Computational Materials Science  
  
Title: Five Degree-of-Freedom Property Interpolation of Arbitrary Grain Boundaries via Voronoi Fundamental Zone Framework  
Ref. No.: COMMAT-D-21-01089  
  
  
Dear Mr. Baird,  
  
Your above-mentioned manuscript has been assessed by referees knowledgeable in the subject of study. The referee(s) raised points that require clarification/revision should you wish your paper to be considered further. Please see these comments below. When revising your paper please consider these comments.   
Your resubmission is due within 60 days. When resubmitting your paper, we require that you also submit an itemized list of responses to each point the referee (or Editor) has made in the report. This helps us assess your revision faster.   
When submitting your revised manuscript, please ensure that you upload the source files (e.g. Word) to expedite the typesetting process should your paper be accepted for publication.   
  
Editor's/Reviewers' comments:  
  
Reviewer #1: The authors present - and make available - a number of computational tools for interpolating grain boundary properties throughout the full five-dimensional space of grain boundary parameters. There are two main contributions - an algorithm to approximate a fundamental zone and an algorithm to approximate the distance between two points in the five-dimensional space. Overall, it is a good contribution and people working in this domain may find these tools useful. I have a few technical comments that might improve the paper/presentation.  
  
1. p. 4, o\_ref cannot be selected to coincide with a symmetry operator. This is brought up later, but is important enough to be mentioned immediately.

We changed “arbitrary” to “random” and added a sentence describing the coincidence with symmetry operators immediately upon introduction of o\_ref.

2. In Fig. 6, for the arc AB, what are the end points crystallographically? In other words, what is the axis or misorienation, angle, and plane normal? This would make it a little more concrete.

Tables with approximate misorientation quaternion / BP normal pairs have been added to the supplementary information and referenced in the main text. Also, disorientations have been plotted in Rodrigues space in supplementary information.  
  
3. To make it less abstract, it would be useful to plot some of the properties in a crystallographic reference frame. For example, the energies of Fe or Ni GBs as a function of grain boundary plane normal for one or more fixed misorientations.

We agree that this will help make the work more concrete, and we plan to address this thoroughly in a follow-up manuscript. Nonetheless, a figure for Ni GBEs for a fixed, Sigma 3 / low GBE misorientation (varying BP normal) has been added.  
  
4. A lot of text is spent comparing various interpolation methods, of which GPR is superior in most ways. Personally, I would be satisfied if the authors said, we tried the following methods (see comparisons in the supplemental information) and GPR was the best. This would shorten the manuscript and make it more accessible.

We appreciate and have considered this suggestion. In order to preserve some computational aspects of our approach (consideration of various machine learning algorithms, comparison of trade-offs between accuracy, runtime, and dataset size), we have decided to leave the other three algorithms (NN, barycentric, and IDW) intact in the manuscript.  
  
5. The conclusion recounts what was done and much of the details is repetitive. I recommend distilling this down to the few most important conclusions of the research.

We shortened the conclusion and condensed the bulleted list of future work topics.  
  
Typographic errors  
p. 3 line 29: "Section S0.2" not a section.

This has been corrected to Section S2 (and corrected in the supplementary information as well).

p. 4 line 11: (k-the) -> k^th  
  
This has been corrected.  
  
Reviewer #2: The authors present a novel method of discretizing the five parameter space (described here as the S7 octonian space) that describes grain boundary properties by implementing a Voronoi fundamental zone framework. The work is extremely thorough description of the methods, usage, advantages and disadvantages, and it should have a significant impact on the field. The inclusion of readily available software packages is to be highly commended as well. There are relatively few minor optional comments that I would suggest before publication:  
  
1 - The authors use a significant number of abbreviations and acronyms. If within the bounds of the journal format, I would recommend inclusion of a table at the begging of the article of "Abbreviations used in this article". If this does not fit within the format of the journal to include within the body of the article, this would be a very valuable appendix.

The minimum required frequency for a phrase to be displayed with acronym formatting has been increased to reduce the number of acronyms. The glossary at the end of the article has been renamed to “List of Acronyms”, and to improve visibility, has been moved to directly after the Conclusions section.  
  
While this is a stylistic choice, I would recommend redefining any acronyms from the abstract in the main body of the article. It does not seem appropriate to have to reference the abstract for the definition.

Acronyms that were defined in the abstract have been redefined in the main body of the article.  
  
2 - The numerical method of constructing the VFZ has a significant appeal in its numerical simplicity, especially in the context of computations. However, the authors spend relatively little time explaining a choice of the o\_{ref} within the octonion space. The reviewer understands that the point only needs to avoid any of the FZ boundaries, but given the relative novelty of the octonion space, these may not be obvious. Providing a reasonable could be quite helpful (and if understood correctly, a wise choice for O\_h would be applicable to all symmetry systems?)

The supplementary information has been updated with an additional section (Choice of Reference GBO), two tables, and a figure to provide more context. The tables and section are now referenced in the main text under “Defining the VFZ”.  
  
3 - The authors claim that they are using an "active" convention for this work. This is an unfortunate nomenclature that the reviewer suggests should be reconsidered. Commonly, active rotation operations are considered those that rotate an object, while passive operations rotate reference frames.   
The original work referenced (Ref 75) regarding the active and passive notation was doing this from a point of confusion of some that worked with various angle/axis representations, wherein many forgot that there was an implicit expectation that the rotation operations should be describing passive operator.   
  
I think the authors would agree that the symmetry operators, or the quaternion/octonion operators within this work are not changing the actual directions of the octonions, only the reference frame that they are expressed within. Thus is it not appropriate to refer to their convention as being "active".   
  
Where the authors differ from previous work (Ref 55 & 75 in particular) is their use of the crystal frame as being the reference frame for the quaternions and octonions descriptions, while many others use the sample frame as the reference. Indeed in my own work, I use the same representation of the quaternion operator listed in Appendix A (eq. 5), with P = +1, but would define the misorientation between two crystals (Eq. 4) as q\_m = q\_A {q\_B}^{-1}.   
  
It should be acknowledged that setting P=-1 when using the authors code base would make it compatible with reversing the choice of reference frame, but this should not be confused with the "active" and "passive" nomenclature. Thus if someone that uses the sample frame as reference was going to use the authors code-base, they would keep their own operators working with P=+1, but would use P=-1 when using the author's programs. Both sets of code agree that the operators are passive, they disagree on the reference.   
  
The reviewer proposes changing the wording in the article to annotate the assumed reference frame for the quaternion/octonion operations, and use Appendix A to suggest how the P term in eq 5 can be used to easily reverse the reference frame choice (a fortuitous by-product of the use of the P parameter).

Where appropriate, references directly to the two equations in the appendix are given explicitly. Appendix A has been renamed to “Rotation Conventions”, and we describe that P can be negated to flip the choice of rotation convention. There is a reference to Rowenhorst 2015 for further information.  
  
Editor note: There is a previous publication that may be relevant to this work. It is: D. Olmsted, Acta Mater., 57, 2793 (2009).  
  
We agree this is a relevant prior work. A reference to *D. Olmsted, Acta Mater., 57, 2793 (2009)* was added in the text when discussing a typical correlation length of 10 degrees. Additionally, this work is already referenced indirectly via:

Morawiec, A. On Distances between Grain Interfaces in Macroscopic Parameter Space. *Acta Materialia* **2019**, *181*, 399–407. <https://doi.org/10.1016/j.actamat.2019.09.032>.

which compares many different distance metrics.