"IN SITU" SYNCHROTRON WHITE BEAM X-RAY TOPOGRAPHIC STUDY OF GRAIN BOUNDARY MIGRATION

J. Gastaldi, C. Jourdan, G. Grange

To cite this version:

HAL Id: jpa-00230326
https://hal.archives-ouvertes.fr/jpa-00230326
Submitted on 1 Jan 1990

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
"IN SITU" SYNCHROTRON WHITE BEAM X-RAY TOPOGRAPHIC STUDY OF GRAIN BOUNDARY MIGRATION

J. GASTALDI, C. JOURDAN and G. GRANGE

CRMCZ-CNRS, Campus de Luminy, Case 913, F-13288 Marseille Cedex 9, France

Abstract - The methods of observation used so far (optical microscopy, electron microscopy...) do not give all the details necessary for the comprehension of mechanisms of grain boundary migration. The purpose of this work is to show that further information can be obtained by Synchrotron White Beam X-Ray Topography. As this method permits continuous monitoring of grain boundary displacement as well as species and density of localized defects "in situ" and in Real Time, it has been possible to establish that the grain boundaries which are moving during the recrystallization of a grain formed upon annealing of a deformed single crystal of aluminium have velocities and structure depending on their inclination relatively to the growing grain lattice, submitted to internal stresses ranging from $10^{-7}$ G (G being the shear modulus) to the limit necessary for the production of lattice dislocations. The third point has been confirmed by experiments on polycrystals of aluminium and of β titanium. All these results are in favour of a grain boundary migration mechanism involving atom transferring across the boundary. They also indicate that the mechanism can be disturbed by stresses spreading out locally in the boundary.

1 - INTRODUCTION

Despite considerable efforts made to elucidate them, the mechanisms of grain boundary migration (GBM) remain unknown. There is a lot of experimental evidences /1,2,3/ which prove that besides driving force F, temperature T and impurity concentrations C_i, both misorientation θ of the grains and inclination φ of the grain boundary (GB) plane, influence the GB velocity V. This means that any quantitative expression of V has to take into account the GB structure, which is now commonly accepted, insofar as this structure has been satisfactorily worked out for static GB at low temperature /4,5,6/ and as possibilities of phase transformation at high temperature have been considered /7,8/. Accordingly several expressions of GB velocity in pure materials, which
include parameters related to the features of static GB such as ledge and kink distances and vacancy concentration, have been given /9,10/. They are all deduced from the absolute rate theory and can be expressed in the following general form

\[ \Delta Q = \frac{D_0 \Delta F}{n kT} \left( \frac{\lambda_p, \lambda_k, \lambda_p}{C_V} \right) \]

\[ V = \frac{DO}{n kT} \cdot \frac{\Delta F}{\Delta Q} \left( \frac{\lambda_p, \lambda_k, \lambda_p}{C_V} \right) \]

where \( D_0 \) is the frequency factor, \( \Delta F \) is the driving force, \( \lambda_p \) and \( \lambda_k \) are the ledge and kink distances, \( C_V \) is the vacancy concentration, \( k \) is the Boltzmann constant, \( T \) is the absolute temperature, \( n \) is the number of atoms per unit boundary area and \( \Delta Q \) is the activation energy for GBM. Therefore, it is quite clear that the remaining problem is the determination of quantities associated with the GB structure which cannot be worked out without a better knowledge of the structure of moving GB and of the connected GB kinetics.

Several direct observations of moving GB have been attempted by using Conventional Transmission Electron Microscopy (C.T.E.M.) and High Voltage Electron Microscopy (H.V.E.M.) /11,12,13/. Besides sample thickness and irradiation damages which disturb the phenomenon /14,15/, the difficulties with these experiments are increased by the inability to monitor continuously features of moving GB. As these features (for example GB dislocations) can be clearly seen only before and after migration /16/. Therefore it has been impossible to determine precisely the part played respectively by steps and GB dislocations in the atom transferring from the absorbed grain to the growing one. As far as we know, "in situ" High Resolution Electron Microscopy (H.R.E.M.) investigations of GBM, such as those performed during Solid Phase Epitaxial Regrowth in Si and GaAs /17/, for example, have not yet been reported. Only a preliminary atomic-level study of the displacement of GB by Field Ion Microscopy has been carried out /18/. As for C.T.E.M. and H.V.E.M., difficulties with sampling and imaging have impede the observation of the actual process.

The most reliable information on this phenomenon has been inferred, so far, from GBM kinetics, by optical microscopic measurements performed at room temperature, after sample cooling. Thus this technique allowed determination of the above mentioned dependence of GB velocity on the various parameters \( F, T, C_i \) (see equation 1). However, it is obvious that the dependence on GB structure remains imprecise and there is a need of additional GB velocity measurements as a function of GB parameters \( \Theta \) and \( \Phi \) to clarify the relation \( V = f(\lambda_p, \lambda_k, \lambda_p, C_V) \). Furthermore, it can be noticed in equation (1) that \( \Delta Q \) is in turn a function of \( \lambda_p, \lambda_k \) and \( C_V \), which means that velocity measurements must be performed as a function of temperature for various GB structures.

GB kinetics and moving GB structure appear in any case tightly linked together. As they both suffered from discontinuous investigations, further progress can be expected from combined "in situ" studies /19,20/. An experiment which would permit "in situ" atomic level observation and concomitant kinetic data tracing, without phenomena disturbance, would be welcome, but has been, so far, somewhat unfeasible. Actually one has to choose between High Resolution and Low Disturbance. Developments in the former way are in the domain of electron microscopy and are presented at this symposium by other authors /21/. Progress can also be achieved following the second way and by using novel synchrotron X-ray sources. The purpose of this conference is to point out the results which have been obtained by Synchrotron White Beam X-Ray Topography (SWBXRT). The possibilities of conducting "in situ" and Real Time experiments in bulk samples by this method are described in section II. Afterwards (section III), SWBXRT studies of the kinetic and of the structure of moving GB in monocrystalline aluminum samples are related together with the features (stress, defects) which have been observed during the course of GBM in polycrystalline aluminum and titanium. Finally (section IV), existing mechanisms of GBM are reviewed in the light of all these results, before conclusion (section V).

(1) a preliminary "in situ" HRTEM observation of moving grain boundary in polysilicon at high temperature has in fact been presented at this congress: see this issue the paper at H. Ichinose, T. Kizuka and Y. Shida.
II - EXPERIMENTS

II.1 Possibilities of GBM investigations by SWBXRT

As previously reported /22,23/, SWBXRT derives basically from the LAUE diffraction technique and consists in analysing diffraction spots produced by a crystalline sample placed in front of the synchrotron X-ray beam. More precisely, owing to the special properties of this radiation (high intensity, high collimation, polychromatism and large cross section), these spots are real X-ray topographs (they have the same spatial resolution as those obtained by conventional X-ray topography \( \simeq 5 \mu m \)) and can be recorded either intermittently on High Resolution photographic plates (complete diffraction patterns) or continuously with a video camera (single spot) /24/. This is very convenient to characterize growing grains in bulk samples (thickness \( \simeq 1 \) mm for aluminum) at each stage of their growth. Information on the shape, the velocity \( V \), the nature (misorientation \( \theta \), inclination \( \phi \)) and the macrostructure of GB, as well as the matrix dislocations which they emit, can be deduced from these topographs with a fairly good precision (2° for \( \theta \) and \( \phi \), \( \Delta V/V = 0.01 \)) /25/. Instantaneous GB velocity determination can therefore be performed by SWBXRT, with continuous monitoring of evolutive parameters such as \( \theta \), \( \phi \), \( F \) (matrix dislocation density of the growing grain) and of the moving grain boundary macrostructure /26/ which satisfy nearly all the conditions recalled above (see section I) for "in situ" and Real Time studies of GBM.

II.2 Development of experiments for "in situ" and Real Time SWBXRT study of GBM

"In situ" and Real Time SWBXRT studies of GBM in aluminum and in titanium have been carried out /1/ with a multipurpose heating camera designed in our laboratories. This camera which is composed of an Ultra-High-Vacuum Chamber permeable to X-rays and inside of which is a furnace, has been extensively described in a previous paper /24/. Hereafter, we will only describe the procedures we followed on one hand to measure GB velocities, and on the other hand to avoid an increase of the low level of disturbances which characterizes the method (see section I).

II.2.1 Measurements of GB velocities

As it is desirable (see section I), GBM velocities have been measured as a function of \( T \), \( \theta \) and \( \phi \). They were determined "in situ" and in Real Time in faceted grains expanding into lightly deformed, annealed and indented monocrystalline aluminum of two grades (99.99 wt % and 99.999 wt %). Measurements were made on topographs perpendicularly to the traces of moving GB on external surfaces and give velocities which differ from the conventional migration velocities by \( \frac{1}{\cos \phi} /25/ \). Care was taken to perform these measurements only during the period within which the inclination \( \phi \) of the GB remains constant. This was checked instantaneously by gauging the width of GB projection on the topographs displayed by a video monitor and determined precisely afterwards on topographs /25/.

II.2.2 Experimental device reducing phenomenon disturbances

Phenomenon disturbances can arise from thermal and mechanical stresses generated either by the heating system or by the sample holder. These stresses have been reduced by using a resistance furnace inside of which the sample is held between two screws. They have been minimized by making some cuttings, with a sparking machine, around the bearing surfaces of the screws (figure 1,1) and (2)). This sample device was designed for an "ex situ" study of recrystallization and successfully tested by exposing X-ray topographs in the region of screw bearings of two recrystallized single crystals : one of them being cut and the other not (figure 1, 3) and (4)) /27/. It is clear that GB can migrate in cut sample without any influence of the long range stresses generated by the screws (figure 1, (4)).

(2) Synchrotron X-Ray topographs have been performed at the L.U.R.E. at Orsay (France).
Fig. 1 - Experimental device reducing phenomenon disturbances: (1) stirrup piece with two screws for sample fastening, (2) sample with cuttings around the screw bearings, (3) conventional X-ray topograph of a monocrystalline aluminum sample without cutting, (4) conventional X-ray topograph of a monocrystalline aluminum sample having cuttings. (From /27/)

III - RESULTS

As the recrystallized grains formed upon annealing of deformed single crystals of aluminum exhibit well delineated anisotropic growth /2/, "in situ" studies of GBM have been conducted mainly in these crystals. Faceting evolution, structure and kinetics of moving GB as well as stress concentration in their vicinity, have been thereby investigated. Further evidence of this stress concentration has been acquired by observing grain growth in polycrystalline aluminum and β titanium.

III.1 GBM in monocrystalline aluminum samples

III.1.1 Evolutionary faceting

SWXRT identifies as how grains growing near the indentations made on monocrystalline samples which have been previously deformed and annealed become progressively faceted. Although the low resolution of the method (a few μm) impedes the observation of the nucleation stage /28/, it was clearly seen that, at the beginning of the growth stage, the grains display rapidly a three-dimensional faceted shape /29/ (figure 2). Afterwards the three-dimensional faceted growth transforms into a two-dimensional one as soon as the size of the growing grains becomes as large as the sample thickness (figure 3). Inclinations of faceted GB have been determined during both the three and the two-dimensional growths. It appears that fast growing GB have random inclination and that the remaining growing GB are inclined parallel to dense crystallographic planes such as {100}{110} and {111}. This faceting disappears when several grains impinge. Thereby curved GB arise from the grain joining. It can be noticed that during further annealing, these curved boundaries straighten again as a result of the reduction of GB energy, during the
so-called grain growth stage which follows recrystallization.

III.1.2 Structure and kinetics of moving GB

As mentioned above (see section II), structure and kinetics of faceted moving GB have been investigated "in situ" and in Real Time by SWBXRT. Moving GB structure appears on synchrotron X-ray topographs as a long range periodic structure (1-50 μm) and is coarser than that of static GB evidenced by electron microscopy /26/. For a constant grain misorientation, it varies, like boundary migration rate, with the inclination of the GB relatively to the crystallattice of the growing grains. The most rapid (V > 10^-2 mm/s) GB are randomly inclined (see section III.1.1) and have a blurred structure (the time of response of existing high resolution detector becoming too slow ≥ 1 s). Among the slowest GB, those which are vicinal to {110} or to {100} exhibit parallel lines and move faster than those which are vicinal to {111} which have no parallel lines but only isolated dislocations and depth fringes (figure 4). Although it has been impossible to characterize fully the parallel lines /26/, it can be said undoubtly that this type of contrast corresponds to GB defects which have long range stress fields extending up to the limit of the resolution of SWBXRT (a few μm). These defects, which are more numerous in fast moving GB, indicate that the structure of these boundaries are more disturbed than those of slow moving one. Therefore the defective structure of moving boundaries can be considered as extending further than that of static boundaries which means that the GB movement increases considerably the number of extrinsic defects inside of the boundary.

The displacements x of well-defined facets have been plotted as a function of time t at various temperatures T. In general, x is a linear function of t and it is easy to verify the aforementioned relationship between the GBM rate V and the inclination of the boundary relatively to the lattice planes of the growing grains. Typical results for a Σ7 (θ = 38° <111>) boundary are presented in figure 5, wherein V of three grain boundary facets, approximately parallel to the crystallographic planes (111), (100) and (011) can be compared. It is noteworthy that V(011) > V(100) > V(111). Curves ln V = f(1/T) plotted for various GB facets, grain misorientations and impurity level, yield activation energy ΔQ ranging from 56 to 105 Kcal/mole and preexponential factors V₀ (intercepts at 1/T = 0) ranging from 10^10 to 10^23 cm/s. No systematic variation of slope and/or intercept for various GB inclinations (facets) and specimen purities was observed /30/.
Fig. 3 - Synchrotron X-Ray Topographs recorded during the two-dimensional growth of a faceted aluminum grain. Stress concentration in moving GB (depth fringe disturbance and linear kinematic contrasts) and matrix dislocation emission from these boundaries can be noticed.
Fig. 4 - Synchrotron X-ray topographs of GB moving in a monocrystalline sample aluminum: the GB planes of these boundaries are close to a) (101), b) (100), d) (111) planes of the growing crystal. (From /29/)

Fig. 5 - Plots of the displacement $x$ as a function of time $t$ of three well defined facets of a grain growing in a monocrystalline aluminum sample.
III.1.3 Stress concentration in moving GB

Transient contrast variations were observed in/or close to/moving GB during the "in situ" and Real Time study of GBM by SWBXT. These variations affect either kinematic or dynamic contrasts. An illustration of these variations can be seen in the series of SWBXT Topographs displayed in figure 3 and which were recorded during the growth of an aluminum grain. It can be noticed successively that, while the grain is expanding, 1°) depth fringes associated to the well developed boundary of faceted grain (figure 3a) become blurred (figure 3b), 2°) lateral boundaries are progressively underlined by kinematic contrasts (figure 3c), 3°) matrix dislocations are emitted at the level of kinematic contrasts (figure 3d), 4°) both linear kinematic contrasts and matrix dislocations leave the grain which recovers its previous quality (figure 3e). The detailed study of dynamic and kinematic variations of contrast has been reported elsewhere /31/. We have shown, in analysing the local variations of the interference order in the depth fringe network of a moving GB, that stresses of about $10^{-7}$ G (G being the shear modulus) can act perpendicularly to the moving GB plane /31/. These stresses are not sufficient to generate lattice dislocations which agree with the lack of such defects in the vicinity of dynamic contrasts (figure 3b). However, in the case of kinematic contrasts, although it is difficult to quantify the underlying stresses, the presence of lattice dislocations behind them (figure 3d,e) prove that these stresses are considerably higher (in the range of G/30). A mechanism taking into account the direction of stresses which has been determined (perpendicularly to the migrating GB plane) and the geometry of the observed defects which are mainly screw dislocations /32/ has been proposed to explain the generation of lattice dislocations by moving GB. According to the precautions which have been taken to avoid all external stress effects in the samples during the anneal (see section II.2.2), it can be stated that all the defects observed in the grains during the GBM are related to this phenomenon.

III.2 GBM in polycrystalline aluminum and titanium samples

Kinematic contrasts have also been observed in the vicinity of moving GB during the recrystallization of polycrystalline aluminum and β titanium /33, 34/. These contrasts were spreading farther than in monocrystalline samples, and were mainly detected when moving GB were slowing down and taking a highly curved shape by passing round an obstacle. At that time, many matrix dislocations were emitted and screw dislocations with a $a/2 \langle 110 \rangle$ Burgers vector have been identified in aluminum /33/.

IV - DISCUSSION

Results of "in situ" and Real Time observations of GBM in aluminum and titanium may be divided in two groups.

1°) those which are in favour of a GB mechanism involving an atom transferring across the boundary,

2°) those which indicate that whatever the mechanism may be, it can be disturbed during the GB displacement.

IV.1 Results in favour of a mechanism by atom transferring across the boundary.

It has been observed that GB which migrate in monocrystalline samples were always faceted and that the inclination, the structure and the migration rates of faceted boundaries are mainly related to the crystal lattices of growing grains (see sections III.1.1 and III.1.2). Accordingly, grain growth in monocrystalline samples appear to be like crystal growth from an isotropic phase (vapour or liquid) /35/. Although the analogy cannot be considered as complete /26/, it can be argued nevertheless that GBM involve also atomic jump across the boundary. Moreover, as GB displacements depend linearly on time (see section III.1.2), it can be added that GBM is interface-controlled and thereby is rate limited by the atomic jumps.

This mechanism may be substantiated by the large values of activation energies $\Delta Q$ and preexponential factors $V_0$ stemming from our investigations. Values of
AQ are three or four times greater than those previously reported /36/ which were in the range of activation energy of GB diffusion (15-20 Kcal/mole). Values of $V_0$ are several orders of magnitude larger than those of about $10^5$ cm/s generally found and which correspond to a single atom transferring across the boundary /36/. Therefore as these large values are not lowered when a purer aluminum is used (99,999 % instead of 99,99 %), it can be said that they are related to correlated atomic jumps /36/. Thus, in so far as we made kinetics measurements for well delineated faceted boundaries moving rather slowly at the end of the growth stage (see section III.1.2), we can expect that the displacement of these boundaries is facilitated by coordinated jumps parallelly to the GB plane whereas randomly inclined moving GB observed at the beginning of the growth (see section III.1.1) or previously investigated /36/, migrates by single atomic transferring perpendicularly to the grain boundary plane.

Although frequent variations of inclination preclude measurements of displacements of fast moving faceted grain boundaries, SWBXRT provides verification that grain growth proceeds continuously in these two regimes. Sometimes faceted GB slowed down and became curved, which indicates that elemental mechanisms were disturbed.

IV.2 Results which indicate disturbances of GB displacement

Stresses ranging from $10^{-7}$ G up to the limit necessary to create new dislocations inside a crystal (G/30) have been measured in the plane of moving faceted boundaries of grains growing in monocrystalline aluminum samples (see section III.1.3). The larger stresses have also been detected during the recrystallization of polycrystalline aluminum and β titanium. Accordingly, whatever the actual mechanism of GBM, it is not unreasonable to assume that, under the influence of the driving force, plastic and elastic incompatibilities can arise in the grain boundary plane at each step of the migration; that is, there is an interaction between the growing grain and the absorbed one which gives rise to local internal stresses which can relax instantaneously in the moving GB or in their vicinity. Thereby both the crystalline qualities of the moving GB and that of the grains may be changed according to the intensity of stresses. Small stresses are supposed to produce just depth fringe displacements and a concomitant increase of the interference order, middle stresses can explain the long range periodic structure and large stresses can generate matrix dislocations.

All these stresses can disturb the atom transferring occurring in the GBM mechanism proposed above. The number of extrinsic GB dislocations and of matrix dislocations intersecting the moving GB can be increased and thereby can activate locally GB displacements as to produce the bulges which were occasionally observed.

V CONCLUSION

SWBXRT allows us to establish that the GB which are moving during the recrystallization of a grain formed upon annealing of a deformed single crystal of aluminum:

1°) present an evolutionary faceting;

2°) have velocities and structure depending on their inclination relatively to the growing grain lattice;

3°) are submitted to internal stresses ranging from $10^{-7}$ G (G being the shear modulus) to the limit necessary for the production of matrix dislocations. The third point has been confirmed by experiments on polycrystalline aluminum and titanium. (G/30)

All these results are in favour of a GBM mechanism involving atom transferring across the boundary. They also indicate that this mechanism can be disturbed by stresses spreading out locally in the boundary.

ACKNOWLEDGEMENTS

This work was partly supported by the Centre National de la Recherche Scientifique and the National Science Foundation under Grants INT 84-13891 and DMR 85 506 436. The authors are grateful to professor C.L. Bauer for valuable discussion of the results and thank P. Marzo for his technical assistance and S. Hanania and F. Quintric for the preparation of the manuscript.
REFERENCES

/11/ Sehgal, H.K., Butler, E.P. and Swann, P.R., Scripta Met. 9 (1975) 165.
/12/ Rae, C.M.F., Phil. Mag. 44 (1981) 1395.
/14/ Roberts, W. and Lehtinen, B., Phil. Mag. 26 (1972) 1153.
/15/ Hutchinson, W.B. and Ray, R.K., Phil. Mag. 28 (1973) 953.
/16/ Rae, C.M.F. and Smith, D.A., Phil. Mag. 41 (1980) 477.
/21/ Bauer, C.L. and Scholz, R., this issue.