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.   
  
To submit a revision, please go to <https://www.editorialmanager.com/commat/>   
Forgotten your password?  
Select "Login" from the top menu and click 'Forget your Password?' at the bottom of the next screen.   
Click on "Submissions Needing Revision" to find your submission.   
  
  
NOTE: Upon submitting your revised manuscript, please upload the source files for your article. For additional details regarding acceptable file formats, please refer to the Guide for Authors at: <http://www.elsevier.com/journals/computational-materials-science/0927-0256/guide-for-authors>  
  
When submitting your revised paper, we ask that you include the following items:  
  
Manuscript and Figure Source Files (mandatory)  
  
We cannot accommodate PDF manuscript files for production purposes. We also ask that when submitting your revision you follow the journal formatting guidelines. Figures and tables may be embedded within the source file for the submission as long as they are of sufficient resolution for Production. For any figure that cannot be embedded within the source file (such as \*.PSD Photoshop files), the original figure needs to be uploaded separately.Refer to the Guide for Authors for additional information.  
<http://www.elsevier.com/journals/computational-materials-science/0927-0256/guide-for-authors>  
  
Highlights (not mandatory if Graphical Abstract submitted)  
  
Highlights consist of a short collection of bullet points that convey the core findings of the article and should be submitted in a separate file in the online submission system. Please use 'Highlights' in the file name and include 3 to 5 bullet points (maximum 85 characters, including spaces, per bullet point). See the following website for more information   
<http://www.elsevier.com/highlights>  
  
Graphical Abstract (mandatory)  
  
Graphical Abstracts should summarize the contents of the article in a concise, pictorial form designed to capture the attention of a wide readership online.   
Refer to the following website for more information:   
<http://www.elsevier.com/graphicalabstracts>   
  
If you do not wish to upload a Graphical Abstract, then submit a Word or other text format file at the prompt for the graphical abstract that contains the statement "no graphical abstract". In this way, the EM system will allow you to complete the submission but you will not be obliged to provide this content.  
  
Please note that we allow 60 days for author revisions.  
  
Please note: While submitting the revised manuscript, please double check the author names provided in the submission so that authorship related changes are made in the revision stage. If your manuscript is accepted, any authorship change will involve approval from co-authors and respective editor handling the submission and this may cause a significant delay in publishing your manuscript.  
  
NOTE: Upon submitting your revised manuscript, please upload the source files for your article. We cannot accommodate PDF manuscript files for production purposes. We also ask that when submitting your revision, you follow the journal formatting guidelines. For additional details regarding acceptable file formats, please refer to the Guide for Authors at: <https://www.elsevier.com/journals/computational-materials-science/0927-0256/guide-for-authors>  
  
Include interactive data visualizations in your publication and let your readers interact and engage more closely with your research. Follow the instructions here: <https://www.elsevier.com/authors/author-services/data-visualization> to find out about available data visualization options and how to include them with your article.  
  
Yours sincerely,  
  
Jeffrey Hoyt, Ph.D.  
Associate Editor  
Computational Materials Science  
  
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.  
  
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).

Reference to “active” or “passive” convention has been removed, and 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 using P=-1 changes the reference frame. 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. This work is 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. However, a reference to *D. Olmsted, Acta Mater., 57, 2793 (2009)* was added in the text when discussing a typical correlation length of 10 degrees.

Data in Brief (optional):   
  
We invite you to convert your supplementary data (or a part of it) into an additional journal publication in Data in Brief, a multi-disciplinary open access journal. Data in Brief articles are a fantastic way to describe supplementary data and associated metadata, or full raw datasets deposited in an external   
  
repository, which are otherwise unnoticed. A Data in Brief article (which will be reviewed, formatted, indexed, and given a DOI) will make your data easier to find, reproduce, and cite.   
  
  
You can submit to Data in Brief when you upload your revised manuscript. To do so, complete the template and follow the co-submission instructions found here: [www.elsevier.com/dib-template](http://www.elsevier.com/dib-template). If your manuscript is accepted, your Data in Brief submission will automatically be transferred to Data in Brief for editorial review and publication.   
  
  
Please note: an open access Article Publication Charge (APC) is payable by the author or research funder to cover the costs associated with publication in Data in Brief and ensure your data article is immediately and permanently free to access by all. For the current APC see: [www.elsevier.com/journals/data-in-brief/2352-3409/open-access-journal](http://www.elsevier.com/journals/data-in-brief/2352-3409/open-access-journal)   
  
Please contact the Data in Brief editorial office at [dib-me@elsevier.com](mailto:dib-me@elsevier.com) or visit the Data in Brief homepage ([www.journals.elsevier.com/data-in-brief/](http://www.journals.elsevier.com/data-in-brief/)) if you have questions or need further information.