Double-inverse grain size dependence of deformation twinning in nanocrystalline Cu

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While a Hall-Petch-type dependence is known for deformation twinning (DT) in Cu and other metals of conventional grain sizes ($D > 1\ \mu$m), with D decreasing into the nanocrystalline regime, the propensity for DT turns around to increase, exhibiting an inverse grain size dependence. This trend is inversed again at still smaller grain sizes, returning to the behavior of increased difficulty in DT with D going further down. This double-inverse behavior with respect to the normal Hall-Petch D dependence is demonstrated here for nanocrystalline Cu films, deformed in tension at room temperature and slow strain rates. The nonmonotonic D dependence of DT is explained by modeling the competing grain size effects on the emission of the first partial dislocation and the plane-to-plane promotion of partial dislocation slip afterwards.

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I. INTRODUCTION

Deformation twinning (DT) occurs in face-centered-cubic (fcc) metals such as Cu, especially at low temperatures and high strain rates ($\dot{\varepsilon}$).[1–3] The stress required for DT (σ_{DT}) increases with decreasing grain size D, following a Hall-Petch (H-P)-type relationship,[4,5] in the regime of $D > 1\ \mu$m where conventional twin growth mechanisms (i.e., bulk-based pole mechanisms[6]) operate. While in this "conventional regime" DT becomes increasingly difficult with smaller D, in nanocrystalline (NC) grains DT has been found to be a major contributing deformation mechanism again, even at room temperature (RT) and low $\dot{\varepsilon}$.[5–9] For example, the propensity of DT in NC-Ni was observed to increase with decreasing D for $D < \sim 90$ nm.[8] This D effect, in our opinion, can be termed an inverse Hall-Petch dependence as it is opposite the trend known for coarse grains. Interestingly, a second inverse D dependence[10] has been reported recently, when D was reduced to $< \sim 35$ nm for NC-Ni. The DT propensity was found to decrease again with decreasing D, resulting in a peak twinning propensity at this "inversion grain size" (as termed in Ref. 10), $D_{DT}^{inv} = 35$ nm. Such a nonmonotonic, double-inverse grain size dependence is rather intriguing and the underlying mechanism has not been understood.

Here we report experimental observations of a similar grain size effect in tensile-tested NC-Cu films, at RT. The propensity for DT also exhibits a peak in the NC regime but at a D_{DT}^{inv} considerably larger than that for NC-Ni. This behavior is explained based on the "stimulated slip" concept[6] of DT, but with grain boundary (GB) acting both as (a) the emitter of the first partial dislocation, and (b) the plane-to-plane promoter of partial slip afterwards, with opposing D dependence.

II. EXPERIMENTAL METHODS

500-nm-thick Cu thin films were deposited on dog-bone-shaped 125-μm-thick polyimide substrate via dc magnetron sputtering. The as-deposited films were *in situ* annealed at 150 °C to stabilize the microstructure. X-ray diffraction showed a strong (111) peak, followed by (200) and (220) peaks, indicating that the majority of the grains have these out-of-plane orientations. The D distribution was examined in a transmission electron microscope (TEM), revealing a D range spanning 200–20 nm (average ~ 110 nm), as shown in the plan-view [Fig. 1(a)] and cross-sectional [Fig. 1(b)] TEM micrographs. Samples were cut from the as-prepared films for uniaxial tensile testing at RT using a MTS® Tytron 250 at $\dot{\varepsilon} = 10^{-4}$ and 10^{-2} s^{-1}. The tensile samples had a total length of 65 mm and a gauge section of 30 mm × 4 mm and were strained to ~20% total elongation, without removing the polyimide substrate. Postmortem TEM examinations provided an estimate of the number of grains that contained deformation-induced twins and stacking faults (referred to as DTs and DSFs hereafter), respectively, following the procedures in Ref. 10. We examined ~1800 grains for each tensile-deformed specimen (100 grains for each D). Figure 1 shows the presence of twins and SFs; e.g., grain A contains multiple twins, see Fig. 1(c). A high-resolution image of the boxed region is shown in Fig. 1(d), together with its Fourier transform indicating the twinning relationship. Since before tensile pulling there were already some grains that contained initial growth/annealing twins (ITs) and initial stacking faults (ISFs), only the increment (see below) will be taken as the DTs and DSFs. Still, one has to keep in mind that not all twins/stacking faults are visible in a plan-view TEM image.

III. RESULTS AND DISCUSSION

Figure 2 displays the histograms, sorted for grains of different sizes, showing the number of grains containing ITs and ISFs before deformation, and the number of grains containing twins (ITs+DTs) and stacking faults (ISFs+DSFs) after tensile straining, for two strain rates. The number of grains containing DTs and DSFs generated during deformation is then determined from the difference between the two numbers before and after deformation,[10] and the fraction (percentage) of DT (and DSF) containing grains is obtained by normalizing against the total number of grains (100) at the particular D (each D is for the grain sizes within a 10 nm bin width).

As seen in Figs. 2(a) and 2(b), for ITs and ISFs, respectively, the number of grains containing ITs (and ISFs) before

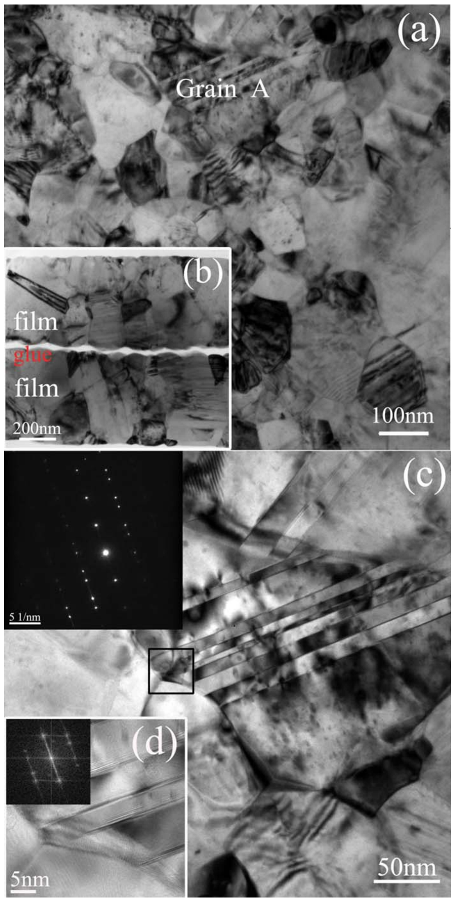

FIG. 1. (Color online) (a) Plan-view and (b) cross-sectional TEM micrograph showing the microstructure of the NC-Cu film after tensile straining at $10^{-4}/\mathrm{s}$; (c) is a magnified view of grain A in (a), showing twins and the corresponding diffraction pattern (inset); (d) is the high-resolution TEM image of the region containing the twins [boxed area in (c)] and its Fourier transform (inset).

deformation is low and varies only slightly with D over the entire range from 200 to 20 nm. After straining at $10^{-4}/\mathrm{s}$, however, the numbers of twin (or stacking fault) containing grains are significantly larger, and depend obviously on D. The same behavior is seen for the sample before and after straining at another strain rate, $10^{-2}/\mathrm{s}$, in Figs. 2(c) and 2(d) for ITs and ISFs, respectively. For both strain rates, there is a general trend that the fraction of grains containing DTs first increases and then decreases with decreasing D. This is similar to what was observed in NC-Ni,[10] as discussed in Sec. I. There appears to exist a double inverse D dependence of DT in the submicron-NC regime for fcc metals. The propensity of conventional DT is very small above $D \sim 200$ nm (Fig. 2), as expected from the Hall-Petch-type behavior of conventional DT (difficult at RT for Cu, and even more so at small D). But DT propensity then rises as D is reduced to below $D_{\mathrm{DT}}^{\max} \sim 160$ nm, and it falls back down again with decreasing D when an opposite grain size effect gradually takes over. We use a solid curve in the figures to represent the general trend of the DT behavior across the entire grain size range studied, from which an apparent peak in twinning propensity is observed at a $D_{\mathrm{DT}}^{\mathrm{inv}}$ in the vicinity of 80 nm. The error bar for the number of twinned grains, which are in double digit numbers out of the ~ 100 grains we examined for each grain size, is believed to be much smaller than the difference seen in the figure for the various grain sizes, so the peak observed appears to be real.

From Fig. 2 one observes a maximum grain size that contains DSFs ($D_{\mathrm{SF}}^{\max}$) and DTs ($D_{\mathrm{DT}}^{\max}$). $D_{\mathrm{SF}}^{\max}$ moves to lower D while $D_{\mathrm{DT}}^{\max}$ and $D_{\mathrm{DT}}^{\mathrm{inv}}$ remain similar at the higher $\dot{\varepsilon}$, suggesting that DSFs are more likely to be converted into DTs at higher $\dot{\varepsilon}$. Also, DT is clearly more favorable at higher $\dot{\varepsilon}$ as expected.[1–5,8] When comparing the present results at $\dot{\varepsilon} = 10^{-4}/\mathrm{s}$ with those in NC-Ni at $\dot{\varepsilon} = 3 \times 10^{-3}/\mathrm{s}$,[10] NC-Cu has a larger $D_{\mathrm{DT}}^{\max}$ (~ 160 nm for Cu vs ~ 60 nm for Ni), a higher $D_{\mathrm{DT}}^{\mathrm{inv}}$ (~ 80 nm vs 35 nm), and a much wider D range favorable for DT in terms of $D_{\mathrm{DT}}^{\max} - D_{\mathrm{DT}}^{\mathrm{inv}}$ (~ 80 nm vs ~ 35 nm). This is consistent with the general understanding that DT is easier in Cu than in Ni because the former has a lower stacking fault energy (SFE).

We next provide a semiquantitative explanation to the double-inverse D dependence of DT in NC-Cu. We point out again that while a $D_{\mathrm{DT}}^{\mathrm{inv}}$ was observed before for NC-Ni,[10] the mechanism was not understood. Compared to ordinary (full) dislocation plasticity, DT is a highly correlated inelastic shearing process, during which many adjacent atomic planes slip in the same manner. We believe this occurs by "stimulated slip:"[6] the slip of partial dislocation with Burgers vector $\mathbf{b}_{\mathrm{p}}$ on atomic plane n can "stimulate" plane $n+1$ to slip in the same way. Such plane-to-plane "stimulation" or "infection" is made possible by promoter defects, i.e., pole dislocations in the bulk with screw character.[6] In this conventional DT regime for Cu,[4,5] the H-P slope is 0.7 MPa m$^{1/2}$, larger than that for ordinary full dislocation slip, ~ 0.35 MPa m$^{1/2}$.[11] Due to this steep rise in σ_{DT} with reducing D, for small-D Cu grains conventional DT is difficult and not expected at RT.

However, with D approaching the nanoscale, dislocations (including twinning partial dislocations) no longer operate from sources in the interior of the grains, but instead emit from GBs.[12,13] This direct emission of partial dislocations triggers DSFs and DT nonconventionally.[12,13] The probability to nucleate one such partial (with smaller Burgers vector than that of full dislocation) in lieu of the full dislocations scales with the difference in the required stresses (τ),[7,9]

$$P_{\mathrm{partial}}^{\mathrm{emission}} \propto \tau_{\mathrm{full}} - \tau_{\mathrm{partial}} = \left[\frac{2\alpha\mu(mb_{\mathrm{F}} - b_{\mathrm{P}})}{D} - \frac{\gamma_{\mathrm{sf}}}{b_{\mathrm{P}}} \right] = \frac{\gamma_{\mathrm{sf}}}{b_{\mathrm{P}}} \left(\frac{D_{\mathrm{C}}}{D} - 1 \right), \quad (1)$$

where μ is the shear modulus (48 GPa for Cu), γ_{sf} is the SFE (41 mJ/m^2 for Cu), the parameter α is on the order of unity and reflects the character of the dislocation and contains the scaling factor between the radius of curvature of the dislocation and D, m is a stress concentration factor,[9] and b_{F} and b_{P} are the magnitudes of the Burgers vectors of the full dislocation and the Shockley partial dislocation, respectively. There is a critical D_{C}, which can be determined by setting Eq. (1) to zero ($D_{\mathrm{C}} \sim 200$ nm for Cu,[9] near the observed $D_{\mathrm{DT}}^{\max}$) below which partial dislocation emissions dominate, leaving behind DSFs and also setting the stage for subsequent DT (see below). The switch-on of this partial dislocation mechanism explains the DSFs below D_{C}, as well as the

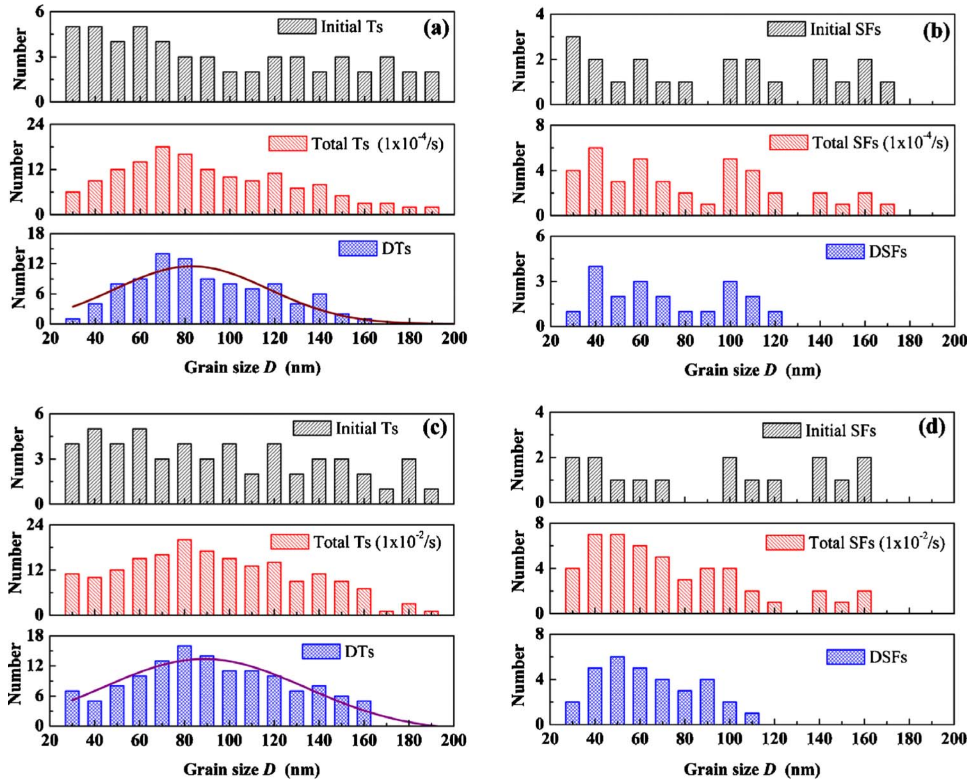

FIG. 2. (Color online) [(a) and (c)] Grain size effect on the formation of deformation twins and [(b) and (d)] stacking faults in Cu thin films tensile tested at two strain rates, 10^{-4} and 10^{-2} s^{-1}. A solid line is drawn as a guide to the eye to indicate the general trend of the DT propensity. See text for details.

increasing DT propensity (P_{DT}) with decreasing D seen in Fig. 2. The smaller the D is below D_C, the larger the stress difference in Eq. (1), and the higher probability to directly activate partials rather than full dislocations, i.e., the higher the $P_{partial}^{emission}$, as it quickly approaches unity (all the GB-emitted dislocations are partials), as schematically illustrated in Fig. 3.

However, the nucleation of one partial, while accounting for the DSFs, is a necessary but not a sufficient condition for DT. Uncorrelated random emissions of individual partials cannot accidentally form a twin tens, or even hundreds of atomic layers thick, as those seen in Fig. 1. The GB emissions of partial dislocations need to be spatially, and perhaps temporally, correlated. This layer-by-layer promotional effect may well be due to the inertia of a plane n partial dislocation loop (rebound-promotion mechanism), which can move very fast (~speed of sound) and accumulate huge kinetic energy after nucleation, driven by the high stress in grain interior, with net energy several times the stationary dislocation self-energy as it hits the GB, thus satisfying energy conservation for the creation of a partial dislocation on plane $n+1$. The multiplane generalized stacking fault energy[14] of the underlying crystal also favors emissions of identical partial on plane $n+1$, rather than full dislocations. This is because $\gamma_{sf} \approx 2\gamma_{ctb}$, where γ_{ctb} is the coherent twin boundary energy, so no extra *planar* energy is needed to *thicken* the twin.[14] That is to say, right next to an existing stacking fault or twin, an additional Shockley partial acts as if a "full dislocation," but with smaller Burgers vector. This is the root cause for the promotional effect in stimulated slip, which would promote correlated slip even without the kinetic-energy argument.

Nonetheless, elastic compatibility and other requirements may still demand the GB sites that can successfully promote stimulated slip to be rather "special," and the configurational density n_s of such sites should be proportional to the circumference of the slip plane—GB intersection, that is, $n_s \propto D$. Specifically, on a (111) plane in a Cu grain, the partial slip needs to be "promoted" to the next (111) plane, such that twinning partials become available on successive planes one after another in a highly correlated fashion to thicken the twin.[6,15] To sustain this plane-to-plane "infection" required for stimulated slip, the partial running on the (111) plane need to hit special GB sites where a twinning partial can be created (see discussion above). Alternatively, the promotion

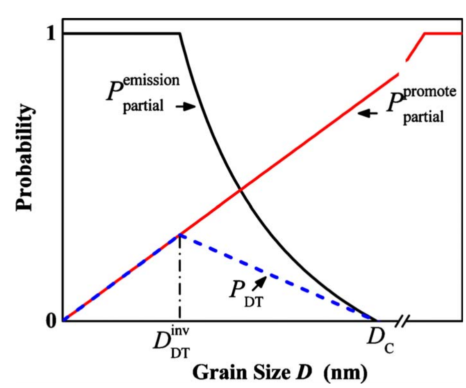

FIG. 3. (Color online) Schematic illustration of the D effects on the DT propensity P_{DT}, and its constituent $P_{partial}^{emission}$ and $P_{partial}^{promote}$ contributions, that lead to D_C and D_{DT}^{inv}.

could be achieved if the partials emit from certain GB sites where proper dislocation reactions occur to induce a suitable twinning partial on the next atomic plane.[15] In either case, this "promotion-to-the-next-layer" probability is

$$P_{\text{partial}}^{\text{promote}} = n_s P_{\text{reaction}}^{\text{GB}} = kDP_{\text{reaction}}^{\text{GB}}, \tag{2}$$

where n_s is the number of available GB sites with the right configurations such that the local site is potentially capable of the right type of dislocation reaction,[15] with or without the aid of the kinetic energy of the incoming dislocation; n_s scales with the length of the intersecting perimeter of the (111) plane with the GB. $P_{\text{reaction}}^{\text{GB}}$ is the probability to accomplish the dislocation reaction (sometimes aided by a hit of the incoming partial) and successfully promote identical partial slip on plane $n+1$ at the GB site, and k is a scaling constant. From Eq. (2), the smaller the D, the less available the proper sites and the less likely for the partials to come off or hit the right spots for realizing a promotion event to infect the next layer for DT.

The two probabilities [Eqs. (1) and (2)] are drawn schematically in Fig. 3. The product of the two competing terms determines the overall P_{DT} (dashed line in Fig. 3), which exhibits a behavior consistent with the double-inverse D dependence in the NC regime. The competition between the two probabilities naturally leads to a peak in the overall P_{DT}, which is likely to be responsible for the observed $D_{\text{DT}}^{\text{inv}}$. Going away from the peak, at D much below $D_{\text{DT}}^{\text{inv}}$ DT becomes increasingly less likely because of the falling $P_{\text{partial}}^{\text{promote}}$, while $P_{\text{partial}}^{\text{emission}}$ and the DSFs (no need for $P_{\text{partial}}^{\text{promote}}$) remain steady.[10] Above $D_{\text{DT}}^{\text{inv}}$, promotion is more proficient, and most of the DSFs get converted to DTs (Fig. 2). But if the D is too large, although $P_{\text{partial}}^{\text{promote}}$ gets closer to unity (Fig. 3) $P_{\text{partial}}^{\text{emission}}$ becomes a problem and would reduce the overall P_{DT}. Beyond $D_{\text{DT}}^{\text{max}}$, the GB-emitted partials are no longer dominant and full dislocation slip takes over. In this case, DT no longer relies on the emission and promotion of partial dislocations at GB, and the conventional mechanism then leads to the normal Hall-Petch-type D dependence,[5] as explained in Ref. 6. This sequence of mechanisms across the entire grain size range can thus account for the double-inverse dependences observed with respect to the known Hall-Petch behavior,

For NC-Ni, its SFE (110 mJ/m^2) is larger than Cu. This could move the $P_{\text{partial}}^{\text{emission}}$ curve to the left according to Eq. (1), rendering smaller $D_{\text{DT}}^{\text{max}}$ and $D_{\text{DT}}^{\text{inv}}$. A higher $\dot{\varepsilon}$ (higher stresses and dislocation speed) enhances the emission and promotion of the partials, leading to elevated P_{DT} across the board (Fig. 2).

IV. CONCLUSION

In the nanocrystalline grain size regime, the propensity for deformation twinning can exhibit the so-called "inverse grain size effect,"[10] deviating from the normal monotonic Hall-Petch-type dependence for deformation twinning promoted by bulk dislocation poles. This is demonstrated here for nanocrystalline Cu deformed in tension at room temperature and slow strain rates. We have shown that the deformation twinning propensity exhibits not just one, but two, inverse D dependences, and explained this phenomenon by considering competing factors that influence DT. Specifically, the emission of the first partial dislocation, and the plane-to-plane promotion of partial dislocation slip afterwards, have opposite grain size dependences. The combination of the two effects governs the overall DT propensity, leading to a nonmonotonic behavior (with a peak or double inversion) in the NC grain size regime, in lieu of the conventional Hall-Petch-type dependence.

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