J Mater Sci (2010) 45:6166–6169
DOI 10.1007/s10853-010-4703-1

On the rate-controlling mechanism during the plastic deformation of nanocrystalline Cu

Hans Conrad · Di Yang

Received: 25 March 2010 / Accepted: 7 June 2010 / Published online: 22 June 2010

Abstract Data in the literature on the effect of grain size from micrometers to nanometers on the flow stress, dislocation density, apparent activation volume and strain rate sensitivity parameters for Cu are analyzed to determine the rate-controlling mechanism. Accord occurs for the intersection of dislocations over the entire grain size range from 10 nm to 150 μm. The dislocation density and its structure and the related Friedel factor are important parameters.

Introduction

Three grain size (GS) regimes have been identified in the effect of GS (d) on the plastic deformation flow stress (σ) of FCC metals at low homologous temperatures; see Fig. 1. Grain size hardening occurs in Regimes I and II, GS softening in Regime III. The transition from I to II ($d \approx 0.5$ μm) occurs when the predicted size of the dislocation cells which develop in Regime I becomes larger than the existing GS, the transition from II to III ($d \approx 10$ nm) when the dislocation elastic interaction spacing is larger than the GS. Because of the high stresses which occur at the II–III transition, a significant number of stacking faults and twins may develop when $d = 10$–50 nm [2, 3]. Important regarding the mechanical properties of nanocrystalline metals ($d = 10$–100 nm) is a knowledge of the mechanism(s) governing their plastic deformation kinetics, i.e., the effects of strain rate $\dot{\varepsilon}$ and temperature T on the flow stress σ.

It is generally considered that the intersection of dislocations is rate-controlling in Regime I. Two rate-controlling mechanisms, which are based on the stress concentration *due to the pile-up of dislocations* at the grain boundary, have been proposed for FCC metals with GS in Regime II: (a) generation of dislocations in the adjacent grain by cross-slip [4–6] and (b) grain boundary shear [7–9]. The finding which led to the latter mechanism was the observed "anomalous" increase in the apparent activation volume $v = kT\partial \ln \dot{\varepsilon}/\partial\sigma$ with decrease in temperature (and the corresponding increase in σ) in ultrafine-grained Cu [10–12]. However, as shown in [13] an anomalous increase in v can also occur if the rate-controlling mechanism in the submicron GS range is the intersection of dislocations in the grain interior. The objective of the present paper was therefore to examine further the idea that the rate-controlling mechanism in FCC metals with a submicron grain size (including nanocrystalline) is the intersection of dislocations. Cu was chosen for the study because pertinent data have become available for this metal.

Analysis and discussion

There exists considerable evidence that the rate-controlling mechanism at low homologous temperatures and modest strains and strain rates in FCC metals with GS in the micron range is the intersection of dislocations [14, 15]. The resolved shear strain rate for this mechanism is given by

$$\dot{\gamma} = \rho_m b^2 \nu_D \exp\left(-\frac{\Delta G(\tau)}{kT}\right) \quad (1)$$

where ρ_m is the mobile dislocation density, b the Burgers vector, ν_D the Debye frequency and $\Delta G(\tau)$ the Gibbs free

H. Conrad (✉) · D. Yang
Materials Science and Engineering Department, North Carolina State University, Raleigh, NC 27695-7907, USA
e-mail: hans_conrad@ncsu.edu

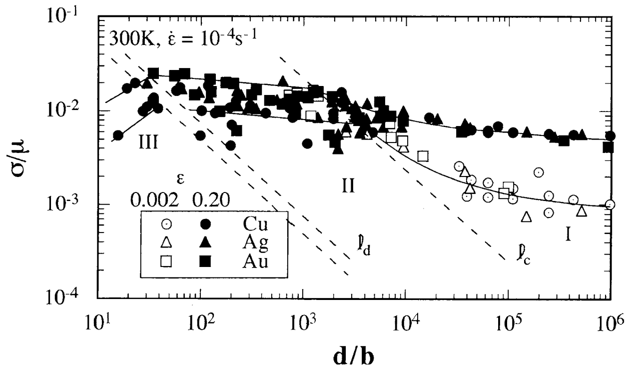

Fig. 1 Modulus-normalized flow stress σ/μ vs. the Burgers vector-normalized grain size *d*/*b* for the plastic deformation of Ag, Au, and Cu at 300 K. From Conrad and Jung [1]

energy, which is a decreasing function of the applied resolved shear stress τ. Neglecting the bowing of dislocations due to their line tension, the activation volume for the process is [16]

$$v^* = -\partial\Delta G/\partial\tau = l^* b x^* = kT\frac{\partial \ln\dot{\gamma}}{\partial\tau} = MkT\frac{\partial \ln\dot{\varepsilon}}{\partial\sigma} = M\,v \tag{2}$$

where l^* is the dislocation forest contact spacing along the gliding dislocations, x^* the thermally activated intersection distance, $\dot{\gamma}$ the resolved shear rate and $M \approx 3$ the Taylor orientation factor and $v = kT\partial \ln\dot{\varepsilon}/\partial\sigma$ the apparent activation volume. Further, in the micrometer GS range it has been established that the flow stress is proportional to the dislocation density according to [14, 17]

$$\tau = \alpha\mu b\rho^{1/2} = \sigma/M \tag{3}$$

where μ is the shear modulus, ρ the total dislocation density and $\alpha \approx 0.3$. The direct proportionality between τ and $\rho^{1/2}$ indicates that dislocations are responsible for both the long-range and short-range obstacles to dislocation motion.

In keeping with the mechanisms proposed for the effect GS on ρ, namely $\rho = \beta/bd$ [18–21], Fig. 2 presents a plot

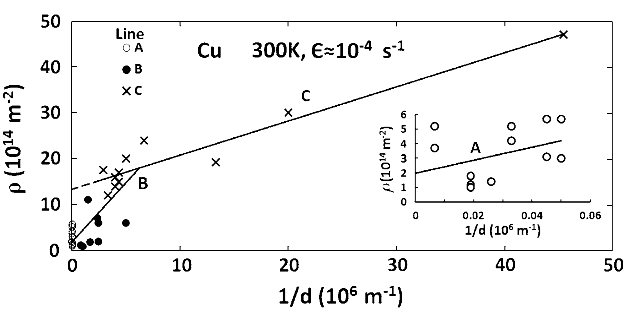

Fig. 2 The dislocation density ρ in Cu (strain $\varepsilon \approx 0.10$) vs. the reciprocal of the grain size d^{-1} showing three regions. *Open circle* region A, *filled circle* region B, and *times* region C. Data from [17, 22–32]

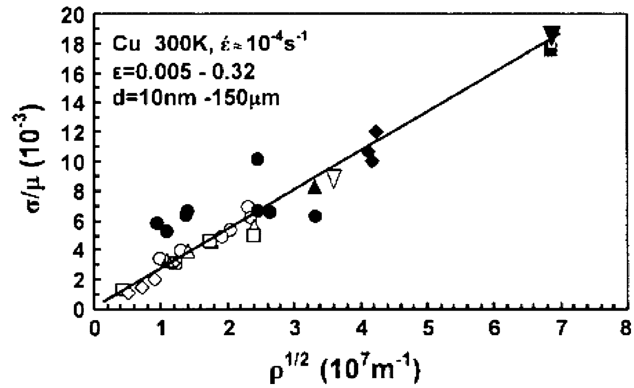

Fig. 3 Modulus-normalized flow stress of Cu with grain size from $d = 150$ µm down to 10 nm vs. the square root of the dislocation density. Open data points are for $d > 200$ nm, filled for $d < 200$ nm. Data from [17, 22–32]

of ρ vs. d^{-1}. Three regions (A, B, and C) are indicated. The slope of line A($1.0b^{-1}$) is in accord with the average slip distance mechanism [19, 20], that of line B($0.2b^{-1}$) with the geometric dislocations mechanism [21] and that of line C with the GB ledge mechanism [18]. To be noted is that ρ increases with decrease *d* in the submicron GS range as well as in the micron range.

The plot of σ/μ vs. $\rho^{1/2}$ in Fig. 3 includes data covering the entire GS range from 10 nm to 150 µm. The fit to a single line through the origin suggests that the same mechanism is rate-controlling in the submicron GS range as in the micron range. Moreover, the direct proportionality between σ/μ and $\rho^{1/2}$ (with slope giving $\alpha = 0.33$) is in accord with the intersection of dislocations as the rate-controlling mechanism for the entire GS range.

That a single mechanism is rate-controlling over the entire GS range comprising Regimes I and II is further indicated by the variation of the product $vm\rho^{1/2}$ as a function of grain size, where $m = \partial \ln\sigma/\ln\dot{\varepsilon}$ is the so-called "strain rate sensitivity parameter". Rearranging the equation for m and multiplying both sides by *kT* one obtains

$$\frac{kT}{m} = \sigma v \tag{4}$$

Further, substituting $M\alpha\mu b\rho^{1/2}$ for σ and normalizing *v* by b^3 one obtains

$$m(v/b^3)\rho^{1/2} = kT/M\alpha\mu b^4 \tag{5}$$

Equation 5 gives that at a constant *T* and $\dot{\varepsilon}$ the product $m(v/b^3)\rho^{1/2}$ should be independent of GS if the rate-controlling mechanism remains constant. A plot of this product vs. *d* for tests at 300 K is presented in Fig. 4. It is see that the product is relativity constant over the entire GS range from 10^1 to 10^4 nm. The line through the data points gives $kT/M\alpha\mu b^4 = 3.7 \times 10^7\ \mathrm{m}^{-1}$, which is in reasonable accord

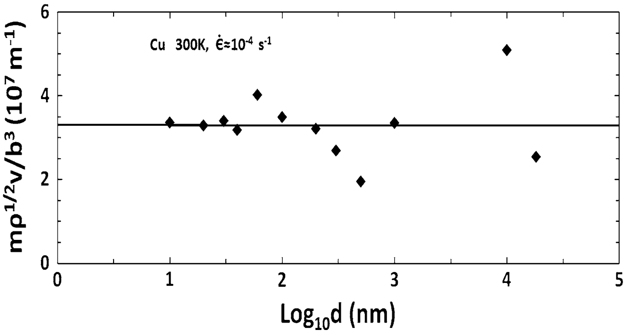

Fig. 4 The product $m\rho^{1/2}v/b^3$ vs. log *d* for the plastic deformation of Cu at 300 K. Data for *v* and *m* from [10–15, 33, 34]; those for ρ from Fig. 2

with the calculated value $2.3 \times 10^7\ \mathrm{m}^{-1}$ employing Eq. 5, in view of the fact the data were from a number of sources.

Friedel [35] has pointed out that the dislocation forest contact spacing *l** depends on the dislocation line tension and the thermal component of the applied stress. Hence, we will take

$$l^* = (\Phi\rho_f)^{-1/2} = [\Phi(3\rho/4)]^{-1/2} \quad (6)$$

where Φ is here termed the Friedel factor, ρ_f is the average dislocation forest spacing, and ρ the total dislocation density. The fraction ¾ arises from the existence of four non-parallel slip planes in FCC metals, one being the specific glide plane under consideration. Substituting Eq. 6 for *l** into Eq. 2 gives for the activation distance

$$\frac{x^*}{b} = \frac{3}{4}Mb\Phi^{1/2}(\upsilon/b^3)\rho^{1/2} \quad (7)$$

since ΔG and in turn x^* are constant for plastic deformation at a constant *T* and $\dot{\varepsilon}$, Eq. 7 gives for the variation of Φ with GS

$$\frac{\Phi_1}{\Phi_2} = \left\{\frac{((\upsilon/b^3)\rho^{1/2})_2}{((\upsilon/b^3)\rho^{1/2})_1}\right\}^2 \quad (8)$$

where the subscripts 1 and 2 refer to two respective GSs. Taking values of (v/b^3) as a function of *d* from the literature [10–15, 33, 34] and the corresponding value of ρ from Fig. 2, the ratio $\Phi_d/\Phi_{18\mu m}$ calculated using Eq. 8 is plotted vs. log *d* in Fig. 5. To be noted is that the ratio increases by two orders of magnitude with decrease in GS from 18 µm to 10 nm. Relatively little change in the ratio occurs between 18 µm and 0.5 µm, an order of magnitude jump at ~0.5 µm (transition between GS Regimes I and II), an irregularity between 500 and 100 nm, followed by a rapid increase between 100 and 10 nm.

For a *uniform* density of forest dislocations Friedel [35] calculated that

$$l^* = (2\Gamma l_f^2/\tau^* b)^{1/3} \quad (9)$$

where Γ is the dislocation line tension, l_f the average spacing of the forest dislocations and $\tau^* = \tau - \tau_\mu$ is the

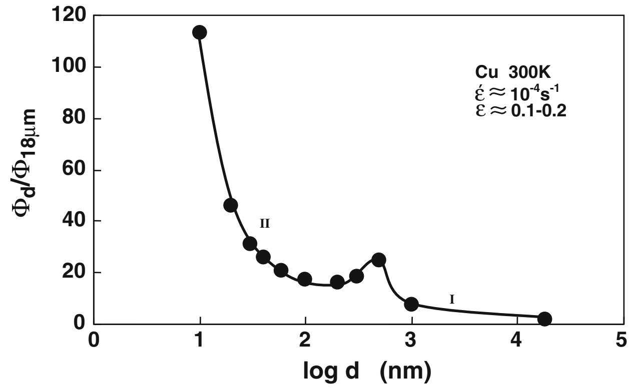

Fig. 5 The Friedel factor ratio $\Phi_d/\Phi_{18\mu m}$ vs. log *d* for the plastic deformation of Cu at 300 K

thermal component of the shear stress, τ_μ being the long-range thermal component. Taking $\Gamma = \mu b^2/2$, $l_f = \rho_f^{-1/2} = (a\rho)^{-1/2}$ and $\tau^* = c\tau = c\sigma/M = c\mu b\rho^{1/2}/M$ and inserting into Eq. 9 one obtains

$$l^* = (M/ac)^{1/3}\rho^{-1/2} = (\Phi_F\rho)^{-1/2} \quad (10)$$

where ρ is the total dislocation density, *a* and *c* are proportionality constants which can vary with dislocation density and structure and Φ_F is the corresponding Friedel factor. Taking $a = 3/4$ and $c = 1/3$ [15] and inserting these values into Eq. 10 gives $\Phi_F = 0.19$ for a uniform forest dislocation distribution. The dislocation structure which resembles that considered by Friedel (namely, a relatively uniform dislocation density) is that which occurs with $d = 100$–500 nm. Inserting $\Phi_F = 0.19$ and the corresponding values of v/b^3 and $\rho^{1/2}$ from the literature into Eq. 7 gives $x^*/b = 0.32$–0.38 for this grain size range, which value is reasonable for the intersection of dislocation in Cu [15].

Of further interest is the magnitude of the total force *f* required for the intersection process, which is given by

$$\frac{f}{\mu b^2} = \frac{\tau b l^*}{\mu b^2} = (M\varphi_F^{1/2})^{-1} \quad (11)$$

Taking $M = 3$ and $\Phi_F = 0.19$, Eq. 11 gives $f/\mu b^2 = 0.76$. The force can also be determined from the work ΔW performed by the applied stress during the intersection process, which is given by

$$\Delta W = fx^* = \sigma\upsilon = kT/m \quad (12)$$

Rearranging Eq. 12 gives

$$\frac{f}{\mu b^2} = \frac{kT}{mx^*\mu b^2} \quad (13)$$

Taking $m = 0.018$ (the average value for $d = 100$–500 nm) from the literature and $x^* = 0.35$, and inserting into Eq. 13 gives $f = 0.93$, which is in reasonable accord with that obtained using Eq. 11. Further taking $\tau^* = \tau/3$

for Cu deformed at 300 K [15], one obtains $\frac{f^*}{\mu b^2} = 0.28 - 0.31$, which along with the values of x^* are reasonable for the intersection of dislocations in Cu [14, 15, 35–37].

The above analysis gives that the intersection of dislocations is the rate-controlling mechanism in both GS Regimes I and II. There then exists the need to explain the anomalous increase in v which occurs with decrease in deformation temperature (or increase in σ) in Cu with GS in Regime II [10–12], compared to the decrease in v normally obtained for FCC metals with GS in the micron range [14, 15]. According to Eq. 2 an increase in v can result from an increase in either l^* or x^*, with the former being the more likely. According to Eq. 6 an increase in l^* will occur if either Φ or ρ decrease. The reduction in Φ with decrease in GS between 500 and 100 nm shown in Fig. 5 could contribute to the increase in the slope $dv/d\sigma$ with decrease in d reported for this GS range [12]. Further, a decrease in ρ with decrease in temperature can occur if the dislocation multiplication is a thermally activated process, such as for example by double cross-slip. Needed for further resolution of the question regarding the anomalous increase in v are observations and measurements on the dislocation density and arrangement (structure) in Cu with $d = 200$–500 nm.

Summary and conclusions

The existing plastic deformation kinetics data in the literature on Cu are in accord with the intersection of dislocations as the rate-controlling mechanism over the entire grain size range from micrometers down to 10 nm. It is expected this applies as well to other FCC metals. The increase in the dislocation density with decrease in GS and the related Friedel factor are important considerations. It is suggested that the anomalous increase in the apparent activation volume with decrease in temperature, or stress, observed in Cu specimens with a submicron grain size can result from either a reduction in the thermally generated dislocation density, or from a decrease in the Friedel factor. Observations and measurements of the dislocation density and structure in the submicron grain size regime are needed to validate this suggestion.

References

1. Conrad H, Jung K (2005) Mater Sci Eng A406:78
2. Shen YF, Lu L, Lu Q, Jin A, Lu K (2005) Scripta Mater 52:989
3. Lu L, Chen X, Huang, Lu K (2009) Science 323:607
4. Armstrong RW (1970) In: Herman H (ed) Adv Mater Res. Wiley, New York, p 101
5. Armstrong RW (1972) In: Vasu KI, Raman K, Sastry D, Prasad Y (eds) Defect structures in solids. Indian Institute of Science, Bangalore, p 306
6. Armstrong RW, Rodriguez P (2006) Phil Mag 86:5787
7. Conrad H (2003) Mater Sci Eng A341:216
8. Conrad H (2004) Metall Mater Trans A 35A:2618
9. Conrad H (2007) Nanotechnology 18:325701
10. Embury JD, Lahaie DJ (1993) In: Natasi M et al (eds) Mechanical properties and deformation behavior of materials having ultrafine microstructure. Kluwer-Academic, Dordrech, p 287
11. Conrad H, Yang Di (2002) J Electron Mater 33:304
12. Conrad H, Jung K (2005) Scripta Mater 53:581
13. Conrad H, Jung K (2006) J Mech Behav Mater 17:337
14. Conrad H (1965) In: Zackay VF (ed) High-strength materials. John Wiley, New York, p 436
15. Conrad H, Cao W-d (1996) In: Arsenault RJ et al (eds) The Johannes Weertman Symposium. TMS, Warrendale, PA, p 321
16. Conrad H, Meester B, Yin C, Doner M (1975) In: Mukerjee AK, Li JCM (eds) Rate processes in plastic deformation of materials. ASM Materials Park, OH, p 175
17. Doner M, Cheng H, Conrad H (1974) J Mech Phys Solids 22:555
18. Li JCM (1963) Trans TMS AIME 227:239
19. Conrad H (1963) In: Thomas G, Washburn J (eds) Electron microscopy and strength of crystals. Interscience Publication, New York, p 299
20. Conrad H (1970) In: Burke J, Reed MG, Weiss V (eds) Ultrafine-grain metals. Syracuse University Press, New York, p 213
21. Ashby MF (1970) Philos Mag 21:399
22. Gordon P (1955) Trans AIME 203:1043
23. Clareborough LM, Hargreaves ME, Lorretto MH (1958) Acta Metall 6:725
24. Bailey JE (1963) Philos Mag 8:223
25. Staker MR, Holt DL (1972) Acta Metall 20:569
26. Hanson N, Ralph B (1982) Acta Metall 30:411
27. Hommel M, Kraft O (2001) Acta Mater 49:3935
28. Ungár T, Oth S, Sanders P, Borbely A, Weertman JR (1998) Acta Mater 446:3693
29. Zheng Y, Tao N, Lu K (2008) Acta Mater 56:2429
30. Balogh L, Ungár T, Zhao Y, Zhu Y, Horita Z, Yu C, Langdon T (2008) Acta Mater 56:809
31. Brandstetter S, Derlet PM, Van Petegem S, Van Swygenhoven H (2008) Acta Mater 56:165
32. Li YS, Zhang Y, Tao N, Lu K (2009) Acta Mater 57:761
33. Carreker RP, Hibbard WR (1935) Acta Mater 1:656
34. Lu L, Dao M, Zhu T, Li J (2009) Scripta Mater 60:1062
35. Friedel J (1967) Dislocations. Pergamon Press, New York
36. Hirth JP, Lothe J (1968) Theory of dislocations. McGraw-Hill, New York
37. Hull D, Bacon O (1984) Introduction to dislocations, 3rd edn. Pergamon Press, New York