***In Situ* Observation of Dislocation Behavior in Nanometer Grains**Lihua Wang,¹ Xiaodong Han,^{1,*} Pan Liu,¹ Yonghai Yue,¹ Ze Zhang,^{1,2} and En Ma³¹*Institute of Microstructure and Properties of Advanced Materials, Beijing University of Technology, Beijing, 100124, China*²*Department of Materials Science, ZheJiang University, Hangzhou, 310008, China*³*Department of Materials Science and Engineering, Johns Hopkins University, Baltimore, Maryland 21218, USA*

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Using a newly developed nanoscale deformation device, atomic scale and time-resolved dislocation dynamics have been captured *in situ* under a transmission electron microscope during the deformation of a Pt ultrathin film with truly nanometer grains (diameter $d < \sim 10$ nm). We demonstrate that dislocations are highly active even in such tiny grains. For the larger grains ($d \sim 10$ nm), full dislocations dominate and their evolution sometimes leads to the formation, destruction, and reformation of Lomer locks. In smaller grains, partial dislocations generating stacking faults are prevalent.

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The generation, motion, interaction, storage, and annihilation of dislocations are well understood for plastic deformation of conventional polycrystalline metals [1,2]. However, when the grain size (diameter d) is refined to nanometer scale ($d < \sim 10$ nm), how the dislocations behave becomes an unsettled issue. It has been proposed that in this d regime the strength of metal would exhibit an inverse Hall-Petch d dependence [3] and during plastic deformation dislocation activities subside, giving way entirely to the sliding of grain boundaries (GBs) [3–7]. There is so far no conclusive experimental evidence that this is indeed the case. Experimentally, most previous transmission electron microscopy (TEM) studies examined the so-called “nanocrystalline” grains, whose sizes ($d < 100$ nm) are typically of the order of 20 nm or larger. Some of these TEM observations suggest that dislocations do run across the small grains, but usually have little chance to interact, and leave no debris behind for post-mortem observations after the applied stress is removed [8–14]. One exception is that after large (cold rolling) strains in confined geometries [15], Lomer-Cottrell locks were observed in the ($d = 20$ nm) grain interior. While the $d < \sim 10$ nm regime is rarely accessed in experiments, it is the realm of molecular dynamics (MD) simulations [4–7], which did predict persistent dislocation activities, as well as their interactions and storage including the formation and destruction of Lomer and Lomer-Cottrell locks. It is therefore of interest to acquire experimental evidence for these, and observe *in situ* during deformation, the evolution of dislocations in truly nanometer-sized grains.

Models and MD simulations [4–7,16] have also suggested that at sufficiently small d , such as nanometer grains with $d < 10$ nm, dislocations emitted from GBs would be partial dislocations in lieu of full dislocations, leaving stacking faults (SFs) in their wake (sometimes nucleating twinning). This, however, was not observed in a nanocrystalline Ni with $d \sim 10$ nm [14]. There have also been doubts that the MD-observed presence of only the leading partial dislocations are due to the limited MD time scale,

which precluded the emergence of the trailing partial [17,18]. While many TEM observations found SFs and twins that may be used to infer partial dislocation activities [16,19,20], direct experimental evidence for the switch from full dislocation to partial dislocation with decreasing d has not been forthcoming. Note that the two-dimensional thin films have two bottom and top free surfaces. Thus, the dislocation behavior can be different from those of bulk materials. However, understanding the dislocation mechanisms in thin film samples has merit in its own right, although caution should be exercised when extrapolating them to bulk nanocrystalline metals.

In this work, from a previously developed technique for *in situ* tensile experiments in scanning electron microscope [21], we have developed a new *in situ* tensile testing device for TEM [22], which can slowly and gently deform an electron-transparent film, and at the same time retain the double-tilt capability to perform high-resolution TEM (HRTEM) observations. Using this device, we have monitored dislocation dynamics in a nanograined Pt ultrathin film with an average d of ~ 9 nm. We report Lomer lock formation through the reaction of full dislocations and their destruction processes at the atomic scale, as well as the direct and statistical evidence of the switch to partial dislocations as d goes below a critical value.

The bimetallic strips making up the sheets have different thermal expansion coefficients. The two plates were arranged to bend slowly in opposite directions, Fig. 1(a), actuating the uniaxial pulling of the Pt film at an estimated strain rate of $\sim 10^{-4} \text{ s}^{-1}$ (see supplementary material [29] for details). This setup allows the double-tilt capability during deformation, such that grains can be oriented appropriately for high-resolution imaging and determination of the Burgers vectors of dislocations. The real-time evolution of the film was captured *in situ* along with deformation in a JEOL-2010 field emission TEM operating at 200 kV.

As shown in the bright-field TEM image of Fig. 1(b), the Pt film has nanometer-sized and equi-axed grains, with no

FIG. 1 (color online). (a) Homemade testing device used for *in situ* tensile pulling of the Pt thin film in a TEM. (b) Bright-field TEM image of the film, with a selected area diffraction pattern in the inset. (c) HRTEM image showing grains separated by high-angle GBs. (d) Statistical distribution of the grain size.

obvious texture (see the selected area diffraction pattern in the inset). The HRTEM image in Fig. 1(c) shows that most grains are separated by high-angle GBs, with no porosity or microcracks. The distribution of grain size, as measured for 100 grains, is shown in Fig. 1(d). Most of the grains have d below 14 nm, with an average of ~ 9 nm.

For the “larger” grains, e.g., the $10 \text{ nm} \times 13 \text{ nm}$ grain A, upon loading we observed obvious movements and interactions of full dislocations. Figure 2(a) and 2(b) display two HRTEM images, taken 180 seconds apart, and Fig. 2(c) and 2(d) are the enlarged HRTEM images corresponding to the framed regions in (a) and (b), respectively. Two trapped dislocations are seen in Fig. 2(c), and local Burgers circuits were drawn to identify the Burgers vector. The inverse fast Fourier-filtered (IFFT) image is given in Fig. 2(e). Extra half planes are seen for both the $(\bar{1}11)$ and the $(1\bar{1}1)$ planes. This configuration represents a Lomer Dislocation (LD) [23], exhibiting the Burgers vector of a $(a/2)[011]$ full dislocation. The LD is formed by the interaction of two nondissociated full dislocations with Burgers vectors $(a/2)[\bar{1}01]$ and $(a/2)[110]$, respectively, moving under applied stress on two intersecting slip planes, $(1\bar{1}1)$ and $(\bar{1}11)$ [23,24]. The dislocation reaction can be written as

$$\frac{1}{2}[\bar{1}01] + \frac{1}{2}[110] \rightarrow \frac{1}{2}[011]. \quad (1)$$

The two reacting dislocations mutually knit with each other to reach a low energy configuration [2,25].

Atomistic and dislocation dynamics simulations have shown before that the Lomer and Lomer-Cottrell locks have practically the same strength [24,26–28], and these stable junctions are dislocation obstacles and therefore a source of strain hardening. The LD junction above is expected to be sessile since it can glide in neither of the

FIG. 2 (color). *In situ* TEM observations of dislocation activities in a $10 \text{ nm} \times 13 \text{ nm}$ -sized grain, grain A in (a). Two Lomer dislocations are found in (c); see details in the IFFT shown in (e). The corresponding frames taken 180 seconds later are displayed in (b), (d), and (f), depicting the unzipping of the LD. (g) At a later stage of deformation, LD reforms in this grain, as seen in the IFFT image in (h). See text for more illustrations.

slip planes of the reactant dislocations. High stresses are needed to break the Lomer lock and its destruction process was only observed before in MD simulations [24,27,28]. Here, a major advantage of our *in situ* experiment is that it can unravel the temporal evolution of the (same) dislocations and in the same location (grain A in this case). For the first time, our observations [see Fig. 2(d) and 2(f)] reveal the LD destruction process at the atomic scale. The process left two dislocations, as shown in Fig. 2(d). The Burgers circuits show that both of them are 60° full dislocations since only one set of (111) planes have an extra half plane inserted; the details are shown in Fig. 2(f). Comparing with the configuration in Fig. 2(c) and 2(e), the LD is clearly unlocked, as it can be destroyed by splitting into two 60° dislocations under high stresses in the reaction

$$\frac{1}{2}[011] \rightarrow \frac{1}{2}[\bar{1}01] + \frac{1}{2}[110]. \quad (2)$$

For each LD, one of the split dislocations has escaped and the other remained trapped in the grain.

We further discovered the reformation of Lomer lock in the same region in the same grain A, with continued straining. Figure 2(g) shows that the trapped dislocation has again become a Lomer lock, formed by the reaction between two full dislocations as described by Eq. (1). The sequence of LD formation, destruction, and reformation has never been experimentally observed in nanometals.

These are direct atomic scale evidence of the dynamic dislocation activities in ~ 10 nm grains. The dislocations are not only very active in their generation and motion, but also proficient in their interaction and reaction. The locks and full dislocations can be stored in the tiny grains and can also be liberated and driven away by the high stresses. The investigated grains are subjected to very high stresses because they are near the crack tip. From the HRTEM image [Fig. 3(a)], the perfect dislocation density is as high as $2.3 \times 10^{16} \text{ m}^{-2}$ (many grains even higher). As indicated in Figure S3 of the supplementary material [29], the force direction is nearly along $[100]$ direction of the tensile grain. The Schmid factors of the 12 slip systems are shown in Table S1 of the supplementary material [29]. Four of the slip systems $\langle 110 \rangle \{111\}$ of the tensile grain have very large Schmid factors of 0.4. These large Schmid factors insured at least two sets of slip systems can be activated, making it possible for the interaction of two dislocations to form Lomer locks. See more discussions in the supplementary material (Fig. S3 and Table S1).

Furthermore, dislocation activities were also observed in many grains far away from the crack tips and some dislocation activities were observed before the crack initiation. Figure S4 of the supplementary material [29] shows an example of *in situ* observation of perfect dislocation nucleation in a grain before the crack initiation. This indicates that the full dislocation activities are prevalent in these small grains, and are not solely induced by the crack.

Figure 3 is the schematic view which illustrates the formation and destruction process of the Lomer lock under applied stress. The geometry can be understood from the Thompson tetrahedron shown in Fig. 3(a). We consider the case of two interacting $a/2\langle 110 \rangle \{111\}$ dislocations in Fig. 3(b). The first dislocation stays in the horizontal

ACD plane in Fig. 3(a) and the vector $\vec{CD}$ represents its Burgers vector. If this dislocation interacts with another dislocation which is in the inclined slip plane ABC (with a Burgers vector $\vec{BC}$) under applied stress, then the LD can be formed and the Burgers vector of the junction segment is the sum of the initial vectors. From the Thompson tetrahedron, it is obvious that this vector is $\vec{BD}$ which is in neither of the initial glide planes and the Lomer lock formation can be written as

$$\vec{BC} + \vec{CD} \rightarrow \vec{BD}. \quad (3)$$

The Lomer lock can be destroyed if a critical stress is reached, as seen in MD simulations [26,27]. Figure 3(d) illustrates the unzipping of the LD. With the increasing stress, the length of the Lomer segment decreases and then the junction breaks under high stress.

We next examine the smaller grains in our d distribution. It has been predicted that [16], for Pt with an SF energy of 0.27 to 0.373 J/m² [30,31] and a shear modulus of 65.4 GPa [32], there can be a critical d of about 7–9.5 nm. Above this d , full dislocation nucleation requires lower stress and would be preferred over the partials. Below this demarcation d , partial dislocations become favorable. Figure 4(b) is a HRTEM image taken from the red framed region of Fig. 4(a). The stacking fault resulting from a partial dislocation is captured in a 7 nm $\times$ 9 nm-sized grain.

The difference of dislocation behavior between the larger and the smaller grains in our d distribution is more obvious and meaningful, after a statistical survey of the results. For grains relatively large in our distribution ($> \sim 10$ nm), we examined more than 20 grains at the end of the pulling, and found 70% of them have full dislocations inside. SFs are rarely seen. In contrast, for the smaller grains we examined ($< \sim 8$ nm, also 20 grains), a large fraction of them (40%) contain SFs, and few of them contain full dislocations and LD locks. This provides statistical evidence that the full dislocations and the SFs are the dominant and stored defects for these two groups, respectively. There is indeed a trend for the predicted switch from full dislocations (stored inside the grains and sometimes inducing LDs) and partial dislocation (depositing SFs), as d decreases across this grain size regime.

In conclusion, using our own *in situ* deformation device, we have captured many atomic scale images of the dynamic dislocation processes in strained Pt nanograins of the ultrathin films. Our work focuses on a grain size regime ($< \sim 10$ nm) hitherto rarely explored in experiments. Dislocations are highly active, even for these truly nanometer grains. Even for such tiny grains, we observed dislocation storage, and the dislocation types are different depending on d . Full dislocations and the Lomer locks are frequently observed for larger grains (~ 10 nm), whereas partial dislocations generating SFs are prevalent in the smaller grains. The observations of the generation, motion, interaction, and storage of full and partial disloca-

FIG. 3 (color online). (a) Thompson tetrahedron showing the Burgers vectors of the interacting dislocations. (b),(c) Schematic view of the Lomer lock formation: the Lomer lock is formed by the interaction of two nondissociated full dislocations. (d) The Lomer dislocation is unlocked under stress.

FIG. 4 (color online). (a) Low-magnified TEM images showing the deformation region. (b) HRTEM image taken from the framed region of (a), showing a partial dislocation in a $7\text{ nm} \times 9\text{ nm}$ grain. A magnified view illustrating the stack fault is given in the inset.

tions in the $d < 10\text{ nm}$ regime, including the dynamic process of Lomer lock formation, destruction, and reformation, shed light on the role of dislocation mechanisms in the plasticity of nanometer-grained ultrathin films, and should have implications for the design and integrity of ultrahigh strength thin films used in microelectronics and microelectromechanical systems.

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