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Viewpoint Paper

Size dependence of rate-controlling deformation mechanisms in nanotwinned copper

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Abstract—Nanotwinned metals exhibit an unusual combination of ultrahigh strength, considerable ductility and enhanced rate sensitivity. We find that a Hall–Petch-type relationship closely fits the experiment data of activation volume as a function of twin spacing. The results suggest a transition of the rate-controlling mechanism from intra-twin- to twin-boundary-mediated processes with decreasing twin lamellar thickness. These findings provide insights into the possible routes for optimizing the strength and ductility of nanostructured metals by tailoring internal interfaces.

Keywords: Twin boundary; Size dependence; Rate-controlling mechanism; Activation volume

The strength, ductility and many other mechanical characteristics of metals and alloys are strongly dependent upon their micro- or nanoscale structures. In the classic case of grain refinement strengthening, grain boundaries (GBs) provide barriers to dislocation motion. The increase in strength is proportional to the reciprocal square root of the grain size (d), i.e. the characteristic spacing of barriers to dislocation slip, as in the well-known Hall–Petch (H–P) relationship [1,2]. Nanocrystalline (nc) metals, with d typically finer than 100 nm, follow the H–P relation [3–5].

An analogous strengthening mechanism is twin boundary (TB) strengthening. Recent experiments have shown that the yield strength varies with the twin spacing in the same manner as the grain size for ultrafine-grained Cu (with a grain size of several hundred nanometers) containing layered nanotwins (typically tens and hundreds of nanometers in thickness). The H–P relationship has been experimentally verified to be applicable to strength vs. twin lamellar thickness λ ($\lambda^{-1/2}$ dependence) [6]. However, nanotwinned Cu has increased strength with preserved high tensile ductility, while severely compromised tensile ductility is often observed in nc metals. These results suggest that there exists a fundamental difference between TBs and GBs in controlling ductility, whereas they are both effective in strengthening the material by blocking dislocation motion.

In order to understand the mechanistic role of the internal interfaces (TB and GB) in ductility, it is important to first determine the rate-controlling deformation mechanisms. Investigating the sensitivity of flow stress to the applied loading rate is an effective technique to probe the active deformation mechanism. This is because the rate sensitivity index m and the associated activation volume v^* can vary by orders of magnitude for different rate-controlling processes, so that m and v^* can be taken as effective kinetic signatures of deformation mechanisms [7].

The size-dependent rate sensitivity and activation volume have been reported for face-centered cubic (fcc) metals [8,9]. Specifically, grain refinement from the micrometer to the nanometer scale leads to an increase in m by up to one order of magnitude and a concomitant decrease in v^* by two orders of magnitude [10]. It has been proposed that bulk dislocation–dislocation interac-

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tion (forest hardening) is the rate-controlling mechanism when the grain size is in the micrometer range [5,9], and that the dominant rate mechanism changes to GB-mediated dislocation activity when the grain size is reduced to the nanometer scale [5,11,12].

In this paper, based on a number of recent developments [13–16], we briefly review the twin density (size) dependence of rate sensitivity and activation volume for nanotwinned Cu (nt-Cu). The underlying dislocation mechanisms are discussed in terms of transmission electron microscopy (TEM) observations [17,18], as well as atomistic simulations and crystal plasticity modeling [13,14,16,19]. We find that an H–P-type relation closely fits the experiment data of activation volume vs. twin lamellar thickness. The implications of the dominant deformation mechanisms and their consequent effects on ductility are discussed. Processing routes are suggested to optimize strength and ductility.

We synthesized ultrafine-grained Cu containing nanoscale growth twins by the pulsed electro-deposition technique. The sample consisted of grains of irregular shape and random orientation, with an average size of 400–500 nm [17,20]. The grains contained a tunable high density of twin boundaries; λ can be varied from 100 to 15 nm by modifying the electro-deposition parameters. TEM and high-resolution (HR) TEM images show that most TBs are coherent in the as-deposited samples; a few interfacial and lattice dislocations could be detected in most lamellae, as indicated in Figure 2a [15].

Figure 1a summarizes the experimental results on m as a function of λ for nt-Cu [17,18,21]. When λ is reduced to about 15 nm, m can be nearly one order of magnitude higher than that for samples with micron-sized grains. In addition, the m values for nc-Cu are also shown in Figure 1a, with d replacing λ as the characteristic structural length scale. It is seen that m increases slightly with d reducing from the micrometer to the sub-micrometer scale, while an obvious "take-off" appears when d is further reduced to below 100 nm or so. It is interesting to note that the dependence of rate sensitivity on λ is very close to that on d in the size range measured for nc-Cu [5]. Figure 1b shows the effects of λ and d on activation volume, v^*, measured in units of b^3, where b is the Burgers vector magnitude of a perfect dislocation. v^* decreases from $1000b^3$ to only tens of b^3 when d (or λ) decreases from the micrometer to the nanometer range.

Figure 2a shows the as-deposited samples containing few lattice and TB dislocations. In contrast, Figure 2b shows the deformed sample with fine twins (about 50 nm thick), where a large amount of dislocation debris is accumulated near TBs. The HRTEM image in Figure 2d indicates that these debris dislocations are Shockley partials. Figure 2c reveals dislocation structures in wider twins; one can see that a number of lattice dislocations and dislocation tangles have accumulated inside the lamellae, as well as the much-strained TBs containing a high density of boundary dislocations. Such complicated dislocation configurations suggest that in general the intra- and inter-twin mechanisms co-evolve with deformation, contributing to strain hardening in a cooperative manner. This observation motivates the crystal plasticity modeling, as detailed later.

Atomistic simulations have been performed to understand the TB-mediated dislocation mechanisms [16,19]. These atomically detailed studies are central to understanding the role of TBs in controlling strain hardening and ductility. Various mechanisms have been revealed in molecular dynamics (MD) simulations. As shown in Figure 3a, there are generally two types of dislocation–TB interaction, i.e. the screw and the non-screw cases. As a screw dislocation impinges on a TB, it may either

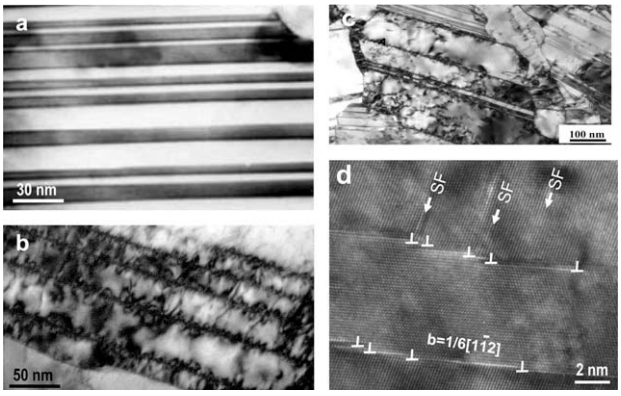

Figure 2. TEM images of nanotwinned Cu. (a) As-deposited sample; the coherent TBs are straight and defect-free. (b) In deformed samples with fine twins (about 50 nm thick), dislocations are mostly accumulated near TBs. (c) With increasing twin width, a number of lattice dislocations and dislocation tangles are accumulated inside lamellae. (d) HRTEM image showing Shockley partials at the TBs [15].

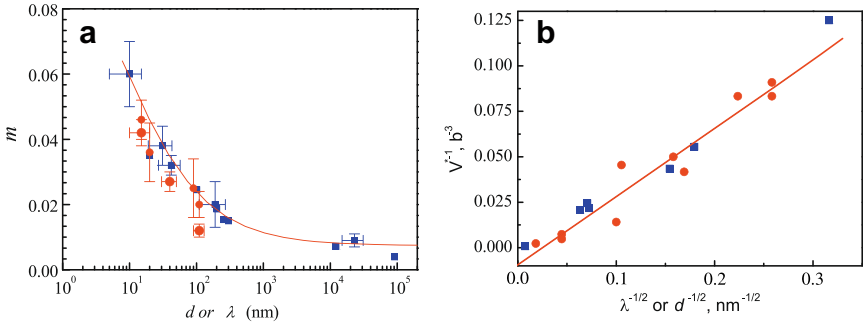

Figure 1. Effect of twin lamellar thickness (λ on (a) rate sensitivity and (b) activation volume (measured in unit of b^3) for nanotwinned Cu (red symbols) [17,18,21]. For comparison, the rate sensitivity and activation volume are also plotted as functions of grain size (d) for nanocrystalline Cu (blue symbols) [5]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(i) be absorbed into the TB, with two Shockley partials propagating along the TB in the opposite directions; or (ii) transmitted into the adjoining twin by cutting through the boundary without leaving any residual incompatibility in the TB [19]. The non-screw dislocation similarly involves absorption and transmission. However, the situation is much more complicated. Consider transmission as an example. As shown in Figure 3b, when a 60° non-screw dislocation of type $\vec{b}_1$ impinges on the TB, it may be transferred directly through the boundary into the adjoining twin accompanied by emission of an additional partial dislocation along the TB (as indicated in Fig. 2c). In contrast, when a 60° dislocation of type $\vec{b}_2$ proceeds, a 30° leading Shockley partial can be released from the TB, and a long stacking fault ribbon is left behind. The remaining partial is pinned down at the boundary, which can be identified as a Hirth lock of 1/3[0 0 1] type, as seen in Figure 3c. Such a sessile dislocation in the TB opposes the subsequent transmission across the TB as well as the glide of nearby partials within the TB [16].

Whereas MD simulations provide physical insights into the TB-mediated dislocation mechanisms, one may wonder whether a quantitative connection can be established between atomistic modeling and experimental measurement. It should be emphasized that MD is limited by the extremely high strain rate or high stress, which is very different from experimental conditions. In order to determine the deformation mechanisms relevant to laboratory experiments, Zhu et al. [14] used a novel approach of reaction pathway exploration to analyze the slip transfer reactions between the lattice dislocations and TB. Specifically, using the free-end nudged elastic band method, they determined the minimum energy paths for reactions of absorption, desorption and slip transmission of a screw dislocation at the TB. Figure 3d shows as an example the saddle-point state of absorption, involving a cross-slip process with the constriction and simultaneous bow-out. Based on the three-dimensional saddle-point states, they further predicted the yield stress, activation volume and rate sensitivity, which were consistent with experimental measurements. This agreement demonstrates that the slip transfer reactions at TBs are the rate-controlling mechanisms when the twin lamellar thickness is around or below 20 nm. Along the same lines, Asaro and Kulkarni [13] recently performed a dislocation mechanics analysis of the rate sensitivity and strength mediated by cross-slip in nanotwinned fcc metals. They pointed out that the resolved shear stress is proportional to the reciprocal square root of twin lamellar thickness, $\lambda^{-1/2}$, via clear and specific mechanisms for dislocation/TB interaction [13]. Their results are consistent with those of Zhu et al. [14]. It should be emphasized that, for wider twins, the apparent activation volume measured in experiments needs to be connected to the physical activation volume at the TB through a size-dependent model, such as the one given by Eq. (1) below.

Based on the TEM observations and atomistic modeling results, a twin-boundary affected zone (TBAZ) model was proposed [15]. A two-dimensional [15] and, most recently, a three-dimensional [22] continuum crystal plasticity model were formulated. The predicted stress–strain behavior, strain-rate sensitivity and failure initiation limit captured the experimentally observed trends very well in terms of twin thickness λ. These continuum models took into account the size- and orientation-dependent dislocation blocking and absorption at the twin boundaries. An important underlying assumption of the TBAZ-based crystal plasticity model is that

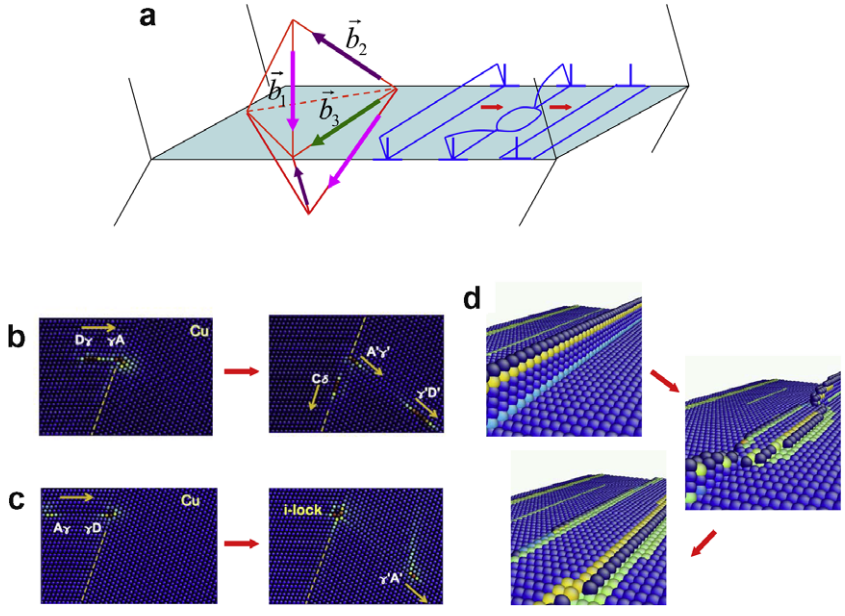

Figure 3. Atomistic modeling of the dislocation–TB interaction. (a) Thompson's tetrahedron showing the Burgers vectors of incoming and outgoing dislocations; also shown is a schematic of the reaction pathway of dislocation absorption into the TB. (b) and (c) show, respectively, MD simulation of slip transmission of the non-screw 60° dislocation of type $\vec{b}_1$ and type $\vec{b}_2$ [16,19]. (d) Atomic structure from reaction pathway calculation showing the absorption of a screw dislocation by cross-slip [14].

there are co-existing deformation mechanisms operating simultaneously, including TB-mediated dislocation activities and orientation-dependent lattice dislocation activities, as schematically shown in Figure 4a and b. Although, in most cases, the TB-mediated dislocation activities are considered to be the dominant deformation mechanism in nt-Cu, other possible operating mechanisms, such as lattice dislocation interactions, are also important and cannot be ignored. Only by combining these different contributing factors instead of exclusively considering the dominant deformation mechanism can a fairly good match between model predictions and experimental results be achieved for the size (λ)-dependent trends of strength, rate sensitivity and ductility.

An H–P-type relationship for v^{*-1} has been previously observed for nc fcc metals, i.e. v^{*-1} varies linearly with $d^{-1/2}$ [12]. This size dependence was rationalized by combining the dislocation pile-up model and the thermal activation analysis of plastic flow rate. Considering the similar role of TB and GB in opposing dislocation motion, here we generalize the H–P-type relationship for v^{*-1} to include both the GB and TB effects

$$\frac{1}{v^*} = \frac{1}{v_0^*} + \left[\frac{k}{2m\tau_{GB}^c v_{GB}^*}\right] d^{-1/2} + \left[\frac{k}{2m\tau_{TB}^c v_{TB}^*}\right] \lambda^{-1/2} \qquad (1)$$

where v_0^* is the activation volume associated with the intra-grain or intra-twin dislocation mechanism, i.e. intersection of lattice dislocations; v_{GB}^* and v_{TB}^* are the activation volumes associated with the GB- and TB-mediated mechanisms, respectively. In Eq. (1), the local shear resistance τ_{GB}^c or τ_{TB}^c is determined from the classic H–P k value ($k \propto \sqrt{\tau_{GB}^c}$) [23] in accordance with the dislocation pile-up model. A value of $\tau_{GB}^c = 77.7$ MPa was obtained for cross-slip at the GBs to sustain plastic flow [24]. Here m is the Taylor factor ($m \sim 3.1$) and we assume $\tau_{GB}^c = \tau_{TB}^c$. In Figure 1b, we plot the fitting curve for v^{*-1} based on Eq. (1), matching the experimental data. This demonstrates that the H–P-type relation for v^{*-1} could be generally used for both the nc and nt metals.

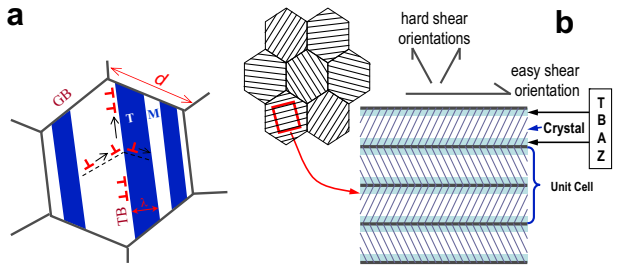

Figure 4. Schematic drawings of the crystal plasticity modeling of TB-mediated deformation mechanisms. (a) A grain containing TBs with an average twin spacing of λ, and possible strengthening mechanisms arising from interactions of dislocations with TBs. The TBs provide sufficient room for the accommodation and pile-up of interfacial dislocations; TBs gradually lose coherency during plastic deformation and contribute to ductility. The interfacial dislocations along the TBs can also be blocked by the GBs. (b) The crystal plasticity implementation of the proposed TBAZ model with both soft- and hard-mode shearing [15].

The size dependence of activation volume shown in Figure 1b, along with its agreement with the dislocation pile-up model of Eq. (1), suggests a size-dependent transition of the rate-controlling mechanism from the intra-twin to inter-twin processes with decreasing twin lamellar thickness. At large λ, the dislocation mechanism within the twin lamellar dominates, giving a characteristically large activation volume of about $1000b^3$, which arises because of bulk dislocation–dislocation interactions. In the limiting case of λ around or less than 20 nm, which is approximately the limited equilibrium distance for two dislocations pile-up at the TBs, the stored lattice dislocation tangle can hardly be stabilized inside the twin lamella. The TB-mediated dislocation activity is then expected to dominate. In between, both mechanisms are operative; the relative contribution of each mechanism is controlled by the twin thickness, as indicated by Eq. (1). The cooperation and transition of the intra- and inter-twin mechanisms are also consistent with previous discussions of TEM experiments, atomistic simulations and crystal plasticity modeling.

The foregoing analysis demonstrates that the size dependence of rate sensitivity, the activation volume and the associated rate-controlling mechanisms in nanotwinned systems can be similarly modeled and understood as nc metals. How, then, can we explain their striking difference in ductility? Tensile ductility is often limited by the onset of necking instability. This has been well studied at the continuum level [25,26]. The central result is that the high strain-rate sensitivity and the high strain hardening coefficient can both help delay the onset and development of necking, thereby promoting near-uniform tensile deformation. Since the TBs and GBs play a similar role in mediating the rate sensitivity and activation volume, the ductility difference can be largely attributed to the higher strain hardening capacity and damage tolerance in nt-Cu [14]. To understand the beneficial effect of TBs on strain hardening, we note that the TEM observations and atomistic simulations have shown a high density of interfacial dislocation debris at the TB. This suggests that, compared to the random GBs, the coherent TBs are more adaptable for creating and accommodating dislocations before cleavage failure. As a result, the coherent TBs are much more hardenable when they accumulate interfacial dislocations and gradually lose coherency during the initial stage of plastic deformation. This gives rise to an increased strain hardening (relative to nc) and a consequent delay in the onset of necking that lead to the significantly improved tensile ductility [14].

As discussed above, attention so far has been focused on the effect of twin lamellar thickness. The ultrahigh strength with preserved high ductility was mostly achieved in the nanotwinned materials with a fixed grain size ($d \sim 500$ nm). In fact, the grain size can also play an important role in nanotwinned materials, considering that the intersections between GBs and TBs can act as effective dislocation sources and sinks. Furthermore, the grain size controls the length scale in the easy glide direction parallel to the TB. If λ is fixed at the nanometer scale, decreasing d into the nanometer range would result in higher strength, but compromise ductility. This

predicted trend is consistent with the experiment showing a high yield strength (1.05 GPa) and a considerable loss of ductility in a sputter-deposited Cu foil with a columnar grain size of 43 nm and a twin spacing of several nanometers [27]. We anticipate that increasing the length of twin lamellae with the grain size is likely beneficial for the ductility of nanotwinned metals. Therefore, a high density of nanoscale twins in a large grain may well be an optimal choice for the microstructure design. However, it is a great challenge to produce a high density of twins (either growth or deformation twins) inside large grains (in the sub-micrometer or even micrometer regime).

In summary, we have demonstrated the twin spacing (size) dependence of strain-rate sensitivity and activation volume in nanotwinned copper. An H–P-type relationship is found between activation volume and twin thickness. This result indicates that TB-mediated dislocation activities play an increasingly dominant role with decreasing twin lamellar thickness. We have addressed the strength and ductility trade-off in terms of the cooperation and transition of the intra- and inter-twin mechanisms. Our findings suggest that the strength and ductility can be further optimized by exploring the microstructure design dimension involving both the grain and twin sizes.

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