ABSTRACT

In-situ straining experiments on nanocrystalline (nc) and ultra fine grain (UFG) Aluminum were performed at room and intermediate temperatures. Both materials exhibit significant stress assisted grain growth. The strain induced by grain boundary motion has been measured in UFG Al, and was found to be on the order of a few percents. These results cannot be interpreted solely within the framework of Displacement Shift Complete (DSC) dislocation motion. We propose here that GB motion occurs via both shuffling and secondary DSC dislocation motion.

INTRODUCTION

Ultrafine grains (UFG) and nanocrystalline (nc) metals have exceptional mechanical properties such as very high yield stresses. However their structure is unstable and tends to evolve by grain growth even at relatively low temperature. Grain growth seems also favoured by the application of a mechanical stress as shown during fatigue [1] or monotonic tensile experiments [2]. However few studies have focused on the elementary mechanisms that promote this instability and on the role of grain growth in the overall deformation process. Such mechanisms should indeed be crucial in the plasticity of materials where grain boundaries constitute a large volume fraction. For the last few years, in situ transmission electron microscopy (TEM) deformation experiments have provided a well designed tool for probing these mechanisms at the nanoscale. Indeed, it is supposed that, according numerical simulations [3], plasticity mechanisms in nc metals which should imply extensively fast grain boundaries and dislocations motion, could be probe at a timescale and length scale only reached by in-situ TEM. In this article we present the results of in situ straining experiments on self standing nc-Aluminium films at room temperature and on aluminium ultra fine grains (UFG) at moderate temperature. The choice of UFG Aluminum at this given condition was dictated by first, the fact that it should be easier to analyze grain boundary (GB) motion in UFG than in nc materials and second, by the fact that grain boundary motion should be strongly enhanced at higher temperature in association with a lower dislocation activity.

EXPERIMENT

180nm thick nc aluminum films were deposited on Si substrate by DC pulsed magnetron sputtering. Once patterned and released from the substrate by reactive ion and XeF_2 etching, they form electron transparent free standing films. Aluminum UFG were produced by equal channel angular pressing (ECAP) after 8 passes. The mean grain size was around 40-90nm for nc and 800nm for UFG. In situ experiments were carried out in JEOL 2010 microscope operated at 200kV. Rectangular UFG samples were cut in massive ingots and prepared by electro-chemical polishing for TEM observations. Grain boundary motion was monitored by means of DVD/HD recording using a MEGAVIEW II camera.

RESULTS AND DISCUSSION

Figure 1 shows dark field (DF) micrographs extracted from a video sequence in nc Al. They were taken close to a crack tip (not visible at about one micron on the left) that concentrates the stress. Between the two pictures, the sample was strained uniaxially (the straining axis is vertical). It can be clearly seen on the grain in contrast that its lower part is growing. The speed of the moving GB is around 25nm/s. The fact that the grain observed here is larger than the original average mean size indicates that grain growth has already occurred before. This means that grain growth is a faceted growth which occurs repetitively and sequentially on all the facets of the grain. Faint dislocation contrasts can be observed as well in the grain. The dislocations seem to move rapidly toward the lower part of the grain where they stop. However it seems that dislocation nucleation and GB motion are processes occurring separately. It was observed that growing grains are first free from dislocations. Then, larger grains are more prone to dislocation nucleation. This situation is typical in all investigated grains in nc.

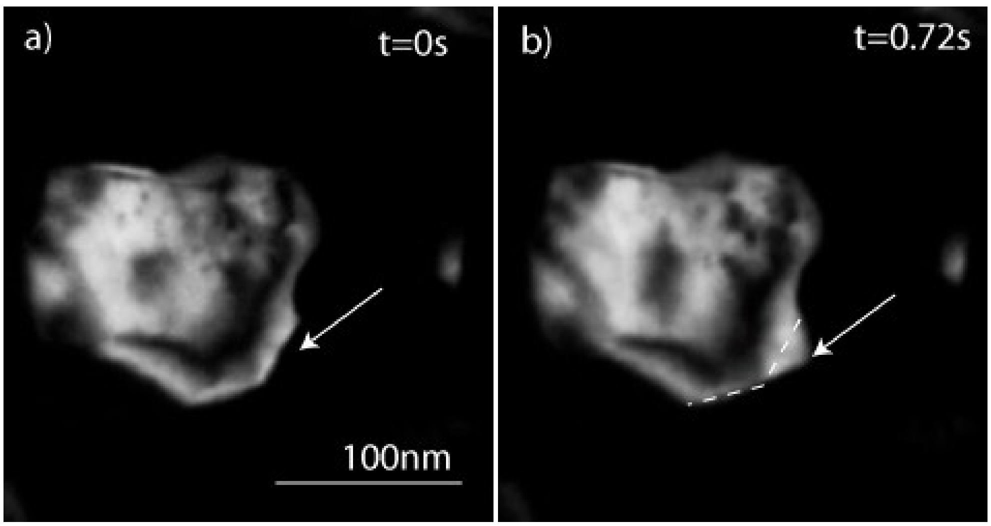

Figure 1. TEM DF micrographs taken in nc-Al deformed in-situ. Note the fast motion of the lower part of the grain. The dashed line indicates the position of the GB in a).

Figure 2 shows pictures extracted from a video sequence taken during the deformation of UFG Al at 370°C. At this temperature, recrystallization started to occur. These pictures were taken in non-recrystallized area. In that condition, we suppose that an additional stress can be sufficient to promote grain growth. The figure shows 4 grains (labeled A-D) after a substantial growth of grain A. Under stress, the grain A, free of dislocation move toward grains B, C and D. The growth starts when the stress is applied and eventually stops as soon as the stress is completely relaxed. Small defects attached to the surface due to sample preparation are marked as references in the 4 grains (labeled X0-X3). After a few minutes, grains B, C and D were swept by grain A (fig. 2d). A maximum GB speed of 60nm/s was recorded.

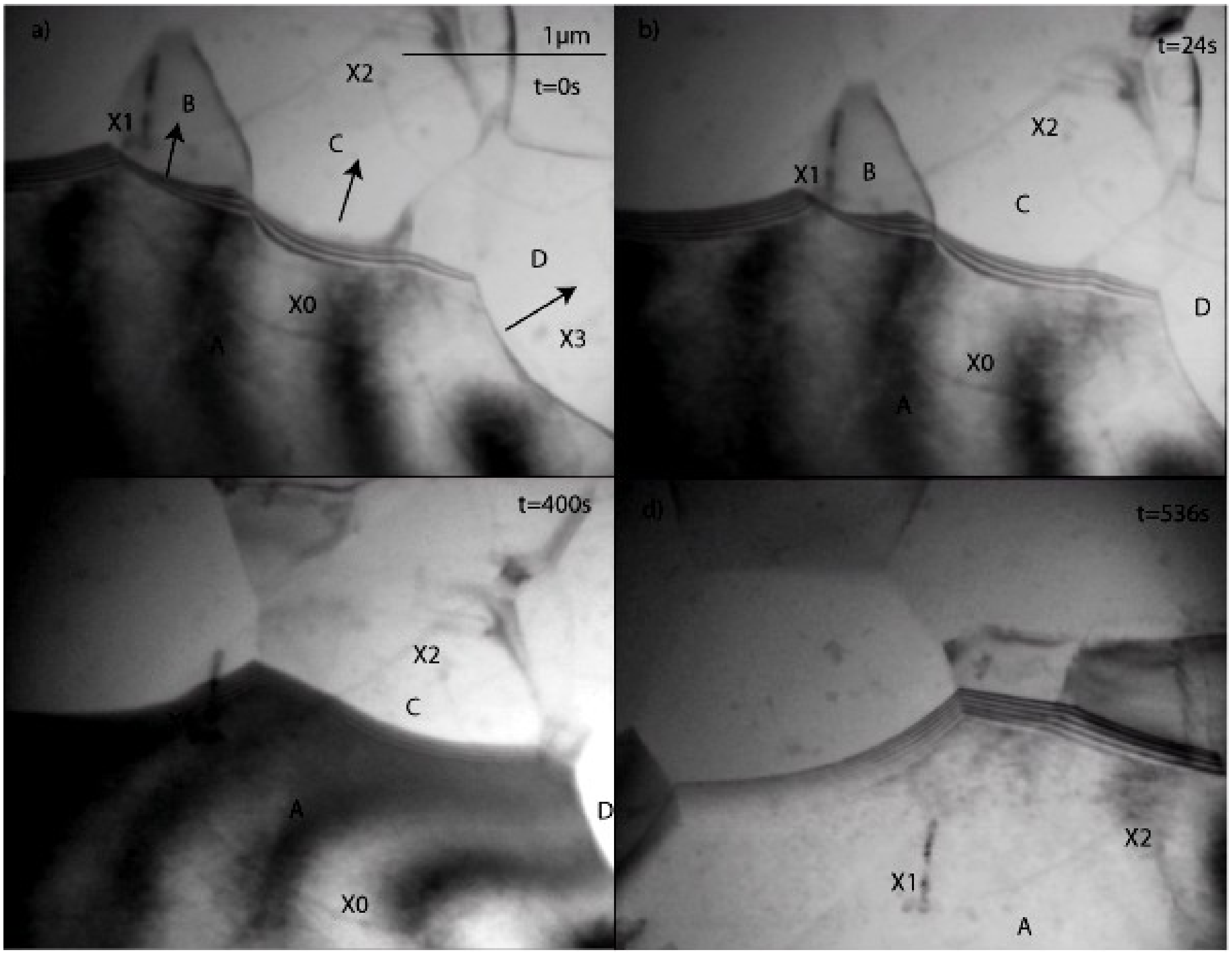

Figure 2. Stress assisted growth of grain A in UFG Al at 370°C. Note the progressive disappearing of grains B, C, D. Note the presence of the markers X0-X3 attached to the sample surface.

In order to get quantitative data about GB motion, pictures taken before and after motion were superimposed with a maximum coincidence between the two pictures in the grain A area (fig. 3). In that purpose, the contrast of one of the picture was reversed in order to get a minimum contrast in grain A (see for example the low contrast of X3). The fact that markers (X1-X3) do not superimpose indicates that GB motion induced deformation in grains B, C and D (note that marker contrast is enhanced on figure 3). The arrows joining markers before (in black) and after (in white) GB motion on figure 3 indicate the amplitude and the direction of the deformation. It can be seen that the deformation is mainly parallel to the GB plane corresponding to a grain shear. The deformation can be obtained by measuring the ratio

$$\varepsilon = d_{//}/d_{\perp} \qquad (1)$$

where $d_{//}$ refers to the shear amplitude parallel to the interface and $d_{\perp}$ the distance swept by the GB between its initial position and the marker (see fig. 3). From figure 3, the measured deformation is on the order of 10%.

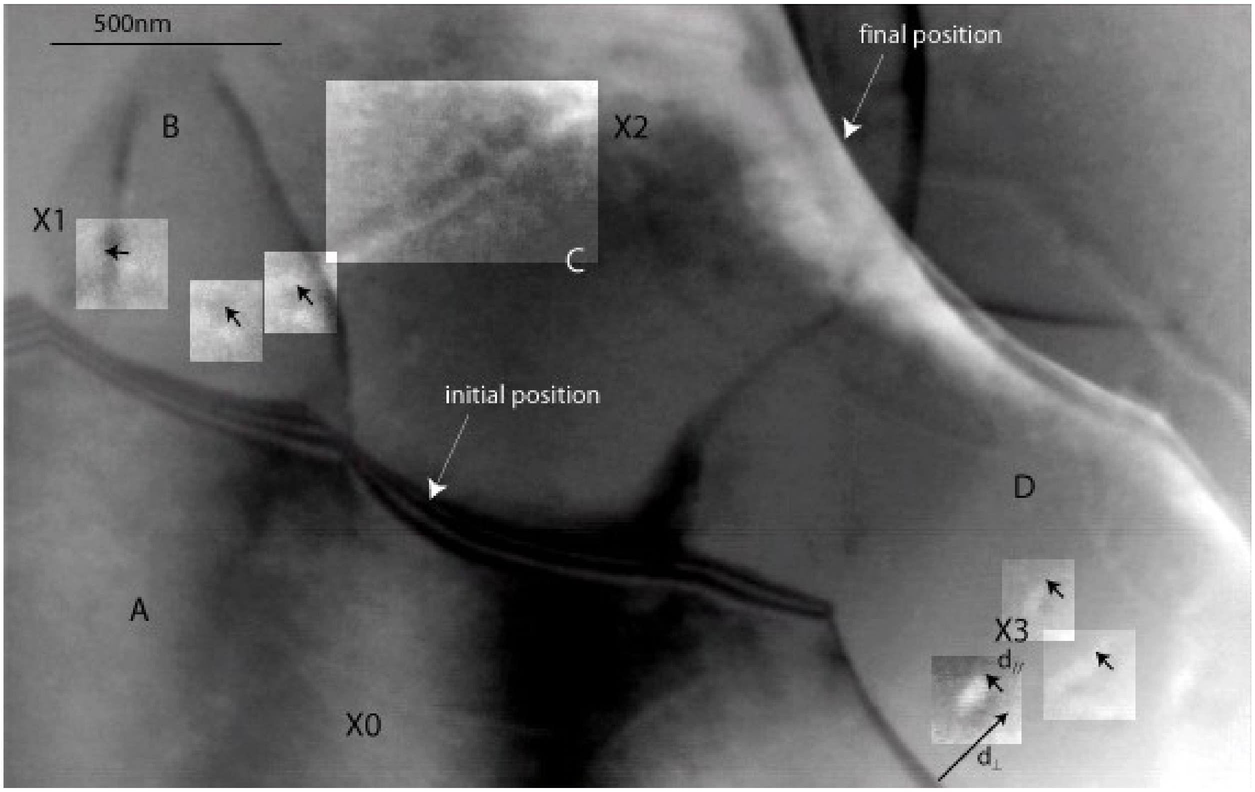

Figure 3. Superposition of micrographs from the same area before and after GB motion (with reversed contrast). The minimum contrast (i. e. maximum coincidence) is made in the grain A. Note that the contrast of the markers in grains B, C and D do not superimpose indicating that a deformation has occurred (marker contrast is enhanced).

According to the model proposed by Cahn et al. [4], which is equivalent to a Displacement Shift Complete (DSC) dislocation model, the corresponding deformation induced by the motion of a tilt GB is given by:

$$\varepsilon = 2 \tan(\theta/2) \qquad (\theta < 90°)$$
$$\varepsilon = -2 \tan(\pi/4-\theta/2) \qquad (\theta > 90°) \qquad (2)$$

where θ is the misorientation angle. To determine the nature of the GBs, electron diffraction was used to orientate grains A, B and C. It shows that GB A/B can be decomposed as a rotation of 36.87° around [100] (Σ5) plus a Δθ = 5° residual rotation close to [135]. Grain boundary A/C can be decomposed as a rotation of 129.52° around [110] (Σ11) plus a Δθ = 10° residual rotation around [-112]. According to (2), the large tilt component of GB A/B has is expected to produce a deformation of the order of magnitude of 66% which is far greater than measured here. The GB A/C is almost a twist boundary and is thus expected to produce a much smaller deformation. It can be concluded here that another mechanism has to be invoked to explain how GB motion produce deformation. According to Babcock and Baluffi [5], GBs can move by spontaneous atomic shuffling without changing its structure, i. e., without any deformation. Migration analyses using molecular dynamic of a Σ5 tilt grain boundary were performed recently [6] and show atomic hopping within the coincident site lattice (CSL) unit cell.

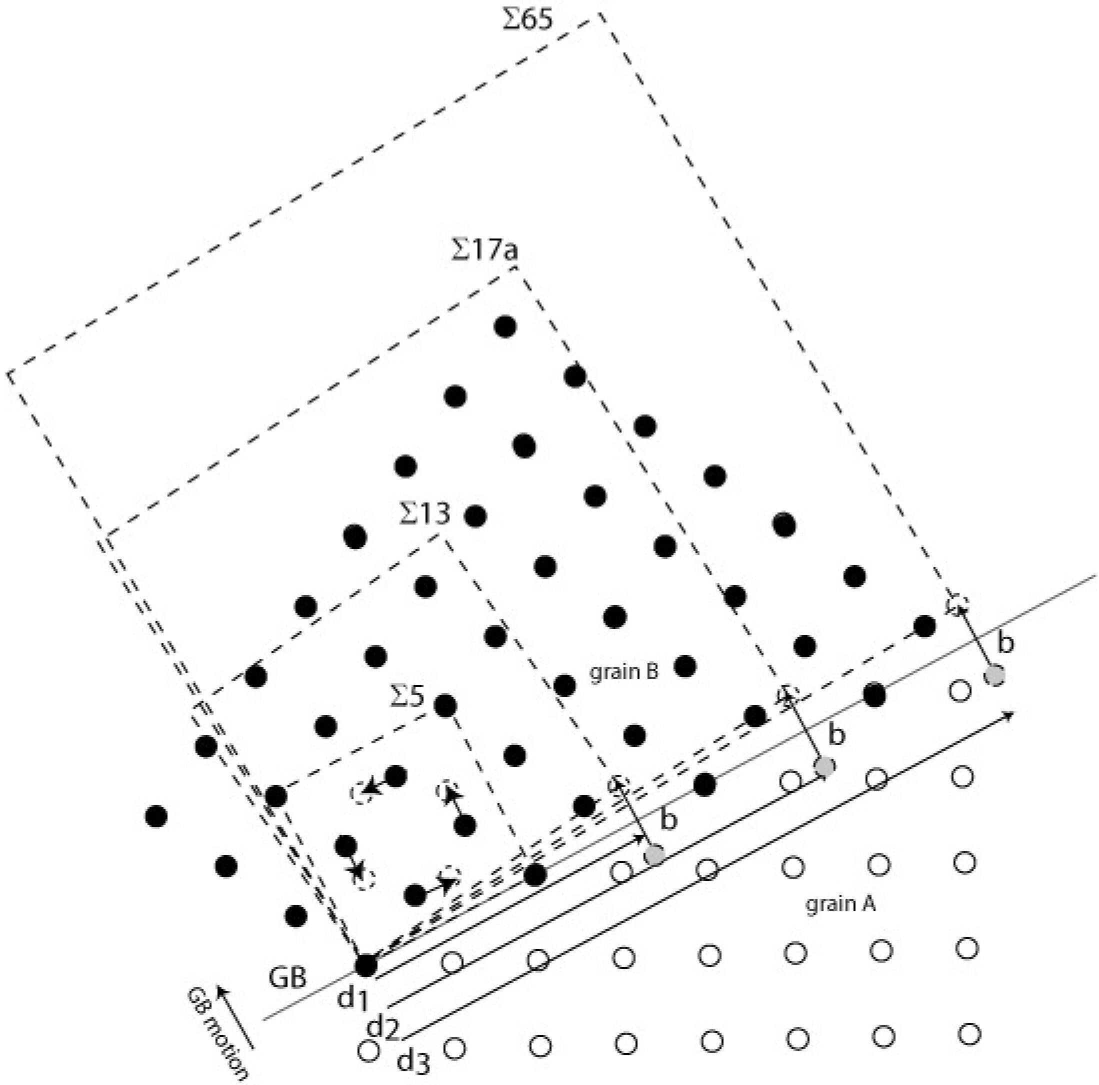

Figure 4. Slight deviation from a Σ5 [100] GB giving rise to higher coincidence GBs can be described by a set of DSC dislocations with a Burgers vector perpendicular to the GB plane spaced by the distance d . Dashed white dots correspond to the lattice extension of grain A once the GB has moved upwards.

Figure 4 shows a Σ5 [100] tilt boundary between two lattices represented with white and black spots. Atomic shuffling is schematized by arrows pointing from black (grain B) to white dashed dots (growing A grain) and corresponds to a rotation within an elementary CSL unit cell. Considering that atomic shuffling occurs in every cell probably sequentially, the GB is moving globally upward by a distance equal to the CSL lattice parameter. Figure 4 shows deviations from Σ5 with a misorientation angle $\Delta\theta$ =14.25°, 8.8° and 6.36° which corresponds respectively to Σ13, Σ17 and Σ65. It can be seen that this deviation can be accommodated by a set of DSC dislocations with a burgers vector **b** perpendicular to the GB, respectively spaced by the distances labeled d_1, d_2 and d_3. Smaller misorientation angles can be obtained by an adequate set of DSC dislocations with a larger spacing. This means that a general tilt boundary around [100] can be decomposed into a perfect Σ5 GB and a set of DSC dislocations. This has already been pointed out by Fukutomi et al. [7]. We can now assume that since a perfect Σ5 GB is able to move by atomic shuffling, then the deformation produced by the motion of a slightly misoriented GB is given by the motion of the appropriate set of DSC dislocations. Applying (2) leads then to

$$\varepsilon = 2 \tan (\Delta\theta/2) \quad (3)$$

In the case of GB A/B, equation (3) with $\Delta\theta = 5°$, gives $\varepsilon = 8\%$, which is of the same order of magnitude than measured. This result seems to give a more qualitative agreement than the model proposes by Cahn. The latter however received a good agreement in the case of a GB motion in a bicrystal [8]. In our case, the fact that GB motions have to be a cooperative process where strain incompatibilities can arise between adjacent grains, could require more extensive atomic shuffling.

CONCLUSIONS

Stress-assisted grain growth was unambiguously established in nc Al at room temperature and in UFG Al at moderate temperature using in-situ TEM experiments. It was shown that GB motions induce strain of the order of few percents, smaller than values obtained from a pure DSC dislocation motion. However, a good qualitative agreement is found as soon as we assume that a general GB can be decomposed as a GB of a low coincidence index which migrates by atomic shuffling, and a set of DSC dislocations with a Burgers vector perpendicular to the interface plane forming a low angle tilt boundary. Therefore, the real deformation only results from the motion of this low angle boundary.

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REFERENCES

1. H. Mughrabi, H.W. Hoppel, M. Kautz, *Scripta Mater* **51**, 807 (2004).
2. D.S. Gianola, S. Van Petegem, M. Legros, S. Brandstetter, H. Van Swygenhoven, K.J. Hemker , *Acta Mat* **54**, 2253 (2006).
3. D. Farkas, A. Froseth, H. Van Swygenhoven, *Scripta Mater* **55**, 695 (2006).
4. J. W. Cahn, Y. Mishin, A. Suzuki, *Acta Mat.* **54**, 4953 (2006).
5. S. E. Babcock and R. W. Baluffi, *Acta Metall*. **37**, 2367 (1989).
6. H. Zhang and D. J. Srolovitz, *Acta Mater* **54**, 623 (2006).
7. H. Fukutomi, T. Iseki, T. Endo and T. Kamijo, *Acta Metall Mater* **39**, 1445 (1991).
8. D. A. Molodov, V. A. Ivanov, Gottstein, G., Acta Mater **55**, 1843 (2007).