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Dislocation characterization in fatigued Cu with nanoscale twins

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Previous studies have shown that strain-controlled cyclic stability was maintained in bulk Cu samples with highly oriented nanoscale twins. In order to explore the underlying fatigue mechanism, transmission electron microscopy observations under two-beam diffraction condition were utilized to characterize the dislocation configurations in the twin/matrix layers of as-fatigued nanotwinned Cu. It was clarified that the threading dislocations with Burgers vector parallel to twin boundaries are mainly active during fatigue. A three-dimensional stereo projection was re-configured for demonstrating the special structure of dislocations in nanoscale twins.

Nanotwinned metallic materials have attracted widespread attention due to their unique tensile properties, such as high strength, considerable ductility, enhanced work hardening and so on [1–4]. From view of engineering application, apart from tensile properties, fatigue properties and cyclic deformation of nanotwinned materials are of vital importance. Actually, investigations on the fatigue properties and cyclic deformation of nanotwinned metals under cyclic loading were carried out gradually in recent years [5–8]. Stress-controlled tension-tension fatigue tests of nanotwinned Cu (nt-Cu) film synthesized by magnetron sputtering have shown that their high-cycle fatigue life and fatigue limit are greatly improved over that of coarse grained Cu (CG-Cu) [5]. A vast amount of the original nanoscale twin boundaries (TBs) were unstable and disappeared after fatigue [6]. Instead, numerous dislocation walls formed. The soft de-nanotwinned regions led to surface depressions, which acted as sites for crack initiation.

Bulk nt-Cu samples containing highly aligned nanoscale twins were synthesized by means of a direct-current electrodeposition technique and their low-cycle fatigue properties have been investigated in tension-compression fatigue tests under strain control [9]. The low-cycle fatigue life of nt-Cu was comparable with that of CG-Cu, longer than that of ultrafine/nanograined (UFG/NG) metals [10,11]. More interestingly, cyclic stability with the stable hysteresis loops is maintained in nt-Cu, which is fundamentally distinct from cyclic softening of UFG [12,13] and cyclic hardening of CG counterparts [14–16]. Furthermore, the stable microstructure of nt-Cu under strain-controlled fatigue is also different from the unstable structures associated with detwinning in nt-Cu films [6,7].

Schmid factor analysis reveals that threading dislocations with the Burgers vector parallel to TBs but the slip plane inclined to TBs would be preferentially activated in the nanoscale twin lamellar channels when the twin plane is parallel to the loading axis [8,9]. However, the direct characterization of the threading dislocation by the conventional bright/dark field image under transmission electron microscope (TEM) is quite difficult. On one hand, no trace of threading dislocations will be left behind on coherent TBs, due to their Burgers vector parallel to TBs. On the other hand, threading dislocations within twin/matrix layers at nanometer scale will overlap with the edge-on coherent TBs and become invisible in TEM under a perfect [110] incident electron beam. This is because threading dislocations are severely curved under the confinement of the twin/matrix layer at nanometer scale. In this study, the Burgers vector and morphology of threading dislocations in strain-controlled polycrystalline Cu with highly oriented nanoscale twins will be specifically characterized with an unique observation direction with respect to TBs by means of TEM analysis, i.e. under two-beam diffraction condition.

Bulk (~3.4 mm in thickness) high-purity copper plates containing highly oriented nanoscale growth twins were synthesized by a direct-current electro-deposition technique in an electrolyte of $CuSO_4$. Microstructural observations indicated that the as-deposited nt-Cu sample was composed of numerous columnar-shaped grains separated by clear grain boundaries (GBs). The grain size displays a wide distribution from 1 to 20 μm, with an average value of ~7 μm, which is approximately an order of magnitude

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larger than that of magnetron sputtered nt-Cu films [5–7]. The columnar grains are embedded with the twin lamellae which are roughly perpendicular to the growth direction. The twin lamellae thickness has a broad distribution from several tens to ~500 nanometers, but majority of them are in the nanometer scale, with an average value of ~75 nm. More details about the sample preparation and microstructures of the nt-Cu sample are described in [8,9]. Tensile tests reveal that nt-Cu sample displays a yield strength (σ_y) of 302 MPa and an ultimate tensile strength (σ_{uts}) of 360 MPa, much higher than that (σ_y ~ 69 MPa, σ_{uts} ~160 MPa) of twin-free CG-Cu with an average grain size of 9 μm.

The fatigue specimens with a gauge cross-sectional area of 4 mm × 3.4 mm and a length of 12 mm were cut from nt-Cu plates by an electrical spark machine. Uniaxially symmetric tension-compression tests under plastic strain control were performed on an Instron 8874 testing machine by using a dynamic strain gauge extensometer with a gauge length of 10 mm at room temperature. The cyclic loading axis was roughly parallel to the majority of the twin planes. It should be noted that the fatigue test was terminated when the cyclic stress amplitude dropped 70% relative to the largest stress amplitude. In this case, cyclic stability with a stress amplitude of 325 MPa is maintained in nt-Cu at the uniaxial plastic strain amplitude ($\Delta\varepsilon_{pl}/2$) of 3.16×10^{-3}, which is distinct from cyclic hardening of CG-Cu from 66 MPa at the first cycle to 165 MPa at the 700th cycle. Moreover, the final fatigue life of nt-Cu is approximately 1500 cycles, much longer than that (~770 cycles) of CG-Cu.

The dislocation features of the as-fatigued nt-Cu were studied by an FEI Tecnai F20 TEM operated at 200 kV. A two-beam diffraction technique in TEM [10,17] was used to accurately characterize the Burgers vector of the dislocations in the twin interiors.

Fig. 1 reveals a general bright-field TEM image of a deformed grain (far away from the GBs) of the nt-Cu with 1500 cycles. The confined threading dislocations within nanoscale twins cannot be detected due to most parts of them overlapping with the edge-on TBs under a perfect [110] incident electron beam, hence Fig. 1 was taken under the incident electron beam slightly deviated from [110] to present the dislocations clearly. Clearly, the TBs after fatigue are intact, straight and mutually parallel, which are obviously different from the numerous areas of detwinning and grain coarsening in fatigued nt-Cu films [6,7]. No dislocation patterns, such as persistent slip bands (PSBs) or dislocation cells are observed in twin interiors. Instead, numerous dislocations are universally detected. Those dislocations consist of short bowing out heads and long tails in the length of 90–150 nm, which are similar with the hairpin dislocations observed in nanoscale thin films and multilayer materials [18,19].

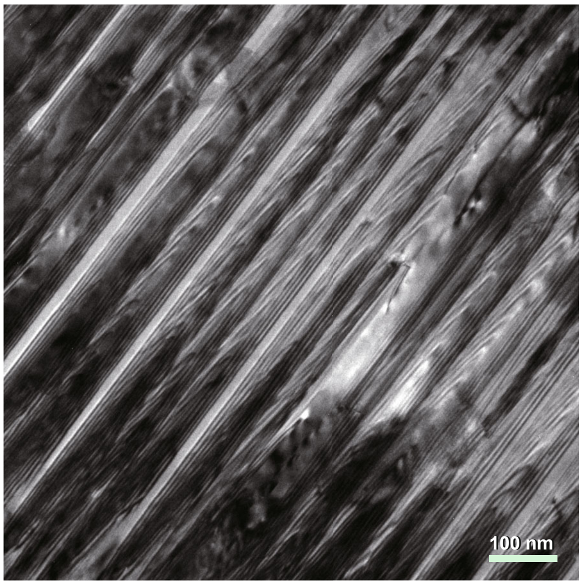

Figure 1 Cross-sectional TEM image of a fatigued grain (far away from the GBs) of nt-Cu samples with 1500 cycles in the slight deviated [110] incident electron beam, showing numerous dislocations with long tails confined by the TBs.

Subsequently, a two-beam diffraction technique in TEM [10,17] was adopted to identify the Burgers vector (*b*) and character of dislocations in the twin interiors, based on the extinction rule of the dislocation which is visible under $g{\cdot}b = \pm 1$, and vice-versa [17]. The dislocation features in the nanoscale twins are similar under $g = [-111]_M$ and $[-200]_M$ from matrix ($[-111]_T$ and $[-200]_T$ from twin). So the dislocation features in nanoscale twins under $g = [-111]_M$ are shown in Fig. 2a as an example. Under $g = [-111]_M$, the twin planes, which are inclined to the incident electron beam, are projected to be the regions with moiré fringe, as indicated by the arrow in the white square 1. Typical short and straight dislocation segments overlapping with the moiré fringes are prevalently seen, which suggests that they are located in the vicinity of TBs. These dislocation segments in the vicinity of TBs are parallel to each other, implying that they probably belong to the same single-slip system during cyclic loading.

In addition, as denoted by the black triangle in square 2, some slightly curved dislocation segments are also detected in the middle of twin/matrix interiors. They are nearly perpendicular to TBs or at a large inclined angle with respect to TBs. Thus, there are two types of dislocation segments in the twin interior and in the vicinity of TBs. Nevertheless, these dislocation segments become invisible under $g =$

$[111]_{M/T}$ either in matrix (M) or in adjacent twin lamellae (T), as shown in Fig. 2b. Based on the extinction rule of the dislocation [17], it is indicated that the Burgers vector of these two types of dislocation segments is parallel to TBs.

Cross-sectional scanning electron microscopy (SEM) observations of the as-fatigued nt-Cu samples have revealed that the slip bands are mutually parallel to each other in the grain interiors, but inclined to TBs [8,9]. The slip bands on both sides of TBs are mirror symmetric relative to TBs, which implies that they come from a single slip system with the slip planes inclined to TBs. Both SEM and TEM observations under two-beam diffraction conditions definitely demonstrate that the dislocation segments in nt-Cu belong to the slip system with Burgers vector parallel to TBs but the slip plane inclined to TBs.

However, the scenario of the short dislocation segments in the vicinity of TBs and in the twin interior in Fig. 2 is clearly distinct from the hairpin-like dislocations with long tails in Fig. 1. The lengths of dislocation segments in Fig. 2 are only 30–60 nm in average, which is much shorter than those (90–200 nm) in Fig. 1.

In order to further clarify whether dislocations in Figs 1 and 2 are consistent, a three-dimensional (3D) stereo projection of the whole threading dislocations in the nanoscale twin is re-configured, by means of simulating TEM images under two-beam diffraction conditions. Under the stereo projection rule with g_M, the twin planes are projected to be the grey regions, rather than a sharp boundary in the TEM image, like the moiré fringes observed in Fig. 2a. Due to the severe confinement by the nanoscale twins, the threading dislocations in the lamellar channels possess curved heads and relatively long tails, which are postulated to be parallel to Burgers vector (indicated by *b* and an arrow) and TBs. However, only part of threading dislocations can exist in the nanometer-thick TEM foils because the grain size of nt-Cu is in the micrometre scale. Most possibly, partial tails of the threading dislocations, i.e. the blue solid line on the slip plane in Fig. 3, can be detected in the TEM sample, comparing with the nonexistent short heads (denoted by the dashed lines on the slip plane in Fig. 3). Under simulated two-beam diffraction condition with g_M, the tails are projected to be short, straight and parallel segments (i.e. the blue solid line) overlapping with the grey regions in TEM image, while the heads are projected to the dislocation segments in the middle of the twin interior (as indicated by the red solid line in Fig. 3), both of which are in accordance with the segments in Fig. 2.

The length of the dislocation segments observed in the TEM image with g_M depends on the thickness of TEM samples and the projection direction relative to the slip plane (i.e. the degree of tilting TEM foil). For example, the larger tilt angles in the thinner TEM foils, the shorter dislocation segments will be observed. This could explain why the dislocation segments in Fig. 2 are much shorter than that in Fig. 1. Consequently, above TEM images as well as the 3D stereo projection analysis further confirm that single-slip system dislocations with Burges vector parallel to TBs but slip direction inclined to TBs are preferentially activated in the nt-Cu samples when twin planes are parallel to the cyclic loading axis.

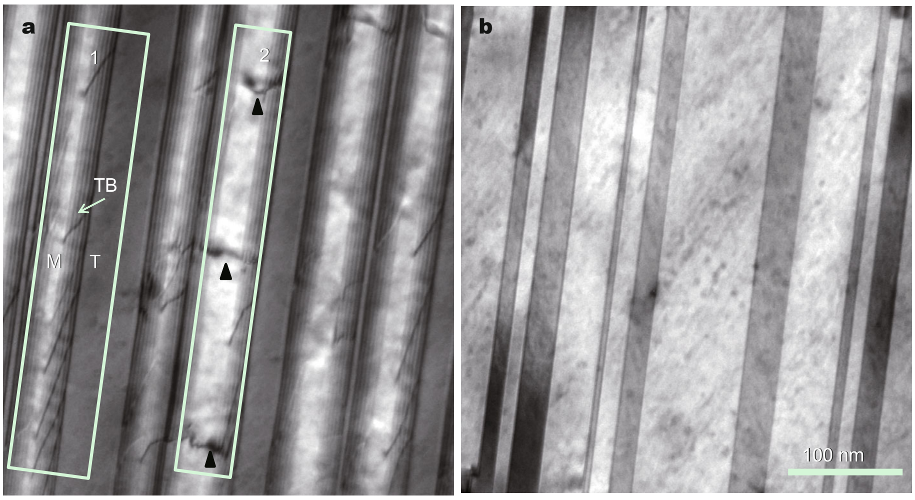

Figure 2 Cross-sectional bright-field TEM images of nt-Cu samples with 1500 cycles under two-beam conditions (a) with $g = [-111]_M$, showing the twin planes are projected to be the regions of moiré fringe. Numerous short dislocation segments in the vicinity of TBs which are parallel and overlapped with the moiré fringes are shown in square 1 and some dislocation segments in the middle of twin interiors, nearly perpendicular with the TBs are shown in square 2. (b) All dislocation segments in (a) are invisible with $g = [111]_{M/T}$.

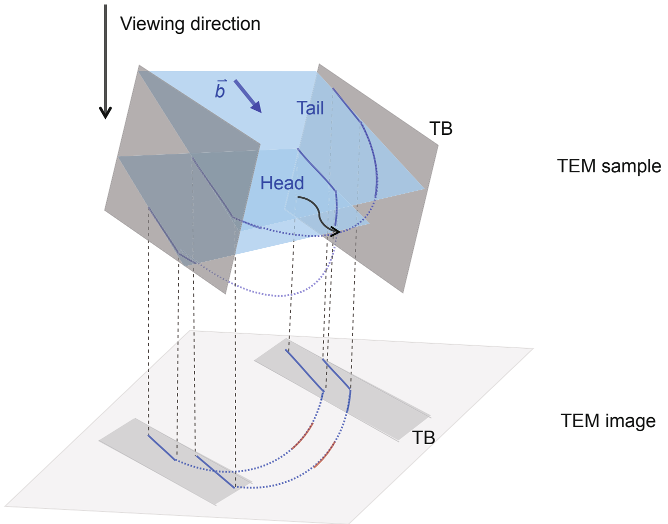

Figure 3 Schematic illustration of a 3D stereo projection for the whole threading dislocations in nt-Cu. The Burgers vector of the dislocations is indicated by *b* and an arrow. In the TEM sample, the blue solid line on the slip plane between the twin planes (dark grey color) denotes the existing dislocation segments, while the short dashed line denotes the nonexistent part. In the TEM image, the blue solid line overlapping with TBs (grey color) denotes the visible part while the short dashed line denotes the invisible part under g_M.

Usually, cyclic stress response of metallic materials during fatigue depends on their cyclic deformation mechanism. For instance, cyclic hardening is pervasively observed in CG metals as a result of the multiple-slip induced dislocation cells [9,14] while cyclic softening occurs in UFG/NG metals due to the unstable microstructure and abnormal grain coarsening/shear banding [12,13]. Even though the grain size and twin spacing of nt-Cu samples are in the range of several microns and nanometers, respectively, no cyclic hardening/softening, but cyclic stability is detected in strain-controlled nt-Cu samples. Because single-slip system of threading dislocations with the maximum Schmid factor are preferentially activated in nanoscale twins when the twin planes are parallel to the cyclic loading axis. Apart from the grain orientation, the nanoscale twins also facilitate the activation of single-slip threading dislocations in nanotwin interiors by restricting the activation of secondary or multiple slip systems [15,20]. In this case, forming immobile dislocation locks or tangles in nanoscale twin interiors and accompanying hardening become difficult in nt-Cu during fatigue, compared with conventional multiple slips in CG metals [10].

The saturated stress of strain-controlled nt-Cu is mainly determined by the nanoscale twin confinement of threading dislocation motion in the twin/matrix layers, based on the confined layer slip model [9]. Because the grain size of nt-Cu is two-order of magnitude larger than the twin thickness, the influence of grain size on saturated stress is minor compared with the influence of nanoscale twin thickness. Therefore, the higher saturation stress can be reached in the thinner twin lamellae. Similarly, higher saturated stress of nt-Cu fatigued at larger plastic strain amplitude are possibly correlated with the activation of threading dislocations in smaller twins, even in some grains with relatively hard orientations.

However, we have to emphasize that the microstructural stability of nt-Cu sample in the fatigue tests is significantly influenced by its grain size. The GB regimes of nt-Cu usually undergo a much larger plastic strain than that sustained by grain interiors [4]. Dislocation cells/subgrains gradually appear in the original GB regime with increasing tensile strain, accompanied by elimination or thickening of the nanotwins. The concentrated GB deformation would become more serious in nt-Cu samples with a smaller *d*, since the GB volume fraction is approximately inverse to *d*. This possibly interprets the instability of magnetron sputtered nt-Cu films in the stress-controlled high-cycle fatigue tests [6,7]. Therefore, the larger grain size of nt-Cu in this study can effectively suppress the inhomogeneous deformation adjacent to GBs and ensure the stability of microstructure and property in the strain-controlled low-cycle fatigue tests.

Guo *et al.* [21] investigated the influence of twin thickness on the dislocation configuration during cyclic deformation of polycrystalline Cu with a wide distribution of twin thicknesses. It demonstrated that no dislocation patterns, such as PSBs or cell structures, could be formed when the twin thickness is less than 1 μm. Therefore, single slip system activation of threading dislocations inside nanotwin lamellar channels and relatively stable TBs and GBs are two key responsible factors for the steady state of nt-Cu samples under all strain amplitudes. The detailed possible mechanism for the cyclic stability or saturation on nanotwinned Cu sample is still under exploration.

In summary, the most significant results of the present study are the threading dislocation configurations in strain-controlled fatigued nanotwinned Cu elucidated by TEM observations under two-beam diffraction conditions. Numerous short parallel dislocation segments are observed in the vicinity of TBs while some curved segments in the middle of twin interiors. Systematic analysis based on SEM, TEM and 3D stereo projection reconstruction further confirmed that both of them are threading dislocations with slip plane inclined to the TBs but Burgers vector parallel to the TBs.

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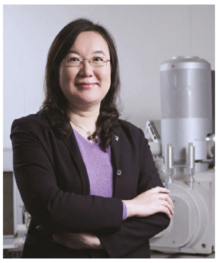
Author contributions Lu L and Pan Q designed the project and the experiments. Pan Q performed the experiments. Lu L and Pan Q analyzed the results and wrote the paper.

Conflict of interest The authors declare that they have no conflict of interest.

Qingsong Pan received his PhD degree in materials physics and chemistry at the Institute of Metal Research, Chinese Academy of Sciences, in 2014, and joined Professor Lei Lu's group as an assistant professor. His research focuses on the fatigue behavior of nanotwinned metals.



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中文摘要 块体择优取向纳米孪晶Cu在应变疲劳时保持循环稳定. 为了研究其本征疲劳机制, 本文利用透射电子显微镜在双束条件下研究了纳米孪晶Cu疲劳后孪晶片层内位错形貌特征. 结果表明纳米孪晶Cu的疲劳变形多由受限于孪晶片层内部的滑移方向平行于孪晶界面、滑移面倾斜于孪晶界面的单滑移贯穿位错主导. 通过三维投影模型重构了贯穿位错在纳米孪晶片层内的空间分布.