#### MATERIALS SCIENCE

## Revealing extreme twin-boundary shear deformability in metallic nanocrystals

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Metals containing abundant coherent twin boundaries (TBs) are able to sustain substantial plastic deformation without fracture due to shear-induced TB migration and sliding. Retaining ductility in these metals, however, has proven difficult because detwinning rapidly exhausts TB migration mechanisms at large deformation, whereas TB sliding was only evidenced for loading on very specific crystallographic orientations. Here, we reveal the intrinsic shear deformability of twins in nanocrystals using in situ nanomechanical testing and multiscale simulations and report extreme shear deformability through TB sliding up to 364%. Sliding-induced plasticity is manifested for orientations that are generally predicted to favor detwinning and shown to depend critically on geometric inhomogeneities. Normal and shear coupling are further examined to delineate a TB orientation-dependent transition from TB sliding to TB cracking. These dynamic observations reveal unprecedented mechanical properties in nanocrystals, which hold implications for improving metal processing by severe plastic deformation.

#### INTRODUCTION

Twin boundary (TB), a predominant planar defect with perfect symmetry and coherent structure, often yields an unparalleled combination of superior mechanical and physical properties in metallic materials (1, 2), including high strength (3-5), well-retained room-temperature ductility (6), improved fatigue resistance (7-9), good electrical conductivity (10, 11), and radiation damage tolerance (12, 13). In particular, the simultaneous high strength and ductility arises from TB-dislocation interactions such as pile-up, transmission, and nucleation (14-17). Because of the strong plastic anisotropy of TBs (18), dislocation activities in nanotwinned face-centered cubic (FCC) metals are typically divided into two hard modes and one soft mode (19). Both hard modes (transverse and confined-layer slips) involve direct intersection of dislocations with TBs, leading to the strengthening of nanotwinned materials (16, 18, 20, 21). In contrast, the propagation of twinning partials nucleated from the grain boundaries (GBs) induces TB migration, leading to the softening of nanotwinned materials (22). At large deformation, detwinning associated with TB migration exhausts the strain hardening capability of nanotwinned materials.

Yet, the inherent deformability of coherent TBs remains largely unexplored, because decoupling this behavior from other geometric factors (e.g., GB confinement) presents significant experimental challenges. Therefore, metallic nanocrystals provide an ideal platform, owing to the controllable TB density, loading orientation, and unconfined geometry (23–25). It has been observed both experimentally and by atomistic simulation that free surfaces can facilitate shearinduced TB sliding and TB density–governed shear localization Copyright © 2021 The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. No claim to original U.S. Government Works. Distributed under a Creative Commons Attribution NonCommercial License 4.0 (CC BY-NC).

(21, 26, 27), which are expected to play critical roles in ductility. However, all evidence of shear-localized TB sliding was obtained by uniaxial tension/compression of metallic micropillars that impose both normal and shear stresses on TBs, whereas the plastic behaviors of coherent TBs under shear loading remain unknown so far.

Here, we report an extreme shear plasticity of coherent TBs in metallic nanocrystals dominated by sliding up to a strain of 364%, using state-of-the-art in situ transmission electron microscope (TEM) nanomechanical testing, supported by atomistic and finite element simulations. Such TB-induced shear deformation occurs on a range of crystallographic orientations beyond the specific cases reported in the literature, including those predicted to favor TB migration, which is due to geometric inhomogeneities and proven to be insensitive to small differences in surface structures. In contrast, conspicuous TB cracking is activated toward orthogonal loading. A comprehensive map of TB deformation is thus established on the basis of the underlying competition between the nucleation of TB dislocation and the evolution of TB cleavage in theory, which matches well with our experiments. Our findings provide atomistic insights into the intrinsic shear deformability of TBs, which holds great promise for developing metallic nanocrystals with extreme mechanical properties.

#### RESULTS

#### Extensive TB sliding under shear loading

A high-quality [111]-oriented Au nanocrystal (diameter of ~20.6 nm) containing orthogonal coherent TBs was fabricated via in situ nanowelding (see Materials and Methods). Shear loading parallel to the TBs, rather than the commonly adopted uniaxial loading (21, 26, 28), was applied to the as-fabricated Au nanocrystal (see movie S1). As shown in Fig. 1A, four parallel TBs (i.e., TB<sub>1</sub> to TB<sub>4</sub>) are delineated by the dashed lines, and several lattice defects preexisted near TB<sub>1</sub>, which arose from the shear deformation that occurred along TB<sub>1</sub> before recording the movie (see fig. S1). Upon shearing, numerous dislocations and stacking faults (SFs) were nucleated from the free surface and propagated parallel to the TBs (Fig. 1B), in stark contrast to the threading/transverse dislocations that dominate in twin lamella under tensile/compressive loading (18, 21). With increased

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**Fig. 1. Extensive shear deformability of a nanotwinned Au nanocrystal under simple shear loading.** (**A**) A [111]-oriented nanotwinned Au nanocrystal under shear loading, where four parallel TBs (TB<sub>1</sub> to TB<sub>4</sub>) existed in the nanocrystal, as delineated by the dashed lines. Shear load was applied to the bottom of this nanocrystal, as shown by the yellow arrow. (**B**) Different slip systems activated between TB<sub>1</sub> and TB<sub>2</sub>, as indicated by the light blue arrows. (**C**) Dynamic twinning and detwinning induced by the Shockley partials gliding on TB<sub>3</sub> and TB<sub>4</sub>. The blue arrow indicates the shear offset along TB<sub>1</sub>. (**D**) Increasing number of atomic steps (red arrows) at the right surface arising from the annihilation of Shockley partials gliding parallel to the TB. (**E**) Elimination of TB<sub>3</sub> and TB<sub>4</sub> after complete detwinning. Slight shear offset (marked by the blue arrow) occurred along TB<sub>2</sub>. (**F**) Large shear deformation before fracture, with a shear offset of 14.4 nm along TB<sub>1</sub>. TB<sub>1</sub> and TB<sub>2</sub> were selected as the references for quantitative analysis since they hardly migrated during the deformation. Scale bar, 5 nm.

shear strain, the nanoscale twins exhibited substantial shear deformability through the shear offset along the TB, as shown by the successive expansion of surface steps at the TB1-surface intersection [marked by the blue arrows in Fig. 1 (C and D)]. Meanwhile, dislocation activities between TB1 and TB2 led to detwinning or left atomic steps at the right free surface (Fig. 1, C and D). After the complete elimination of defects between TB<sub>1</sub> and TB<sub>2</sub> via detwinning (Fig. 1E), TB sliding was accelerated significantly, as evidenced by the dominant shear offset at the TB1-surface intersection (Fig. 1F). A maximum shear strain of approximately 364% was achieved mainly through the sliding of TB<sub>1</sub>, while TB<sub>2</sub> exhibited comparatively limited activities (see discussion S1), which resulted in a marked geometry change before the fracture of this nanocrystal. This geometric change can result in deviation of shear loading away from the TB plane, which may trigger occasional nucleation of threading or transverse dislocations in the nanocrystal.

The deformation mechanism was thereby analyzed at atomic scale to uncover the physical origin of the extensive shear deformability of coherent TBs, as shown in Fig. 2. Theoretically, four slip modes can be activated in nanotwinned FCC metals (19), which

correspond to the TB (full) dislocation, parallel Shockley partial, transverse dislocation, and threading dislocation, as schematically demonstrated in Fig. 2A. Our in situ observations unambiguously revealed these four slip modes upon shearing (see Fig. 2, B to E, and detailed Burgers circuit analysis in fig. S2). The TB full dislocation proceeded via the successive emission of leading and trailing partials on the TB, which led to TB sliding and expansion of surface steps (Fig. 2B). In some cases, the SF bound with leading partial was not instantly removed by the trailing partial, inducing TB migration with a distance of one atomic layer, i.e., twinning or detwinning. Shockley partials gliding parallel to the TB were observed inside nanotwin lamellae as well (Fig. 2C), especially during the initial stage of the shear deformation, and the annihilation of these partials led to the formation or enlargement of surface steps. Fig. 2D shows a typical transverse leading partial emitted from an atomic step on TB<sub>2</sub>, with the trailing SF bound at this step. Detailed structural analysis resolved a full dislocation near this step, which partly released the local stress concentration near the TB step (shown by the evident lattice distortion). Unlike the previous dislocation modes, threading dislocations moving on the inclined {111} planes between



Fig. 2. Atomistic mechanism of extensive shear deformation in Au nanotwinned nanocrystal. (A) Schematic showing the four dislocation modes during the deformation of nanotwins, including TB dislocation (red), matrix Shockley partial (blue), and transverse and threading dislocations (purple). (B to E) Deformation snap-shots showing the structural configuration and dynamic evolution of four types of dislocations. The projected Burgers vectors of each dislocation mode were determined from the Burgers loops in (B) to (D), and the dislocation line is indicated by the white arrows in (E). The red arrowheads indicate the formation or expansion of surface steps after the annihilation of dislocations. (F) Cumulative shear strain that arises from each dislocation mode with increasing loading time. The red, blue, and purple curves exhibit the incremental contributions of TB full dislocation, Shockley partial, and transvers/threading dislocations, respectively. The black dashed curve presents the evolution of total shear strain with loading time. Scale bar, 2 nm.

two neighboring TBs cannot be observed directly in high-resolution TEM (HRTEM). Nevertheless, the projection of the slightly bow-out dislocation segment yields a featured contrast in HRTEM snapshots (18), which can be readily identified (as exemplified in Fig. 2E). Under shear loading, the deformation induced by TB full dislocations and parallel Shockley partials contributes to most of the shear plasticity of nanotwinned crystals. In contrast, the contribution of transverse and threading dislocations to shear plasticity is comparatively limited. This prediction was verified by tracking and quantifying the dislocation activities during shear deformation, as shown in Fig. 2F. In principle, each type of dislocation (Fig. 2A) contributes to a specific amount of shear strain, and the cumulative statistic of each dislocation mode under shear loading can be obtained by tracking the evolution of deformation structures (as schematically shown in fig. S3). Fig. 2F shows that the extreme shear plasticity of nanotwins mainly originated from the gliding and annihilation of TB full dislocations (the original statistics of activated dislocations is presented in fig. S4). Here, the representations of both transverse and threading dislocations (i.e., hard modes) were combined into one category owing to their scarce appearance throughout the shear deformation, in comparison with those gliding parallel to or along the TB. During the

initial shear stage (before 250 s), the contribution of these three categories of dislocations was comparable, and the shear strain increased steadily with deformation. Subsequently, TB full dislocations and Shockley partials started to dominate the shear deformation of the nanotwinned crystal, consistent with the large resolved shear stress parallel to the TB. After complete detwinning (250 to 580 s), shear offset along the TB (corresponding to TB dislocations) became dominant, while Shockley partials inside nanotwin lamellae were seldom activated. At the final stage (beyond 580 s), TB sliding was accelerated markedly (as shown in Fig. 1F), which contributed to ~78% of the extensive total shear strain of approximately 364% in the nanotwinned nanocrystal. Notably, the exceptional TB sliding proceeded steadily despite a marked change in nanocrystal geometry. It underscores the key role of coherent TBs in stabilizing dislocation activities toward extreme shear deformation. In contrast, shear loading on nanoscale Au single crystal without TBs yielded very limited lattice shear offset due to delocalized surface nucleation, which caused dislocations on different slip planes to lock inside the crystal (fig. S5A). Moreover, the shear loading parallel to the (111) plane of Au single crystal often resulted in the formation of nanotwins, followed by TB sliding-dominated deformation (fig. S5B).

#### Geometry dependence of TB sliding

To fundamentally understand the governing factors for extensive TB sliding, molecular dynamics (MD) simulations of shear deformation of prototypical Au nanotwinned nanocrystals were performed, with a particular focus on characterizing the influence of nanoscale geometric inhomogeneities. Here, this atomistic simulation study was motivated by our TEM observations indicating that the nanocrystals did not have a perfectly constant cross section in the vicinity of TBs, due to the inherent zigzag surface faceting promoted during free growth of nanotwinned crystals (21, 29). Fig. 3A1 shows an ideal cylindrical model of Au nanocrystal containing two TBs, with a similar size as that in our experiments in Fig. 1. The superimposed atomic von Mises stress (Fig. 3A2) demonstrates a uniform stress distribution axially along the side surface after energy minimization at 0 K, where no stress concentration is evidenced at the TB-surface junctions before shear loading. Under shear loading along [101], exceptional shear offset was induced via TB sliding (Fig. 3A3 and movie S2), which is confined to only one of the parallel TBs with more intense step evolutions, rather than distributed uniformly among multiple TBs, in agreement with our experimental observations. This localized TB sliding is proven to retain with different TB densities as well (fig. S6, A and B). Moreover, the large shear offset (Fig. 3A) can be fully recovered under reversed shear loading (fig. S6C). Quantitative analysis further reveals a transformation of TB-mediated deformation mechanism with in-plane shear orientation in which TB sliding and migration dominate under (101) and (112) shear loadings,

respectively (see discussion S2). The underlying physical origin relies on the Schmid factors for leading and trailing partials on the TB, which is consistent with previous reports in uniaxial tension/compression (27). The simulated engineering stress-strain curves under [101] and [112] shear loadings exemplify the fundamental difference in mechanical behaviors between the two mechanisms (see fig. S7). In addition, we explored the potential effects of SF energy by performing MD simulations on nanotwinned Ni and Al nanocrystals with the same geometry. Nearly identical shear deformation through TB sliding prevailed in these metals with much higher SF energies (fig. S8).

Beyond an ideally smooth and cylindrical surface, facets and inhomogeneous cross-sectional variations that commonly exist in nanocrystals can induce high surface stress concentrations, reducing the energy barrier for defect nucleation (*30*). MD simulations of simple shear deformation of Au nanotwinned crystals with smooth zigzag (111) surface facets are shown in Fig. 3 (B and C), where each facet is connected to two coherent TBs. Fig. 3B1 indicates that the nanotwinned crystal subjected to shear loading along the [112] direction favors TB migration. After relaxation and before shear loading, larger atomic stress concentrations (~4.69 GPa) are present at the TB-surface intersections, as shown in Fig. 3B2, which is known to balance the residual stresses between the crystal surface and interior. The stress concentration on the surface facilitates the dominant TB migration with extensive detwinning under [112] shear loading (shown in Fig. 3B3 and movie S3), which is consistent



**Fig. 3. Geometry dependence of shear deformation of nanotwins by TB sliding versus detwinning.** (A1 to A3) MD simulation snapshots showing the dominant TB sliding in an ideal cylindrical nanotwinned Au under [101] shear loading. (A2) Distribution of atomic-scale von Mises stress after energy minimization. (B1 to B3) Deformation of nanotwinned Au with zigzag surfaces under [112] shear. (B1) The surfaces of nanotwinned samples consist of conjugate {111} facets. (B2) Distribution of atomic-scale von Mises stress after energy minimization, where substantially higher atomic stresses appear on the TB-surface intersection sites (shown by the red arrows). (B3) TB migration and resultant detwinning under [112] shear. (C1 to C3) TB sliding under [112] shear in a nanotwinned Au with nonuniform cross sections. (C1) The central twin lamella was enlarged by 2 times along [110] direction and by 1.5 times along [112] direction. (C2) Substantially higher atomic stress of ~5.94 GPa at the TB-surface intersection sites, as indicated by the red arrows. (C3) Extensive sliding along TB<sub>2</sub> and TB<sub>3</sub> in association with detwinning in the top and bottom twin lamellae.

with the theoretical prediction of deformation mechanism based on homogeneous surface without facets. When the shear loading direction was changed to [101] direction, extensive TB sliding occurred (see fig. S9). Therefore, TB-mediated deformation mechanisms are relatively insensitive to surface faceting as long as the crystal cross section is constant.

Unexpectedly, however, we found that a nonuniform change in cross section along the axial direction near TBs (as commonly observed experimentally) produced a higher stress concentration at the TB-surface intersection, resulting in a definitive shift of deformation mechanism from TB migration to TB sliding under [112] shear loading. Specifically, we have modeled a nanocrystal with identical twin and surface facet structure as that in Fig. 3B1, except that the cross section of the central twin lamella was enlarged by 2 times along [110] direction and by 1.5 times along [112] direction, as shown in Fig. 3C1. This approach resulted in a substantially higher atomic stress of ~5.94 GPa at the TB2-surface intersection across different-sized lamella (Fig. 3C2). Consequently, extensive sliding along TB<sub>2</sub> was induced under [112] shear (Fig. 3C3 and movie S4). We attribute this unexpected shift from TB migration to TB sliding to the abrupt geometry variation that confines the dislocation activities along the TB and significantly increases the energy barrier for TB migration in the axial direction of the crystal. The unpredicted TB sliding with [112] shear loading was still found to occur when the geometric inhomogeneity was moved to the top or bottom TBs, as shown in fig. S10. These findings, therefore, strongly suggest that TB sliding can be stimulated under most crystallographic directions where local geometric inhomogeneities are present, including those otherwise predicted for TB migration in ideally smooth specimens (discussion S2).

#### Coupling between normal and shear loading

In addition to in-plane shear, the effects of the out-of-plane loading orientation were investigated to gain in-depth insights into the plastic deformation of nanotwins. The TB sliding-mediated shear offset acted as a general deformation mode when increasing the slanted angle  $\varphi$  (the angle between TB and loading direction) from 0° to 21°, 36°, 49°, and 52° (see Fig. 4 and fig. S11), respectively. The deformation snapshots in Fig. 4 (A and B) show extensive shear offset before fracture at  $\varphi = 0^{\circ}$ , which is consistent with the shearinduced TB sliding in Fig. 1. Similar TB sliding-mediated deformation of nanoscale twins occurred at  $\varphi = 21^{\circ}$  (fig. S11A). When  $\varphi$  was increased to 36°, combined TB migration and sliding were activated sequentially, as shown in Fig. 4 (C and D) (the deformation process of the entire Au nanocrystal is presented in fig. S11B). Likewise, the TB-dominated plasticity was unambiguously presented in nanoscale twins at  $\varphi = 49^{\circ}$  and  $\varphi = 52^{\circ}$  (fig. S11, C and D). A coherent TB enables exceptional shear deformation even in single crystals under uniaxial tensile loading. As exemplified in an [001]-oriented Au single crystal (fig. S12), lattice sliding along the (111) plane was activated first, which simultaneously induced the formation of a nanotwin near the surface. Subsequent tensile deformation was accommodated by stable TB sliding, which resulted in a pronounced shear offset nearly three times larger than that via lattice sliding.

Further increase in the slanted angle, however, gives rise to cracking along the TB, as demonstrated by the nanotwinned crystal with a slanted angle of 78° in Fig. 4 (E to G) and movie S5. Under the longitudinal uniaxial tensile loading, a premature crack was

nucleated from the TB-surface intersection site (Fig. 4F) and propagated rapidly along the TB as the deformation proceeded (Fig. 4G). Similar brittle behaviors have been reported in nanotwinned nanowires with  $\varphi = 90^{\circ}$  that, however, are combined with the effects of extremely high TB densities (21, 23). To rationalize the TB orientation dependence of the deformation mechanism transition from TB sliding to cracking with orthogonal loading, we propose a simplified model based on an individual TB and a preexisting crack on the TB plane (see Materials and Methods). A theoretical Peierls shear factor  $\chi$  is defined accordingly to predict the propensity for TB shearing versus cracking from the competition between dislocation nucleation ahead of the crack tip and direct crack propagation along the TB, which is quantified as follows (31)

$$\chi = \frac{2(1-\nu)m^2}{(1-\nu)(1+m^2)+n^2} \frac{\gamma_s}{\gamma_{usf}}$$

where v is the Poisson's ratio;  $m = K_{II}/K_I$ ,  $n = K_{III}/K_I$  ( $K_I$ ,  $K_{II}$ , and  $K_{III}$ ) are the stress intensity factors associated with mode I, II, and III cracks, respectively);  $\gamma_s$  and  $\gamma_{usf}$  denote the surface energy and unstable SF energy of Au, respectively (see discussion S3). The crack tip in our analysis acts as a stress concentrator to initiate dislocation nucleation or cracking, which generally exists in real nanocrystals due to the presence of surface roughness or structural inhomogeneity. On the basis of this model, a comprehensive shear deformation map that addresses the transition of TB-mediated deformation mechanism can be established, as shown in Fig. 4H, which is verified by experimental observations of various nanotwinned nanostructures. Finite element analysis (FEA; see Materials and Methods and discussion S3) quantifies the changes in *m* and *n* values at the crack tip with the out-of-plane loading orientation (i.e., slanted angle  $\varphi$ ), which demonstrates a parabolic trend of  $\chi$  with the loading orientation. When  $\chi > 1$ , dislocation activities act as the predominant deformation mode ahead of the crack tip; however, when  $\varphi$  increases to cause a level of  $\chi < 1$ , TB cracking becomes predominant, which is consistent with our experimental observations (Fig. 4, E to G). To quantify the loading orientation effects on the shear deformability of nanoscale twins, experimentally observed TB sliding behaviors in different nanotwinned metals are normalized using the sliding distance versus the total TB length (denoted as normalized shear offset), as summarized in Fig. 4H. Here, the blue marks represent the shear deformation outcomes in our work, while those with other colors document the previous reports on different FCC nanostructures including Cu nanopillars (27) and Au or Cu nanowires (21, 26). We can see that the maximum shear offset often occurs under pure shear loading along the TB ( $\phi = 0^{\circ}$ ), which diminished gradually with the increasing slanted angle of the TB. Associated with the reduced tendency of TB sliding was the increasing contribution of transverse and threading dislocations to the deformation (18). Near  $\phi \sim 45^\circ$ , however, the contribution of TB sliding became important again due to the large resolved shear stress on TB. At even larger slanted angles ( $\phi \sim 78^\circ$ ), TB cracking dominates the deformation of nanotwinned Au crystals, which is consistent with our theoretical prediction of  $\chi < 1$  when  $\phi > 78^{\circ}$ . This TB cracking is similar to the brittle-like fracture in  $\langle 111 \rangle$ -oriented ( $\phi \sim 90^{\circ}$ ) nanotwinned nanowires/pillars (21, 23).



**Fig. 4. Loading orientation dependence of TB-mediated plasticity.** (**A** and **B**) TB sliding dominated deformation of nanotwinned Au nanocrystal under shear loading, with a slanted angle  $\varphi$  of 0°. (**C** and **D**) TB sliding accompanied with TB migration in a nanotwinned Au under uniaxial tensile loading with  $\varphi = 36^\circ$ . The white dotted line delineates the initial position of TB<sub>1</sub> in (C), and the red arrows indicate the step evolution associated with TB sliding. (**E** to **G**) TB cracking in a nanotwinned Au nanocrystal with  $\varphi = 78^\circ$ . A crack nucleated from the TB-surface intersection and then propagated along the TB. (**H**) Deformation map summarizing the loading orientation–dependent (illustrated by the inset schematics) shear factor ( $\chi$ ) of nanotwinned Au nanocrystal and the deformation mechanisms of different FCC nanotwinned structures (*21, 23, 26, 27*). The red curve shows the parabolic trend of shear factor  $\chi$  with respect to the loading orientations. The shear deformability among different nanostructures in Au and Cu is quantitatively characterized by the normalized shear offset, which is defined as the TB sliding distance divided by the total TB length. Two competitive deformation mechanisms including TB sliding and TB cracking (shown by the triangular and diamond marks) dominate over different ranges of loading orientations. NT, NW, and NP denote nanotwin, nanowire, and nanopillar, respectively. Scale bars, (A and C) 2 nm and (E) 5 nm.

#### DISCUSSION

Despite the widely discussed deformation mechanisms in nanotwinned metals and nanotwinned metallic nanowires/nanopillars under uniaxial loading (18, 19, 21, 23), the intrinsic shear deformability of nanoscale twins has been rarely investigated. Taking advantage of the successful development of in situ shear testing and controlled preparation of nanotwinned metallic nanocrystals, we revealed an extreme shear deformability of nanoscale twins via the stable sliding along TB. Note that this extreme shear deformability was enabled by the elimination of inevitable geometric constraints on TBs using simple shear loading of nanotwinned nanocrystals, which cannot be realized by conventional uniaxial loading. Moreover, the shear and normal deformation mechanisms are readily decoupled in the present study. In essence, TB sliding is governed by the consecutive nucleation and propagation of Shockley partial pairs on TB, where the TB moves back and forth by one atomic layer along the plane normal. Since the lattice strain from the leading partial can always be removed by the propagation of the trailing partial, no evident stress accumulation or dislocation entanglement appears in the nanocrystal, which is in contrast to that in nanoscale single crystals with limited plasticity under shear loading (fig. S5A). As a result, a maximum shear strain of approximately 364% was mainly realized (Fig. 1) in which TB sliding accounted for approximately 78% of the total strain, substantially pushing the shear deformability to extreme. Extrinsically, the maximum amount of TB sliding is determined by the local geometry change (e.g., cross-sectional area) of nanoscale twins and the loading orientation, since the deviation of shear loading from the TB at latter stage may activate other deformation modes (particularly, transverse slip) and thus impair the continuous shear deformability. Nevertheless, the large shear offset reflects the continuous and extreme deformability of TBs by sliding, which indicates

an unparalleled accommodation of shear plasticity for nanotwinned metals. In metallic single crystals, coherent TBs are also proven to facilitate extensive shear deformability by preferential nucleation of nanotwins before substantial sliding or shear offset under either shear or uniaxial loadings (figs. S5B and S12).

With regard to the diverse geometries commonly seen in nanocrystals, we explored the two key factors of surface facets and shape homogeneity. Unexpectedly, TB sliding appears to be insensitive to surface morphology (smooth or with zigzag facets), which validates the theoretical predictions of shear deformation mechanisms on the cylindrical model with ideally smooth surfaces (see discussion S2). In contrast, local TB area inhomogeneity seems to play a critical role in the deformation of irregularly shaped nanotwinned crystals, where dislocation activities can be effectively confined within the TB plane. This phenomenon can be attributed to geometric constraints imposed by the sudden change of nanocrystal cross sections, as well as the marked increase of true shear stress on the TB plane, leading to TB sliding even under loading directions that were supposed to favor TB migration in view of the Schmid factors. The TB-dominated shear deformability revealed by combined in situ testing and atomistic simulations in this work advances our current understanding on the critical role of coherent TBs in accommodating large shear plasticity in metallic nanocrystals, even with different SF energies (fig. S8).

Besides the exceptional TB sliding under shear loading, our in situ testing demonstrates similar TB-dominated plasticity in metallic nanocrystals over a wide range of loading orientations ( $\phi$ ), which governs the overall deformation mechanism of metallic nanocrystals. Theoretically, the out-of-plane TB orientation  $\phi$  mainly governs the competition between twinning partials and transverse/threading dislocations that interact with TBs. It is widely acknowledged that twinning partials lead to TB-dominated plasticity (32, 33), while transverse/threading dislocations intersecting TBs predominantly contribute to the strengthening of nanotwinned materials (5, 21). Therefore, this transition of deformation mechanisms can critically influence the mechanical properties of nanotwinned materials. Our experimental investigations show that TB sliding can be activated in a wide range of loading orientations, as summarized in the deformation map in Fig. 4H. The maximum shear deformability was realized at  $\varphi = 0^{\circ}$  (i.e., under shear loading), and the increasing  $\varphi$  leads to an intensified competition between TB sliding and transverse/ threading dislocations. The diminished sliding tendency associated with the increasing  $\phi$  arises from the larger Schmid factors of transverse and threading dislocations that nucleate from TB or free surface (18). These dislocation modes can effectively dissipate the accumulative shear strain for the continuous TB sliding, and only a small amount of TB sliding retained (Fig. 4, A and B). When  $\varphi$ keeps increasing to ~45°, TB sliding was reactivated due to the rebounding Schmid factors toward a local maximum of 0.5. Similar TB sliding was also reported in nanotwinned FCC metallic nanowires (26, 28) and nanopillars (27) with a larger diameter range of 100 to 200 nm and different SF energies. However, our findings differ from the very specific cases for TB sliding predicted in the literature, because the stress concentration present at the TB-surface intersections is found to be a critical factor that governs TB sliding beyond the slip geometry or nanocrystal size (27, 28). When approaching the upper limit of loading orientation (i.e., orthogonal loading), however, TBs serve as preferential pathways for cracks that nucleated from the TB-surface intersections. This phenomenon arises from the deformation mode transition from dislocation activities ahead of the crack tip to rapid crack propagation along the TB, which is quantitatively characterized by our theoretical model beyond the critical loading orientation of 78°. Compared with previous reports on nanowires/pillars with orthogonal TBs ( $\varphi = 90^{\circ}$ ) (21, 23), our experimental evidence at  $\phi \sim 78^\circ$  (Fig. 4, E to G) extends the orientation range for TB-assisted brittle fracture, which indicates an earlier ductile-to-brittle transition in nanotwinned metallic nanostructures.

Although our in situ experiments and MD simulations were carried out on nanoscale metals, the impact of these findings could probably be extended to the behavior of bulk nanotwinned materials. For example, TB sliding can be expected during the coordinated shear deformation across multiple grains subjected to friction, given that GBs serve as nucleation sites for deformation twins (34, 35). Nevertheless, the amount of TB sliding should be significantly reduced due to the geometric constraints imposed by neighboring grains, whereas coherent TBs inside grains are able to accommodate shear deformation by providing different pathways for dislocation motion (36). Moreover, the nanotwins can deflect intergranular cracks via TB sliding (see an example in fig. S13), which could potentially impede catastrophic intergranular fracture. Consistent with TBdominant deformation in metallic nanocrystals, TB cracking in bulk bicrystals is critically dependent on loading direction during cyclic loading as well (8). The correlation between loading orientation and TB cracking obtained in metallic nanocrystal should presumably be retained in nanotwinned materials, although the localized loading condition in bulk polycrystalline materials becomes increasingly complex. Additional work is required to quantitatively explore the TB shear behaviors and the corresponding load condition effects in nanotwinned bulk materials.

In conclusion, we have uncovered an extreme shear deformability of nanoscale twins, which is dominated by dislocation-mediated TB sliding. Through systematic experimental and theoretical investigations, we revealed that TB-dominated plasticity is the most efficient under shear loading and remains highly effective for a wide range of loading orientations, until the TB cracking dominates over dislocation activities on TBs beyond the loading orientation of approximately 78°. An accessible deformation map has been established to clarify the transition of intrinsic deformation mechanisms of coherent TBs in metallic nanocrystals, which opens new avenues for predicting and tuning the deformation behaviors of nanostructured metals and for materials processing by severe plastic deformation in general.

#### **MATERIALS AND METHODS**

#### In situ TEM nanomechanical testing

Fabrication and in situ nanomechanical testing of the nanotwinned Au nanowires were conducted inside a FEI F20 field-emission gun TEM, equipped with a PicoFemto TEM electrical holder from Zeptools Co. In a typical experiment, two Au rods (with a diameter of 0.25 mm) were fractured by a wire cutter to obtain nanoscale tips on the fracture surface, which were loaded onto the static and probe sides of the TEM-STM (scanning tunneling microscope) holder, respectively. During experiment, the Au STM probe was manipulated by a piezo-controller to contact a nanoscale tip on the fracture surface of the Au rod on the static side. An Au nanocrystal was fabricated by nanowelding, where a voltage potential of 1.5 V was preapplied to the STM probe to improve the weldability. Nanotwins with different orientations can be induced by this nanowelding procedure. In situ shear and uniaxial testings were realized by steadily moving the Au STM probe laterally or backward with an average strain rate of  $\sim 10^{-3}$  s<sup>-1</sup> using the piezo-controller. The TEM was operated at 200 kV with low beam intensity to minimize the beam effects on deformation, and the structure evolutions of metallic nanocrystals were recorded by a charge-coupled device in real time with a frame rate of  $\sim 0.3$  s per frame.

#### **MD** simulations

MD simulations were carried out on an Au nanotwinned model with a total of ~150,000 atoms using the Large-scale Atomic/Molecular Massively Parallel Simulator (37) and embedded-atom-method potentials for Au (38). The overall size of the cylindrical sample without surface facets was about 13.6 nm in diameter and 15 nm in height. The uniform samples with zigzag facets were set with a width/height/thickness of 21.6 (22.5) nm by 15.5 nm by 16.2 (16.0) nm for (112) ((110)) shear, while a twin lamella in the middle of the nonuniform sample was enlarged to a width and thickness of 31.9 and 32.1 nm, respectively. The matrix/twin/matrix structure was constructed by stacking (111) atomic layers from which 20 atomic layers were set for the twin. Periodic boundary conditions were applied along the longitudinal direction, while the radial directions were set free. During the shear deformation, a constant shear velocity  $v = 1 \text{ ms}^{-1}$  parallel to the TB plane was applied on the fixed area of the bottom grain at 300 K while fixing a few atom layers at the top of the nanocrystal, and a typical MD time step of 1 fs was used. The common neighbor analysis in the visualization software Ovito (39) was used for the structure of the nanotwinned models. FCC atoms were removed, while hexagonal close-packed atoms at the TB and the others on the surface were marked in yellow and blue,

respectively. Dislocation extraction analysis was adopted to identify different dislocation configurations during deformation. The distribution of atomic von Mises stress of nanotwinned Au was obtained by color coding in Ovito after energy minimization at 0 K to avoid thermal disturbance of atoms. The higher level of atomic stresses across the TB was obtained after relaxation, by calculating the numerical average of the von Mises stress of each atom along the atom column at the TB-surface intersection. The atomic von Mises stress was derived by {[( $\sigma_x - \sigma_y$ )<sup>2</sup> + ( $\sigma_y - \sigma_z$ )<sup>2</sup> + ( $\sigma_z - \sigma_x$ )<sup>2</sup> + 6( $\tau_{xy}$ <sup>2</sup> +  $\tau_{yz}$ <sup>2</sup> +  $\tau_{zx}$ <sup>2</sup>)]/2}<sup>1/2</sup>/atom volume. To clearly demonstrate the stress concentration at the TB-surface intersections in zigzag models, the cutoff von Mises stress in the map was set as 4 GPa. In all simulations, temperature and pressure were controlled by Nose-Hoover and Parrinello-Rahman methods, respectively.

#### Finite element analysis

The FEA software package ANSYS was used to estimate the stress intensity factors (K) at different loading orientations ( $\varphi$ ) with the assumption that a preexisting crack (due to the presence of a typical surface facet) is available on the TB. A cylindrical model with a diameter of d and a height of 2.65d was created to mimic the geometry of our experimental samples. A sharp edge crack with a depth of  $5 \times 10^{-2} d$ and a height of  $2.5 \times 10^{-3} d$  was implanted on the symmetric plane of the cylinder (fig. S14). The upper surface of the crack was set to be horizontal and coincides with the TB. A local coordinate system was established at the center of the crack tip to facilitate the analysis of the stress concentration factors. The material is assumed to be isotropic for convenience, and the material constants for Au are shown in table S1. Solid186 elements and the curvature size function were used to generate the mesh. To make the results more accurate, elements with sizes as small as  $7.5 \times 10^{-3} d$  were applied in a spheroidal area with a radius of 0.4d from the crack tip. The maximum size of the elements far away from the crack tip is chosen as 0.15d. The crack was analyzed as a premeshed crack. The top end of the model was fixed by applying the fixed support in ANSYS, where no displacement was imposed to the mass points on this surface. Homogeneous loading with different orientations was applied to the free-standing bottom end. The magnitude of the tensile loading was 10 MPa for every loading orientation ( $\varphi$ ). The fracture tool in ANSYS was used to output the stress intensity factors  $K_{\rm I}$ ,  $K_{\rm II}$ , and  $K_{\rm III}$  at the crack tip.

#### SUPPLEMENTARY MATERIALS

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# **Science**Advances

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