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Observations of grain boundary protrusions in static recrystallization of high-purity bcc metals

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Protrusions at migrating high-angle grain boundaries were observed following partial recrystallization of high-purity body-centred cubic refractory metals. Their morphology is similar to that previously observed in aluminium and is in good agreement with the predictions of a kinetic model based on the sinusoidal variation of the local driving pressure along the boundary. The heterogeneous dislocation structure intersecting the boundary is considered to cause this sinusoidal variation and, therefore, the growth of protrusions in the high-purity metals.

Keywords: Serrated boundaries; Grain boundary migration; Recrystallized microstructure; Static recrystallization

Protrusions, also termed serrations or undulations, are frequently observed at migrating grain boundaries in static and dynamic recrystallization. Perhaps the first observation of protrusions is the one shown in Figure 1, adapted from the classical work of Beck and Sperry [1] on the static recrystallization of high-purity aluminium. Subsequently, protrusions were also observed by other authors in the static recrystallization of high-purity aluminium without [2,3] and with additions of 50 ppm of B [3], and of 0.1% and 0.3% Mn [4]. In the dynamic recrystallization (sometimes called geometric dynamic [5]) protrusions were also seen in high-purity aluminium [6,7], aluminium alloys [8–11] and other face-centred cubic (fcc) [12–14] and hexagonal close-packed metals [15–18]. They have also been found in minerals, such as quartz [19] and haematite [20], following dynamic recrystallization. However, to the best of the authors' knowledge, observations of protrusions in body-centred cubic (bcc) metals have not been reported in the literature. The purpose of this paper is to show that protrusions similar to those seen in fcc metals form in the static recrystallization of high-purity bcc metals due, probably, to the same mechanisms.

While in alloys the growth of protrusions could be a result of the pinning of parts of the boundary due to solutes or second-phase particles, this mechanism does not apply to high-purity metals. Kassner and McMahon [6], following Drury and Humphreys [8], and Bailey and Hirsch [21], suggested that protrusions probably develop at the junctions between subgrain boundaries (dislocation walls) and high-angle grain boundaries. At these junctions, there is a larger local driving pressure that pushes the high-angle boundary locally, overcoming the opposing pressure stemming from the boundary curvature and creating the protrusion. The dislocation walls of the subgrain structure could have formed during deformation or recovery, depending on the stacking fault energy and deformation conditions.

Recently, the development of protrusions during static recrystallization of commercially pure aluminium was observed in situ by Schmidt et al. [22] with a novel three-dimensional X-ray diffraction technique. Despite the presence of particles, those authors concluded that the dragging force due to particle pinning was negligible when compared with the driving force from the stored energy of plastic deformation (due to dislocations).

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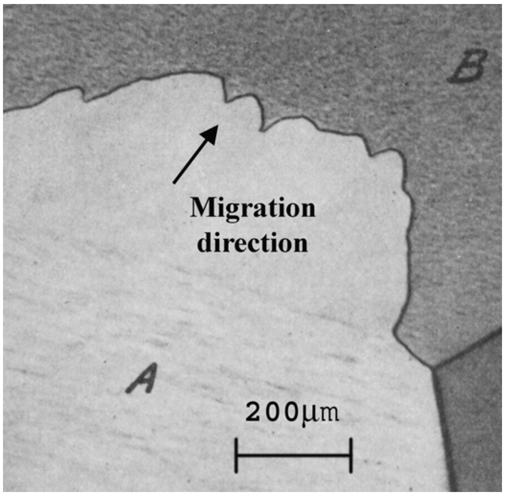

Figure 1. Optical microscope micrograph of a grain boundary with protrusions migrating by the mechanism of strain induced grain boundary migration in the static recrystallization of high-purity Al (from Beck and Sperry [1], with adaptations). The boundary migration direction is indicated.

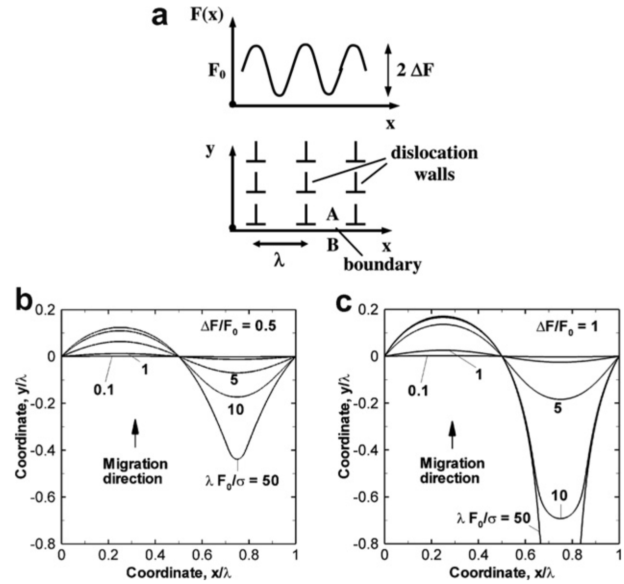

Figure 2. (a) Dislocation walls separated by interspacing λ intersect the initially flat two-dimensional boundary between two grains (A and B), one of which (A) has not yet recrystallized, as assumed by Martorano et al. [23]. The dislocation walls cause a sinusoidal driving pressure F on the boundary, of average F_0, amplitude ΔF and wavelength λ. Calculated steady-state shapes of the boundary are presented for various values of the ratio $\lambda F_0/\sigma$ and for: (b) $\Delta F/F_0 = 0.5$; (c) $\Delta F/F_0 = 1$ (adapted from Martorano et al. [23]).

Schmidt et al. [22] also agreed that protrusions were probably caused by variations in the driving pressure as a result of the non-uniform distribution of dislocations, arranged in dislocation walls, on the side to which the high-angle grain boundary migrated.

Although protrusions have been observed very frequently at migrating grain boundaries in recrystallization, only recently has their growth been modelled by Martorano et al. [23]. They proposed a novel form of the classic kinetic equation to describe, in two-dimensions, the movement of a grain boundary of constant mobility and energy subjected to a driving pressure due to boundary curvature (energy) and to a "bulk" driving pressure F (the excess strain energy density in the matrix) that varied sinusoidally along the boundary owing to parallel equidistant dislocation walls, perpendicular to the initial straight boundary (Fig. 2a). In their simulations, Martorano et al. [23] integrated the kinetic equation for the local boundary velocity [24,25] to show that protrusions develop at an initially flat boundary and, if the dislocation walls are long enough, reach a steady-state shape that depends on the dimensionless parameters $\Delta F/F_0$ and $\lambda\ F_0/\sigma$, as shown in the examples of Figure 2b and c. F_0 and ΔF ($\Delta F < F_0$) are the average bulk driving pressure and its amplitude, respectively; λ is the wavelength of the driving pressure, which is related to the distance between dislocation walls; and σ is the boundary energy per unit area. The results obtained for the steady-state regime show that the difference $\Delta\kappa$ between the absolute values of the curvatures at the valleys (κ_v) and peaks (κ_p) of the protrusions is positive and equals

$$\Delta\kappa = |\kappa_v| - |\kappa_p| = 2\frac{\Delta F}{\sigma} \tag{1}$$

As Figure 2b and c shows, $\Delta\kappa$ increases as $\Delta F/F_0$ increases at fixed $\lambda F_0/\sigma$. From Eq. (1) it can also be concluded that an increase in boundary energy, σ, decreases $\Delta\kappa$ at fixed ΔF, with no effect of F_0.

In the present investigation, selected microstructures showing grain boundaries migrating during the static recrystallization of two high-purity bcc metals (niobium and tantalum) were selected for a comparison with fcc aluminium (see Fig. 1) and also for comparison with the results of the Martorano et al. model [23]. High-purity tantalum and niobium ingots obtained by multiple electron-beam melting were chosen to perform this investigation. In the ingots, grain sizes ranged from 50 to 300 mm. Their interstitial impurity contents were as low as 20 wt.-ppm O (oxygen), 10 wt.-ppm N for Nb, and 65 wt.-ppm O (oxygen), 5 wt.-ppm N for Ta. Slabs of sizes $1 \times 7 \times 20$ and $3 \times 18 \times 50$ cm were cut out from the tantalum and niobium ingots, respectively, and were rolled to reductions of 33% (Nb) and 70% (Ta) in multiple passes at room temperature. From the rolled slabs, a metalographic sample was extracted at random and vacuum annealed for 1 h at 800 °C for the Nb sample and at 900 °C for the Ta sample. Static recrystallization was only partial in both cases. The migrating boundaries depicting protrusions at the recrystallization front were imaged using light optical microscopy (LOM) and scanning electron microscopy (SEM) in the backscattered electron mode (BSE) to increase the orientation contrast among neighbouring regions.

Protrusions observed at migrating grain boundaries, which formed during the static recrystallization of high-purity tantalum and niobium, are shown in the micrographs of Figures 3 and 4, respectively. In both figures, as well as in Figure 1 for high-purity aluminium, the migration direction, pointing to the boundary side of high dislocation density, is indicated. The morphologies of the protrusions in Figures 1, 3 and 4 are comparable

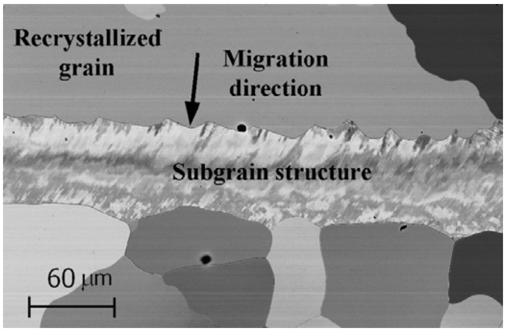

Figure 3. Grain boundary with protrusions migrating into a recovered region in the static recrystallization of high-purity tantalum (SEM, BSE). The boundary migration direction is indicated.

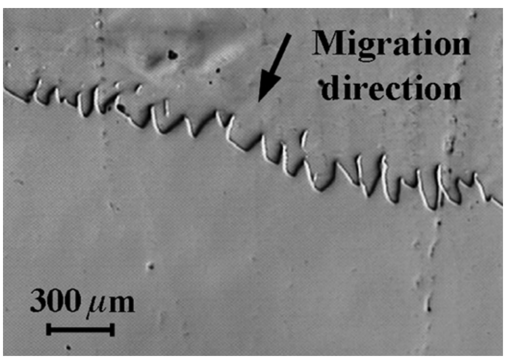

Figure 4. Grain boundary with protrusions migrating into a recovered region in the static recrystallization of high-purity niobium (LOM, Nomarski). The boundary migration direction is indicated.

with those calculated from the two-dimensional model for values of $(\lambda F_0/\sigma)(\Delta F/F_0) = (\lambda \Delta F/\sigma) \gtrsim 10$ (Fig. 2b and c). These values are within the range estimated by Martorano et al. [23] for common metals, e.g. $5 < (\lambda F_0/\sigma) < 250$ and $0 < \Delta F/F_0 < 1$. In addition, the calculated values of $\Delta y/\lambda$ (where $\Delta y = y_{\max} - y_{\min}$ is the amplitude of the protrusion) agree well with the experimental observation. This similarity between the calculated and observed protrusions strongly indicates that the observed protrusions are caused by an inhomogeneous microstructure that originates a varying bulk driving pressure on the migrating boundary. Since the aforementioned metals (aluminium, tantalum and niobium) are all of high-purity, the varying driving pressure was probably due to an inhomogeneous dislocation structure of cells and subgrains intersecting the boundary.

It is also worth noting that the portions of the migrating boundary with smaller curvature are in most cases at the migration front, as concluded above from the two-dimensional model. In their work on the dynamic recrystallization of quartz, Nishikawa et al. [19] also observed boundary protrusions caused by an inhomogeneous dislocation structure. These protrusions consisted of sharp cusps pointing to one side of the boundary and gently curved segments between the cusps bowing out to the other side, strongly resembling the protrusions and its curvature difference between peaks and valleys calculated by Martorano's et al. model [23]. It should be noted, however, that the curvature of normal sections of the boundary surface at any point varies between two extremes (the principal curvatures) as the section rotates. Therefore, any conclusion about curvature differences along the boundary from observations at two-dimensional sections should be drawn carefully.

The similarity of the morphology of the protrusions in the bcc metals and in fcc aluminium is apparent when Figures 1, 3 and 4 are compared, suggesting the presence of similar dislocation structures in both cases, in spite of the different crystalline structures and dislocation properties. Protrusions were found at only about 10% of the recrystallization boundaries observed in the present work, probably because an appropriate subgrain structure with fairly sharp and long dislocation walls formed in only a few cases. Note that protrusions, when formed, can be detected for most inclinations of the observation section plane relative to the boundary.

Protrusions were observed, for the first time to our knowledge, at migrating grain boundaries in the static recrystallization of bcc metals (tantalum and niobium). The experimental observations of protrusions were confronted with the findings of the two-dimensional model of Martorano et al. [23] based on a sinusoidal variation of the bulk driving pressure along the boundary. The shapes and sizes of the protrusions calculated with that model agree reasonably well with those observed in sections of the three-dimensional samples examined in this work. In particular, the prediction of smaller curvature at the protrusion peaks is largely confirmed by the experimental observations. The high-purity of the metals studied suggests that protrusions form because of the variable bulk driving pressure along the boundary caused by an inhomogeneous dislocation structure. The variable driving pressure has fairly sharp peaks at the dislocation walls that connect to the boundary, as suggested by Kassner and McMahon [6], Drury and Humphreys [8], and Bailey and Hirsch [21]. Both solute and particle dragging effects are absent in these high-purity metals. The protrusion shapes and sizes observed in the bcc metals resemble those observed by other authors in fcc aluminium, indicating similarities between the dislocation structures formed during plastic deformation or recovery. The fact that only a small fraction of the recrystallized boundaries observed in the present work exhibited protrusions can be attributed to an incipient development of a network of dislocation walls with fairly long cells and narrow cell walls of high stored energy, i.e. sub-boundaries of large misorientation angle.

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