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On "The five-dimensional parameter space of grain boundaries" by Sutton, Banks & Warwick

Abstract: In their recent paper [*Proc. Roy. Soc. A* **471**, 2181 (2015)], AP Sutton, EP Banks and AR Warwick criticized previous metrics in the space of macroscopic boundary parameters and proposed a new function instead. However, some aspects of their analysis need to be clarified. In particular, reasons for introducing one of the criticized metrics are explained, and deficiencies of the new function are indicated.

Keywords: grain boundary, interface, metric, crystal symmetry, microstructure

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Sutton, Banks & Warwick have recently published an article about metrics in the space of macroscopic grain boundary parameters [1]. The authors deserve credit for bringing the subject to the attention of the scientific community. This comment is intended to correct some issues raised in [1]. Particularly important is the categorical claim made by the authors that a metric in the boundary space "must" satisfy certain conditions and the observation that the metric χ used in [2] does not fulfill them.[1] The erroneousness of χ would have far-reaching implications because a number of papers about grain-boundary populations (see, e.g., [3] and references therein) tacitly rely on a measure derived from this metric.

1. It is worth explaining the original context and initial reasons for introducing the boundary-space metric. Some years ago, experimental developments in automatic characterization of polycrystalline materials opened the opportunity to study frequencies of occurrence of macroscopic boundary types [4]. Two formal issues needed to be resolved to depict and interpret these frequencies: first, it was necessary to determine the symmetry-caused equivalence of boundary parameters, and second – as in all problems concerning geometrical probabilities – one needed to specify a measure (or 'random' distribution or 'volume element') in the boundary parameter space. Solutions to these problems for both homo- and hetero-phase boundaries were given in [5] and than described in more details in [6] and [7]. The construction of the volume element was based on the metric tensor, and that was the primary purpose of introducing the metric. Only later the distance in the space of macroscopic boundary parameters arose as an independent subject [8, 9].

[1] The symbol $\Delta^{(M)}$ used in [1] denotes χ^2.

2. As was explicitly stated in [5], the measure in the boundary space is not unique. Also the choice of a metric from which the measure is derived is a matter of convention, but because of the physical interpretation, intuitive simplicity or computational convenience some metrics are better than the others. On the other hand, the problem of equivalence between boundary parameters has an unambiguous solution. Because of crystal symmetries, different points in the boundary parameter space correspond to macroscopically identical boundary configurations. In the case of homo-phase boundaries, the additional grain exchange symmetry may arise. The equivalence of boundary representations induces symmetries in the space. (Alternatively, the actual boundary space can be seen as comprised of classes of equivalent boundary representations [7], but this concept was not used in [1] and will be omitted here.) Functions determined on the boundary space exhibit symmetries in the sense that values of a given function at equivalent points are equal.

3. A metric as a (two-argument) function on the boundary parameter space must reflect the symmetries of the space. More precisely, the transformations of the space by symmetry operations need to be isometries, i.e., if $\mathbf{b}$ represents a boundary, and $\mathcal{S}(\mathbf{b})$ is the representation equivalent to $\mathbf{b}$ due to the symmetry $\mathcal{S}$, then the distance $\widetilde{d}\,(\mathbf{b}_1, \mathbf{b}_2)$ between boundary representations $\mathbf{b}_1$ and $\mathbf{b}_2$ is expected to be equal to the distance between $\mathcal{S}(\mathbf{b}_1)$ and $\mathcal{S}(\mathbf{b}_2)$

$$\widetilde{d}\,(\mathbf{b}_1, \mathbf{b}_2) = \widetilde{d}\,(\mathcal{S}(\mathbf{b}_1), \mathcal{S}(\mathbf{b}_2)) \ . \tag{1}$$

A simple method of constructing the desired metric in the actual symmetry-affected boundary space is by defining an auxiliary metric in a simpler provisional space devoid of some symmetries, and then by extending it to a metric in the fully symmetric space. Preferably, any physical meaning, intuitive simplicity or computational convenience of the auxiliary metric should be passed to the extended metric in the fully symmetric space.

4. The auxiliary metric χ denounced in [1] was designed to be used in the case of homo-phase boundaries in the presence of both crystal symmetry and grain exchange symmetry [5]. It is described in [1] as "inconsistent" because of the "lack of separation" between the contributions from misorientations and boundary normals. In a more formal language, χ is not a product metric although its domain is a Cartesian product; this obvious fact was explicitly expressed in [7].

The "lack of separation" is a feature of general spaces. In a space which is a Cartesian product, having the property of "separation" is appealing, but it is *not* a necessary requirement for a function to be referred to as a metric. The reason for endorsing χ was

that it naturally satisfies eq.(1) with respect to both crystal symmetry and grain exchange symmetry; see eq.(31) in [7]. Therefore, the step of extending χ to a metric in the symmetric boundary space is straightforward [7].

5. The authors of [1] use a boundary parameterization with misorientations described by Rodrigues parameters ($\boldsymbol{\rho}$) and a vector ($\hat{\mathbf{N}}$) determined by boundary normals. With this parameterization, an auxiliary metric Δ ($= \Delta_{12}$) in the provisional space is introduced. It has the appealing attribute of being a product metric. Just like χ [6], the metric Δ breaks down at zero misorientation, but besides that, Δ has the disadvantage that if both the misorientation and the boundary planes are changed in such a way that $\hat{\mathbf{N}}$ is kept constant, only the misorientation change contributes to the growing distance. This effect is strongly pronounced when the misorientation angles approach π; in this case, the distance Δ between very different boundaries can be negligibly small. (See Appendix A.) Thus, besides the singularity at the point of zero misorientation, Δ fails near the surface of half-turns in the misorientation space.

The metric Δ has the property described by eq.(1) with respect to the grain exchange symmetry, but it does not have it with respect to some crystal symmetries; see Appendix B. To account for all equivalences, the authors extend the auxiliary metric Δ by taking the "minimum value of Δ_{ij}" [1], i.e., for given two boundaries, the extended function, say $\widetilde{\Delta}$, is the smallest of distances Δ of particular representations of the first boundary from particular representations of the second boundary. The problem with this approach is that the so-defined extension $\widetilde{\Delta}$ violates the triangle inequality; see Appendix C. Thus, the extended Δ proposed in [1] is not a metric in the true boundary space influenced by crystal symmetry.

6. Summarizing the above, the metric χ is not a product metric, but it satisfies eq.(1), and this allows for a simple extension to the actual boundary space. On the other hand, Δ is a product metric, but it does not fulfill eq.(1), and this complicates the step of extending it.

As was explained in [7], there is a general way of extending an auxiliary metric to the space with classes of equivalent representations by using properly adapted metrics applicable to sets. This can be the Hausdorff metric in the form given in [7], and the concept can be applied to Δ. Although the Hausdorff-extended Δ does not take into account the structure of the equivalence classes and lacks any physical interpretation, it formally satisfies all necessary conditions for being a metric in the symmetry-affected boundary space.

7. In relation to the metric, the paper [1] deals with the problem of equivalent specifications of grain boundaries. Sutton, Banks & Warwick ignore previous accounts on this subject [5–7] and present it as if it had not been considered before, although appropriate rules are briefly reviewed in [2] – the paper they critically examined in [1]. Moreover, the transcription of the rules from the matrix formalism of [2] to the Rodrigues-parameters formalism of [1] is imprecise. No distinction is made between the grain exchange symmetry and the crystal symmetry, and the case of symmetries involving proper rotations (represented by Rodrigues vectors) is mixed with symmetry operations in general (including improper rotations, which are not represented by Rodrigues vectors). The imprecision is manifested in arbitrariness of vector directions in [1]. (It adds to the confusion that at the outset only non-enantiomorphism of crystals is assumed, thus allowing for non-enantiomorphic *polar* crystals.) In a correct approach, any change of a vector direction should be linked to a symmetry operation, and the application of the symmetry to a boundary normal should be accompanied by application of this symmetry to misorientation parameters and vice versa.

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Appendix A: Example of arbitrarily small distance Δ between different boundaries.

All symbols except those defined in this note have the meaning specified in [1]. With $i = 1, 2$, let $\mathbf{b}_i = \left(\mathbf{N}^{(i)}, \boldsymbol{\rho}^{(i)}\right)$, where $\boldsymbol{\rho}^{(i)} = (0, 0, \cot \epsilon_i)$, $\mathbf{N}^{(i)} = (\epsilon, 0, 1)$, and ϵ and ϵ_i (in radians) are such that $1 \gg \epsilon_1 \gg \epsilon \gg \epsilon_2 > 0$. The corresponding boundary normals are

$$\mathbf{n}^{(1)} = (\epsilon, \epsilon \cot \epsilon_1, 1) \approx (0, 0, 1), \qquad \mathbf{n}'^{(1)} = (\epsilon, -\epsilon \cot \epsilon_1, 1) \approx (0, 0, 1),$$
$$\mathbf{n}^{(2)} = (\epsilon, \epsilon \cot \epsilon_2, 1) \approx (0, 1, 0), \qquad \mathbf{n}'^{(2)} = (\epsilon, -\epsilon \cot \epsilon_2, 1) \approx (0, -1, 0).$$

The distance Δ between $\mathbf{b}_1$ and $\mathbf{b}_2$ equals $2(\epsilon_1 - \epsilon_2) \approx 2\epsilon_1$, i.e., it is small despite very different boundary planes; $\mathbf{b}_1$ represents a near-180°-twist boundary, whereas $\mathbf{b}_2$ corresponds to a near-180°-tilt boundary.

Appendix B: Δ does not satisfy eq.(1) with respect to crystal symmetry.

The example below is based on two boundaries considered in [1] and expressed via direction indices of the cubic lattice: $\mathbf{b}_1 = \left(\mathbf{N}^{(1)}, \boldsymbol{\rho}^{(1)}\right) = ([1\,1\,1],\ [1\,1\,1]/3)$ and $\mathbf{b}_2 = \left(\mathbf{N}^{(2)}, \boldsymbol{\rho}^{(2)}\right) = ([1\,1\,1],\ [2\,3\,4]/7)$. Let $\mathcal{S}$ correspond to the crystal symmetry represented by $\sigma = [0\,0\,\infty]$ and acting on the left side of $\boldsymbol{\rho}^{(i)}$, i.e., the parameters of misorientations $\boldsymbol{\rho}^{(i)}$ and boundary normals $\mathbf{n}^{(i)}$ are changed to $\sigma \star \boldsymbol{\rho}^{(i)}$ and $\sigma \star \mathbf{n}^{(i)} \star (-\sigma)$, respectively. Hence, one gets the equivalent specifications $\mathcal{S}(\mathbf{b}_1) = \left([0\,0\,1],\ [1\,\overline{1}\,\overline{3}]\right)$ and $\mathcal{S}(\mathbf{b}_2) = \left([1\,\overline{2}\,7],\ [3\,\overline{2}\,\overline{7}]/4\right)$. The distance between $\mathbf{b}_1$ and $\mathbf{b}_2$ differs from that between their equivalents $\mathcal{S}(\mathbf{b}_1)$ and $\mathcal{S}(\mathbf{b}_2)$: $\Delta\left(\mathcal{S}(\mathbf{b}_1), \mathcal{S}(\mathbf{b}_1)\right) = \Delta(\mathbf{b}_1, \mathbf{b}_2) + \arccos(7\sqrt{6}/18)$.

Appendix C: $\widetilde{\Delta}$ violates of the triangle inequality.

Let the crystal point group be C_{2h} with the two-fold axis along z. In this case, the only non-trivial proper rotation among crystal symmetry operations corresponds to $\sigma = [0\,0\,\infty]$. With the grain exchange symmetry taken into account, a general boundary has 16 equivalent representations in the space being a Cartesian product of proper rotations and the sphere of unit vectors. The 'distances' $\widetilde{\Delta}$ between the boundaries $\mathbf{b}_1$ and $\mathbf{b}_2$ listed in Appendix B, and $\mathbf{b}_3 = \left(\mathbf{N}^{(3)}, \boldsymbol{\rho}^{(3)}\right) = \left([0\,1\,1],\ [\overline{1}\,\overline{1}\,0]/4\right)$ are $\widetilde{\Delta}(\mathbf{b}_1, \mathbf{b}_2) \approx 23°$, $\widetilde{\Delta}(\mathbf{b}_2, \mathbf{b}_3) \approx 92°$ and $\widetilde{\Delta}(\mathbf{b}_3, \mathbf{b}_1) \approx 55°$. Since $\widetilde{\Delta}(\mathbf{b}_2, \mathbf{b}_3)$ exceeds the sum of $\widetilde{\Delta}(\mathbf{b}_1, \mathbf{b}_2)$ and $\widetilde{\Delta}(\mathbf{b}_3, \mathbf{b}_1)$, the triangle inequality is violated. For simplicity, the lowest non-trivial crystal symmetry with inversion was assumed above, but numerical tests show that the conclusion is also true for higher symmetries including the cubic holohedry O_h considered in [1].