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### Authors

Wang, Y. M.  
Hodge, A. M.  
Biener, J.  
[et al.](#)

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## Deformation twinning during nanoindentation of nanocrystalline Ta

Y. M. Wang,<sup>a)</sup> A. M. Hodge, J. Biener, and A. V. Hamza  
Lawrence Livermore National Laboratory, Nanoscale Synthesis and Characterization Laboratory,  
Livermore, California 94550

D. E. Barnes and Kai Liu  
Department of Physics, University of California, Davis, California 95616

T. G. Nieh  
Department of Materials Science and Engineering, The University of Tennessee, Knoxville,  
Tennessee 37996

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The deformation mechanism of body-centered cubic (bcc) nanocrystalline tantalum with grain sizes of 10–30 nm is investigated by nanoindentation, scanning electron microscopy and high-resolution transmission electron microscopy. In a deviation from molecular dynamics simulations and existing experimental observations on other bcc nanocrystalline metals, the plastic deformation of nanocrystalline Ta during nanoindentation is controlled by deformation twinning. The observation of multiple twin intersections suggests that the physical mechanism of deformation twinning in bcc nanocrystalline materials is different from that in face-centered cubic (fcc) nanocrystalline metals. © 2005 American Institute of Physics. [DOI: 10.1063/1.1883335]

Most recent studies on deformation physics of nanocrystalline (nc, grain sizes <100 nm) materials have focused on face-centered cubic (fcc) metals and alloys.<sup>1–3</sup> Various grain boundary mediated processes (grain boundary sliding,<sup>4,5</sup> grain rotation,<sup>6</sup> grain coalescence and growth<sup>7</sup>) and partial dislocation mechanisms (stacking fault, deformation twinning<sup>8–12</sup>) have been proposed theoretically and demonstrated experimentally in fcc nc metals.<sup>10–12</sup> On the other hand, we have very little knowledge about the deformation mechanisms of body-centered cubic (bcc) nanocrystalline materials.<sup>13,14</sup> To date, almost all experiments documenting the deformation behavior of bcc nanocrystalline metals have been performed on samples prepared through the powder metallurgy route that is chronically affected by impurity and porosity issues. So far, shear banding appears to be the only deformation mode observed in these particulates.<sup>13</sup> In this letter, we demonstrate that the deformation of bcc nanocrystalline tantalum with grain sizes of 10–30 nm proceeds via a twinning mechanism. We selected  $\alpha$ -tantalum as a model bcc system as it is widely used in the microelectronic industries as capacitor and diffusion barrier layer material. The deformation behavior of nanoscale tantalum is thus important for the reliability of microelectronics.

Nanocrystalline tantalum (99.95%) films with thickness of  $\sim 0.1 \mu\text{m}$  were prepared by magnetron sputtering onto oxidized silicon substrates at a deposition rate of  $\sim 0.5 \text{ \AA/s}$ . To prevent the formation of columnar grain structures, the deposition was interrupted for an interval of 10 s after deposition of every 5  $\text{\AA}$  (10 s). Both impurity and texture of the as-deposited films are negligible, as indicated by energy dispersive x-ray spectroscopy (EDS), x-ray diffraction measurement, and the transmission electron microscopy (TEM) selected area diffraction pattern. TEM study in Fig. 1 confirms that the as-deposited nanocrystalline Ta has a dominant  $\alpha$ -phase and narrow grain size distribution in the range of 10–30 nm.

The mechanical response of the as-deposited nanocrystalline Ta was probed using a Hyston Triboindenter with a Berkovich diamond tip. The depth-sensing nanoindentation allows us to deform materials locally in a well-controlled fashion and thus has major advantage over other uncontrollable deformation techniques such as ball-milling, rolling, and grinding.<sup>10,11,15</sup> After indentation, the residual impressions of indents were examined by scanning probe microscopy (SPM) equipped with the tribo-indenter and scanning electron microscopy (SEM) to check out for the evidence of microcracks, slip, and pileup/sinkins.

For high-resolution transmission electron microscopy (HRTEM) experiments, a series of microsized indentations (100  $\mu\text{m}$  apart in both  $x$ - and  $y$ -direction) were made on the Ta film. The substrate was then mechanically removed and ion milled to transparency from the silicon side, using a Gatan high precision ion polishing system (Model 691) with  $\text{Ar}^+$  accelerating voltage of 4 kV at a glancing angle of  $3^\circ$ . The HRTEM investigation was performed with a Philips

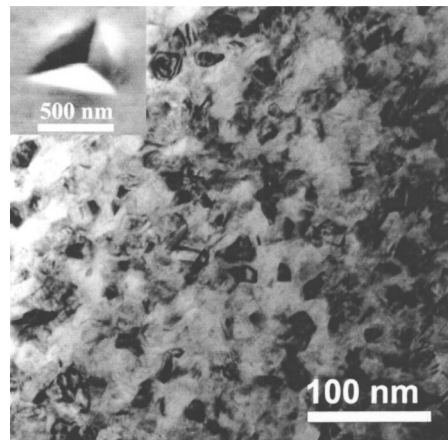


FIG. 1. Transmission electron micrograph of nanocrystalline Ta with grain sizes in the range of 10–30 nm. The inset is a scanning electron microscopy image of an indentation, showing no evidence of microcracking or shear banding.

<sup>a)</sup>Electronic mail: ymwang@llnl.gov

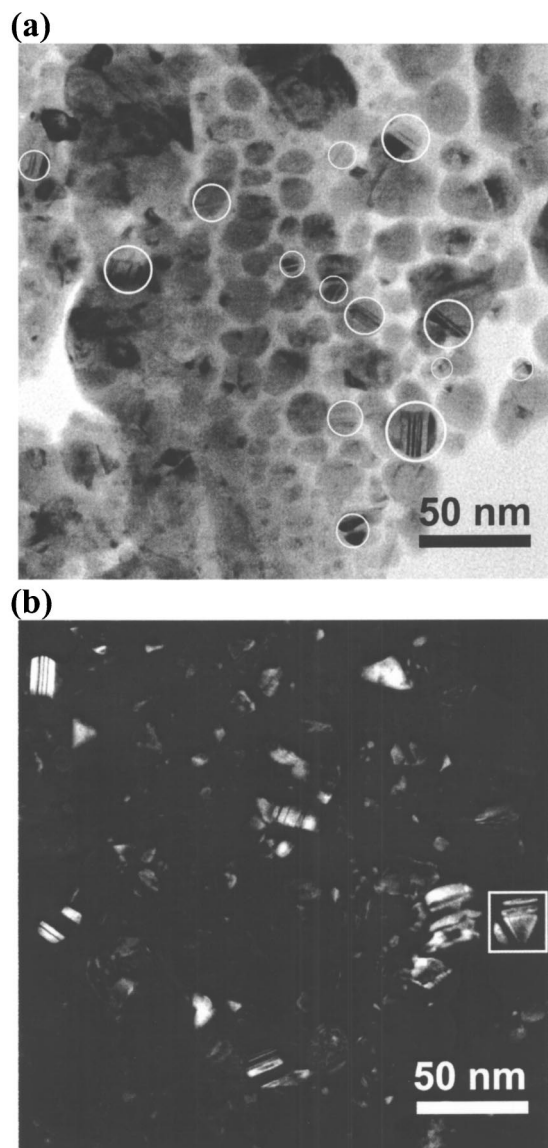


FIG. 2. (a) Bright field, and (b) dark field transmission electron micrograph of the deformation microstructure in nanocrystalline Ta after indentation. Deformation twins are often observed near the edges and corners of the indents.

CM300 FEG microscope operating at 300 kV with a point-to-point resolution of 1.8 Å. The extraction voltage for field emission gun was 4.2 keV. The high-resolution images were recorded with a 2048 K × 2048 K charge-coupled-device camera.

The inset of Fig. 1 shows a typical SEM image of an indent on nanocrystalline Ta after unloading at a maximum load of 3 mN. The threefold symmetry of the indenter is clearly visible, with no evidence of microcracking or shear banding even at the corners of the indent where the sample experiences the highest tensile stress. The smooth pileup of the materials around the indent demonstrates the fully plastic flow during nanoindentation of nanocrystalline Ta.<sup>16</sup>

A transmission electron micrograph of the deformation microstructure in nanocrystalline tantalum from a region in the vicinity of an indent is shown in Fig. 2(a); abundant deformation twins (DBs) are identified, as accentuated in the white circles where the twins can be seen with two characteristic flat interfaces parallel to each other. The DBs are preferentially located near the edges or corners of the in-

dent. It is evident from the TEM image that twinning occurs in all the grain sizes of up to ~30 nm. The width and orientation of the twin lamellas vary from grain to grain, which is better illustrated in a TEM axial dark-field image, Fig. 2(b). In some cases, two twin bands with different orientations [one of them is marked with a white frame in Fig. 2(b)] coexist in one single grain. The twin density is not uniform but reaches up to  $4 \times 10^7/\text{m}$  in certain regions. According to the twin density shown in Fig. 2(a), the total strain contribution from the deformation twinning is estimated as ~3.7%. The ubiquitousness of the deformation twins implies that twinning is the primary deformation mode during nanoindentation of nanocrystalline Ta.

A close examination of some deformation twins in Fig. 2 further reveals the intersection of multiple twin bands in the middle of a single grain, one example of which is shown in Fig. 3(a). The different orientations of the twin bands are indicated with white arrows. The observation of twin intersection in bcc nanocrystalline metals is rather interesting, considering that such phenomenon has not been reported in fcc nc metals, where most deformation twins are coplanar in nature.<sup>10–12,15</sup> This observation strongly suggests that the twinning behavior of bcc nc metals is different from that of fcc nc metals. The former may be related to the unique dislocation feature in bcc metals that three {112}-type deformation twins could intersect along a common  $\langle 11\bar{1} \rangle$  direction, the simplest type of twin-twin interaction in bcc metals. In addition, the grain rotation and/or sliding mechanism in refractory bcc nc metals could be relatively difficult at room temperature that is merely a small portion of their melting temperatures, which in turn helps to promote multiple twin interactions.<sup>15</sup>

To further elucidate the atomic mechanism of the deformation twinning in bcc nc Ta, Fig. 3(b) displays a high-resolution transmission electron micrograph of an individual grain, imaged with the electron beam parallel to  $\langle 111 \rangle$  direction and featuring a twin. The twin boundaries are determined parallel to one set of {112} planes and marked with two white arrows. Similar to many deformation twins seen in fcc nc metals, the twin band observed is only a few atomic layers thick.<sup>12</sup> The corresponding fast Fourier transformation (FFT) patterns of the matrix [Fig. 3(c)] and the twinned area [Fig. 3(d)] confirm the twinning relationship between the matrix and the twin band. At the bottom of the image, Fig. 3(b), another type of twin is marked by the white frame. The wavy twin boundary suggests that this type of deformation twin may be formed through a mechanism similar to that discussed in Ref. 12.

With respect to the deformation twinning in bcc metals, many models have been proposed for conventional coarse-grained sizes, all of which involve the repeated expansion of twinning partial  $1/6 \langle 111 \rangle$  on {112} type planes. Similar to dislocation slip, it is believed that the critical stress required to nucleate twins also obeys a Hall–Petch (HP) relationship, with a slope  $K_T$  that is much higher than the slope  $K_s$  for slip.<sup>17</sup> For this reason, coarse-grained bcc metals normally deform via a dislocation slip mechanism unless at low temperatures and/or very high strain rates. The critical twinning stress has been linked to the dislocation pileup against the grain boundaries.<sup>17</sup> It is conceivable that such twinning-HP relationship would fail when the grain sizes decrease into nanometer regime where the dislocation pileup becomes unstable,<sup>18</sup> and the formation of partial dislocations becomes

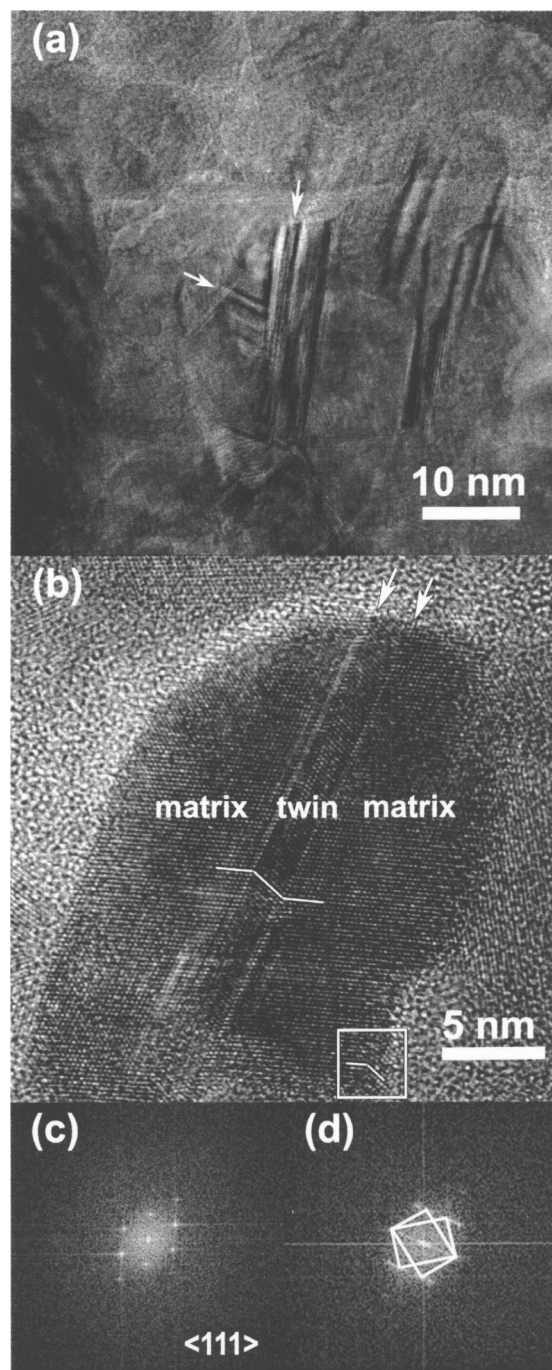


FIG. 3. (a) Transmission electron micrograph of the intersecting twins in nanocrystalline Ta. The twinning directions are marked with two white arrows; (b) high-resolution TEM image of a nanograin, featuring a deformation twin, and the fast Fourier transformation (FFT) patterns of (c) matrix, and (d) twin band. The twinning relationship can be identified through atomic image in (a) and the FFT pattern in (d).

the preferred deformation mode.<sup>14</sup> This image is consistent with our TEM results that no full dislocation was observed in the grains smaller than 20 nm. The twinning-HP relationship breakdown has recently been reported in fcc nanocrystalline materials,<sup>12</sup> but the current results first demonstrate such a breakdown in bcc nc metals. The preference of deformation twinning observed here suggests that the extrapolation of twinning-HP relationship into nanocrystalline regime may need to be thoroughly reexamined.<sup>17</sup>

The observation of deformation twinning in nanocrystalline Ta is remarkable in several respects. First, the deformation

twinning, which was predicted and experimentally verified in fcc nanocrystalline metals (Al, Cu, Pd),<sup>10–12,15,19</sup> has never been observed in bcc nanocrystalline metals before, nor has it been foreseen from the only reported atomistic simulation in bcc nc metals.<sup>14</sup> This suggests that more experimental and theoretical effort is needed in order to better understand the deformation mechanisms of bcc nanocrystalline materials. Second, conventional coarse-grained Ta does not deform by twinning except at very high strain rates and/or low temperatures.<sup>20</sup> The occurrence of deformation twinning in nc Ta, subjected to well-controlled and slow (quasistatic) indentation, can thus be directly related to the grain size effect, indicating a transition of deformation mechanisms in nanocrystalline bcc metals as the grain sizes decrease to tens of nanometers. Third, some deformation twins observed in nc Ta appear to be noncoplanar in nature even when the grain sizes are very small [ $\sim 20$  nm, Fig. 3(a)], opposite of what has been reported in fcc nc metals.<sup>10–12,15</sup> This implies that multiple twinning systems are operative during deformation of bcc nc grains. Finally, it is worth pointing out that the twinning-HP relationship in bcc metals may not be applicable in the nanocrystalline regime, where twins could form via partial dislocation emission from grain boundaries (heterogeneous nucleation) instead of traditional pole mechanism. A more thorough examination of the twinning-HP relationship in bcc nanocrystals is clearly interesting but transcends the scope of this letter.

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