding other obstacles. For instance, with grain sizes larger than a few microns, dislocations interactions inside the grains also contribute to the storage rate. Therefore, introducing dislocation mean free paths for interactions between several active slip systems and the related storage rate [22, 23] allows extending the domain of validity of present model up to coarse-grained polycrystals.

However, the output of the present simulations yields an unrealistically large value for the coefficient α. For instance, $\alpha \approx 1.2$ at the maximum strain instead of about 0.35. The reason is that for the small strains investigated here, some grains are still poorly deforming plastically, in contrast to what is implicitly assumed by the Taylor relation. For a usual proof stress value of 0.2%, and assuming uniform deformation (α = 0.35), one has $k_{HP} \approx 0.115$ MPa.m^{-2}, whereas $k_{HP} \approx 0.158$ MPa.m^{-2} according to experiment. To reach this last value, the proof stress must be of the order of 0.38%, which gives an indication of the strain needed to reach homogeneous deformation. Thus, Eq. (3) cannot describe the low strain behavior, which is governed by source activation and this early deformation stage has to be modeled separately.

Grain Rotations. Finally, the rotations of individual grains during plastic flow were investigated. They are plotted in Fig. 5 for the largest grain size and can be compared with the prediction of the two traditional models by Sachs (Fig. 5–*a*) and Taylor (Fig. 5–*b*). As illustrated by this figure, the grain rotations are globally determined by the activity of the slip system with highest Schmid factor. This explains why many grains behave as if they were deforming in single slip conditions with their orientation evolving toward the $[1\,\bar{1}\,1]-[100]$ zone axis. This feature would be in agreement with the Sachs model, but deviations are clearly observed, for instance toward the [100] orientation. This last aspect, which is consistent with the Taylor model, illustrates the influence of plastic strain incompatibilities, even for strain as small as 0.1%. As a general trend, it is observed that the more the polycrystal is deformed, the more the simulation results tend to be in agreement with the predictions of the Taylor model. Total agreement is, however, never found with either the Sachs or the Taylor model and each of these seems approximately consistent with the present results in different areas of the stereographic plot. In spite of the small plastic strains and the small grain sizes, this last result is qualitatively similar to observations made by Winther [24] on coarse-grained copper polycrystals deformed up to 6%.

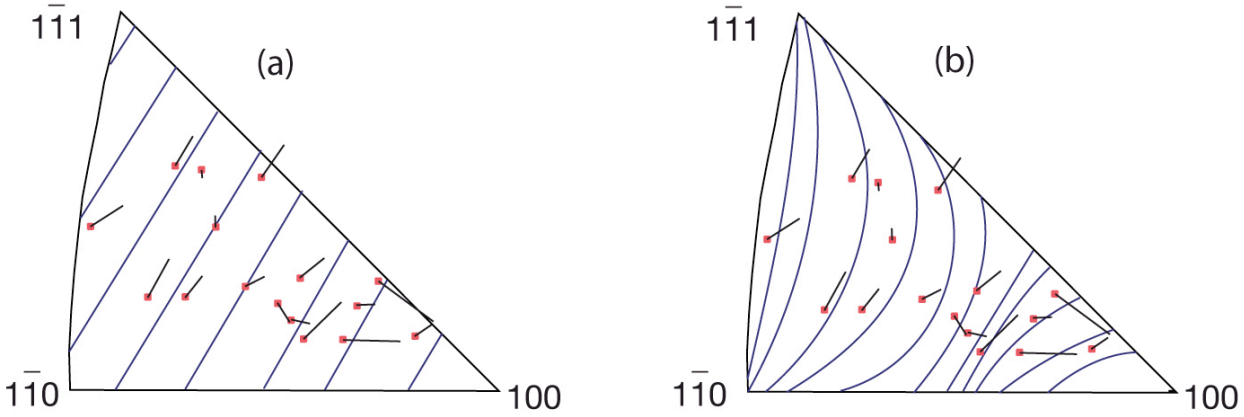

Figure 5. Grain rotations in the simulated polycrystal at ε_{max} (color online). The grain size is d = 1.875 mm. For each grain, a small filled symbol marks the initial position in the stereographic plot and the evolution of the orientation is amplified by a factor of 500. (a) – Comparison with the prediction of the Sachs Model (long straight blue lines). (b) – Comparison with the prediction of the Taylor Model (long curved blue lines).

Concluding Remarks

The 3D DD simulations of microcrystalline copper carried out in this work show that combining a periodic assembly of space-filling polyhedra with the assumption that grain boundaries are impenetrable obstacles for dislocations provides a reasonable base for investigating grain boundary strengthening. In addition, the number of grains, sixteen in the present case, does not need to be extremely large to obtain a reasonably realistic size effect.

The microstructures developed in the grains are not compatible with the prediction of the pile-up model. The disappearance of dislocation pile-ups for grain sizes of about 1 μm confirms a general expectation as well as the 2D simulation results by Lefebvre et al. [19]. The stored density increases like d^{-1}, in agreement with a dislocation mean free path limited by the grain size. This shows that the compatibility stresses between grains increase with decreasing grain size. As a result, the cross-slip probability increases too and promotes the homogenization of strains inside the grains. In parallel, and for a given axial strain, the intergranular deformation becomes increasingly heterogeneous for small grain sizes. As a consequence, the Taylor relation yields a too large value for the coefficient α as long as the deformation is not sufficiently large. In the present case, the exponent of the Hall-Petch relation has not yet stabilized to its expected value at the largest strain that could be reached, $\varepsilon_{\max} = 0.1\%$.

Dislocation multiplication via a fixed initial density of sources or by double cross-slip, plays a major role in the homogenization of the deformation, and hence on the transition between an initial regime governed by source operation and a regime controlled by dislocation storage. For decreasing grain sizes, a larger dislocation density is, indeed, needed to reach this transition [25]. With decreasing strains, the grain size exponent increases and tends toward -1. This scaling is characteristic of the confinement of the source segments, in good agreement with a recently published compilation of experimental results [26].

Information about the generation of dislocations at grains boundaries can only be obtained at atomic scale and is presently lacking. It seems likely that this mechanism is operative at the smaller grain sizes investigated here. If it were so, the transition between the d^{-1} and $d^{-1/2}$ grain size exponents may occur at smaller stresses. Thus, some difficulties encountered in the present simulations are similar to those met upon attempting to interpret experimental data. What is also missing to obtain fully realistic 3D DD simulations is an increased computing power, which will hopefully become available in the coming years. This will allow checking more precisely the view according to which the Hall-Petch relation originates from dislocation storage at the grain boundaries, hence from the strain incompatibilities between the deforming grains.

Acknowledgement

One of the authors (C.de S.) gratefully acknowledges Prof. T. Hoc (Ecole Centrale Paris) for help and advice at some critical points of this work.

References

[1] E.O. Hall, Proc. Phys. Soc. London, B64, 747 (1951).

[2] N.J. Petch, J. Iron Steel Inst., 174, 25 (1953).

[3] P.G. Sanders, C.J. Youngdahl and J.R. Weertman, Mat. Sci. Eng. A234-236, 77 (1997).

[4] S. Cheng et al., Acta Mater., 53, 1521 (2005).

[5] J. Schiøtz and K.W. Jacobsen, Science, 302, 1357 (2003).

[6] A. Lasalmonie and J.-L. Strudel, J. Mat. Sci. 21, 1837 (1986).

[7] N. Hansen, Scripta Mater. 51, 801 (2004).

[8] K.S. Kumar, H. Van Swygenhoven and S. Suresh, Acta Mater., 51, 5743 (2003).

[9] H.-H. Fu, D.J. Benson and M.A. Meyers, Acta Mater. 49, 2657 (2001).

[10] J. Eshelby, F. Frank and F.R.N. Nabarro, Phil. Mag. 42, 351 (1951).

[11] J.C.M. Li and Y.T. Chou, Metall. Mater. Trans. B, 1, 1145 (1970).

[12] J. Friedel: *Dislocations*, Pergamon Press, Oxford (1964).

[13] M. Ashby, Phil. Mag., 21, 324 (1970).

[14] A.A. Johnson, Phil. Mag., 7, 177 (1962).

[15] U.F. Kocks, Metall. Trans. 1, 1121 (1970).

[16] J. Embury: in *Strengthening Methods in Crystals*, edited by A. Kelly and R.B. Nicholson, Applied Science Publishers, London, p. 331 (1971).

[17] U.F. Kocks and H. Mecking, Progr. Mater. Sci., 48, 171 (2003).

[18] S. Biner and J. Morris, Phil. Mag. 83, 3677 (2003).

[19] S. Lefebvre, B. Devincre and T. Hoc, J. Mech. Phys. Solids, 55, 788 (2007).

[20] D.S. Balint et al., Int. J. Plas. 24, 2149 (2008).

[21] C. de Sansal: *Plasticité et Effet de Taille dans les Polycristaux à Grains Micrométriques: Simulations Mésoscopiques et Modélisation*, PhD thesis, Ecole Centrale Paris.

[22] B. Devincre, T. Hoc and L. Kubin, Science, 320, 1745 (2008).

[23] L. Kubin, B. Devincre and T. Hoc., Acta Mater. (2004), in press.

[24] G. Winther, in: *Evolution of Deformation Structures in 3D*, edited by C. Gundlach et al., Risoe National Laboratory, Roskilde, Denmark, p. 211 (2004).

[25] G. Saada, Phil. Mag. 85, 3003, 2005.

[26] N. Ohno and D. Okumura, J. Mech. Phys. Solids, 55, 1879 (2007).