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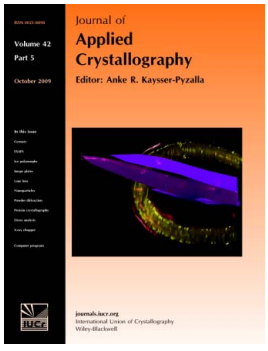

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Models of uniformity for grain boundary distributions

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Progress in experimental methods of serial sectioning and orientation determination opens new opportunities to study inter-crystalline boundaries in polycrystalline materials. In particular, macroscopic boundary parameters can now be measured automatically. With sufficiently large data sets, statistical analysis of interfaces between crystals is possible. The most basic and interesting issue is to find out the probability of occurrence of various boundaries in a given material. In order to define a boundary density function, a model of uniformity is needed. A number of such models can be conceived. It is proposed to use those derived from an assumed metric structure of the interface manifold. Some basic metrics on the manifold are explicitly given, and a number of notions and constructs needed for a strict definition of the boundary density function are considered. In particular, the crucial issue of the impact of symmetries is examined. The treatments of homo- and hetero-phase boundaries differ in some respects, and approaches applicable to each of these two cases are described. In order to make the abstract matter of the paper more accessible, a concrete boundary parameterization is used and some examples are given.

1. Introduction

The analysis of inter-crystalline interfaces constitutes an important area in the investigation of polycrystalline materials. Studies on sharp grain boundaries are frequently focused on special boundaries, for example, specific tilt or twist boundaries, low-angle boundaries, coincidence lattice (CSL) boundaries, or particular orientation relationships for hetero-phase interfaces. However, there are cases in which statistical methods would be more appropriate. Such methods are essential to quantify uncertainties and test hypotheses. Some years ago, statistical analyses were practically limited to misorientation angle distributions, misorientation distributions or frequencies of CSL boundaries. Recently, however, grain boundaries have been studied in a more complete way, for all possible grain misorientations and boundary inclinations (see *e.g.* Saylor *et al.*, 2003; Rohrer *et al.*, 2006; Randle, 2008, and references therein). Concurrently, progress in automatic serial sectioning and orientation determination methods allows for analysis of large sets of boundaries (Spowart, 2006; Rowenhorst *et al.*, 2006). Particularly interesting is the development of the scanning electron microscopy technique combining a focused ion beam unit for serial sectioning with electron backscatter diffraction for orientation determination (Groeber *et al.*, 2006; Uchic *et al.*, 2006; Konrad *et al.*, 2006). With further advancement of these experimental procedures, one may expect many more statistical studies of general boundaries with all macroscopic boundary parameters involved.

A proper formalism constituting a foundation for statistical analysis of experimental misorientation and inclination data requires geometric concepts beyond the classical Euclidean geometry, and it also turns out to be more complicated than in the related field of crystallographic textures. Below, we consider essential aspects of such a formalism. For clarity, it is necessary to start with familiar notions of interface research, and link them to geometric structures added to the boundary space. With these structures, tools intrinsic to the space can be used to study interface data.

The notion of a 'boundary distribution' is central to the statistical analysis of boundaries. The frequency of given boundary types represented by a density function over the macroscopic boundary parameters plays a role similar to that of the 'orientation distribution function' in quantitative texture analysis (*e.g.* Bunge, 1982). In order to define a boundary distribution, a measure in the parameter space must be specified. The measure in the boundary space, however, is not unique, and thus, the boundary distribution depends on its choice.[1] This is different from the situation in textures, where the practical (invariant) 'volume element' is distinctive. The expression for a volume element can be derived from the metric structure determining the distance between points representing the degree of similarity of boundaries. Metrics for the boundary space and the resulting measures are the

[1] In other words, different models of uniformity lead to different statistical results. For an elementary illustration of this issue search the Internet for 'Bertrand's paradox' of geometric probability or see Kendall & Moran (1963).

main subject of this paper. It needs to be mentioned that there is a difference in the geometric description of the hetero- and homo-phase boundaries, with the treatment of the latter being more complex. Both cases are considered in parallel.

We begin with considering a possible topology of the 'boundary space' and some parameterizations. Then, metrics are introduced, and the impact of symmetries is discussed. Finally, we proceed to measures and boundary distributions. Although this paper contains numerous mathematical notions, it is meant to be comprehensible for a materials scientist. In order to make it less abstract, a number of concrete examples are given. The exposition of the mathematical material is somewhat informal, and the text is intended to be read in an informal spirit. Some elementary information available in a concise form in less accessible publications (Morawiec, 1998; Morawiec, 2004) is repeated here for completeness.

2. Topology and parameterizations

Let us start with basic characteristics of the 'space' of boundaries. Only macroscopic boundary parameters identifying misorientations and inclinations are considered here (see *e.g.* Sutton & Balluffi, 1995). With right-handed Cartesian reference systems attached to crystallites, relative orientations of the crystallites correspond to rotations relating the systems. For simplicity, we assume that inversion is a symmetry operation for both crystals, so one does not need to deal with chirality. For two neighboring crystallites, the misorientation M between the first grain and the second one is characterized by a proper rotation. Proper rotations are faithfully represented by special orthogonal matrices with composition of rotations corresponding to matrix multiplication. Thus, misorientations can be identified by elements of $SO(3)$ – the set of these matrices; in what follows, we consider M to be in $SO(3)$.

Besides the misorientation, one needs to specify local inclination of the boundary. For that, one can use a unit vector $\mathbf{m}_1$ normal to the planar boundary (segment), directed (by convention) towards the second grain, with coordinates given in the coordinate system of the first crystallite (Fig. 1). Endpoints of possible normals – or unit vectors in all directions – constitute the sphere S^2.

Already at this basic level there are obvious differences between homo- and hetero-phase interfaces. Although Cartesian crystal reference systems are chosen according to conventions, in the homo-phase case, the crystal reference systems are attached to each crystallite in the same way. Therefore, there exists a natural and unique choice for the reference alignment or reference misorientation represented by the identity matrix $I_3 := \mathrm{diag}\,(1, 1, 1)$. On the other hand, if two crystallites are of different types, there is no natural reference alignment, the reference misorientation is not unique and the misorientation represented by I_3 depends on convention (Fig. 2).

Moreover, within the macroscopic model, two neighboring crystallites of the same type with the reference misorientation $M = I_3$ constitute one crystallite, *i.e.* there is no boundary between them. Thus, this misorientation is not valid in the description of homo-phase boundaries. One may think about the space of misorientations as $SO(3)$ with a 'hole' at I_3, or – if inclinations are taken into account – with a two-dimensional 'cut' through the boundary space. These points can be added to 'complete' the space – and this is necessary in a sense – but, as indicated by Cahn & Taylor (2006), it seems sensible to identify all inclinations corresponding to I_3 because there is no physical difference between them.

Let the symbol $\mathcal{M}_B$ denote the boundary space. Based on the above discussion, a hetero-phase boundary is determined by the pair $(M, \mathbf{m}_1)$. Formally, such pairs constitute a Cartesian product of $SO(3)$ and S^2: *i.e.* the space of hetero-phase boundaries equals $SO(3) \times S^2$. A homo-phase boundary is also characterized by the pair $(M, \mathbf{m}_1)$, except the case of $M = I_3$. If this point was removed from the misorientation space, the boundary space would be $(SO(3) \setminus \{I_3\}) \times S^2$; instead, in practice, theoretical considerations can be based on $SO(3) \times S^2$, and – when physical aspects are considered – the points corresponding to $M = I_3$ may be simply ignored. Finally, if $M = I_3$ is allowed as a misorientation with identified

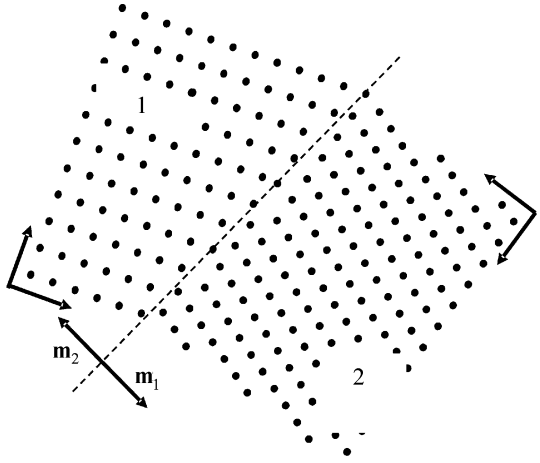

Figure 1
Schematic of an interface between two-dimensional crystallites.

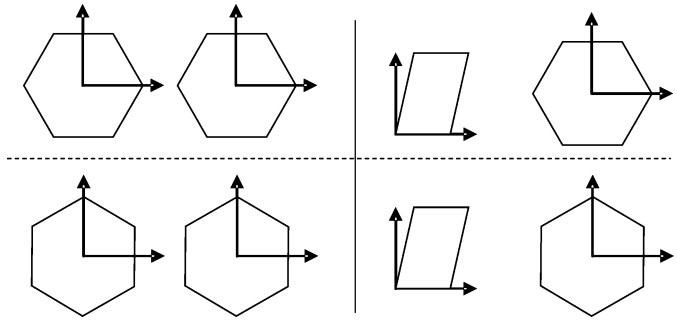

Figure 2
For identical objects like the hexagons on the left, the reference misorientation is the same irrespective of whether the horizontal or vertical axis is perpendicular to an edge of the hexagon; in both configurations all edges are parallel. In the right figure, in which the two objects differ, an edge of the parallelogram is parallel to an edge of the hexagon only in the upper configuration.

inclinations, the 'meridian' $\{I_3\} \times S^2$ of $SO(3) \times S^2$ collapses into a single point leading to a space essentially different from that of $SO(3) \times S^2$.

Digression on boundaries in two dimensions. The boundary geometry in three dimensions is difficult to visualize. It is much easier to deal with boundaries in two-dimensional microstructures. As in three-dimensions, a boundary between two-dimensional 'crystallites' is specified by a misorientation and an inclination (Fig. 1). For simplicity, we allow only proper rotations. Boundary misorientations can be represented by elements of $SO(2)$. The group $SO(2)$ is isomorphic to the group of complex numbers of unit length, and – topologically – $SO(2)$ is the unit circle S^1. On the other hand, all possible normals (unit vectors in all directions) also constitute S^1. The Cartesian product of these circles is a torus $S^1 \times S^1$ (Fig. 3). With inclinations at the reference misorientation collapsed into a single point, the space looks like the one shown schematically in Fig. 4. These are analogies in portrayal of two- and three-dimensional boundaries. However, when symmetry is taken into account (as in §4 below), there is a disparity between the two- and three-dimensional symmetric cases: I_3 is the only element of $SO(3)$ commuting with all other elements, whereas the group $SO(2)$ is commutative. Since in practice only the three-dimensional description is applicable to real polycrystalline materials, we will not pursue the two-dimensional case any further.

Going back to the specification of boundaries in three dimensions, the following question arises: with $(M, \mathbf{m}_1)$ representing the boundary between the first grain and the second grain, what is the representation of the boundary between the second grain and the first one? The misorientation changes from M to M^{T} (where superscript T denotes the transpose of the matrix). The coordinates of the normal to this boundary in the Cartesian coordinate system of the second grain are given by $M^{\mathrm{T}}\mathbf{m}_1$. Thus, the grain boundary between the second grain and the first grain has the form $(M^{\mathrm{T}}, \mathbf{m}_2)$ with the normals $\mathbf{m}_1$ and $\mathbf{m}_2$ related by

$$\mathbf{m}_2 = -M^{\mathrm{T}}\mathbf{m}_1. \tag{1}$$

It is convenient to use a notation 'symmetric' in $\mathbf{m}_1$ and $\mathbf{m}_2$ (Morawiec, 1998). The boundary corresponding to the pair $(M, \mathbf{m}_1)$ is represented by a 4×4 interface matrix,

$$\mathbf{B} = \begin{bmatrix} 0 & \mathbf{m}_2^{\mathrm{T}} \\ \mathbf{m}_1 & M \end{bmatrix}, \tag{2}$$

and $\mathbf{B}^{\mathrm{T}}$ represents the boundary $(M^{\mathrm{T}}, \mathbf{m}_2)$. The matrix $\mathbf{B}$ can be also expressed in the form

$$\mathbf{B} = \begin{bmatrix} 0 & -\mathbf{m}_1^{\mathrm{T}} \\ \mathbf{m}_1 & I_3 \end{bmatrix}\begin{bmatrix} 1 & 0 \\ 0 & M \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & M \end{bmatrix}\begin{bmatrix} 0 & \mathbf{m}_2^{\mathrm{T}} \\ -\mathbf{m}_2 & I_3 \end{bmatrix}, \tag{3}$$

useful for considering the effect of rotations of the reference systems. If a proper rotation represented by a 3×3 orthogonal matrix R_1 is applied to the first crystallite in such a way that features of the second crystallite remain unaffected, the new boundary will correspond to the pair $(R_1M, R_1\mathbf{m}_1)$ and to the interface matrix

$$\begin{bmatrix} 1 & 0 \\ 0 & R_1M \end{bmatrix}\begin{bmatrix} 0 & \mathbf{m}_2^{\mathrm{T}} \\ -\mathbf{m}_2 & I_3 \end{bmatrix} = \mathbf{R}_1\mathbf{B}, \text{ where } \mathbf{R}_1 = \begin{bmatrix} 1 & 0 \\ 0 & R_1 \end{bmatrix}. \tag{4}$$

A similar relation applies to rotations of the second crystallite; rotating it by R_2 transforms the boundary $\mathbf{R}_1\mathbf{B}$ into $\mathbf{R}_1\mathbf{B}\mathbf{R}_2^{\mathrm{T}}$, where $\mathbf{R}_2$ is defined analogously to $\mathbf{R}_1$.

It must be stressed that the above notation – the pairs $(M, \mathbf{m}_1)$ and interface matrices – is applicable only to points of $SO(3) \times S^2$. If inclinations linked to $M = I_3$ are identified, the notation makes no sense at this special point. Below, we use the symbol $\mathbf{B}$ to indicate a boundary as an element of arbitrary $\mathcal{M}_{\mathcal{B}}$; this symbol denotes an interface matrix only if the context implies that $\mathcal{M}_{\mathcal{B}}$ equals $SO(3) \times S^2$.

Example boundary. For homo-phase boundaries between crystals with a cubic (primitive, face centered cubic or body centered cubic) lattice, the $\Sigma 7$ CSL misorientation can be represented by

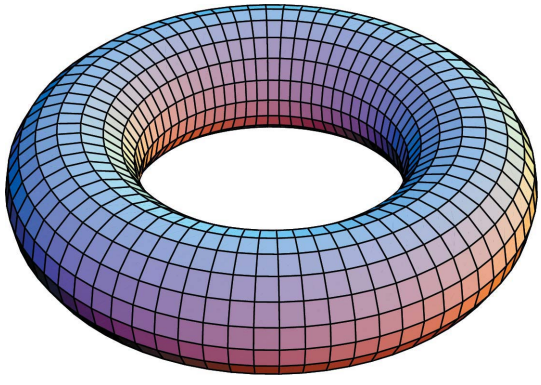

Figure 3
The boundary space $S^1 \times S^1$ for two-dimensional interfaces. Misorientations are assumed to change along the zonal (horizontal) circles, whereas inclinations change along meridional (vertical) circles.

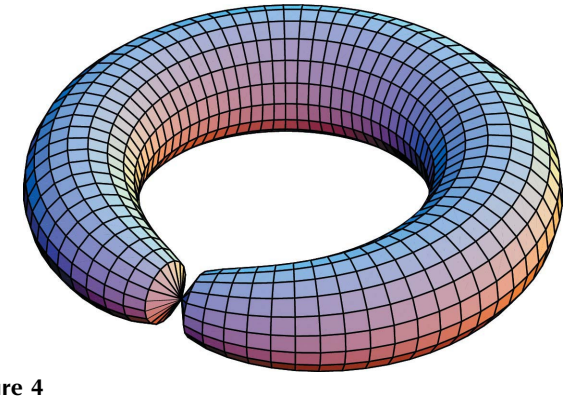

Figure 4
The boundary space with $\{I_2\} \times S^1$ collapsed to a point.

$$M = \frac{1}{7}\begin{bmatrix} -3 & -2 & 6 \\ -6 & 3 & -2 \\ -2 & -6 & -3 \end{bmatrix}. \quad (5)$$

With $\mathbf{m}_1 = [1\ 2\ 3]^{\mathrm{T}}/14^{1/2}$ and $\mathbf{m}_2 = -M^{\mathrm{T}}\mathbf{m}_1 = [3\ 2\ 1]^{\mathrm{T}}/14^{1/2}$, the rotation axis $[\bar{1}2\bar{1}]$ corresponding to M is perpendicular to $\mathbf{m}_1$ and $\mathbf{m}_2$. Thus, the interface matrix

$$\mathbf{B}_{(\Sigma 7,\,[1\,2\,3])} = \frac{1}{14}\begin{bmatrix} 0 & 3(14^{1/2}) & 2(14^{1/2}) & 14^{1/2} \\ 14^{1/2} & -6 & -4 & 12 \\ 2(14^{1/2}) & -12 & 6 & -4 \\ 3(14^{1/2}) & -4 & -12 & -6 \end{bmatrix} \quad (6)$$

represents a $\Sigma 7$ tilt boundary with the (123) face.

Symmetric representation. There is no essential difference between a pair $(M, \mathbf{m}_1)$ and the corresponding interface matrix. On the other hand, there are other ways of identifying boundaries. To give an example confirming the diversity of possible approaches, one can use an even more symmetric but computationally less convenient way of referring to a boundary: Let N be an orthogonal matrix, which corresponds to the rotation about the same axis as M but where the rotation angle is half of that for M. (An additional convention for selection of the axis is assumed if M is a rotation by π and two axes with opposite senses are applicable.) The domain for N is not the complete $SO(3)$ but a part of it with the trace of matrices not smaller than 1 (*i.e.* with rotation angles not exceeding $\pi/2$). With such N, one has

$$NN = M. \quad (7)$$

Let $\mathbf{n}$ be defined by

$$\mathbf{n} := N^{\mathrm{T}}\mathbf{m}_1 = -N\mathbf{m}_2. \quad (8)$$

The pair $(N, \mathbf{n})$ determines a boundary. With $(N, \mathbf{n})$ corresponding to the boundary between the first crystallite and the second one, $(N^{\mathrm{T}}, -\mathbf{n})$ corresponds to the boundary between the second and the first. The interface matrix $\mathbf{B}$ is related to $(N, \mathbf{n})$ *via*

$$\mathbf{B} = \begin{bmatrix} 1 & 0 \\ 0 & N \end{bmatrix}\mathbf{B}_0\begin{bmatrix} 1 & 0 \\ 0 & N \end{bmatrix}, \text{ where } \mathbf{B}_0 = \begin{bmatrix} 0 & -\mathbf{n}^{\mathrm{T}} \\ \mathbf{n} & I_3 \end{bmatrix}. \quad (9)$$

This has a simple interpretation based on equation (4): if the crystallites one and two were rotated by N^{T} and N, respectively, the interface matrix of the boundary would be $\mathbf{B}_0$.

We will not use the symmetric representation but it needs to be kept in mind that some of the schemes introduced below for the pairs $(M, \mathbf{m}_1)$ can also be applied to $(N, \mathbf{n})$ or other constructs of similar type.

In mathematics, a topological space that can be 'charted' is referred to as a topological manifold. Both $SO(3)$ and S^2 can be seen as topological manifolds, and both are ordinarily taken with their additional structures but – at this point – we are only in search of parameterizations. Spherical projections used in crystallography are examples of charts on S^2. As for $SO(3)$, numerous parameterizations are listed by Morawiec (2004). In practice, it is important to know that parameterizations of S^2 and $SO(3)$ have singular points. However, these singularities are inessential; one can use other maps properly covering the area containing points singular on the first map, with each pair of maps 'coherent' in the area of their overlap.

As an example, we will use one of the simplest charts of a part of S^2 – the so-called orthographic projection of the upper hemisphere (with additional maps needed to cover the rest of the sphere). The parameters are the first k^1 and the second k^2 coordinates of a unit vector $\mathbf{k}$ indicating a given point of S^2. The third coordinate k^3 depends on k^1 and k^2 *via* $k^3 = (1 - k^a k^a)^{1/2}$ $(a = 1, 2)$. Here and below, summation over indices that appear twice in a single term is assumed.

The orthographic projection can also be used for parameterizing rotations because $SO(3)$ is topologically identical to the three-dimensional sphere (sitting in four-dimensional space) with antipodal points identified. The complete sphere can be described by a unit four-component vector $\mathbf{q}$. Three components of $\mathbf{q}$, say q^1, q^2, q^3, can be used as parameters of the upper hemisphere and of $SO(3)$. The fourth component, q^0, depends on q^i *via* $q^0 = (1 - q^i q^i)^{1/2}$ $(i = 1, 2, 3)$.[2]

Parameterizations of S^2 and $SO(3)$ are a basis for charting $SO(3) \times S^2$. A boundary is identified by five numbers, say $(x^1, \ldots, x^5)$; in the above case of orthographic projections, one has $(x^1, x^2, x^3, x^4, x^5) = (q^1, q^2, q^3, k^1, k^2)$. With a sufficient number of maps, the five parameters $(x^1, \ldots, x^5)$ can be given for an arbitrary pair of a special orthogonal matrix and a unit vector. This applies to $(M, \mathbf{m}_1)$, $(N, \mathbf{n})$ or any other such pair.

3. Distance functions and Riemannian metrics

Manifolds may have additional structures. One of them is a distance function determining distances between points of a manifold. The distance function is expected to satisfy the conditions of non-negativity, symmetry, identity of indiscernibles and triangle inequality. With the interface manifold provided with such metric structure, boundaries can be compared by checking distances between them; similar boundaries should be close, *i.e.* the distance between them should be small.

A topological manifold is called a differential manifold if it has a globally defined differential structure, which means that, in areas where the charts overlap, the coordinates defined by each chart are differentiable with respect to the coordinates of every other chart. Distances on differential manifolds are determined using a metric tensor (usually denoted by g). For a

[2] The numbers q^i can be seen as components of a unit quaternion. Such quaternions constitute a (two-to-one) representation of rotations but we do not really need this here. The relationships between an orthogonal matrix, say R, and the parameters q^1, q^2, q^3 are given by

$$R_{ij} = [(q^0)^2 - q^k q^k]\delta_{ij} + 2q^i q^j - 2\varepsilon_{ijk} q^0 q^k$$

and

$$q^k = -\varepsilon_{ijk} R_{ij}/[2(1 + R_{ll})^{1/2}],$$

where R_{ij} are components of R, δ is the Kronecker symbol and ε is the permutation symbol (see *e.g.* Morawiec, 2004).

differential manifold possessing a metric tensor (Riemannian manifold), the length of a segment of a curve parameterized by u, from u_1 to u_2, is given by $\int_{u_1}^{u_2}[g_{ij}(\mathrm{d}x^i/\mathrm{d}u)(\mathrm{d}x^j/\mathrm{d}u)]^{1/2}\,\mathrm{d}u$, where $x^i = x^i(u)$ $(i = 1, \ldots, 5)$ are parametric equations describing the curve on a local chart. For a 'complete' Riemannian manifold, the distance between two points is defined as the length of the shortest curve between these points. Furthermore, the opposite occurs: a suitable distance function, say s, determines a metric tensor; however, in general, the Riemannian metric originating from this metric tensor may be different from s. On a Riemannian manifold, there is a unique canonical differential structure (Levi-Civita connection) compatible with the metric of the manifold.

The issue of assigning a metric to the interface space is somewhat 'delicate'. Differently than in textures, there is no distinct metric. Moreover, conditions are different depending on whether hetero- or homo-phase boundaries are considered. In the latter case – as the discussion of Cahn & Taylor (2006) implies – the expectations may be to have a metric continuous through the special point with collapsed inclinations at the misorientation of I_3. It was concluded by Cahn & Taylor (2006) that 'the 'no boundary' singularity in Grain Boundaries might be a clue to appropriate metrics'. A number of metrics are considered below but we focus on practical cases directly linked to concepts familiar from analysis of (mis)orientations. More 'exotic' metrics, some of which are mentioned by Cahn & Taylor (2006), are ignored here.

There exist distinct (up to a constant factor) metrics on $SO(3)$ and S^2; these are the metrics invariant under rotations in the Euclidean space, and proportional to misorientation angles and great-circle distances on the sphere, respectively. The metrics on $SO(3)$ and S^2 can be used to construct a metric on the interface manifold. This approach is in a way more intuitive than working with other admissible metrics. Further on, we will use the markers $\bullet$ and $\circ$ for distances and metric tensors on $SO(3)$ and S^2, respectively.

Generally, to obtain a metric tensor from a finite distance function s in a given parameterization, one needs to calculate the squared infinitesimal distance $\mathrm{d}s^2$ between a given point (x^i) and the point $(x^i + \mathrm{d}x^i)$, which differs by infinitesimal changes of the parameters; the metric tensor $g = g(x^i)$ contains coefficients of quadratic terms in the infinitesimal changes

$$\mathrm{d}s^2 = g_{ij}\,\mathrm{d}x^i\,\mathrm{d}x^j, \tag{10}$$

and higher-order terms are neglected. Let us illustrate this on the unit sphere. The distinct distance $s_\circ$ between points $\mathbf{k}$ and $\mathbf{k}'$ on S^2 is a multiple of the angle between $\mathbf{k}$ and $\mathbf{k}'$:

$$s_\circ(\mathbf{k}, \mathbf{k}') = c_\circ \arccos(\mathbf{k}\cdot\mathbf{k}'). \tag{11}$$

With this, the corresponding metric tensor in the coordinates based on the orthographic projection can be easily calculated. For small $s_\circ^2(\mathbf{k}, \mathbf{k}')$, there occurs

$$s_\circ^2(\mathbf{k}, \mathbf{k}') \simeq 2c_\circ^2\{1 - \cos[s_\circ(\mathbf{k}, \mathbf{k}')/c_\circ]\} = 2c_\circ^2(1 - \mathbf{k}\cdot\mathbf{k}'). \tag{12}$$

This approximation gives the same components of the metric tensor as $s_\circ^2(\mathbf{k}, \mathbf{k}')$ (because appropriate expressions differ by terms higher than quadratic in the infinitesimal changes), while it allows for avoiding trigonometric functions. For the vectors $\mathbf{k} = [k^1, k^2, (1 - k^a k^a)^{1/2}]^{\mathrm{T}}$ and $\mathbf{k}' = \{k^1 + \mathrm{d}k^1, k^2 + \mathrm{d}k^2, [1 - (k^1 + \mathrm{d}k^1)^2 - (k^2 + \mathrm{d}k^2)^2]^{1/2}\}^{\mathrm{T}}$ differing in k^a by $\mathrm{d}k^a$, one has

$$\begin{aligned} 2(1 - \mathbf{k}\cdot\mathbf{k}') = {} & \big[1 + (k^1/k^3)^2\big](\mathrm{d}k^1)^2 + \big[1 + (k^2/k^3)^2\big](\mathrm{d}k^2)^2 \\ & + 2\big[k^1k^2/(k^3)^2\big]\mathrm{d}k^1\mathrm{d}k^2 \end{aligned} \tag{13}$$

with accuracy to quadratic terms in $\mathrm{d}k^a$. Thus, for the coordinates k^1 and k^2, the metric tensor is given by

$$g^\circ_{ab} = c_\circ^2\big[\delta_{ab} + k^a k^b/(k^3)^2\big], \tag{14}$$

where $a, b = 1, 2$.

The distinct distance on $SO(3)$ is a multiple of the smallest angle of the rotation necessary to transform R into R'. That rotation is $R'R^{\mathrm{T}}$, and since the cosine of the rotation angle is directly related to the trace of the rotation matrix *via* [tr(matrix) − 1]/2 (*e.g.* Morawiec, 2004), one has

$$s_\bullet(R, R') = c_\bullet \arccos\big\{[\mathrm{tr}(R'R^{\mathrm{T}}) - 1]/2\big\}. \tag{15}$$

On the basis of this function, one can calculate the squared infinitesimal distance $\mathrm{d}s_\bullet^2$ and the metric tensor. For the coordinates q^i, the latter is given by an expression similar to equation (14):

$$g^\bullet_{ij} = 4c_\bullet^2\big[\delta_{ij} + q^i q^j/(q^0)^2\big], \tag{16}$$

where $i, j = 1, 2, 3$ (Morawiec, 2004).

The most natural way of constructing a metric on the product manifold $SO(3) \times S^2$ is to use metrics of $SO(3)$ and S^2. With R, R' in $SO(3)$ and $\mathbf{k}, \mathbf{k}'$ in S^2, the product metric on $SO(3) \times S^2$ is defined by $s[(R, \mathbf{k}), (R', \mathbf{k}')] := [s_\bullet^2(R, R') + s_\circ^2(\mathbf{k}, \mathbf{k}')]^{1/2}$. In the parameterization described above, the squared infinitesimal distance is $\mathrm{d}s^2 = g^\bullet_{ij}\mathrm{d}q^i\mathrm{d}q^j + g^\circ_{ab}\mathrm{d}k^a\mathrm{d}k^b$, and the metric tensor has the simple form

$$g = \begin{bmatrix} g^\bullet & 0 \\ 0 & g^\circ \end{bmatrix}, \tag{17}$$

where g° and $g^\bullet$ are given by equations (14) and (16), respectively.

If this metric is applied to $(R, \mathbf{k}) = (M, \mathbf{m}_1)$, we denote it by the symbol s_{A}, *i.e.* $s_{\mathrm{A}}^2[(M, \mathbf{m}_1), (M', \mathbf{m}_1')] := s_\bullet^2(M, M') + s_\circ^2(\mathbf{m}_1, \mathbf{m}_1')$. For brevity, we replace the pairs $(M, \mathbf{m}_1)$ and $(M', \mathbf{m}_1')$ by the corresponding interface matrices $\mathbf{B}$ and $\mathbf{B}'$ as arguments of the distances, *i.e.* $s_{\mathrm{A}}[(M, \mathbf{m}_1), (M', \mathbf{m}_1')] \equiv s_{\mathrm{A}}(\mathbf{B}, \mathbf{B}')$. It is easy to verify that

$$s_{\mathrm{A}}(\mathbf{R}_1\mathbf{B}\mathbf{R}_2^{\mathrm{T}}, \mathbf{R}_1\mathbf{B}'\mathbf{R}_2^{\mathrm{T}}) = s_{\mathrm{A}}(\mathbf{B}, \mathbf{B}'), \tag{18}$$

i.e. 'synchronized' rotations of crystallites and boundary planes are isometries on $SO(3) \times S^2$ with the metric s_{A}. In the case of hetero-phase boundaries, since the reference misorientation is not unique, the relationship [equation (18)] needs to be satisfied because otherwise distances and distance-based quantities would depend on the convention defining that misorientation; with the above requirement fulfilled, s_{A} has a convention-independent meaning.

As was already explained, in the case of homo-phase boundaries, boundary inclinations have no physical sense for the misorientation represented by I_3. If this misorientation is allowed with the additional requirement that all inclinations coupled to it are identified as one point, the expressions for metrics used above are not applicable at this particular point. Some distance functions behaving properly at that point were proposed by Cahn & Taylor (2006). This case is actually well known in differential geometry as the so-called warped geometry. A product of two Riemannian manifolds has such geometry if its (warped product) metric can be written in the form

$$\mathrm{d}s^2 = g^{\bullet}_{ij}\,\mathrm{d}q^i\mathrm{d}q^j + f(q^i)\,g^{\circ}_{ab}\,\mathrm{d}k^a\mathrm{d}k^b, \tag{19}$$

where $f > 0$ is a smooth function of coordinates q^i of the first manifold, and it is independent of the coordinates k^a of the second manifold. Thus, in the considered case, the geometry 'almost' decomposes into a Cartesian product of geometries of $SO(3)$ and S^2 but that of S^2 is scaled by a function of the coordinates on $SO(3)$. What matters here is that, instead of a positive scaling function, we take f vanishing at I_3. This would violate the condition of 'identity of indiscernibles' on $SO(3) \times S^2$ but a proper metric arises with S^2 at $M = I_3$ collapsed to a single point.

Example warped metric tensor. With $\mu > 0$ and $\underline{s}_{\bullet} \equiv s_{\bullet}/c_{\bullet}$, the distance function

$$s^2[(R, \mathbf{k}), (R', \mathbf{k}')] = s^2_{\bullet}(R, R') + [\underline{s}_{\bullet}(R, I_3)\,\underline{s}_{\bullet}(R', I_3)]^{\mu}\, s^2_{\circ}(\mathbf{k}, \mathbf{k}') \tag{20}$$

leads to the metric tensor

$$g = \begin{pmatrix} g^{\bullet} & 0 \\ 0 & f g^{\circ} \end{pmatrix}, \tag{21}$$

where g° and $g^{\bullet}$ are given by equations (14) and (16), respectively, and $f = f(q^i) = \arccos^{2\mu}(1 - 2\,q^i q^i)$. When the above is applied to $(M, \mathbf{m}_1)$, we will refer to the distance [equation (20)] as s_{C}. Rotations are not isometries of s_{C}.

With such warped geometry, there is a particular point on the boundary manifold differing topologically from the remaining points. Moreover, this geometry has an essential shortcoming of the arbitrariness of the scaling function. Besides the condition $f(I_3) = 0$, one may expect that f depends only on the $SO(3)$-based distance from I_3, and the function is smooth, monotonically increasing and concave. There are no other explicit restrictions on the form of f.

4. Equivalent boundary parameters

Owing to crystal symmetries, different points of the interface manifold $\mathcal{M}_{\mathcal{B}}$ correspond to geometrically identical interfaces. Consequently, the boundary parameters are not unique, *i.e.* a number of different sets of parameters stand for the same geometrical arrangement at the boundary. With this, a question arises about the relations between equivalent parameters.

Let C_1 and C_2 be special orthogonal matrices representing point symmetry operations of crystals 1 and 2, respectively. The misorientations M and $C_1 M C_2^{\mathrm{T}}$ are equivalent (Pospiech *et al.*, 1986). It turns out that similar relations apply to boundaries in the representation by $\mathbf{B}$ matrices (Morawiec, 1998). From the expressions for the result of rotating the reference systems [equation (4)], it follows that, with 4×4 matrices $\mathbf{C}_1$ and $\mathbf{C}_2$ defined by

$$\mathbf{C}_1 = \begin{bmatrix} 1 & 0 \\ 0 & C_1 \end{bmatrix} \quad \text{and} \quad \mathbf{C}_2 = \begin{bmatrix} 1 & 0 \\ 0 & C_2 \end{bmatrix}, \tag{22}$$

the matrices $\mathbf{B}$ and $\mathbf{C}_1\mathbf{B}\mathbf{C}_2^{\mathrm{T}}$ represent the same boundary. Consistently, the pair $(M, \mathbf{m}_1)$ is equivalent to $(C_1 M C_2^{\mathrm{T}}, C_1\mathbf{m}_1)$.

Example symmetry operation. With octahedral $m3m$ crystal symmetry, the 120° rotation about the [111] direction and the 180° rotation about $[10\bar{1}]$ are symmetry operations. They correspond to orthogonal matrices

$$C_1 = \begin{bmatrix} 0 & 1 & 0 \\ 0 & 0 & 1 \\ 1 & 0 & 0 \end{bmatrix} \quad \text{and} \quad C_2 = -\begin{bmatrix} 0 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{bmatrix}, \tag{23}$$

respectively. Because of these symmetries, the representation

$$\mathbf{C}_1\mathbf{B}_{(\Sigma 7,\,[1\,2\,3])}\mathbf{C}_2^{\mathrm{T}} = (1/14) \times \begin{bmatrix} 0 & -14^{1/2} & -2(14^{1/2}) & -3(14^{1/2}) \\ -2(14^{1/2}) & 4 & -6 & 12 \\ -3(14^{1/2}) & 6 & 12 & 4 \\ -14^{1/2} & -12 & 4 & 6 \end{bmatrix} \tag{24}$$

is equivalent to $\mathbf{B}_{(\Sigma 7,\,[1\,2\,3])}$.

We assumed at the outset that inversion is a symmetry operation of the crystals involved. Application of the inversion to two such crystals gives the equivalence of $(M, \mathbf{m}_1)$ and $(M, -\mathbf{m}_1)$. In terms of the interface matrices, the above means that $\mathbf{B}$ corresponding to $(M, \mathbf{m}_1)$ is equivalent to

$$\mathbf{B}^{-} = \begin{bmatrix} 0 & -\mathbf{m}_2^{\mathrm{T}} \\ -\mathbf{m}_1 & M \end{bmatrix}, \tag{25}$$

i.e. in the assumed case, $\mathbf{B}$ and $\mathbf{B}^{-}$ represent the same boundary.

Finally, if there is no discrimination of individual crystallites, a homo-phase boundary between grains 1 and 2 may be considered to be equivalent to that between 2 and 1. This interchange of the order of grains gives the equivalence between $\mathbf{B}$ and $\mathbf{B}^{\mathrm{T}}$. The potential presence of this equivalence is another important feature differing for homo- and hetero-phase interfaces.

Symmetries applicable to a boundary constitute a finite group (in the algebraic sense); let it be denoted by $\mathcal{G}$. It is a product of proper subgroups of point groups of the neighboring crystals, a two-element group involving the operation given in equation (25), and – if relevant – a two-element group

of the interchange of grains. $\mathcal{G}$ acts on $\mathcal{M}_\mathcal{B}$. The set of points of $\mathcal{M}_\mathcal{B}$ to which a given point can be transformed by the symmetry operations is an equivalence class. Summarizing previous paragraphs, the equivalence class $[\mathbf{B}]$ of $\mathbf{B}$ in $SO(3) \times S^2$ consists of

$$\mathbf{C}_1\mathbf{B}\mathbf{C}_2^{\mathrm{T}}, \quad \mathbf{C}_1\mathbf{B}^{-}\mathbf{C}_2^{\mathrm{T}}, \quad \mathbf{C}_1\mathbf{B}^{\mathrm{T}}\mathbf{C}_2^{\mathrm{T}} \quad \text{and} \quad \mathbf{C}_1\mathbf{B}^{-\mathrm{T}}\mathbf{C}_2^{\mathrm{T}}, \tag{26}$$

with the last two terms included only if the interchange of the order of grains is applicable, and with $\mathbf{C}_1$ and $\mathbf{C}_2$ covering all proper elements of point groups. The quotient set $\mathcal{M}_\mathcal{B}/\mathcal{G}$ is the set of all equivalence classes in $\mathcal{M}_\mathcal{B}$ by the equivalence relation $\mathcal{G}$. Thus, because of the presence of symmetries, boundaries are determined not on $\mathcal{M}_\mathcal{B}$ but on $\mathcal{M}_\mathcal{B}/\mathcal{G}$.

The number of elements in equivalence classes is not the same. Let the symbol $\mathcal{S}_i$ $(i = 1, \ldots, |\mathcal{G}|)$ denote the ith element of $\mathcal{G}$. For some $\mathcal{S}_i$, some points may be invariant, *i.e.* there occurs

$$\mathcal{S}_i(\mathbf{B}) = \mathbf{B}. \tag{27}$$

Symmetry operations satisfying the above relation for a given $\mathbf{B}$ constitute an isotropy group $\mathcal{G}_\mathbf{B}$ of $\mathbf{B}$. As in texture analysis, the order of this group $|\mathcal{G}_\mathbf{B}|$ will be referred to as the multiplicity of $\mathbf{B}$. The number of elements in the class $[\mathbf{B}]$ is equal to the ratio $|\mathcal{G}|/|\mathcal{G}_\mathbf{B}|$.

Multiplicity of $\boldsymbol{B}_{(\Sigma 7,\,[1\,2\,3])}$. With C_2 given by equation (23), one has

$$\mathbf{C}_2\big(\mathbf{B}_{(\Sigma 7,\,[1\,2\,3])}\big)^{\mathrm{T}}\mathbf{C}_2^{\mathrm{T}} = \mathbf{B}_{(\Sigma 7,\,[1\,2\,3])}. \tag{28}$$

Besides this operation and the identity element of $\mathcal{G}$, there are no other operations with this property. Thus, the multiplicity of $\mathbf{B}_{(\Sigma 7,\,[1\,2\,3])}$ equals 2, and instead of $24 \times 24 \times 2 \times 2 = 2304$ points given by equation (26), there are only 1152 different points of $\mathcal{M}_\mathcal{B}$ representing the boundary '$(\Sigma 7, [1\,2\,3])$'.

A question arises about parts of the interface manifold in which each geometrically distinct boundary is represented only once, *i.e.* about a domain containing exactly one representative of each equivalence class. In texture analysis, a number of different names (*e.g.* asymmetric domain or unit, symmetrically equivalent area, fundamental zone) are ascribed to an analogous region for (mis)orientations. In geometry, a similar notion of the fundamental region is used; with the interface manifold, it is a closed subset $\mathcal{F}_\mathcal{B}$ of $\mathcal{M}_\mathcal{B}$ such that the images $\mathcal{S}_i(\mathcal{F}_\mathcal{B})$ cover $\mathcal{M}_\mathcal{B}$, and the interiors of the images have no common points. It is easy to see that internal points of a fundamental region have a multiplicity of one, and points of higher multiplicity are located at its boundary.

When it comes down to the determination of a fundamental region, the task is to specify proper bounds on the parameters $(x^1, \ldots, x^5)$. This task can be simplified by the fact that fundamental regions for boundaries are related to asymmetric domains for misorientations, and the later are known for all combinations of crystallographic symmetries (Morawiec, 1997). This allows one to construct fundamental regions for all types of hetero- and homo-phase boundaries when $\mathcal{M}_\mathcal{B} = SO(3) \times S^2$. For that purpose, one may take a Cartesian product of the proper fundamental region for misorientations (this limitation accounts for operations $\mathbf{C}_1\mathbf{B}\mathbf{C}_2^{\mathrm{T}}$ plus $\mathbf{B}^{\mathrm{T}}$ if applicable) and a hemisphere of inclinations (which accounts for $\mathbf{B}^{-}$).

Example fundamental region. For the octahedral symmetry of crystals, with q^i, k^a being parameters of $(M, \mathbf{m}_1)$, the fundamental region for misorientations can be given by the conditions $0 \le q^3 \le q^{1,2} \le q^0(2^{1/2} - 1)$ and $q^1 + q^2 + q^3 \le q^0$. If additionally M is equivalent to M^{T}, then one can take $q^1 \le q^2$. The equivalence between $\mathbf{B}$ and $\mathbf{B}^{-}$ can be accounted for by limiting the parameters on the sphere of inclinations to the upper hemisphere ($k^3 \ge 0$).

Symmetries affect metric properties of the manifold of interfaces. Distances may be invariant under symmetry operations. For example, the transformation '$-$' and the rotations of the crystallographic point groups are isometries of s_A [*cf.* equation (18)],

$$s_\mathrm{A}(\mathbf{B}^{-}, \mathbf{B}'^{-}) = s_\mathrm{A}(\mathbf{B}, \mathbf{B}') = s_\mathrm{A}(\mathbf{C}_1\mathbf{B}\mathbf{C}_2^{\mathrm{T}}, \mathbf{C}_1\mathbf{B}'\mathbf{C}_2^{\mathrm{T}}), \tag{29}$$

but the transposition operation T is not. The last issue does not concern hetero-phase boundaries. For homo-phase boundaries, if there is no identification of inclinations at $M = I_3$, one can define a new distance function (*cf.* Morawiec, 2000)

$$s_\mathrm{B}^2\big[(M, \mathbf{m}_1), (M', \mathbf{m}_1')\big] := s_\bullet(M, M')^2 + \big[s_\circ(\mathbf{m}_1, \mathbf{m}_1')^2 + s_\circ(\mathbf{m}_2, \mathbf{m}_2')^2\big]/2 \tag{30}$$

for which all considered symmetry operations are isometries:

$$\left.\begin{matrix} s_\mathrm{B}(\mathbf{C}_1\mathbf{B}\mathbf{C}_2^{\mathrm{T}}, \mathbf{C}_1\mathbf{B}'\mathbf{C}_2^{\mathrm{T}}) \\ s_\mathrm{B}(\mathbf{B}^{-}, \mathbf{B}'^{-}) \\ s_\mathrm{B}(\mathbf{B}^{\mathrm{T}}, \mathbf{B}'^{\mathrm{T}}) \end{matrix}\right\} = s_\mathrm{B}(\mathbf{B}, \mathbf{B}'). \tag{31}$$

Since s_B is not a product metric, its interpretation is not as simple as that of s_A, and the corresponding metric tensor has a complicated form.

For other metrics (*e.g.* s_C), relations analogous to equations (29) and (31) may not be satisfied, and a question arises how to construe distances between classes of equivalent points. According to Cahn & Taylor (2006) 'Symmetries can always be incorporated by replacing actual distances by the minimum among all representatives of the same equivalence class.' This would formally mean that if s denotes a distance defined on $\mathcal{M}_\mathcal{B}$ (*i.e.* without influence of symmetry), with the presence of symmetry, the distance on $\mathcal{M}_\mathcal{B}/\mathcal{G}$ would be

$$\sigma([\mathbf{B}_1], [\mathbf{B}_2]) = \min_{i,j} s\big[\mathcal{S}_i(\mathbf{B}_1), \mathcal{S}_j(\mathbf{B}_2)\big]. \tag{32}$$

However, in general such σ would violate the triangle inequality (Fig. 5). Appropriately, instead of equation (32), distance functions applicable to point sets need to be used. This could be the Hausdorff metric, which in the considered circumstances has the form

$$\sigma^{\mathrm{H}}([\mathbf{B}_1],[\mathbf{B}_2]) = \max\{\max_i \min_j s\big[\mathcal{S}_i(\mathbf{B}_1),\mathcal{S}_j(\mathbf{B}_2)\big], \max_j \min_i s\big[\mathcal{S}_i(\mathbf{B}_1),\mathcal{S}_j(\mathbf{B}_2)\big]\}. \quad (33)$$

Equation (32) is a metric if all symmetry operations are isometries of s: since

$$s\big[\mathcal{S}_i(\mathbf{B}_1),\mathcal{S}_j(\mathbf{B}_2)\big] = s\big\{\mathcal{S}_j^{-1}\big[\mathcal{S}_i(\mathbf{B}_1)\big],\ \mathcal{S}_j^{-1}\big[\mathcal{S}_j(\mathbf{B}_2)\big]\big\} = s\big[\big(\mathcal{S}_j^{-1}\mathcal{S}_i\big)(\mathbf{B}_1),\mathbf{B}_2\big], \quad (34)$$

the function σ involving symmetry is given by the simple formula

$$\sigma([\mathbf{B}_1],[\mathbf{B}_2]) = \min_i s\big[\mathcal{S}_i(\mathbf{B}_1),\mathbf{B}_2\big] = \sigma^{\mathrm{H}}([\mathbf{B}_1],[\mathbf{B}_2]), \quad (35)$$

and it satisfies the triangle inequality. Taking this into account, it is more convenient to use metrics for which symmetry operations are isometries than any of those violating this condition. As was indicated above, symmetry operations are isometries of $s = s_{\mathrm{A}}$ in the hetero-phase case [equation (29)], and of $s = s_{\mathrm{B}}$ in both the hetero- and the homo-phase cases [equation (31)].

Before closing this section, it needs to be mentioned that the differential structure is also affected by the equivalence of symmetric points: the resulting object is not a differential manifold. This object may be properly endowed with differential structure as a so-called differential orbifold – a generalization of a manifold and of a quotient space of a manifold with respect to a group of transformations (Satake, 1956). To pass on the metric structure of the manifold, the group of transformations must consist of isometries. Thus, one may consider the space of hetero-phase boundaries to be the orbifold $\mathcal{M}_{\mathcal{B}}/\mathcal{G}$, where $\mathcal{M}_{\mathcal{B}}$ is the product manifold of $SO(3) \times S^2$ equipped with the product metric based on s_{A}. The same applies to homo-phase boundaries with the metric determined by the distance function s_{B}.

If the symmetry operations are not isometries (*e.g.* warped geometry for homo-phase boundaries with the metric based on s_{C}), a proper 'space' can be built based on a suitably selected fundamental region $\mathcal{F}_{\mathcal{B}}$; with the rest of $\mathcal{M}_{\mathcal{B}}$ ignored, $\mathcal{F}_{\mathcal{B}}$ is a Riemannian manifold with a boundary. To cover complete ranges of boundary parameters, one may take the images $\mathcal{S}_i(\mathcal{F}_{\mathcal{B}})$ and glue them together along equivalent boundaries. Schematics of $\mathcal{F}_{\mathcal{B}}$ and the covering space are shown in Fig. 6.

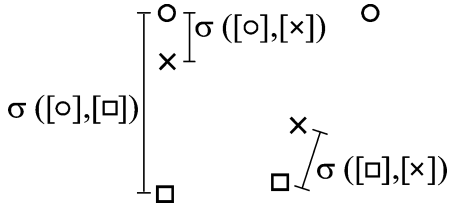

Figure 5
Schematic illustration of σ violating the triangle inequality. Identical symbols represent equivalent points. Assuming Euclidean distance as s, there occurs $\sigma([\circ],[\times]) + \sigma([\times],[\square]) < \sigma([\circ],[\square])$.

5. Volume element and boundary distribution

A volume element is a differential defining a measure on the manifold and provides a means to calculate volumes in a generalized sense. Riemannian manifolds come with canonical volume elements following from their metric structure. In the case of a five-dimensional manifold parameterized by x^i ($i = 1, \ldots, 5$), such a volume element is given by

$$\mathrm{d}V(x^i) = |\det(g)|^{1/2}\,\mathrm{d}x^1\mathrm{d}x^2\ldots\mathrm{d}x^5, \quad (36)$$

i.e. it is obtained from the metric tensor. The element $\mathrm{d}V$ is expected to satisfy the normalization condition $\int_{\mathcal{M}_{\mathcal{B}}} \mathrm{d}V = 1$. For the metrics defined above, this condition can be fulfilled by an appropriate choice of the constants $c_\bullet$ and $c_\circ$.

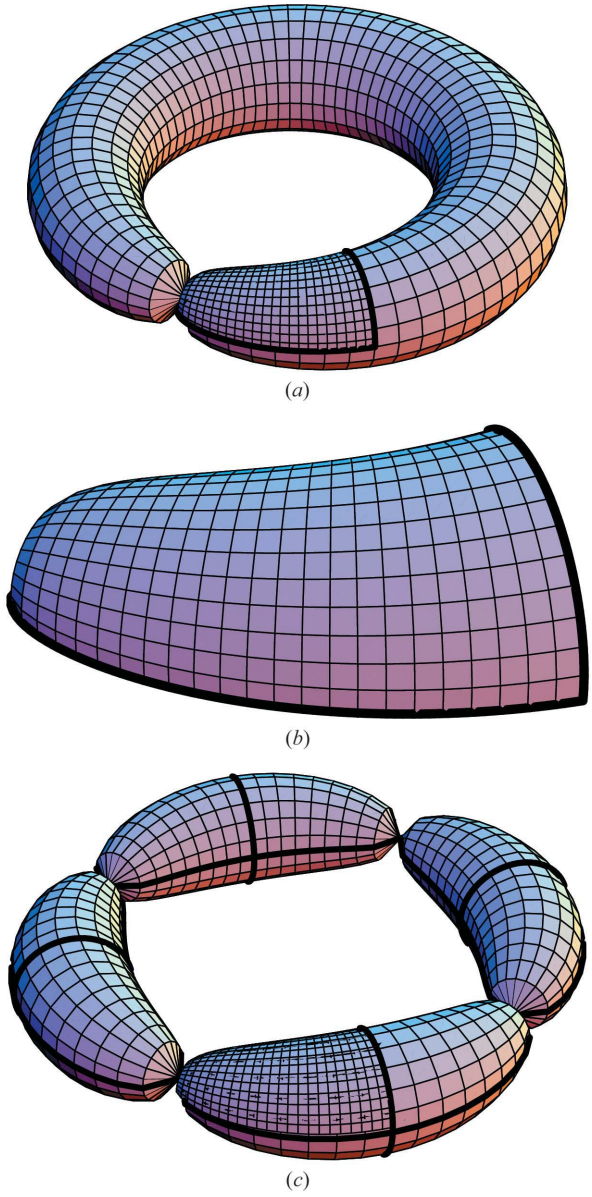

Figure 6
(*a*) Schematic of fundamental region selected on the manifold of Fig. 4. (*b*) The fundamental region detached from the rest of the manifold. (*c*) Unfolded space composed of images obtained by transformation of the fundamental region by symmetry operations.

Example volume element. For the warped product metric [equation (21)], the factor $|\det(g)|^{1/2}$ of the volume element can be expressed as $f|\det(g^{\bullet})|^{1/2}|\det(g^{\circ})|^{1/2}$. This formula also applies to the product metric if f is set to 1; in the case of $s_{\rm A}$ and the parameterization by $(q^1, q^2, q^3, k^1, k^2)$ one has

$$|\det(g)|^{1/2} = 8c_{\circ}^2 c_{\bullet}^3\ (q^0\, k^3)^{-1}. \tag{37}$$

The volume element based on the metric $s_{\rm B}$ is given by

$$|\det(g)|^{1/2} = (8c_{\circ}^2 c_{\bullet}^3 + 2c_{\circ}^4 c_{\bullet})\ (q^0\, k^3)^{-1}, \tag{38}$$

i.e. it differs from equation (37) only by the arrangements of constants $c_{\bullet}$ and $c_{\circ}$. The constants are related by the normalization condition. The integral of $1/k^3$ over the upper hemisphere equals 2π; with additional maps, the integral over the complete sphere is 4π. The integral of $1/q^0$ over $SO(3)$ equals π^2. Thus, the integral of $(q^0 k^3)^{-1}$ over the entire product space $SO(3) \times S^2$ is $4\pi^3$.

The frequency of occurrence of boundaries can be modeled by a density function over the macroscopic boundary parameters. This function is usually referred to as the (five-dimensional or five-parameter) boundary distribution (*e.g.* Randle, 2008). Once the volume element $\mathrm{d}V$ is known, the boundary distribution, say F, can be formally defined. Ignoring symmetries, its value at the point $\mathbf{B}$ times an infinitesimal volume $\mathrm{d}V$ centered at that point is equal to the ratio of the area $\mathrm{d}A$ of boundaries with parameters in $\mathrm{d}V$ to the total area of boundaries A:[3]

$$\mathrm{d}A/A = F(\mathbf{B})\,\mathrm{d}V. \tag{39}$$

With symmetries involved, the boundary distribution must take equal values for equivalent boundary parameters, *i.e.*

$$F(\mathbf{B}) = F[\mathcal{S}_i(\mathbf{B})]. \tag{40}$$

Strictly speaking, F is determined on $\mathcal{M}_{\mathcal{B}}/\mathcal{G}$, *i.e.* $F = F([\,\mathbf{B}\,])$, and the definition of F needs to be suitably modified. As in texture analysis, whether the element $\mathbf{B}$ or the class $[\,\mathbf{B}\,]$ is used as the argument of F may depend on context and convenience. Needless to say, the boundary distribution corresponding to $F = 1$ is considered to be 'random'.

Having a given model for the 'random' boundary distribution, it is essential to resolve how it relates to a boundary distribution of a 'random microstructure'. However, there is no clear definition of the latter. Roughly, this could be a microstructure with randomly oriented equiaxial grains and no particular correlations between grain and boundary characteristics. Generally, one should not expect such a microstructure in real materials. To construct it, one needs clearly defined conditions for microstructure simulation. If one takes Poisson–Voronoï tessellation with orientations randomly assigned to cells [based on invariant measure on $SO(3)$] and random inclinations of cell faces (based on invariant measure on S^2), the boundary distribution will correspond to $F = 1$ based on the product metric $s_{\rm A}$. Such distribution will be uniform also in the space with the volume element obtained from $s_{\rm B}$ because of the proportionality of expressions (37) and (38).

Even in the simple case of $F = 1$, the impact of symmetries needs to be remembered. Because of the symmetries, the number of boundaries of different types will not be the same if their multiplicities differ. Let us consider two balls of the same small radius r centered at $[\,\mathbf{B}_1\,]$ and $[\,\mathbf{B}_2\,]$. Let the volumes of the balls be v_1 and v_2, respectively. With boundaries generated at random, the volume v_i is proportional to the number of boundaries falling within the range r of elements of the class $[\,\mathbf{B}_i\,]$ $(i = 1, 2)$. Thus, v_i is proportional to the number of equivalent elements in the class $[\,\mathbf{B}_i\,]$, which equals $|\mathcal{G}|/|\mathcal{G}_{\mathbf{B}_i}|$. Hence, one has

$$v_1|\mathcal{G}_{\mathbf{B}_1}| = v_2|\mathcal{G}_{\mathbf{B}_2}|, \tag{41}$$

or, in other words, the volume v_i is proportional to the inverse of the multiplicity of $\mathbf{B}_i$.

There is a link between distributions based on warped product metrics with different scaling functions. For the distribution F corresponding to a metric containing the scaling function f, the ratio $\mathrm{d}A/A$ can be expressed as $Ff|\det(g^{\bullet})|^{1/2}|\det(g^{\circ})|^{1/2}\,\mathrm{d}x^1 \ldots \mathrm{d}x^5$. An analogous expression is applicable to the couple F_1 and f^1. Hence,

$$Ff = F_1 f^1. \tag{42}$$

In particular, if a small neighborhood of the misorientation $M = I_3$ is excluded, and $f = 1$ outside the neighborhood, then $F = F_1 f^1$. These relations may serve for comparing the effects of applying various uniformity models.

It is worth noting that the integration of an arbitrary boundary distribution F over all normals (with the weight of $|\det(g^{\circ})|^{1/2}$) gives the conventional misorientation distribution times f. The distribution of normals with respect to the canonical measure on S^2 is obtained by integration of F over misorientations with the weight $f|\det(g^{\bullet})|^{1/2}$.

Finally, it needs to be mentioned that, for analysis and modeling of experimental boundary distributions, model probability density functions are needed. These would be generalizations of the Gauss distribution in Euclidean space. Such generalizations are usually based on Brownian motion, maximum likelihood or maximum entropy principles. In the simple case of the product space $SO(3) \times S^2$, interesting model distributions may be obtained from those used in the field of 'directional statistics' (Mardia, 1972; Prentice, 1986; Schaeben, 1996) or as generalizations of the 'crystallographic exponential family' (van den Boogaart, 2002). More detailed examination of this issue is beyond the scope of this paper.

6. Summary

The present paper contains mathematical foundations and some background needed for defining a five-dimensional boundary distribution. The definition requires a measure in

[3] This function was proposed by Bunge (1982), but no quantitative definition was given except for the very special case when F can be factored with separation of the misorientation (M) and inclination ($\mathbf{m}_1$) variables. Functions similar to F were considered by Adams (1986) and Adams & Field (1992); however, those explicitly defined in these papers differ from F by their arguments.

the space of boundary parameters. The measure, in turn, can be obtained from an assumed metric structure of the interface manifold, and both the metric and the measure are affected by the presence of symmetries.

There is a difference between the formal treatment of homo- and hetero-phase boundaries. For hetero-phase interfaces, the reference misorientation is not unique, but otherwise analysis of such boundaries is straightforward. There is no singularity at the reference misorientation, the Cartesian product $SO(3) \times S^2$ can be taken as the interface manifold, there is no equivalence due to interchange of grains and applicable symmetry operations are isometries of the simple product metric (s_A).

The case of homo-phase boundaries is more complex. Two distinct approaches are possible. First, following Morawiec (1998, 2000), one can assume the interface manifold to be $SO(3) \times S^2$ with a suitable metric (*e.g.* s_B). However, this means that data at $M = I_3$ are meaningless. Moreover, two boundaries – both with misorientations close to I_3 – may be separated by a large distance because their inclinations may differ considerably, whereas – from the physical viewpoint – all 'small angle' boundaries may be considered to be close. Finally, with the first approach, there is no particular 'reference' boundary; this is against the convention of using the misorientation angle – *i.e.* the distance to the reference misorientation of I_3 – for quantitative assessment of boundaries. On the basis of these arguments one may take an alternative approach: following Cahn & Taylor (2006), homo-phase 'boundaries' with the misorientation of I_3 can be identified as one point. For the continuity at $M = I_3$, a warped product metric with the scaling function vanishing at I_3 can be used. This point is distinct from other points and may serve as a 'reference' boundary. However, complications arise because symmetry operations are not isometries of basic warped product metrics (*e.g.* s_C), and 'brute force' symmetrization is needed (*i.e.* the boundary space is obtained by selecting a proper fundamental region and gluing together its images in transformations by symmetry operations). Moreover and more importantly, the choice of the scaling function is rather arbitrary. The choice has an impact on the 'random' boundary distribution; with the volume element depending on the metric, this fundamental distribution is determined by the convention used in selection of the scaling function.

Consequently, a question arises: how significant are the reasons for postulating a warped geometry with identification of inclinations at I_3? Losing a unique reference point may be an inconvenience but it has no other consequences; the lack of it can be accepted like the fact that there is no such point in the case of hetero-phase boundaries. It is true that 'small angle' boundaries have some similar physical characteristics but these characteristics are either not connected to the metric (*e.g.* free energy) or – if they are connected (*e.g.* boundary rate on $\mathcal{M}_B$, *i.e.* roughly its alacrity for changing parameters) – there is no evidence that a warped geometry will be of any help in analysis of these quantities; more likely it will be a nuisance. Taking into account the above arguments for and against each of the two approaches to homo-phase interfaces, we consider the first one to be more convenient.

Generally, other measures – different from those listed above – may be applied but it must be stressed that a given boundary distribution can be discussed only if the measure or model of uniformity is explicitly specified; without that, the boundary distribution is ill-defined and its analysis may lead to inconsistent conclusions. It would be to the advantage of grain boundary research to select a particular model and use it as a standard, so boundary distributions originating from different sources could be easily compared. In our opinion, the measure following from the distance functions s_A and s_B could be a good choice but this does not mean that better approaches may not arise.

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