Journal: Acta Materialia, Paper: A-21-0401
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Title: Five degree-of-freedom property interpolation of arbitrary grain boundaries via Voronoi fundamental zone octonion framework.

The manuscript describes methods for time-efficient commutation of distances between boundaries in the 'space' of macroscopic boundary parameters and for interpolation of grain boundary energy as a function of these parameters. Some statements made in the manuscript are not correct. In my opinion, the manuscript does not meet requirements of Acta Materialia.

A. Key for the computation of distances between grain boundaries is accounting for symmetrically equivalent boundary representations. There are a number of indications that the authors do not fully understand this subject.

1. According to the manuscript, the cardinality of the group O_h is $N_p = 24$ (p.4, l.5), (p.4, l.27, R), (p.18, l.22, R).[1] It is well known that the number of elements in the octahedral point group O_h is 48; see, International Tables for Crystallography. Twenty four is the number of proper rotations in the O_h group.

2. According to the manuscript, the number of symmetrically equivalent representations of a boundary (number of SEOs) is N_p^2 (p.3, l.40, R). This number is not correct. Assuming crystals are invariant with respect to inversion, N_p is the number of proper symmetry operations of the point group, and grain exchange symmetry is allowed, then the number of symmetrically equivalent representations of a boundary is $4N_p^2$.

3. According to the manuscript, the proposed methods "can be applied to any of the 32 crystallographic point groups" (p.1, l.38), (p.21, l.25, R). Such a general approach would have to encompass noncentrosymmetric crystals. No details are given in the manuscript, and I doubt in correctness of that approach.

4. According to the manuscript, "to perform a traditional distance calculation it is necessary to compare all SEOs of o_1 to all of the SEOs of o_2 and take the smallest distance." (p.3, ll.22–25, R). Again, this is not correct. With the considered metric, a transformation between equivalent representations is an isometry, and therefore, to calculate the distance between two boundaries, it is enough to compare a *single* representation of one boundary to all representations of the other boundary. See, e.g., section 7 in [50].

The first three points on the list above can be viewed as mere mistakes. The last point, however, affects large parts of the manuscript devoted to speeding up computations of distances between boundaries.

B. Using Voronoi based fundamental zone in texture related analyzes is not really new. Such fundamental zones are well defined in the case of orientations. In the case of grain boundaries (or misorientations), the construction is not that elegant. Contrary to what is written in the manuscript (p.3, l.25, L), the grain boundary representation taken as the reference point cannot be arbitrary. If the reference point is invariant with respect to a symmetry operation other than identity, then it is at the same smallest distance to *multiple* representatives of another boundary, and the procedure does not work; the resulting Voronoi cell is a multiple of the fundamental zone.

[1] L and R denote left and right columns, respectively.

Moreover, with boundary representatives limited to the fundamental zone, close boundaries may have distant representatives. The authors addressed the problem by performing computations for a number of different reference points, i.e., for a number of different fundamental zones.[2] This approach is novel, but not fully reliable; if the number of different fundamental zones is small, the data may still contain some incorrect distances.

C. The unquestionable new contribution of the manuscript is the use of metric-based all-purpose multivariate interpolation methods to predicting symmetric energy function from known energies of a given set of boundaries. The method is different from and simpler than that described in [66], but the resulting energy functions will be smooth[3] (no cusps).

In conclusion, the manuscript A-21-0401 does report some new science (**C** and **B**), but extensive fragments of it are devoted to solving a non-existent problem (**A**). Therefore, I recommend against publication of A-21-0401 in Acta Materialia.

In case my recommendation is overruled or the manuscript is submitted elsewhere, I would like add the following two comments.

First, the text is long and difficult to read. Here are a couple of measures which (in my opinion) would make the paper more readable.
• I would remove most abbreviations. Some are used only once (3DOF, GBED, kFCV, LOOCV) or twice (BPFZ, TJ), and they do not help. Moreover, I would check for repetitions. E.g., fragments (p.18, ll.30–37, L) and (p.18, ll.26–33, R) carry practically the same message.
• The text is overloaded with overcomplicated expressions. For instance, there is no need to talk about Riemannian manifolds (p.1, l.20), (p.3, l.11, L) as the considered metric is not intrinsic. Or take the word 'octonion' repeated so many times explicitly and in acronyms. Since the octonion product is never used, referring to a pair of unit quaternions as an octonion is a misuse of a well defined mathematical term.[4] The authors followed terminology introduced in other papers, but repeating mistakes of others adds to the confusion. When seeing the word 'octonion' repeated 200 times, a reader reaches the false conclusion that the formalism relies on true octonions.

Second, most parts of the work, in particular considerations concerning interpolation, are independent of boundary parameterization and are applicable to other properly defined metrics. There is no reason for limiting these considerations to a particular grain boundary parameterization or a particular distance function.

[2] The method known to me was to use a larger region = a single fundamental zone plus a symmetry-dependent margin.

[3] This is my guess. No relevant figures are shown.

[4] A simple analogue would be to refer to unit real numbers as complex numbers. Elements of the pair (r_1, r_2) of unit reals satisfy $|r_i| = 1$, i.e., (r_1, r_2) is one of $(\pm 1, \pm 1)$. The multiplication rule for the pairs (r_1, r_2) and (r'_1, r'_2) is $(r_1, r_2) \star (r'_1, r'_2) = (r_1 r'_1, r_2 r'_2)$, whereas for complex numbers it is $(r_1, r_2) \star (r'_1, r'_2) = (r_1 r'_1 - r_2 r'_2, r_1 r'_2 + r_2 r'_1)$. Clearly, using the term 'complex numbers' for the pairs $(\pm 1, \pm 1)$ is out of place. There is no need for complex numbers to multiply, say, 1 by -1.