Dear Editor,

Included in this correspondence is a direct response to all of the points raised by referees. Referee 1 provided an overall endorsement of our paper, and did not request that any changes be made. Referee 2 posed several questions and made recommendations for ways to improve our paper. Ultimately, he/she stated that our work, pending some adjustments, is appropriate for publication in Philosophical Magazine. In addition to the thorough write up that we provided Referee 2 with, we will summarize the points that he/she brought up and our responses to them:

1) Are the phenomena associated with TD universal across supercooled liquids?

* While we do provide the readers with a reference to another study that found similar behaviors in a Zr-based liquid near TD ~= 1.4xTg, we cannot speak to whether or not this is a universal characteristic temperature without conducting extensive simulations of other liquids.

2) Are the timescales at TA and TD sufficiently short so that the simulation is revealing the physics that would arise on a laboratory timescale?

* Yes, we provide a rather detailed discussion of the liquid relaxation timescale and simulation timescale on page 15, which concludes that our results do indeed reflect the behavior of the supercooled liquid that is still in metastable equilibrium (i.e. as it behaves on a laboratory timescale).

3) Clarify what is meant when it is said that TD signals higher-order cooperative rearrangements amongst connected icosahedra.

* We modified both the abstract and conclusion to make clear to the reader that the higher order cooperation that we speak of is the joining of already-formed icosahedra to create larger connected domains of icosahedra that become macroscopically populated as one cools through TD.

4) Explain what is meant by percolation. Ref. 33 seems to suggest that the percolation of the icosahedron network can occur at much higher temperatures than what is indicated in this paper.

* We added an explicit definition of network percolation to page 15. The referee misunderstood the claim being made in Ref. 33. The results of Ref. 33 agrees with our own: the percolation of the icosahedron network occurs just above Tg, near 825K. We provided Referee 2 with a clear discussion of this matter.

5) What are the roles played by the different types of connections formed between icosahedra in the dynamics of the system?

* Some of the authors from this work have previously investigated this matter (Ref. 35) and found that it fell outside the scope of this paper as it is more relevant to the mechanical properties of the glass. We did, however, add a sentence to page 15 that points the reader to two sources on the subject.

6) Can you speak to the relationship, if any, between dynamical and structural correlation lengths in this system?

* We added a discussion to page 13, which points the reader to Ref. 47 and explains that clear correspondence between dynamical and structural length scales, in the context of icosahedral ordering, has been demonstrated in their work.

We believe that we have thoroughly and constructively addressed all of the points raised by Referee 2. It is our hope that, given these revisions, our manuscript can now be published in Philosophical Magazine. Thank you for your consideration.

Sincerely,

Ryan Soklaski, Vy Tran, Zohar Nussinov, Kenneth Kelton, and Li Yang