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Orientation Relationships between Grains and Grain Boundaries in Al–1 wt% Ga at the Beginning of Secondary Recrystallization

By

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The transition from the normal grain growth to the secondary recrystallization (abnormal grain growth) is studied in Al–1 wt% Ga. The orientations of individual grains and grain boundaries are determined with the aid of the selected area channelling method. The area of normal grain growth consists of small uniform grains. In this area almost exclusively twin boundaries are present having low energy and low mobility. Such grain boundaries are not present in the contact area between the abnormally growing large grains and the normal matrix. It is concluded that new grain boundaries with a high mobility are formed during the normal grain growth. Further kinetical selection of such grain boundaries can trigger the onset of the secondary recrystallization.

Изучен переход от нормального роста зерен к вторичной рекристаллизации (аномальному росту) в сплаве Al–1 вес% Ga. Ориентация индивидуальных зерен и границ зерен определялась с помощью каналирования электронов по методу избранной площади. В той части образца, где происходил только нормальный рост зерен, наблюдаются мелкие равноосные зерна. В этой области присутствуют почти исключительно двойниковые границы, обладающие малой энергией и малой подвижностью. Такие границы не наблюдаются в области контакта между аномально растущими крупными зернами и мелкозернистой матрицей. Сделан вывод, что во время нормального роста появляются новые границы зерен, обладающие высокой подвижностью. Кинетический отбор этих высокоподвижных границ может вызвать «запуск» вторичной рекристаллизации.

1. Introduction

The normal grain growth begins during the annealing of a cold deformed material after the end of the primary recrystallization when the deformed matrix is completely consumed by the newly formed grains with a low dislocation density. During the normal growth the sizes of individual grains are relatively uniform and the grain size distribution (i.e. the grain size d normalized by the mean grain size d_0) does not vary with the time t. Normally $d_0 \sim t^n$, where n should be equal to 0.5 as it follows from the theory of dimensions [1, 2]. Experimentally, n is known to lie between 0 and 0.5 [3 to 5]. In some conditions the normal grain growth suddenly changes to the so-called secondary recrystallization or abnormal grain growth [6, 7]. In this case few grains suddenly begin to grow very fast at the expense

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of all other recrystallyzed grains until these are consumed. The secondary recrystallization plays a very important role in defining the microstructure and texture of many technologically important materials [8]. In some cases, like for example Fe–Si magnetic alloys and W alloys for wires, the technologies already exist for many years which permit to control the secondary recrystallization and to achieve, therefore, the necessary magnetic and mechanical properties [9, 10]. In these materials the secondary recrystallization begins after suppressing the normal grain growth by pores or fine particles of a second phase. Such an inhibition of the normal grain growth was assumed to be a necessary condition for the secondary recrystallization.

In the last decade it was shown with the aid of computer modelling that the secondary recrystallization can also occur together with the normal grain growth a) if the energy of a grain boundary (GB) between normal and abnormal grains γ_a is lower than the energy of a GB between normal grains γ_m, b) if the mobility of GBs between abnormal and normal grains μ_a is higher than that of the GBs between normal grains μ_m, and c) if the size of abnormal grains d_a is large enough in comparison with d_m for the normal grains [11]. Later the conditions were analytically formulated for the mobility advantage $M = \mu_a/\mu_m$ and the energy advantage $G = \gamma_a/\gamma_m$ of the abnormal grains necessary for the occurrence of the secondary recrystallization [10, 11].

Unfortunately, the existing experimental data about the first stages of secondary recrystallization are scarce and not unambiguous. They concern mostly systems where the normal grain growth is inhibited by fine particles and the abnormal growth is triggered by the dissolution of such precipitates [14]. The goal of this work is an experimental determination of the crystallographic parameters of the GBs between abnormal and normal grains during the beginning of the secondary recrystallization in a single-phase alloy of high purity. It is well known that the transition from normal to abnormal growth in Al alloys is exceptionally sharp [15]. Recently, it was shown that the temperature of the beginning of the secondary recrystallization in technically important, very pure Al alloys lies by 200 to 300 K lower than usual and depends strongly on the impurity content in the range of a few ppm [14]. Particularly strong is the influence of Ga content on the grain growth in pure Al. Therefore, the Al–1 wt% Ga alloy was chosen for the measurements.

2. Experimental

The Al–1 wt% Ga alloy was produced from nominally 5N Al and 5N Ga as ingots with a mass of about 7 kg. The Ga content in this alloy lies well below the Ga solubility in the Al based solid solution (18 wt% at 0 °C and 10 wt% at 450 °C [16]). This fact and the high purity of the alloy components exclude the formation of any bulk precipitates which can influence the grain growth in the temperature interval studied. The flat plates were then mechanically cut from these ingots and cold rolled to a thickness e of 2.5 mm and 0.6 mm with the same reduction of 60%. During the rolling the plates were periodically cooled in liquid nitrogen in order to conserve their temperature below about −10 °C and to prevent the recrystallization. The cold rolled bands were annealed at 450 °C for 5 min in order to have a fully recrystallized structure without deformed matrix. The microstructure of the bands after this annealing consisted of uniformly recrystallized grains. The about 1 cm wide and 3 to 8 cm long areas corresponding to the rolled old grains were clearly seen. The difference of the mean grain size measured in the different old grains was about 30%. The recrystallized bands were cut into pieces with dimensions about $4 \times 5\ cm^2$. These pieces were then annealed for the grain growth at temperatures of 380, 405, and 425 °C. Each

specimen was annealed several times at the same temperature. After each break the sample was etched for 1 to 2 min in a solution of 10 ml HCl, 50 ml HNO_3, and 50 ml HCl in order to reveal the grain structure. The microstructure was photographed and the mean grain size d_0 was determined on 400 to 500 grains with aid of the intersection method using optical microscopy. The measurements were repeated after each new anneal in the same area in order to diminish the influence of the difference of the starting grain size between the different old grains.

One sample was specially prepared for the investigation of the onset of the secondary recrystallization. In this sample (e = 0.6 mm, annealed at 380 °C for 18 h) the fine grain structure typical for the normal grain growth was still present together with the large abnormal grains. The 3 mm wide and 10 mm long specimens were then cut from this sample in order to investigate their microstructure with the aid of the selected area channelling (SAC) technique [17]. The SAC technique was used in order to determine the mutual misorientations a) between the small "normal" grains in the recrystallized matrix and b) between the large abnormal and small normal grains. In usual X-ray techniques the data come from a large number of grains due to the large diameter of the X-ray beam. In this case the analysis is tedious, time consuming, and indirect because the actual single grain cannot be identified. Transmission electron microscopy (TEM) is the only technique which combines spatial specific diffraction information with fine microstructural details. However, there are known limitations for the application of TEM (small electron transparent region of the foil and the difficulty in relating the view area to the whole specimen). Techniques which occupy a position in between X-ray analysis and TEM are based on scanning electron microscopy. For example, in the SAC technique all grains can be seen and the misorientations are determined in the actual image [18]. It means that many grains can be analyzed, and the overall picture of the misorientation distribution can quickly be obtained. The orientations of the grains were measured "by hand" from a channelling pattern. A grain orientation can be obtained from a screen using only a ruler and standard map. This method was adopted as a simple computer program for routine orientation measurements. It requires the operator to identify poles in the channelling pattern. Then the screen coordinates of three points on each of two pairs of lines and of one pole are the input data into the computer program. These data are compared to a look-up table of rotations and interplanar spacings (band widths) in order to index these lines and thus solve the pattern. The orientation of a grain with respect to the specimen frame may be equally described both by rotation angles and rotation axis or by proper orthogonal matrices. There are 24 geometrically different, crystallographically equivalent rotations for cubic systems. Each of these rotations describes completely the orientational position of a grain in respect to the specimen frame. The direction cosines for the zone axis and for the axes perpendicular to channelling planes have been determined by a primary processing of the patterns. Those with respect to grain frame were obtained via an indexing procedure. In this work the channelling patterns and images were analyzed together. In this case the orientations can be ascribed to individual grains and related to both microscopic and macroscopic features of the specimen. Knowing the orientation of individual grains, the parameters of mutual misorientations of the neighbouring couples of grains were determined.

3. Results

Fig. 1 shows the time dependence of the mean grain size d_0 for three different temperatures. At the beginning of the anneal normal grain growth with diminishing growth rate occurs (solid lines). After about 20 to 40 h the abnormal grain growth begins (dashed lines). A few

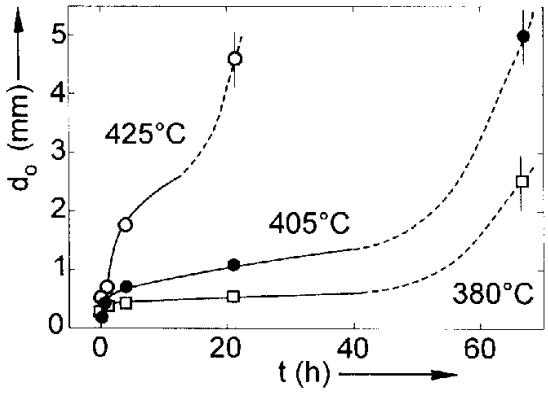

Fig. 1. The time dependence of the mean grain size in Al–1 wt% Ga cold rolled with a reduction of 60% during the normal grain growth (solid line) and secondary recrystallization (dotted line) at three different temperatures

grains begin to grow very fast in the fine-grained recrystallized matrix consuming it. Fig. 2 shows the typical microstructures at the different stages of the grain growth. The stage of the normal grain growth is shown in Fig. 2a when all grains are equiaxial and uniform. The microstructure at the intermediate stage is shown in Fig. 2b when the first abnormally large grains begin to grow in the fine-grained matrix. In Fig. 2c the microstructure is shown

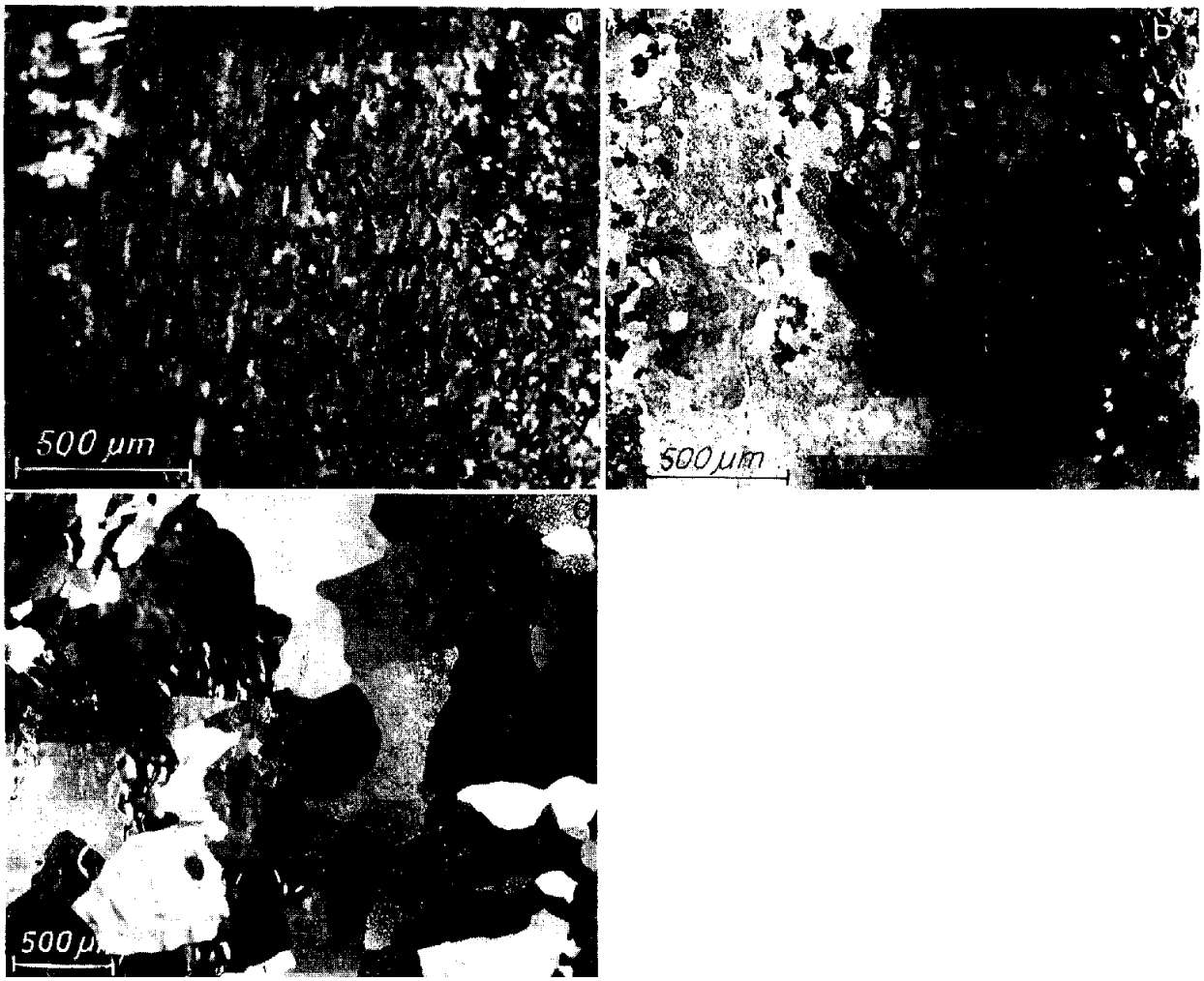

Fig. 2. The microstructure of Al - 1 wt% Ga samples a) after normal grain growth (e = 2.5 mm, 380 °C for 1 h), b) at the transition from the normal grain growth to the secondary recrystallization (e = 0.6 mm, 380 °C for 18 h), and c) at the final stage of the secondary recrystallization (e = 2.5 mm, 405 °C for 187 h)

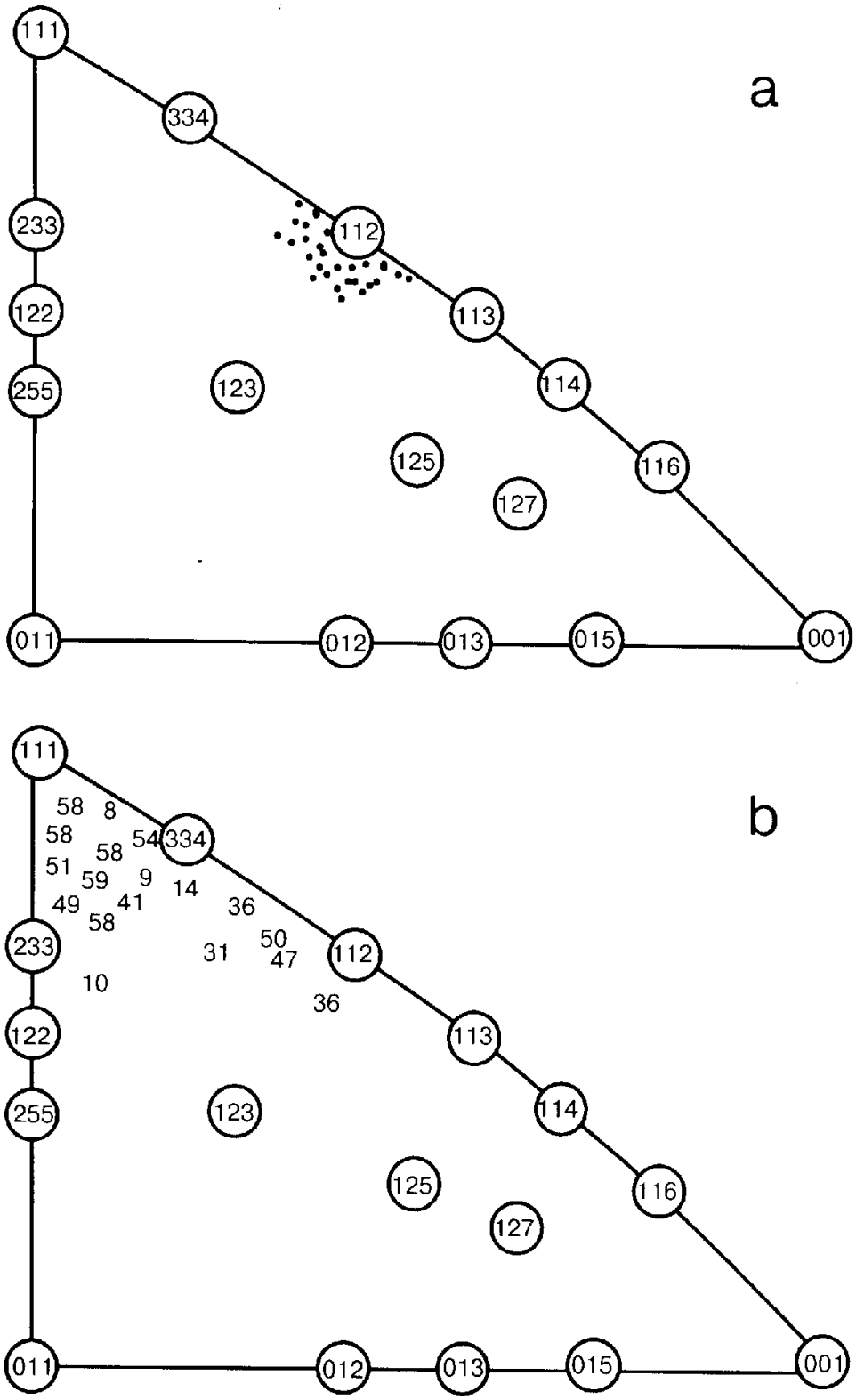

Fig. 3. The orientation parameters of grains and GBs in the fine grained matrix formed after normal grain growth in the sample shown in Fig. 2b. a) Inverse pole figure of grain normals, b) misorientation axes and angles of GBs. The position of the digit marks the misorientation axis of the corresponding GB

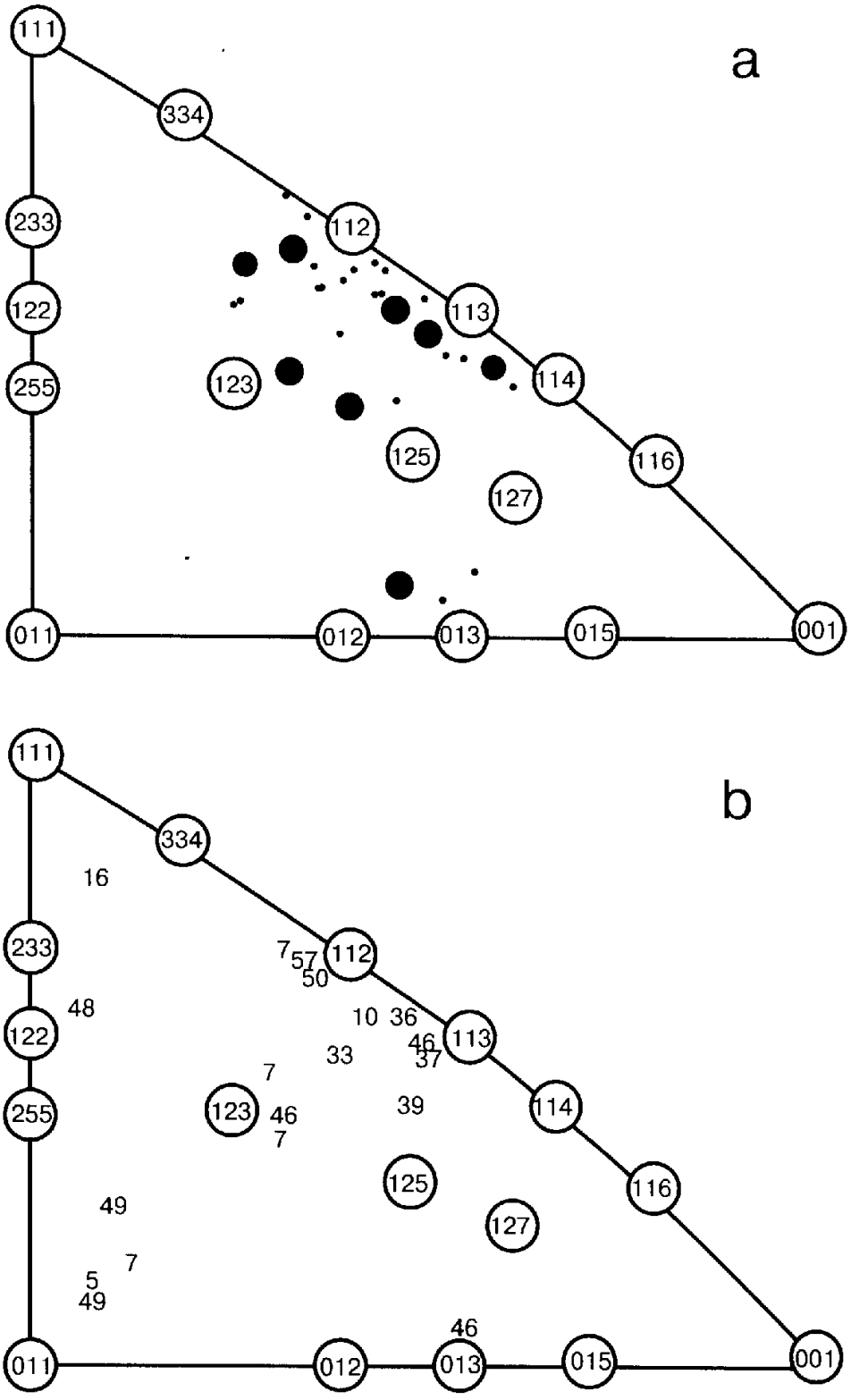

Fig. 4. The orientation parameters of grains and GBs in the contact area of the fine grained matrix formed after normal grain growth (small points) and of the large abnormally growing grains (big points) in the sample shown in Fig. 2b. a) Inverse pole figure of grain normals, b) misorientation axes and angles of GBs. The position of digit marks the misorientation axis of the corresponding GB

which consists almost exclusively of large abnormal grains and the fine-grained "normal" matrix is practically fully consumed.

Fig. 3 and 4 contain the information about the orientation of grains and the misorientation parameters of the GBs in the early stage of the secondary recrystallization when only about 20 to 30% of the fine grained matrix is consumed by the grains growing abnormally fast. This information was obtained with the aid of the SAC method. The microstructure of the sample is shown in Fig. 2. Fig. 3a shows the inverse pole figure of grain normals determined in the fine-grained matrix. In all grains studied the normal to their surface lies very close to the ⟨112⟩ axis. Fig. 3b reveals the GB misorientation axes and angles in the same fine crystalline area. Each number reveals the misorientation angle of the GB, its position marks the misorientation axis. The majority of the GBs studied has misorientation parameters close to those of the twin GBs which have the misorientation axis ⟨111⟩ and the misorientation angle $\varphi = 60°$. Fig. 4 displays the data for the border between 1. the area of large abnormal grains and 2. the area of small equiaxed normal grains. Fig. 4a shows the inverse pole figure for the large grains which grow abnormally fast and for the small grains in the "normal" matrix surrounding the large ones. The large points correspond to the large grains. The small ones reveal the orientations of the small grains. Fig. 4b shows the positions of the misorientation axes and the values of the misorientation angles for the GBs between the large grains and the small ones.

4. Discussion

The results of the SAC measurements show (Fig. 3) that in the fine-grained matrix, which contains the equiaxed grains formed after the primary recrystallization and the normal grain growth, all the grain normals have an orientation very close to the ⟨112⟩ direction. The majority of the GBs in this matrix have misorientation axes near ⟨111⟩ and the misorientation angles are close to 60°. This means that their misorientation parameters lie very close to those of the coherent twin boundaries. The twin GBs in Al and other metals are known to have a very low mobility which is close to that of small-angle BGs and is much lower than the mobility of high-angle GBs, both special and general ones [19, 20]. It is known also that the GBs possess special properties not only exactly at the coincidence misorientation φ_Σ with a low inverse density of the coincidence sites Σ. There is an interval of the misorientation angles $\Delta\varphi = 2|\varphi_c - \varphi_\Sigma|$ near the coincidence misorientation φ_Σ where the properties of the GBs differ very strongly from the properties of the general GBs which have an angle φ far from any φ_Σ [20]. The angular existence interval $\Delta\varphi$ of GBs with special properties decreases fastly with increasing Σ. The twin GBs ($\Sigma = 3$) conserve their special properties, for example the low mobility, in a large misorientation interval $\Delta\varphi$ ($\Delta\varphi_{\Sigma 3} \approx 10°$) [21]. At the border of the named existence interval φ_c the phase transition "special GB–general GB" occurs and the GB loses its special properties [22]. Two neighbouring grains with a misorientation φ close to φ_Σ can also have additional misorientation components. In this case the misorientation axis of two grains does not coincide exactly with the corresponding axis with low indexes. It was experimentally revealed that addition of the twist or the second tilt components affects the special GB properties much weaker than the same deviation of φ from φ_Σ [23]. Therefore, in our case (Fig. 3b) all the GBs with the misorientation axes lying in the triangle ⟨111⟩, ⟨233⟩, ⟨334⟩ and the misorientation angles φ in the interval from 49° to 60° can be treated as twin GBs which have a low energy and a low mobility. However, there is also a minority of grains in this array which cannot be treated as twins.

Let us consider now the area of the contact between the abnormally growing large grains in the "normal" matrix. Like in the previous case, the normals for most grains in this array lie close to the ⟨112⟩ direction, but the scatter of the orientations is now definitely wider (Fig. 4a). Some grains have even orientations close to the ⟨012⟩ and ⟨013⟩ directions. Fig. 4b shows the misorientation axes and angles for the GBs studied. There is one important difference in comparison with Fig. 3b: the GBs with a misorientation axis near ⟨111⟩ and φ near 60° are absent. Therefore, there are no twin GBs among the GBs on the front of the growing abnormal grains. The majority of these GBs have misorientation parameters lying far from the coincidence misorientations. Basing on the experimental data about the orientation dependence of the GB energy and mobility in Al we can conclude that the GBs in the front between the abnormal and normal growing grains have a high energy and high mobility [19, 24 to 26].

Therefore, the data obtained reveal that the GBs dividing the abnormal and normal growing grains have a) the advantage of a higher mobility and b) the disadvantage of a higher energy in comparison with the majority of GBs in the fine-crystalline matrix formed during the normal grain growth [12, 13]. It means that the mobility advantage M predominates over the energy disadvantage G and therefore makes possible the fast growth of the abnormal grains. Comparing Fig. 3 and 4 it can be concluded that the grains which prevail in the stage of the secondary recrystallization exist already in the stage of normal grain growth. During the normal grain growth the new GBs steadily form when the small grains dividing the large growing grains disappear. If the new GB formed in such a way has a misorientation φ far from φ_Σ and, therefore, possesses also a higher mobility, it will predominate over the old GBs during the further grain growth. The selection of such mobile GBs can trigger the abnormal growth.

The transition from the normal growth to the secondary recrystallization in Al was also studied with the aid of local X-ray diffractometry [27]. The authors have not found the nuclei of the abnormal grains at the stage of the normal grain growth. They suppose that the nuclei of the abnormal grains form due to the dissociation of existing GBs. We have studied the onset of the secondary recrystallization with the aid of the SAC method which has a higher spatial resolution in comparison with the local X-ray diffractometry. The data obtained reveal the pre-existence of abnormal grains during the normal grain growth. Therefore, we mean that for the onset of the secondary recrystallization in the Al 1 wt% Ga alloy not the nucleation of new grains is critical but the formation of new neighbourhoods between existing grains and therefore of new GBs having a high mobility.

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