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A representation method for grain-boundary character

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ABSTRACT

As is well known, a grain boundary (GB) is defined by five macroscopic parameters. We propose a method that is useful for representing the GB properties of polycrystalline materials as a function of these five parameters. The properties might include distribution, energy, mobility, segregation and wetting conditions. This method is based on the 'interface-plane scheme', proposed by Wolf and Lutsko, where a GB is characterized by two interface-plane normals and a twist angle ($\mathbf{n}_1, \mathbf{n}_2, \phi$). Considering the equivalent GB descriptions in cubic materials, the 'interface-plane scheme' space ($\mathbf{n}_1, \mathbf{n}_2, \phi$) is reduced to a unit triangle (100–110–111) for $\mathbf{n}_1$, a double unit triangle (100–110–111 and 100–101–111) for $\mathbf{n}_2$ and $0 \leqslant \phi < 2\pi$. All equivalent GBs whose two GB normals are within a given tolerance angle from reference planes are plotted as a function of the twist angle ϕ. This representation method is applied to the GB distributions of an Fe–Mn–Cu polycrystalline alloy. As a result, significantly high frequencies of the GB distribution were observed at (111)(111), $\Sigma = 3$, and small-angle boundaries.

§ 1. INTRODUCTION

In order to characterize a grain boundary (GB), eight geometrical parameters are needed. The eight parameters are divided into five macroscopic parameters and three microscopic parameters (Sutton and Balluffi 1995). The five macroscopic parameters include information on the misorientation of two adjacent grains and the inclination of the GB. The three microscopic parameters include information on the relative translation of the two adjacent grains. We focus here only on the five macroscopic parameters.

Since the choice of the five macroscopic parameters is arbitrary, three different schemes have been proposed. According to Bollmann (1970), a GB is defined by a three-by-three misorientation matrix and a GB normal vector. In his scheme, only three of the misorientation parameters are independent although the matrix has nine variables. This description is very useful because it allows easy mathematical calculations. However, it is not the most convenient method for graphical representations of GB properties. Since a rotation matrix can be converted to the rotation angle and axis, a GB may also be defined by a misorientation angle and axis and a GB normal vector (Goux 1974). As discussed by Wolf and Lutsko (1989), it is difficult to

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recognize the symmetries of GBs from this description, even if the GB is a simple symmetric tilt GB. In view of these disadvantages, Wolf and Lutsko developed an 'interface-plane scheme' where a GB is characterized by two GB normals with respect to the crystal coordinate systems of each of the adjacent two grains, and a twist angle. In their scheme, the GB symmetry with respect to tilt and twist character is represented explicitly (Wolf and Lutsko 1989).

Although the complete description of a GB requires five macroscopic parameters, most experimental studies of GBs in polycrystalline materials have analysed observed behaviour in terms of only three misorientation parameters (Haessner and Sztwiertina 1992, Hirano *et al.* 1998). This is because of the difficulty of measuring GB inclinations in polycrystalline materials (Randle 1998). However, analyses based on three macroscopic parameters are incomplete. For example, it is well known that $\Sigma = 3$ GBs in fcc cubic metals prefer to be bounded by two (111) interfaces. Molecular dynamic simulations have shown that the effects of inclination on the $\Sigma = 3$ GB energy are significant (Shmid *et al.* 1998). Ostuki and Mizuno (1986) have also shown similar effects experimentally in Al bicrystals. Therefore, proper analysis of GB properties in polycrystalline materials requires a complete description in terms of all five macroscopic parameters.

The objective of this study is thus to investigate a representation method for the properties of GBs over the whole space of five macroscopic parameters and to apply the method to the GB distribution of an Fe–Mn–Cu polycrystalline alloy.

§ 2. Theory

2.1. *Review of the 'interface-plane scheme'*

Since we shall adopt the 'interface-plane scheme' for the description of GBs, the basis of the scheme (Wolf and Lutsko 1989) will be reviewed first. We define the two grains on either side of a GB as grain 1 and grain 2. The GB is specified by an orthogonal matrix $\mathbf{g}$ of misorientation and a unit vector $\mathbf{n}$ normal to the GB. The grain orientations g_1 and g_2 of the two grains are specified by appropriate rigid-body rotations with respect to the reference frame (Bunge 1982). The misorientation Δg is written as

$$\Delta g = g_1 g_2^{-1}. \tag{1}$$

Additionally, we specify the unit vector $\mathbf{n}_s$ normal to the GB with respect to the reference frame. The GB normal vectors $\mathbf{n}_1$ and $\mathbf{n}_2$ with respect to the grain 1 and grain 2 frames are obtained by the following equations (note that one grain plane normal is taken to point outward, and the other to point inwards):

$$\mathbf{n}_1 = g_1 \mathbf{n}_s, \tag{2a}$$

$$\mathbf{n}_2 = g_2 \mathbf{n}_s. \tag{2b}$$

Combining these equations, the GB is specified by $(\Delta g, \mathbf{n}_1)$ (Bollman 1970).

A misorientation Δg is decomposed into two rigid-body rotations in the 'interface-plane scheme' as follows:

$$\Delta g = \mathbf{R}(\mathbf{n}_1, \phi) \cdot \mathbf{R}(\mathbf{n}_T, \psi). \tag{3}$$

Here, $\mathbf{R}(\mathbf{n}_1, \phi)$ is referred to as a twist rotation, which is a rigid-body rotation about the GB normal $\mathbf{n}_1$ by an angle ϕ. $\mathbf{R}(\mathbf{n}_T, \psi)$ is referred to as a tilt rotation, which is an

appropriate rigid body rotation about $\mathbf{n}_T$ by an angle ψ (figure 1). The tilt axis $\mathbf{n}_T$, which is perpendicular to $\mathbf{n}_1$ and $\mathbf{n}_2$, and the tilt angle ψ are given by

$$\mathbf{n}_T = \frac{\mathbf{n}_2 \times \mathbf{n}_1}{|\mathbf{n}_2 \times \mathbf{n}_1|}. \qquad (4)$$
$$\cos\psi = (\mathbf{n}_1 \cdot \mathbf{n}_2).$$

Therefore, a GB is specified by $(\mathbf{n}_1, \mathbf{n}_2, \phi)$ in the 'interface-plane scheme' (Wolf and Lutsko 1989).

2.2. *Grain-boundary characterization by the 'interface-plane scheme'*

The misorientation Δg is also described using a rigid-body rotation about the rotation axis $\mathbf{r}$ by the misorientation angle θ as follows (Synge 1960):

$$\Delta g = \mathbf{R}(r,\theta),$$
$$\theta = \cos^{-1}\left(\frac{\Delta g_{11} + \Delta g_{22} + \Delta g_{33} - 1}{2}\right), \qquad (5)$$
$$\mathbf{r} = \frac{1}{2\sin\theta}\begin{pmatrix} \Delta g_{32} - \Delta g_{23} \\ \Delta g_{13} - \Delta g_{31} \\ \Delta g_{21} - \Delta g_{12} \end{pmatrix}.$$

Comparing equations (3) and (5), we obtain the following relations between ϕ, ψ and θ:

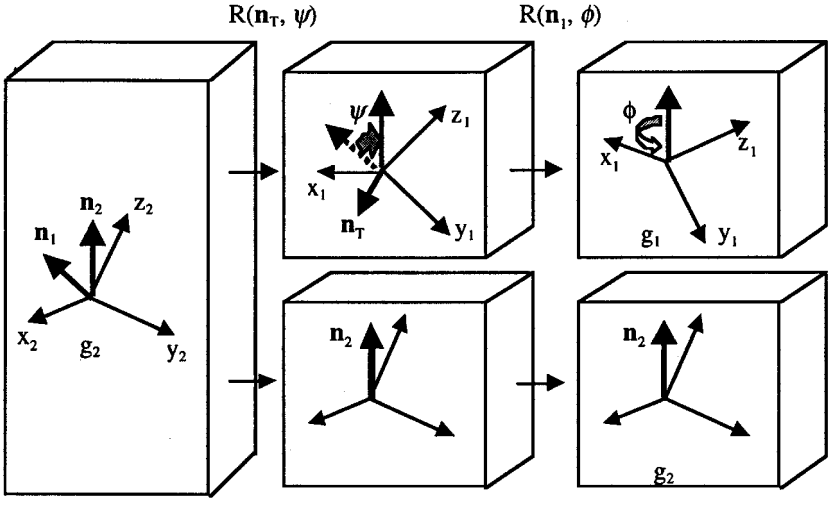

Figure 1. Creation of a GB by the 'interface-plane scheme' (Wolf and Lutsko 1989), that is successive applications of a tilt and a twist, where the twist axis is parallel to the boundary plane normal and the tilt axis lies in the plane of the GB.

$$\phi = \cos^{-1}\left(\frac{2(1+\cos\theta)}{1+\cos\psi} - 1\right), \qquad \text{when } \frac{\mathbf{m}}{|\mathbf{m}|}\cdot\mathbf{n}_1 = 1,$$

$$\phi = 2\pi - \cos^{-1}\left(\frac{2(1+\cos\theta)}{1+\cos\psi} - 1\right), \qquad \text{when } \frac{\mathbf{m}}{|\mathbf{m}|}\cdot\mathbf{n}_1 = -1,$$

$$\mathbf{m} = \begin{pmatrix} h_{32} - h_{23} \\ h_{13} - h_{31} \\ h_{21} - h_{12} \end{pmatrix}, \tag{6}$$

where

$$\mathbf{h} = \Delta g \mathbf{R}(\mathbf{n}_T, \psi)^{-1} = \mathbf{R}(\mathbf{n}_1, \phi).$$

Experimentally, a GB is characterized by g_1, g_2 and $\mathbf{n}_s$. Using equations (1), (2*a*), (2*b*), (5) and (6), a GB $(g_1, g_2, \mathbf{n}_s)$ is specified by $(\mathbf{n}_1, \mathbf{n}_2, \phi)$ in the 'interface-plane scheme'.

2.3. *Review of grain-boundary symmetries*

Since GB symmetries will be used to reduce the 'interface-plane scheme' space, they will be reviewed here. The grain orientations g_1 and g_2 are equivalent to $C_1 g_1$ and $C_2 g_2$ respectively. Here, C_1 and C_2 are symmetry operations. Since the misorientation Δg is defined by $g_1 g_2^{-1}$, Δg is equivalent to $C_1 \Delta g C_2^{-1}$. Therefore $(\Delta g, \mathbf{n}_1)$ is equivalent to $(C_1 \Delta g C_2^{-1}, C_1\mathbf{n}_1)$. For centrosymmetric crystals, $(\Delta g, \mathbf{n}_1)$ is also equivalent to $(\Delta g, -\mathbf{n}_1)$. Moreover, since the identification of the two grains adjacent to the boundary can be interchanged (A $\rightarrow$ B, B $\rightarrow$ A) without altering the boundary, $(\Delta g, \mathbf{n}_1)$ is equivalent to $(\Delta g^{-1}, \mathbf{n}_2)$. For the 24 proper symmetry operators in cubic materials, the GB $(\Delta g, \mathbf{n}_1)$ is equivalent to $(4 \times 24 \times 24 =)$ 2304 GBs (Morawiec 1998):

$$(\Delta g, \mathbf{n}_1) = (C_1 \Delta g C_2^{-1}, C_1\mathbf{n}_1), \tag{7a}$$

$$(\Delta g, \mathbf{n}_1) = (C_1 \Delta g C_2^{-1}, -C_1\mathbf{n}_1), \tag{7b}$$

$$(\Delta g, \mathbf{n}_1) = (C_1 \Delta g^{-1} C_2^{-1}, C_1 \Delta g^{-1}\mathbf{n}_1), \tag{7c}$$

$$(\Delta g, \mathbf{n}_1) = (C_1 \Delta g^{-1} C_2^{-1}, -C_1 \Delta g^{-1}\mathbf{n}_1). \tag{7d}$$

Note that, independent of the choice of symmetry operators in this scheme, the minimum misorientation angle, or disorientation angle, can be found by applying equation (5) to $(C_1 \Delta g^{-1} C_2^{-1})$. This leads, of course to a different choice of C_1 (and C_2).

2.4. *Reduced space of the 'interface-plane scheme'*

In order to represent the GB properties graphically, the 'interface-plane scheme' space $(\mathbf{n}_1, \mathbf{n}_2, \phi)$ should be reduced by the use of the equivalent GBs reviewed in the previous section. From equations (7*a*) and (7*b*), one of the products of $\mathbf{n}_1$ with symmetry operations C_1 or $-C_1$ falls into a single unit triangle, such as (100–110–111). We define the particular product that falls into a single unit triangle (100–110–111) as $\mathbf{n}_1^*$. From equations (2*a*) and (2*b*), the GB normal vector $\mathbf{n}_2$ is written as $\mathbf{n}_2 = C_2 \Delta g^{-1}\mathbf{n}_1^*$. Therefore, $\mathbf{n}_2$ falls into a double unit triangle, consisting of (100–110–111) and (100–101–111). In the same way, from equations (7*c*) and (7*d*), one of

the products of $\Delta g^{-1}\,\mathbf{n}_1$ with symmetry operations C_1 or $-C_1$ falls into a single unit triangle, such as (100–110–111). For the product of $\Delta g^{-1}\,\mathbf{n}_1$ with the particular symmetry operation, $\mathbf{n}_2$ falls into a double unit triangle. Therefore, the 'interface-plane scheme' space $(\mathbf{n}_1, \mathbf{n}_2, \phi)$ is reduced to a unit triangle (100–110–111) for $\mathbf{n}_1$, and a double unit triangle (100–110–111 and 100–101–111) for $\mathbf{n}_2$, which requires consideration of $0 \leqslant \phi < 2\pi$.

2.5. *Representation of grain-boundary properties*

We shall now describe a method that is useful for representing GB properties as a function of the five macroscopic parameters extracted from polycrystalline samples.

First, the reference poles $\mathbf{p}_i$ for the GB normal vectors $\mathbf{n}_1$ are selected in a single unit triangle: (100–110–111). For our purpose, we selected 12 reference poles $\mathbf{p}_i$ $(i = 1,2,\ldots,12)$ (figure 2(*a*)). Second, the reference poles for the GB normal vector $\mathbf{n}_2$ are selected in the first unit triangle (100–110–111) and a neighbouring second unit triangle (100–101–111). We selected 14 reference poles $\mathbf{q}_j$ $(j = 1,2,\ldots,14)$ in the double unit triangle (figure 2(*b*)). Here, $\mathbf{p}_i$ equals $\mathbf{q}_i$ for $i = 1,2,\ldots,12$. The reference poles in the second unit triangle should lie inside that triangle because the poles along the borders of the second unit triangle are equivalent to the corresponding poles on the borders of the first.

After a GB is specified experimentally by $(g_1, g_2, \mathbf{n}_s)$, all equivalent descriptions in the tilt–twist decomposition scheme, $(\mathbf{n}_{1k}, \mathbf{n}_{2k}, \phi_k)$ $(k = 1,2,\ldots,2304)$, are calculated. If the angles between $\mathbf{n}_{1k}$ and $\mathbf{p}_i$ and between $\mathbf{n}_{2k}$ and $\mathbf{q}_j$, are smaller than a preselected tolerance angle β, the GB $(g_1, g_2, \mathbf{n}_s)$ is specified by $(\mathbf{p}_i, \mathbf{q}_j, \phi_k)$.

According to the above discussion on equivalent GBs, the following combinations of $\mathbf{p}_i$ and $\mathbf{q}_j$ are equivalent among all the possible combinations of $\mathbf{p}_i$ and $\mathbf{q}_j$ $(12 \times 14 = 168$ combinations in figure 2):

(i) $(h_1,k_1,l_1)(h_2,k_2,l_2)$ and $(h_2,k_2,l_2)(h_1,k_1,l_1)$, such as (111)(533) and (533)(111);

(ii) $(h_1,k_1,l_1)(h_2,k_2,l_2)$ and $(h_1,k_1,l_1)(h_2,l_2,k_2)$, such as (111)(321) and (111)(312); here, (h_1,k_1,l_1) lies on the border of the unit triangle.

(iii) $(h_1,k_1,l_1)(h_2,k_2,l_2)$ and $(h_2,l_2,k_2)(h_1,l_1,k_1)$, such as (321)(713) and (731)(312); here, (h_1,k_1,l_1) and (h_2,k_2,l_2) lie inside the first and second unit triangles, respectively.

Considering these 87 equivalent GBs, $168 - 87 = 81$ combinations of $\mathbf{p}_i$ and $\mathbf{q}_j$ are needed to represent the properties in the whole space of the GBs. This admittedly

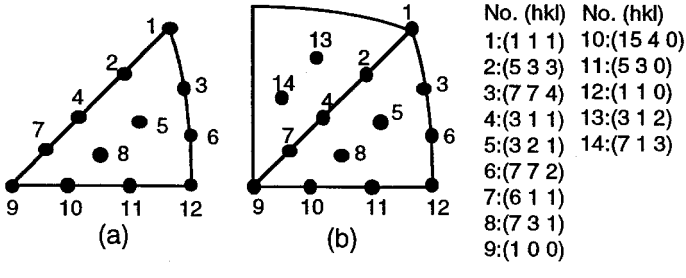

Figure 2. Reference poles for GB normal vectors.

coarse discretization of the space was adequate for an analysis of grain boundary wetting. Other applications may require a more detailed discretization.

§ 3. Application

The representation method described above is now applied to the GB distribution of an Fe–Mn–Cu polycrystalline alloy. In a previous study (Takashima *et al.* 1998), we showed that the GBs in the microstructure are 'wet' (i.e. exhibit a continuous Cu film), 'dry' (i.e. exhibit no Cu film), or 'mixed' (i.e. partly wet and partly dry). However, we focus here only on the GB distribution, without regard to the wetting information. The analysis of the relation between wetting and the five macroscopic parameters will be discussed elsewhere (Takashima *et al.* 1999).

3.1. *Sample preparation*

Fe–30 wt% Mn–10 wt% Cu alloy samples 10 mm thick were cold rolled to 1 mm, annealed at 1120°C in evacuated quartz tubes for 5 h, and quenched.

To determine the five macroscopic parameters of the GBs, not only adjacent grain orientations but also the GB inclinations have to be measured. These were investigated by electron back-scattering patterns (EBSPs) together with serial sectioning, as described below.

The GBs and the Cu-rich wetting phase in the samples were revealed by chemical polishing in 3% HF–97% H_2O_2 followed by chemical etching in 5% HNO_3–95% C_2H_5OH. The microstructure was observed with a scanning electron microscope, and grain orientations were determined with EBSP. After removing a surface 8 μm thick by mechanical polishing, microstructural and EBSP observations were repeated on the same area. The thickness of the layer removed by polishing was determined by the change in the size of microhardness indentations of known aspect ratio, and registry between the first and second sections was obtained by minimizing the mean square deviations of 200 selected reference points. GB inclinations were obtained by comparing GB positions on the first and the second sections. Such data were acquired for a total of 975 GBs.

3.2. *Examples*

The GB distribution was normalized by the distribution of 100 000 simulated random GBs. The random GBs were generated by a combination of random misorientations Δg (Morawiec 1999) and random unit vectors $\mathbf{n}_1$.

The distribution of $(\mathbf{p}_i, \mathbf{q}_j, \phi)$ GBs are plotted as a function of ϕ. Although 81 figures are needed to represent the whole GB space, as discussed above, we show only four figures as examples. The tolerance angle β has been taken to be 9° based on the discretization described above.

The four graphs in figure 3 show examples of the GB distribution, in which the bars represent the frequency of observations relative to the random GB distribution, and lines show the calculated disorientation angle. As noted above, the disorientation angle was found separately from the interface-plane classification, and was used to identify small-angle boundaries. For (111)(111) ϕ GBs, the relative frequency of the observed GB distribution to the random GB distribution was greater than 20 at $\phi = 60°$, 180° and 300°, which correspond to (111)(111), $\Sigma = 3$, twin GBs. For (110)(110) ϕ GBs, the frequencies were high at $\phi = 0°$, 180° and 360°, which correspond to small-disorientation GBs. Although the GBs at $\phi = 70°$, 110°, 250° and 290° are $\Sigma = 3$ GBs, their relative frequencies were essentially zero. For (311)(311) ϕ

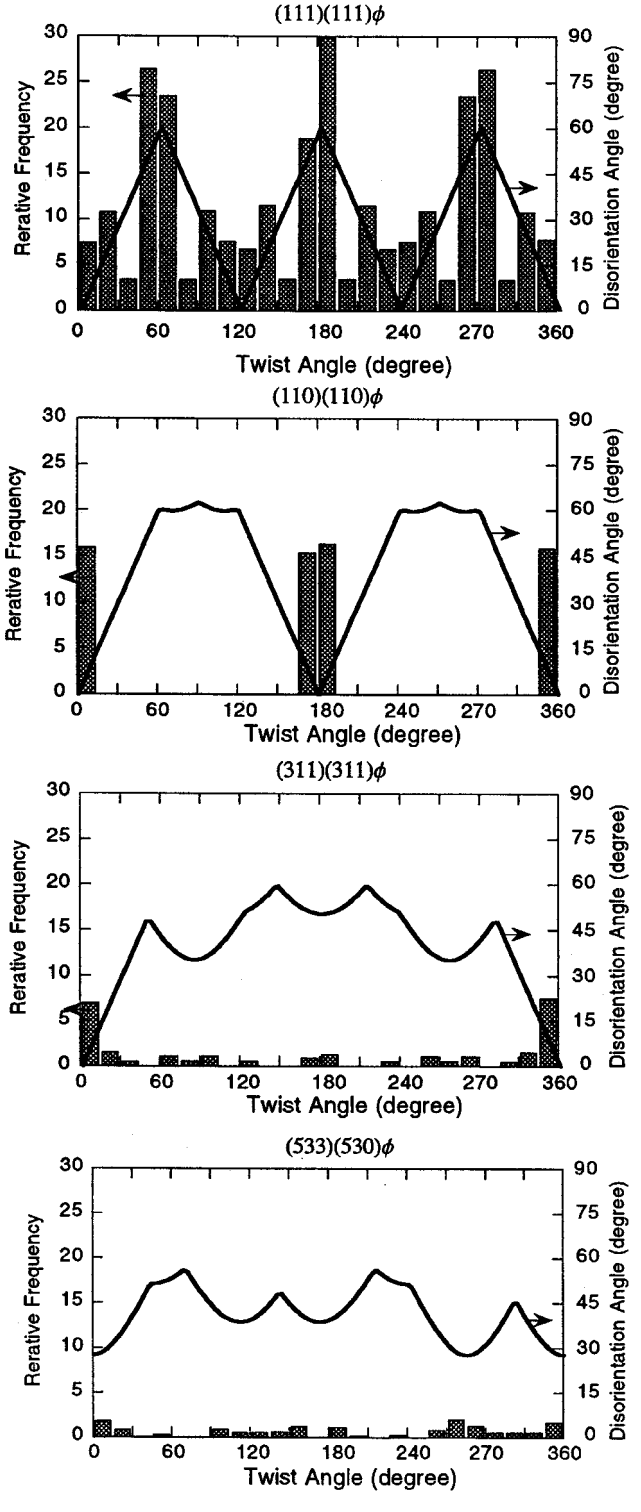

Figure 3. GB distribution displayed as frequency of observation relative to a random GB distribution (bars), and calculated disorientation angles (line) for an Fe–Mn–Cu alloy.

GBs, the frequencies were high at $\phi = 0^\circ$ and 360°, which correspond to small-disorientation GBs. In fcc metals, (311)(311), $\Sigma = 11$ GBs ($\phi = 180^\circ$) are known to display a low energy, like (111)(111), $\Sigma = 3$ twin GBs. However, their frequency was relatively low. No significant high frequencies were found in (533)(530) ϕ GBs at any twist angle.

§4. Conclusions

A method that is convenient for the representation of GB properties in polycrystalline materials, as a function of the five macroscopic parameters, has been proposed. This method is based on the 'interface-plane scheme', where a GB is characterized by two interface-plane normals and a twist angle ($\mathbf{n}_1, \mathbf{n}_2, \phi$).

(1) Equations, which allow calculation of all equivalent GB descriptions with the 'interface-plane scheme', have been provided.
(2) By considering equivalent GB descriptions, the 'interface-plane scheme' space ($\mathbf{n}_1, \mathbf{n}_2, \phi$) is reduced to a unit triangle (100–110–111) for $\mathbf{n}_1$, and to a double unit triangle (100–110–111 and 100–101–111) for $\mathbf{n}_2$ and $0 \leqslant \phi < 2\pi$.
(3) All equivalent GBs with two GB normals which fall within a given tolerance angle from reference planes were plotted as a function of twist angle ϕ.

This representation method was applied to the GB distribution of an Fe–Mn–Cu polycrystalline alloy. Whereas (111)(111), $\Sigma = 3$ and (311)(311), $\Sigma = 11$, GBs are known to have low energies in fcc metals, our observations show that the relative frequency of the (111)(111), $\Sigma = 3$, GB was greater than 20, but that (311)(311), $\Sigma = 11$, GBs only appear at the same frequency as random GBs. In addition, high frequencies were generally observed for small-disorientation-angle GBs. Using this method, the GB properties can be represented over the whole GB space, including asymmetric GBs such as (533)(530) ϕ.

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