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SYMMETRIES OF GRAIN BOUNDARY DISTRIBUTIONS

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Abstract

A function representing the distribution of grain boundaries is needed for a statistical description of the boundaries in polycrystalline aggregates. It is assumed to be determined in the space of macroscopic boundary parameters. Equivalences between points of the space caused by crystal symmetries lead to symmetries of the grain boundary distribution. These symmetries are investigated for both homophase and heterophase grain boundaries. Using known asymmetric domains for misorientation distributions, a method of determining the domains of grain boundary distributions is given for all possible combinations of crystallographic symmetries.

Introduction

Many of the investigations of sharp grain boundaries in polycrystalline materials are case studies but new measurement techniques (see, e.g., [1]) allow a statistical approach. This is, for instance, the case when frequencies of CSL boundaries are considered. However, one would like to be able to analyze grain boundaries statistically in a more complete way, for all possible grain misorientations and boundary inclinations. This is possible by using the distribution (density function) of grain boundaries over the macroscopic boundary parameters. The distribution may play a role similar to that of the orientation distribution in crystallographic texture analysis.

In order to define a grain boundary distribution (GBD), a measure in the parameter space is needed. The measure, however, is not unique, and thus GBD depends on its choice. The situation is different than in the case of the orientation distribution which is determined on the special orthogonal group with a natural choice of the unique invariant volume. Let us also mention that if metric properties of the space are given, they determine the volume element. All these facts have to be taken into account when defining the GBD.

Moreover, because of crystal symmetries, the boundary parameters are not unique, i.e., a number of different sets of parameters represent the same geometrical arrangement at the boundary. The GBD must take equal values for these arguments, and thus, it also exhibits certain symmetries. Its domain can be reduced to an asymmetric domain, i.e., to a part of the parameter space in which each physically distinct boundary is represented only once. The aim of this paper is to investigate possible symmetries of distributions for both homophase and heterophase boundaries.

Definition of GBD

Only macroscopic grain boundary parameters are considered here, i.e., 3 parameters for the misorientation of grains, 2 for the local inclination of the boundary and one discrete parameter for the change of handedness. (See, e.g., [2].) The space in which boundaries are determined consists of (proper and improper) rotations and unit vectors locally normal to the boundary surfaces. The former may be identified with the manifold of orthogonal matrices $O(3)$ and the latter, via Gauss mapping, with the unit sphere S^2 . Thus, in general the domain of the distribution is $O(3) \times S^2$. If enantiomorphic crystals are treated as different phases then improper rotations can be excluded and the space can be confined to the connected component $SO(3) \times S^2$, with $SO(3)$ containing special orthogonal matrices.

A boundary will be specified by an orthogonal matrix g of misorientation and a unit vector \mathbf{n} normal to the boundary. It is assumed that a Cartesian coordinate system is attached to the crystal structure and the components of \mathbf{n} are given in that system, and g relates the systems of two neighboring crystallites. To be more precise, let us consider two neighboring crystallites numbered 1 and 2. All quantities related to the first crystal will have index 1 attached; similarly, 2 will be attached to those related to the second crystal. Let the orientations of the crystallites with respect to an external coordinate system be given by orthogonal matrices g_1 and g_2 (*cf* [3]). The grain boundary between the *first* grain and the *second* one is specified by the misorientation $g = g_1 g_2^T$ and the normal to the boundary \mathbf{n}_1 (by convention) directed towards the second grain, with coordinates specified in the coordinate system of the first crystallite; i.e., the boundary is determined by (g, \mathbf{n}_1) . The coordinates of the normal to the boundary in the Cartesian coordinate

system of the second grain are given by $g^T \mathbf{n}_1$. Thus the grain boundary between the *second* grain and the *first* one has the form (g^T, \mathbf{n}_2) with the normals \mathbf{n}_1 and \mathbf{n}_2 related through

$$\mathbf{n}_2 = -g^T \mathbf{n}_1 . \quad (1)$$

Once the volume element dV is chosen, the GBD, say f , can be defined. Its value at the point (g, \mathbf{n}) times an infinitesimal volume dV centered at that point is equal to the ratio of the area dS of boundaries with parameters within dV to the complete area of boundaries S

$$dS/S = f(g, \mathbf{n}) dV .$$

It is assumed that $\int dV = 1$, and the distribution corresponding to $f = 1$ is considered to be random.¹

As was mentioned in the Introduction, the expression for a volume element follows from a metric structure determining a distance between points of the manifold, i.e., representing the degree of closeness between grain boundaries. With parameters x^i ($i = 1, \dots, 5$) and metric tensor d , the element dV is given by $dV = \sqrt{|\det(d)|} dx^1 dx^2 \dots dx^5$.

In the case of $SO(3) \times S^2$ the most natural choice of metric is the product metric of the unique invariant metric of $SO(3)$ and the unique canonical metric on S^2 inherited from the Euclidean space. For two points (boundaries) (g, \mathbf{n}) and (g', \mathbf{n}') , the distance on $SO(3)$, say χ_\bullet , is related to the smallest angle ω of rotation necessary to transform g into g' , and the distance on S^2 , say χ_\circ , is related to the angle γ between the vectors \mathbf{n} and \mathbf{n}' . It is assumed that $\chi_\bullet^2 = 2(1 - \cos(\omega)) = 3 - \text{tr}(g^T g') = \|g - g'\|^2/2$ and $\chi_\circ^2 = 2(1 - \cos(\gamma)) = 2(1 - \mathbf{n} \cdot \mathbf{n}') = \|\mathbf{n} - \mathbf{n}'\|^2$, where the norm $\|\cdot\|$ of a matrix X is defined by $\|X\| = (\text{tr}(X^T X))^{1/2}$. (Because for ϵ close to 0 one has $2(1 - \cos(\epsilon)) = \epsilon^2 + O(4)$, the local metric properties given by the angles ω and γ are the same as for much simpler $\chi_\bullet(g, g')$ and $\chi_\circ(\mathbf{n}, \mathbf{n}')$, respectively.) Let the finite distance χ between (g, \mathbf{n}) and (g', \mathbf{n}') be determined by

$$\chi^2((g, \mathbf{n}), (g', \mathbf{n}')) = \chi_\bullet^2(g, g') + \chi_\circ^2(\mathbf{n}, \mathbf{n}') . \quad (2)$$

Grain boundaries can be compared by checking their distance χ . Similar boundaries are close in the space and thus their distance is small.

To give an example, let us concentrate on a specific choice of parameters. For the unit sphere S^2 , the spherical coordinates (α, β) with $\alpha \in [0, \pi]$ and $\beta \in [0, 2\pi)$ are defined in such a way that the Cartesian coordinates of \mathbf{n} are $n_1 = \sin \alpha \cos \beta$, $n_2 = \sin \alpha \sin \beta$ and $n_3 = \cos \alpha$. In these coordinates the metric on S^2 can be expressed in the well known form $d\chi_\circ^2 = d\alpha^2 + \sin^2 \alpha d\beta^2$. (It is obtained by determining square of the distance of points with parameters (α, β) and $(\alpha + d\alpha, \beta + d\beta)$.) Thus, the metric tensor is diagonal with 1 and $\sin^2 \alpha$ on the diagonal. With total volume normalized to 1, the volume element dV_\circ on the sphere is $dV_\circ = (4\pi)^{-1} \sin \alpha d\alpha d\beta$.

As for the metric on $SO(3)$, let the coordinates be given by the Euler angles $\varphi_1, \phi, \varphi_2$ (see [3]). The invariant metric on $SO(3)$ in this parameterization has the form $d\chi_\bullet^2 = d\varphi_1^2 + 2 \cos(\phi) d\varphi_1 d\varphi_2 + d\varphi_2^2 + d\phi^2$. One easily gets the invariant volume element dV_\bullet ; with the volume normalized to 1 it is given by $dV_\bullet = (8\pi^2)^{-1} \sin(\phi) d\varphi_1 d\phi d\varphi_2$.

For this particular choice of parameters $(\alpha, \beta, \varphi_1, \phi, \varphi_2)$ the product metric on $SO(3) \times S^2$ is $d\chi^2 = d\alpha^2 + \sin^2 \alpha d\beta^2 + d\varphi_1^2 + 2 \cos(\phi) d\varphi_1 d\varphi_2 + d\varphi_2^2 + d\phi^2$, and the corresponding volume element has the form $dV = (32\pi^3)^{-1} \sin(\phi) \sin(\alpha) d\varphi_1 d\phi d\varphi_2 d\alpha d\beta$ (see [4]). Again, the volume is normalized to 1.

¹The above definition has some features common to that given by Adams [4]. However, in that case, inclinations are referred to external (sample) reference frame and thus the arguments of the function are other than the macroscopic boundary parameters.

This approach, however, has certain deficiency. Let us consider boundaries between grains A_1 and A_2 , and between B_1 and B_2 . One would like the distance between the boundaries A_1/A_2 and B_1/B_2 to be equal to the distance between A_2/A_1 and B_2/B_1 . Does it occur for χ as defined by eq.(2)? The answer is no; but the distance (2) can be modified to make it satisfy that condition. For example, χ can be defined as

$$\chi^2((g, \mathbf{n}_1), (g', \mathbf{n}'_1)) = \chi_{\bullet}^2(g, g') + (\chi_{\circ}^2(\mathbf{n}_1, \mathbf{n}'_1) + \chi_{\circ}^2(\mathbf{n}_2, \mathbf{n}'_2))/2, \quad (3)$$

with the normals are related through eq.(1), i.e., $\mathbf{n}_2 = -g^T \mathbf{n}_1$ and $\mathbf{n}'_2 = -g'^T \mathbf{n}'_1$. The question arises whether this change of the metric affects dV (and thus, the shape of the reference random distribution)? It does not; it can be checked by direct calculation that, disregarding the normalization coefficient, the volume element following from the metric (3) has the same form as the one corresponding to (2).

Symmetries

Due to crystal symmetries, different points of the space $O(3) \times S^2$ may represent geometrically identical interfaces. The presence of the symmetries leads to equivalence between such points. The GBD must take equal values at equivalent points. In relation to this, two problems arise: *i.*) what are the relations between equivalent points, and *ii.*) what is the shape of its asymmetric domain defined as a region in the parameter space in which each geometrically distinct boundary is represented only once, i.e., which contains exactly one representative of each equivalence class.

Let \mathbf{n} be a vector perpendicular to a crystallographic plane with Miller indices $(h k l) = \mathbf{h}^T$. Vector \mathbf{n}^{eq} perpendicular a plane \mathbf{h}^{eq} symmetrically equivalent to \mathbf{h} is related to \mathbf{n} via

$$\mathbf{n}^{eq} = c \mathbf{n}, \quad (4)$$

where $c \in O(3)$ is an orthogonal element of a space group operation.² Because only macroscopic parameters of the boundary are considered, the translations do not play a role here.

Let c_1 and c_2 be point symmetries of crystals 1 and 2, respectively. Thus, the orientation g_1 is equivalent to $c_1 g_1$. Analogously, g_2 is equivalent to $c_2 g_2$. Hence, the misorientations g and $c_1 g c_2^T$ are equivalent.

The question is, when the misorientations $c_1 g c_2^T$ and planes \mathbf{h}_1^{eq} and \mathbf{h}_2^{eq} equivalent to \mathbf{h}_1 and \mathbf{h}_2 , respectively, represent a boundary indistinguishable from the original one. From (4) the normals \mathbf{n}_1^{eq} and \mathbf{n}_2^{eq} to the plane should satisfy $\mathbf{n}_1^{eq} = c_1^x \mathbf{n}_1$ and $\mathbf{n}_2^{eq} = c_2^x \mathbf{n}_2$. We want to know which of the crystal symmetries are acceptable as c_1^x and c_2^x so the relations between the normals remain consistent. Vectors \mathbf{n}_1^{eq} and \mathbf{n}_2^{eq} determine an equivalent boundary if they are related through (1) with g replaced by one of the misorientations $c_1 g c_2^T$ symmetrically equivalent to g , i.e., if $\mathbf{n}_1^{eq} = -c_1 g c_2^T \mathbf{n}_2^{eq}$. This means that $c_1^x \mathbf{n}_1 = -c_1 g c_2^T c_2^x \mathbf{n}_2$ must occur. Taking into account (1) one has $c_1^x g \mathbf{n}_2 = c_1 g c_2^T c_2^x \mathbf{n}_2$,

²To show this let us consider non-coplanar vectors $\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3$ and \mathbf{x}_4 ; first 3 vectors determine the crystallographic plane \mathbf{h} , and the fourth one indicates the sense of the normal. One can write $\mathbf{n} = \mathbf{m} \text{sign}(\mathbf{m} \cdot (\mathbf{x}_4 - \mathbf{x}_1))$ where $\mathbf{m} = (\mathbf{x}_2 - \mathbf{x}_1) \times (\mathbf{x}_3 - \mathbf{x}_1) = \mathbf{x}_1 \times \mathbf{x}_2 + \mathbf{x}_2 \times \mathbf{x}_3 + \mathbf{x}_3 \times \mathbf{x}_1$. An element $(c | \mathbf{t})$ of the crystal space group (\mathbf{t} – translation) transforms \mathbf{x}_i ($i = 1, \dots, 4$) into $\mathbf{x}_i^{eq} = (c | \mathbf{t})\mathbf{x}_i = c\mathbf{x}_i + \mathbf{t}$. Vector \mathbf{n}^{eq} normal to the plane \mathbf{h}^{eq} is determined by $\mathbf{x}_1^{eq}, \mathbf{x}_2^{eq}, \mathbf{x}_3^{eq}, \mathbf{x}_4^{eq}$. There occurs $(\mathbf{x}_4^{eq} - \mathbf{x}_1^{eq}) = c(\mathbf{x}_4 - \mathbf{x}_1)$ and $\mathbf{m}^{eq} = \mathbf{x}_1^{eq} \times \mathbf{x}_2^{eq} + \mathbf{x}_2^{eq} \times \mathbf{x}_3^{eq} + \mathbf{x}_3^{eq} \times \mathbf{x}_1^{eq} = \det(c) c \mathbf{m}$, and hence $\mathbf{n}^{eq} = \mathbf{m}^{eq} \text{sign}(\mathbf{m}^{eq} \cdot (\mathbf{x}_4^{eq} - \mathbf{x}_1^{eq})) = (\det(c))^2 c \mathbf{m} \text{sign}(\mathbf{m} \cdot (\mathbf{x}_4 - \mathbf{x}_1)) = c \mathbf{n}$.

and because \mathbf{n}_2 is arbitrary, one gets $(c_1^T c_1^x)g = g(c_2^T c_2^x)$. For arbitrary g , this equation is satisfied in only two cases

$$c_1^x = c_1, \quad c_2^x = c_2 \quad \text{or} \quad c_1^x = ic_1, \quad c_2^x = ic_2,$$

where i denotes inversion. Because c_1^x and c_2^x must be elements of crystal point groups, the second case occurs only if point groups of both crystals contain inversion. This means that (g, \mathbf{n}_1) is equivalent to $(c_1 g c_2^T, c_1 \mathbf{n}_1)$, and to $(c_1 g c_2^T, -c_1 \mathbf{n}_1)$ if inversion on both sides is involved (and the latter case is already contained in the former). What matters is that the element c_1 preceding \mathbf{n}_1 cannot be decoupled from c_1 preceding g . In agreement with (1) one has $(c_1 g c_2^T)^T c_1 \mathbf{n}_1 = c_2 g^T \mathbf{n}_1 = -c_2 \mathbf{n}_2$.

Concluding, the grain boundary distribution f satisfies

$$f(g, \mathbf{n}_1) = f(c_1 g c_2^T, c_1 \mathbf{n}_1)$$

(for all elements c_1 and c_2 of the point groups). If both crystals are centrosymmetric one has

$$f(g, \mathbf{n}_1) = f(g, -\mathbf{n}_1). \quad (5)$$

The shapes of asymmetric domains for f follow directly from the domains for the misorientation distribution. The later are known for all combinations of crystallographic symmetries [5]. This allows one to construct asymmetric domains for GBD for all types of homo- and heterophase boundaries; for instance, one can take the product of the domain of misorientation distribution (determined by the point groups of the crystals) and a unit sphere (or a hemisphere if (5) occurs).³

Moreover, in the case of homophase boundaries the crystallites are indistinguishable and the GBD takes the same value at (g, \mathbf{n}_1) as at $(g^T, \mathbf{n}_2) = (g^T, -g^T \mathbf{n}_1)$; i.e., there occurs

$$f(g, \mathbf{n}_1) = f(g^T, -g^T \mathbf{n}_1)$$

and the asymmetric domain is additionally reduced in size by half.

It is noteworthy that if one assumes that a given scalar function, say grain boundary energy, depends only on the macroscopic parameters and takes equal values for geometrically identical boundary configurations, analogous symmetries will occur for that function.

It seems to be easier to use the above properties by applying the following notation: let a 4×4 matrices B , C_1 and C_2 be defined by

$$B = \begin{bmatrix} g & \mathbf{n}_1 \\ \mathbf{n}_2^T & 0 \end{bmatrix}, \quad C_1 = \begin{bmatrix} c_1 & 0 \\ 0 & 1 \end{bmatrix}, \quad C_2 = \begin{bmatrix} c_2 & 0 \\ 0 & 1 \end{bmatrix},$$

i.e., the first matrix (with $\mathbf{n}_2 = -g^T \mathbf{n}_1$) determines the boundary (g, \mathbf{n}_1) , and the other two correspond to the crystal symmetries. The matrix related to the boundary (g^T, \mathbf{n}_2) has the form B^T . The equivalency between (g, \mathbf{n}_1) and $(c_1 g c_2^T, c_1 \mathbf{n}_1)$ can be expressed as equivalency between the matrices B and $C_1 B C_2^T$. Thus, the equivalency relations for GBD are analogous to those for misorientation distribution. Let us also mention that $\det(B) = \det(g) = \pm 1$, and that the distance (3) can be expressed as

$$\chi^2((g, \mathbf{n}_1), (g', \mathbf{n}'_1)) = \|B - B'\|^2/2,$$

³Let us mention here that the way of determining the asymmetric domain (fundamental zone) for misorientations in Rodrigues parameters by taking the common to zones of both crystals (as suggested in [2], p.17) is incorrect.

where B' corresponds to (g', \mathbf{n}'_1) . Finally, it has to be taken into account that symmetries affect the finite metric properties of the space; in the symmetric case, the distance between two boundaries is given by the smallest of all values of χ for all representatives of the classes to which the boundaries belong.

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