Hall-Petch analysis of dislocation pileups in thin material layers and in nanopolycrystals

Ronald W. Armstrong[a)]
Department of Mechanical Engineering, Center for Energetic Concepts Development, University of Maryland, College Park, Maryland 20742

(Received 26 October 2012; accepted 8 January 2013)

A potential order-of-magnitude increase in Hall-Petch (H-P)-based strength level for nanoscale grain-size structures is an important enabler of electronic thin film material design applications. Dislocation pileups of smaller lengths in such thin film materials are blocked in a screw orientation at the through-thickness grain boundaries of relatively larger grains. For fully nanopolycrystalline materials, both strength and strain rate sensitivity measurements exhibit complementary H-P reciprocal square root of grain size dependencies. An additional increase in strength level is predicted for transition from a pileup to a single dislocation loop expanding against the grain boundary obstacle. In opposition, disordered grain boundaries are responsible for a reduced H-P stress intensity, k_ε. And at the limiting high stresses reached at lower-limiting nanoscale grain sizes, reversed H-P dependences are obtained both for the strength and strain rate sensitivity.

I. INTRODUCTION

Pioneering work on dislocation pileups against thin (oxide) films began in 1959 as an extension of the Ph.D. thesis researches of Head at Bristol.[1] His work was further extended to a Hall-Petch (H-P) type of analysis for dislocation pileups within elastically anisotropic materials[2] and to catastrophic pile-up releases at obstacle breakthroughs.[3,4] An important concern in current researches is with pileups within the thin films themselves and, especially, within both larger and smaller nanoscale grain structures as well as within counterpart bulk materials. Already there is clear demonstration that the mechanical strength levels of isolated nanopolycrystalline thin layer or bulk materials developed for electronic or other design applications are approximately an order of magnitude higher than that for the same materials with conventional microstructures.[5]

II. DISLOCATION PILE-UP RESULTS/PREDICTIONS

Figure 1 shows adaptation to fracture mechanics crack geometries of model dislocation calculations reported for single-ended, double-ended and circular pileups.[6] In the figure, the effective shear stress acting on the pileups, τ, is divided by the shear modulus, G, and the pile-up length taken to equal a material grain diameter, ℓ, is divided either by the dislocation Burgers vector, b, or the dislocation core radius, r_0. The discrete dislocation character is revealed at small numbers, for example, providing the relation for relative shear strength decrease caused by addition of each dislocation to a single ended pileup of n dislocations of

$$\Delta\tau/\tau = -1/n \quad . \tag{1}$$

The cross-hatched area for each pile-up geometry in Fig. 1 covers the limiting H-P slope values, or stress intensities, that could be fitted to the theoretical dependencies; and, the linear dependencies defined inside the figure area are the fracture mechanics relations for edge crack, internal two-dimensional crack, and circular crack geometries. The top-end scale of the figure gives, with dislocation Burgers vector, b, or dislocation core radius, $r_0 = \sim 0.2$ nm and, say, $\ell/b = 25$, an indicated grain diameter of ~5 nm.

A. One or few grains through the thickness

Figure 2 provides a pioneering determination of H-P results for an Al-1% Si alloy film of 1 μm thickness tested in plane stress by means of a Beams-type bulge test.[7] Brotzen has provided a helpful review of the bulge test method and other test methods for thin film materials.[8] Attention is directed in the figure to the experimental scatter and otherwise indication of low values of the H-P stress intensity, for example, of the reported k_ε (slope) values equal to 0.13, 0.20, and 0.43 MPa $mm^{1/2}$ at increasing strain values. The k_ε values compare with a lowest value of $k_\varepsilon \geq \sim 1.3$ MPa $mm^{1/2}$ for relatively pure Al as shown for a number of results reflecting multicrystal and bulk aluminum (Al) behavior in Fig. 3.[9]

Comparison of the H-P results for the different materials shows that the effect of 1% silicon (Si) in the Al alloy of

[a)]Address all correspondence to this author.
e-mail: rona@umd.edu
DOI: 10.1557/jmr.2013.10

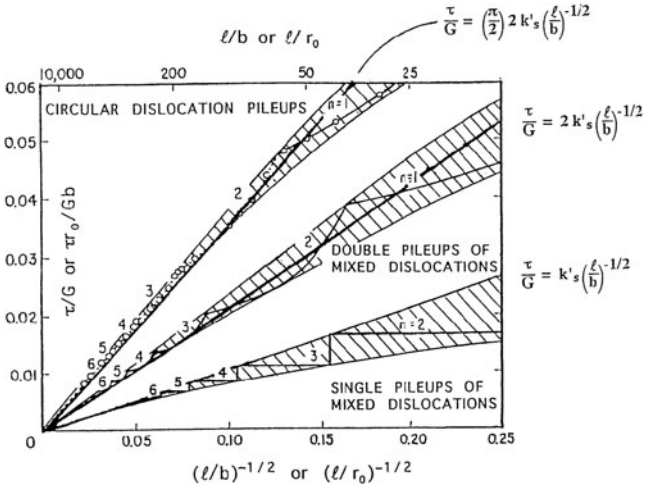

FIG. 1. Model dislocation pile-up calculations at small number of dislocations for single-ended, double-ended, and circular geometries and relation to fracture mechanics crack size-dependent predictions.[6]

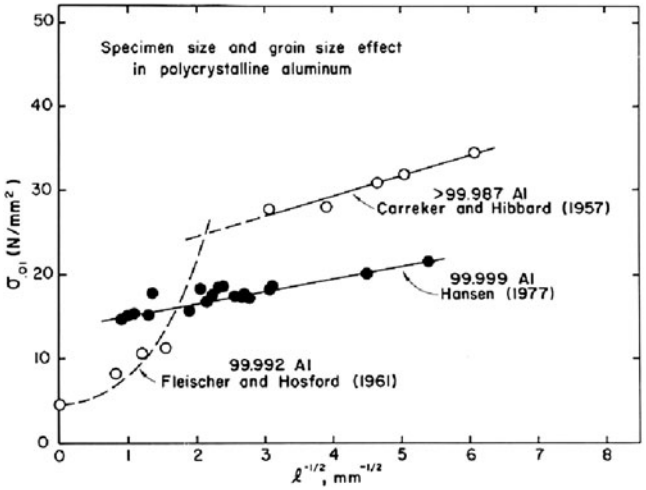

FIG. 3. Specimen size and Hall-Petch grain size dependences for several Al materials of different purities.[10–12]

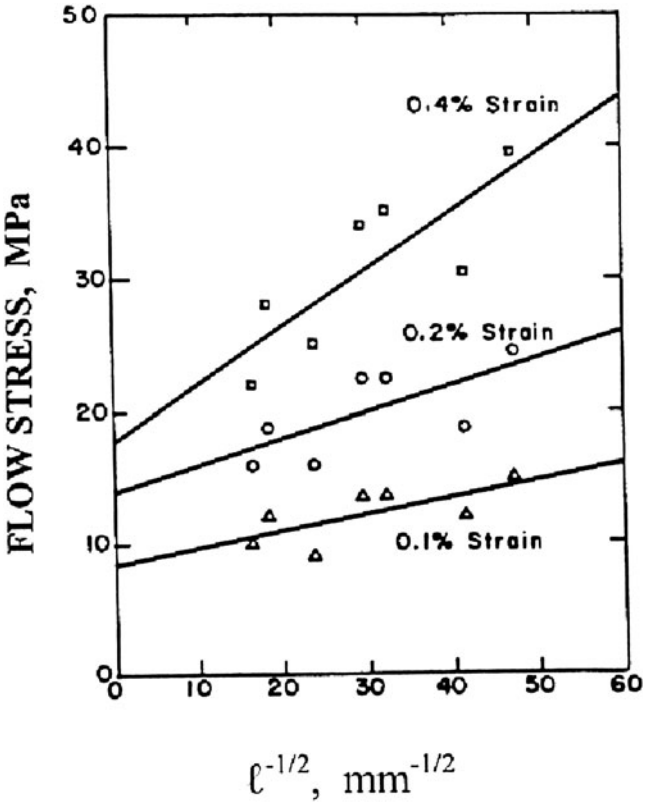

FIG. 2. Hall-Petch flow stress dependence on grain size for 1-μm thick Al-1% Si material as determined in a bulge test system.[7,8]

Fig. 2 has neither increased the experimental friction stress intercept, $\sigma_{0\varepsilon}$, nor k_ε in the H-P relation

$$\sigma_\varepsilon = \sigma_{0\varepsilon} + k_\varepsilon \ell^{-1/2} \quad . \qquad (2)$$

And there does not seem to be any evidence of transition to higher strength level for $\ell^{-1/2} \geq 31.6\ \mathrm{mm}^{-1/2}$ for grain sizes smaller than the film thickness, perhaps because sufficiently smaller grain sizes were not obtained (on the smaller grain size side of $\ell^{-1/2}$) to achieve a condition of true polycrystalline deformation behavior.

Under the plane stress condition for these tested films, there is no shear stress in the surface plane whereas plastic deformation proceeds through the film thickness with expectation of screw dislocation pileups at the through-thickness grain boundaries. Voskoboinikov[13] has reported recently on Head's problem of a screw dislocation near the interface of a coated solid. Perhaps for the results of Griffin et al.,[1,7] there is addition of a small restraining effect of the film surfaces as for Head's concern for an oxide film barrier? Nevertheless, the lesser constraint on grain deformations produces a condition somewhat analogous to the "grains per specimen cross-section" effect indicated in Fig. 3 for the few grains per cross-section shown for the Hosford and Fleischer[10] results as compared with the results reported by Hansen[11] who purposely went to larger specimen cross-sections for his larger grain size material.[12] Keller et al.[14] have reported analogous specimen size-dependent grain size results spanning the range of partially polycrystalline to fully polycrystalline nickel (Ni) materials. The lower k_ε values and greater scatter of experimental measurements in Fig. 2 are in line with concern for influence of a specimen size effect.

B. Micro- to nanopolycrystal H-P results

The strength properties of relatively thicker material layers that have been deposited have often been monitored by hardness testing, for example, as reported for reasonably deep layers of electrodeposited chromium (Cr) material[15] that was shown to exhibit a Hall-Petch-type dependence for diamond pyramid microhardness measurements as

$$H = H_0 + K_H \ell^{-1/2} \quad . \qquad (3)$$

More recently, the same type of nanoindentation hardness dependence has been reported for electrodeposited copper (Cu) films as shown in Fig. 4.[16] In this case, the relatively constant hardness of individual grains was measured at the indicated larger grain sizes and an indentation size effect (ISE) was also revealed. Normally for bulk material behavior, the value of k_H from Eq. (3) is given by $\sim 3k_\varepsilon$ and the value of $k_H = \sim 25$ MPa mm$^{1/2}$ for the Fig. 4 results compares reasonably favorably with a value of $3k_\varepsilon = \sim 15.0$ MPa mm$^{1/2}$ obtained for direct tensile flow stress measurements.[9] Other indentation results have been reported for determining a transition in hardness between layer and substrate, for example, as demonstrated for that of diamond-like carbon thin films and their 316L stainless steel substrates.[17]

Connection between the H-P results of conventional, fully polycrystalline and nanopolycrystalline material is usefully demonstrated on a log/log basis as shown in Fig. 5 for a compilation of selected Al, Cu, and Ni material results. For such logarithmic depiction made for a direct dependence of σ_ε on ℓ, the H-P relation of Eq. (2) gives a level (constant) value of $\sigma_{0\varepsilon}$ at large grain size and an asymptotic straight line dependence of slope $(-1/2)$ at small grain sizes. In Fig. 5, the conventional (open square) grain size results for Al are from Carreker and Hibbard[18] (shown in Fig. 3) as compared with the less pure (filled square), conventional and nanoscale results reported by Tsuji et al.[19] For Cu, the open circle results are from Hansen and Ralph[20] as compared with a compilation of individual filled circle results compiled by Lu et al.[21] For Ni, the indicated larger 0.14 strain, open triangle results are from Keller and Hug[22] whereas the closed triangle results are plotted as H/3 from Torrents et al.[23]

The indicated correlation of results in Fig. 5, with due account taken of their being compiled for different materials and test conditions, provides an important comparison of relative $\sigma_{0\varepsilon}$ and k_ε values for the designated face-centered cubic (fcc) metals. Such fcc metals and alloys had been relegated in importance in the past studies regarding the influence of grain size on determining the polycrystalline strength. And there are two reasons for such disregard: (i) the k_ε values are relatively low when compared with body-centered cubic (bcc) and hexagonal close-packed (hcp) metals; and (ii), as indicated for the Keller and Hug[22] Ni results in Fig. 5, both $\sigma_{0\varepsilon}$ and $d\sigma_{0\varepsilon}/d\varepsilon$ values are relatively high when compared with the magnitude of the $k_\varepsilon \ell^{-1/2}$ term. Broadly representative k_ε values in units of MPa mm$^{1/2}$ are $\sim$24 for steel and $\sim$10 for magnesium (Mg) as compared with $\sim$5 for Cu and Ni, and $\sim$1.3 for Al.[9]

The comparatively lower fcc k_ε values, particularly for Al, have been attributed to the requirement of cross-slip at grain boundaries as compared with strain hardening of the polycrystal grains by dislocation density-controlled build-up of line intersections occurring with increasing strain.[24] The raised $\sigma_{0\varepsilon}$ value for the Keller and Hug nickel results shown in Fig. 5 at $\varepsilon = 0.14$ shows the importance of such fcc strain hardening property. Nevertheless, H-P strengthening in fcc metals is being researched as important at nanoscale grain sizes and also it accounts for an approximate order-of-magnitude increase in strength level. Very interestingly, the results shown by the study of Tsuji et al.[19] for the relatively impure 99.3% Al material in Fig. 5 exhibit a relatively high value of $k_\varepsilon = \sim 4.8$ MPa mm$^{1/2}$ in the nanoscale grain size regime.

C. Transition to one dislocation loop

A natural question to ask about the dislocation pile-up explanation of the H-P relation is what happens as the

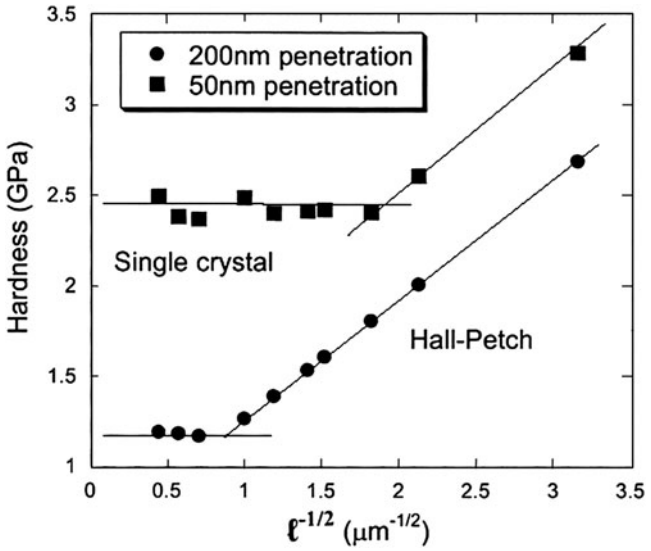

FIG. 4. Nanoindentation measurements for single grain and Hall-Petch hardness dependences.[16]

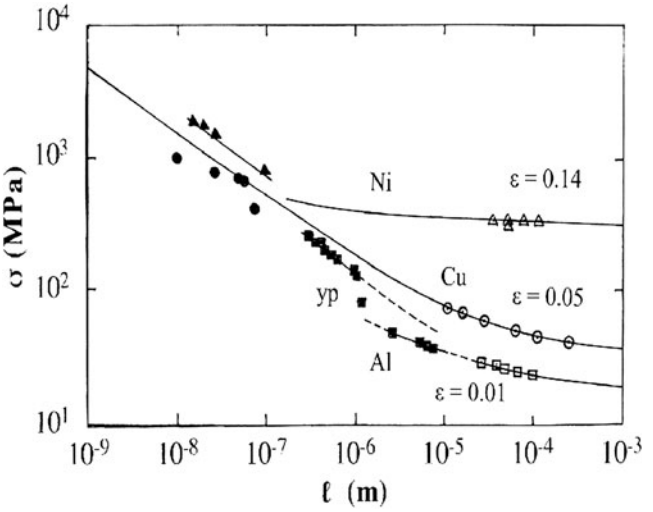

FIG. 5. Log/log type compilation of conventional and nanopolycrystalline Hall-Petch results for Al, Cu, and Ni materials.[12,14,18–23]

number of dislocations in a pileup is squeezed down to the lower limiting case indicated in Fig. 1 of one dislocation loop expanding against the grain boundary resistance. The answer is found by adding τ_C onto the equation for expansion of a single dislocation loop, for example, as[25]

$$\sigma = m[\tau_0 + (3Gb/4\pi\ell)\{(5/6)(\mathbf{ln}[4\ell/b] - 1) - 1/16\} + \tau_C] \quad . \tag{4}$$

In Eq.(4), m is the Taylor orientation factor, τ_0 is the multislip shear stress for deformation within the grain volumes, ℓ is the loop diameter taken equal to the grain diameter, and τ_C is the needed shear stress for penetration of the grain boundary obstacle.

In Eq. (4), the quantity, $m\tau_0 = \sigma_{0\varepsilon}$ in Eq. (2). The value of τ_C may be evaluated from the H-P model evaluation of k_ε that is given as

$$k_\varepsilon = m[\pi m^* Gb\tau_C/2\alpha]^{-1/2} \quad . \tag{5}$$

In Eq. (5), m* is a Sachs orientation factor and α is a factor (of ~0.8) for an average dislocation edge and screw character.[9] For Mg, τ_C had been shown to be controlled by the requirement of prism slip for which the temperature dependence of the resolved shear stress was measured in single crystal experiments.[26] The same type positive comparison was made for Cu between k_ε and τ_{III}, the cross-slip stress.[27] And, as will be utilized later in the present article relating to combination of measurements made for Cu and Ni materials, it is useful to point out that they have very similar k_ε values because the multiplied product of their otherwise very different individual values of G and τ_{III} is essentially equivalent in Eq. (5).[9]

Figure 6 shows for Cu a further connection on a dislocation pile-up model basis of Eqs. (4) and (5), this time involving extrapolation into the ultrafine grain size regime of the H-P results reported by Hansen and Ralph[20] (and shown in Fig. 5). And in Eq. (5), the value of $k_\varepsilon = 5.0$ MPa $mm^{1/2}$ from Hansen and Ralph was used with other known values of the constants to determine τ_C. Thus, with it and τ_0 determined from $\sigma_{0\varepsilon}$ for the Hansen and Ralph results, Eq. (5) could be plotted in the figure. The resultant curve is the lower dotted one beginning from larger grain sizes and transitioning to the solid curve at n = 1.0 as also evaluated on a calculated pile-up basis from

$$n = 2\alpha k_\varepsilon \ell^{-1/2}/\pi mGb \quad . \tag{6}$$

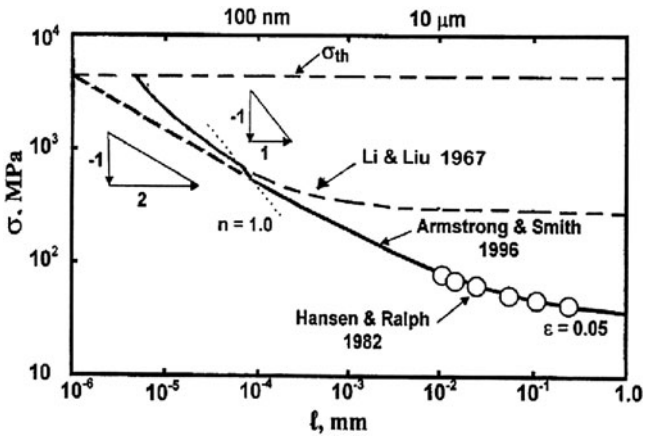

FIG. 6. Hall-Petch model of transition for Cu from a pileup to a single dislocation loop expanding against the grain boundary resistance.[9,20,25]

Thus Fig. 6 shows the transition of a pileup to a single dislocation loop expanding against the grain boundary obstacle for Cu and, very importantly, leads at $\ell < \sim 100$ nm to an increase in stress above the extrapolated H-P line. At the smaller range of grain sizes covered by the solid curve, the relatively high value of τ_C leads to the curve appearing as a shift upwards of a somewhat similar type of $\ell^{-1/2}$ dependence. The most important result of Fig. 6, however, is that transition to a single dislocation loop equation does not lower the material strength and therefore another explanation must be sought for nanoscale experimental measurements showing a lower degree of H-P strengthening. One explanation that has been given for opposite lower strength measurements is based on a disordered grain boundary structure being produced by certain nanoscale material processing methods and in this way leading to reduced values of k_ε through a lower resistance to slip penetrations from within the grain volumes.[28,29] Conrad has provided a review of Cu experimental measurements, most of which fall below expected H-P strength levels and argued for a classification of results into the three grain size-defined regions of: control at larger grain sizes mainly by the dislocation density thus corresponding to $\sigma_{0\varepsilon}$ being dominant; then, an intermediate range of smaller grain sizes where the pileup-induced k_ε represents a more substantial part of the material strength; and last, at the smallest grain sizes, the H-P relation being reversed because of relative grain boundary weakening.[30]

D. The strain rate sensitivity measured by reciprocal value of activation volume

Early measurements demonstrating a grain size dependence of the strain rate sensitivity (SRS) of the hcp metals, Zn and Mg, were associated with a temperature dependence of the H-P k_ε value.[31,32] And for polycrystalline cadmium (Cd) material, whose value of k_ε had been determined to be controlled by pyramidal slip, a grain size-dependent strain rate sensitivity measured by means of reciprocal value of the activation volume, v*, was obtained by differentiating both $\sigma_{0\varepsilon}$ and k_ε terms in Eq. (2) with respect to **ln**(dε/dt) to obtain an H-P type of dependence as

$$(1/v^*) = (1/v_0^*) + (k_\varepsilon/2m\tau_C v_C^*)\ell^{-1/2} \quad . \qquad (7)$$

In Eq. (7), v_0^* is the activation volume determined from $\sigma_{0\varepsilon}$ and v_C^* is that obtained from τ_C^* in Eq. (4). Very importantly, the product $\tau_C v_C^*$was shown to be constant at small strains[31]; and thus, the prediction of Eq. (7) offered a complementary H-P type of dislocation pile-up model dependence analogous to that obtained in Eq. (2).[32] Quantitative verification of Eq. (7) was established later through separate measurements made for cadmium material of the various terms in Eq. (7).[33]

There is application of Eq. (7) to pure fcc metals because of the requirement of thermally-activated cross-slip being needed in the grain boundary regions, in line with match of the temperature and strain rate dependencies of τ_{III} and k_ε measurements for Cu.[27] In addition, compiled SRS measurements have been reported for Cu and Ni v^* values at nanoscale grain sizes with the consequent amplification at much smaller values of ℓ, hence larger values of $\ell^{-1/2}$, as shown in Fig. 7.[34] In the figure, a lesser but definite H-P type of Eq. (7) dependence for conventional grain size Ni material, reported by Narutani and Takamura,[35] has been extrapolated to nanoscale grain sizes for temperatures of 195 and 300 K to encompass a number of compiled v^* measurements reported by Asaro and Suresh.[36] The effect of grain size magnification through Eq. (7) has resulted in an order-of-magnitude increase in v^{*-1}. Through consideration of the two v^* terms in Eq. (7) being for dislocation intersections (in v_0^*) and cross-slip (in v_C^*), Rodriguez[37] was able to resolve difficulty pointed to by Narutani and Takamura in correlating their separate measurements of v^* for the mechanical tests and by means of electrical resistivity.

III. DISCUSSION

The positive H-P results for well-defined nanopolycrystalline materials point to an enabling strength effect in thin films, layers on substrates, and bulk materials under all circumstances in which true nanopolycrystalline material is appropriate, of course, with due regard also to taking into account other material design requirements. From a mechanical strength viewpoint, it seems relevant to discuss the reduction of any H-P k_ε value either due to disorder at the grain boundaries or even because of the reverse in H-P behavior already established for a few materials at their limiting small nanoscale grain sizes, say <10 nm; see Fig. 5. In fact, Lu et al.,[21] while compiling those H-P strengthening measurements shown for Cu in Fig. 5 and confirming the measurements with their own test results, also demonstrated a reverse H-P behavior moving downward in grain size from ~10 nm.

The reverse H-P measurements of Lu et al. can be shown to follow a linear dependence of decreasing stress, as ℓ gets smaller. The same linear dependence had been exhibited in results for Zn reported by Narayan and Conrad.[38] One more or less universal grain size weakening expression that derives from a model of grain boundary shearing during high temperature deformation is[39]

$$(d\varepsilon/dt) = (AD_L Gb/kT)(b/\ell)^p(\sigma/G)^q \quad . \qquad (8)$$

In Eq. (8), A, P, and q are experimental constants; and, D_L is the lattice diffusion coefficient. The linear dependence of decreasing stress on decreasing ℓ for the Zn results leads to an assumption of p = q =1. And then a reciprocal activation volume is easily determined from Eq. (8) as

$$v^{*-1} = (1/kT)[\partial\tau/\partial \mathbf{ln}(d\varepsilon/dt)]_T = \sigma/mkT \quad . \qquad (9)$$

Such dependence has been evaluated for Zn and compared (as filled square points) in Fig. 8 in which comparison may also be made with extrapolation of conventional grain size results reported by Ogawa and Tanaka[40] for a normal H-P dependence of v^{*-1}. Of particular interest on the ordinate axis in Fig. 8 is the filled circle point that corresponds

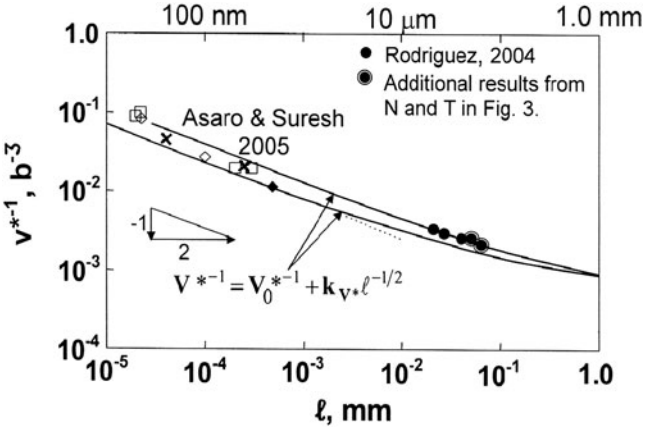

FIG. 7. A Hall-Petch dependence for the reciprocal activation volume, v^{*-1}, for a compilation of Cu and Ni materials covering nanoscale and conventional grain sizes.[34–37]

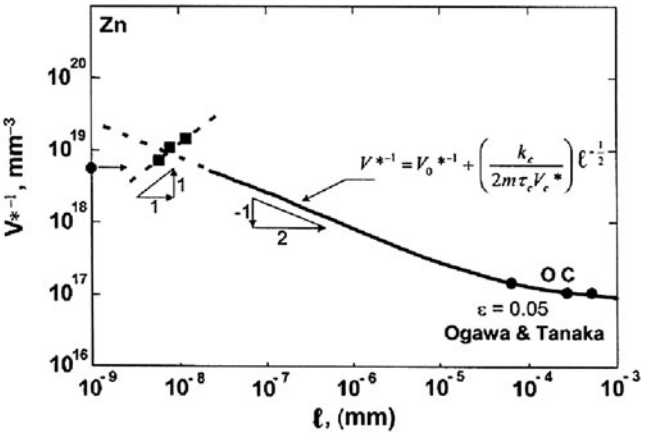

FIG. 8. Prediction of a reversed v^{*-1} Hall-Petch dependence on grain size for the strain rate sensitivity of Zn materials as compared with a normal dependence at conventional grain size.[38,40]

to a smallest dislocation value of b^{-3} for the pyramidal slip established to control τ_C in k_ε. Thus, the v* measurements shown in the figure for a reversed H-P dependence are seen to be smaller than required for any conventional (pyramidal) slip mechanism in the grain boundary regions. The comparison supports a mechanism of smaller displacements for a reversed H-P dependence likely corresponding to shearing of a disordered grain boundary structure.

IV. SUMMARY

The historical background for dislocation pileups against thin film coatings has provided the beginning point for a newer consideration of pileups occurring in thin films or thicker material layers. Electronic and other material applications are enabled by an approximate order-of-magnitude increase in strength level for nanopolycrystalline grain structures. To this end, a description is given for small dislocation pile-up characteristics that lead to the Hall-Petch relation and its connection with relative grain and material layer dimensions and material test conditions, including plane stress testing in the bulge test and of conversion to strength values of hardness test results. Theoretically, strengthening of Cu is predicted to occur for transition of a circular dislocation pileup to a single loop expanding against the grain boundary obstacle. One possibility for the more often made observation of less than extrapolated H-P strength results being obtained is that grain boundary disorder might occur during material production and such disorder should lower the grain boundary resistance. Special advantage is shown for investigating the strain rate sensitivity properties of materials. A complementary H-P type dependence is obtained that, with interpretation on a thermal activation basis, provides connection to the mechanisms of plastic flow within grain volumes and in the boundary regions. Such SRS information is shown to be usefully extended to interpretation of a reverse H-P behavior at lower limiting grain sizes.

ACKNOWLEDGMENTS

Appreciation is expressed to Professor Jay Narayan and colleague guest editors for inviting the present article for the MRS "Focus Issue on Frontiers in Thin-Film Epitaxy and Nanostructured Materials".

REFERENCES

1. A.K. Head: The interaction of dislocations with boundaries and surface films. *Aust. J. Phys.* **13**, 278–283 (1959).
2. R.W. Armstrong and A.K. Head: Dislocation queuing and fracture in an elastically anisotropic material. *Acta Metall.* **13**, 759–764 (1965).
3. A.K. Head: Dislocation group dynamics VI. The release of a pile-up. *Philos. Mag.* **27**(3), 531–539 (1973).
4. R.W. Armstrong, C.S. Coffey, and W.L. Elban: Adiabatic heating at a dislocation pile-up avalanche. *Acta Metall.* **30**, 2111–2116 (1982).
5. R.W. Armstrong: Dislocation pile-up model for the strength of polycrystals, in *International Symposium On Materials For Enabling Nanodevices (ISMEN 2012)*; K-N. Tu, T.F. Liu, and P. Sheng eds.; UCLA, 27–29 Aug., 2012; p. 12.
6. R.W. Armstrong: Dislocation Pile-ups: From {110} cracking in MgO to model strength evaluations. *Mater. Sci. Eng., A* **409**, 24–31 (2005).
7. A.J. Griffin, Jr., F.R. Brotzen, and C. Dunn: Hall-Petch relation in thin film metallizations. *Scr. Metall.* **20**, 1271–1272 (1986).
8. F.R. Brotzen: Mechanical testing of thin films. *Int. Mater. Rev.* **39**(1), 24–45 (1994).
9. R.W. Armstrong: Hall-Petch analysis for nanopolycrystals, in *Nanometals – Status and Perspective*, S. Faester, N. Hansen, X. Huang, D. Juul Jensen, and B. Ralph, eds (Danish Technical University, Risoe Campus, Roskilde, 2012); pp. 181–199.
10. R.L. Fleischer and W.F. Hosford, Jr.: Easy glide and grain boundary effects in polycrystalline aluminum. *Trans. TMS-AIME* **221**, 244–247 (1961).
11. N. Hansen: Effect of grain size and strain on the tensile flow stress of aluminum at room temperature. *Acta Metall.* **25**(8), 863–869 (1977).
12. R.W. Armstrong: On size effects in polycrystal plasticity. *J. Mech. Phys. Sol.* **9**, 196–199 (1961).
13. R.E. Voscoboinikov: Equilibrium of a screw dislocation near the interface in a coated solid. *Phys. Met. Metall.* **113**(5), 448–454 (2012) [in Russian].
14. C. Keller, E. Hug, R. Retoux, and X. Feaugas: TEM study of dislocation patterns in near-surface and core regions of deformed nickel polycrystals with few grains across the cross section. *Mech. Mater.* **42**, 44–54 (2010).
15. C.P. Brittain, R.W. Armstrong, and G.C. Smith: Hall-Petch dependence for ultrafine grain size electrodeposited chromium material. *Scr. Metall.* **19**, 89–91 (1985).
16. S.J. Bull, L. Sanderson, N. Moharrami, and A. Oila: Effect of microstructure on hardness of submicrometre thin films and nanostructured devices. *Mater. Sci. Tech.* **28**(9–10), 1177–1185 (2012).
17. D. Chicot, L. Gil, K. Silva, F. Roudet, E.S. Puchi-Cabrera, M.H. Staia, and D.G. Teer: Thin film hardness determination using indentation loading curve modeling. *Thin Solid Films* **518**, 5565–5571 (2010).
18. R.P. Carreker, Jr. and W.R. Hibbard, Jr.: Tensile deformation of aluminum as a function of temperature, strain rate and grain size. *Trans. TMS-AIME* **209**, 1157–1163 (1957).
19. N. Tsuji, Y. Ito, Y. Saito, and Y. Minamino: Strength and ductility of ultrafine-grained aluminum and iron produced by ARB and annealing. *Scr. Mater.* **47**(12), 893–899 (2002).
20. N. Hansen and B. Ralph: The strain and grain size dependence of the flow stress of copper. *Acta Metall.* **30**, 411–417 (1982).
21. L. Lu, X. Chen, X. Huang, and K. Lu: Revealing the maximum strength in nanotwinned copper. *Science* **323**, 607–610 (2009).
22. C. Keller and E. Hug: Hall-Petch behavior of Ni polycrystals with a few grains per thickness. *Mater. Lett.* **62**, 1718–1720 (2008).
23. A. Torrents, H. Yang, and F. Mohammed: Effect of annealing on hardness and the modulus of elasticity in bulk nanocrystalline nickel. *Metall. Mater. Trans. A* **41**, 621–630 (2010).
24. R.W. Armstrong: Dislocation queuing analysis for the plastic deformation of aluminum polycrystals, in *Physics of Materials; A Festschrift for Dr. Walter Boas on the Occasion of His 75th Birthday*, edited by D.W. Borland, L.M. Clarebrough, and A.J.W. Moore (University of Melbourne, Australia, 1979); pp. 1–11.
25. J.C.M. Li and G.C.T. Liu: Circular dislocation pileups: 1. Strength of ultrafine polycrystalline aggregates. *Philos. Mag.* **15**, 1059–1063 (1967).

26. R.W. Armstrong: Theory of the tensile ductile-brittle behavior of polycrystalline hcp materials; with application to beryllium. *Acta Metall.* **16**, 347–355 (1968).
27. R.W. Armstrong: Strength and ductility of metals. *Trans. Indian J. Met.* **50**, 521–531 (1997).
28. R.W. Armstrong, H. Conrad, and F.R.N. Nabarro: Meso- to nanoscopic polycrystal/composite strengthening, in *Mechanical Properties of Nanostructured Materials and Nanocomposites*, edited by I. Ovid'ko, C.S. Pande, R. Krishnamoorti, E. Lavernia, and G. Skandan (Mater. Res. Soc. **791**, Warrendale, PA, 2004); pp. 69–77.
29. R.W. Armstrong: Grain boundary structural influences on nano-polycrystal strength and strain rate sensitivity. *Emerg. Mater. Res.* **1**(S1), 31–37 (2012).
30. H. Conrad: Grain-size dependence of the flow stress of copper from millimeters to nanometers. *Metall. Mater. Trans. A* **35**, 2681–2695 (2004).
31. R.W. Armstrong: Thermal activation strain rate analysis (TASRA) for polycrystalline metals. *J. Sci. Indust. Res.* **32**(11), 591–598 (1973) [in Indian].
32. Y.V.R.K. Prasad and R.W. Armstrong: Polycrystal versus single-crystal strain rate sensitivity of cadmium. *Philos. Mag.* **29**(6), 1421–1425 (1974).
33. P. Rodriguez, R.W. Armstrong, and S.L. Mannan: The dependence of activation area on grain size in cadmium. *Trans. Indian Inst. Met.* **56**(3), 189–196 (2003).
34. R.W. Armstrong and P. Rodriguez: Flow stress/grain size/strain rate coupling for fcc nanopolycrystals. *Philos. Mag.* **86**, 5787–5796 (2006).
35. T. Narutani and J. Takamura: Grain size strengthening in terms of dislocation density measured by resistivity. *Acta Metall. Mater.* **39**(8), 2037–2049 (1991).
36. R.J. Asaro and S. Suresh: Mechanistic models for the activation volume and rate sensitivity in metals with nanocrystalline grain sizes. *Acta Mater.* **53**, 3369–3382 (2005).
37. P. Rodriguez: Grain size dependence of the activation parameters for plastic deformation: Influence of crystal structure, slip system and rate-controlling dislocation mechanism. *Metall. Mater. Trans. A* **35**(9), 2697–2705 (2004).
38. H. Conrad and J. Narayan: Mechanisms for grain size hardening and softening in zinc. *Acta Mater.* **50**, 5067–5078 (2002).
39. T.G. Langdon: Grain boundary sliding revisited: Developments in sliding over four decades. *J. Mater. Sci.* **41**, 597–609 (2006).
40. K. Ogawa and K. Tanaka: Effect of grain size on the strength of polycrystalline zinc, in *Proceedings of 23rd Japan Congress on Materials Research* (Soc. Mater. Sci., Japan, 1980); pp. 39–45.