**Response to the referees**

In the following, we provide in italics the comments made by the referees, and address them in the order in which they were raised

**Referee 1**

*The material is presented clearly, and while one may wonder how general the phenomena are, such further investigations are likely to enhance the citations of this paper. This is of definite interest to the community, and I recommend publication.*

We appreciate Referee 1’s positive feedback and overall endorsement of our paper.

**Referee 2**

We appreciate Referee 2’s careful review of our paper, and his/her insightful questions. We are happy to provide a point-by-point reply, as requested.

*1 ) They state that TA~2Tg is universal for all liquids. TD~1.4 Tg may be universal as well? If so, perhaps they could add a comment as to why all liquids behave so similarly? If the structural indicator identified here works for Cu64Zr36, what happens to a stronger liquid? In that case the*

*structural evolution would be different. As an example, at some alloy compositions there may be no dominant motif such as the icosahedra discussed here[...]*

The idea that TD may also be a universal characteristic temperature of supercooled liquids is certainly enticing. Towards this end, on page 8 in the second paragraph, we cite reference [46], which identified a similar phenomenon in a Zr-based liquid near TD. Additionally, we have conducted a preliminary analysis of in-house experimental data for liquids, including strong ones, and have found indications that TD is a crossover temperature for a wide range of liquids. That being said, we cannot yet claim that TD is universal. We would first have to conduct detailed structural studies, similar to this paper, of other liquids to understand what occurs when there is not an obvious dominant structural motif. We suspect that there are in fact structural motifs at play, but that these are not easily detected by local structural metrics (e.g. Voronoi analysis or Honeycutt-Andersen indexes). This is certainly a topic of future work for us and for our readers.

*2) In experiments, the time scale is orders of magnitude longer than the MD one in the current paper. Ordering and connection of motifs have much more time to develop. I guess they assume that all these key actions still commence at 2Tg or 1.4Tg, because the shift of their onset in T is minor (and they use ~2Tg and ~1.4Tg, with approximate sign anyway)? A comment in the discussion may help the readers.*

As pointed out by the referee, it is imperative to consider the role of timescales when assessing MD simulations of liquids. We found that the liquid relaxation timescales at both TA and TD are sufficiently short, relative to our overall simulation timescale, so that our results reflect the metastable equilibrium behavior that one would observe on laboratory timescales. We provide a discussion of this matter in the section of our paper named “FLD Fluctuations and Falling Out of Equilibrium”

Specifically, in the first paragraph of page 15, we explain that at 900K, which is below TA and TD, our simulation timescale is still four orders of magnitude larger than the liquid relaxation time. Thus we are confident that the structural motifs that we find are a manifestation of a metastable liquid that is thoroughly exploring phase space. Additionally, we provide the reader with a discussion, and relevant references, of the effects of thermalization and slower quench rates in our system, all of which support our conclusions. Ultimately, we conclude that TA and TD reside at sufficiently high temperatures such that our results would also manifest on laboratory timescales.

*3) The authors argue that TD signals “higher-order cooperative rearrangements amongst connected icosahedra”. This description is a bit too vague. Could they make it more transparent to the readers, as to exactly what the critical event is (including in the abstract and/or in the conclusion)?*

We have revised our abstract and conclusion to emphasize that the higher order cooperativity that we speak of is: the formation of connected domains of icosahedra that are so large that they possess a macroscopic number of the liquid’s icosahedra. The formation of these extensive, Zr-rich structures at TD causes the decoupling of diffusivities of Cu and Zr atoms.

*4) Explain “eventually percolate before the glass is formed”, and define “percolate” for the readers, so they know how to picture the evolution*

We have added the following sentence to the end of the first paragraph of page 15, to clarify the meaning of percolation to the reader:

“A structure is said to percolate a material when a singly-connected domain of that structure spans the material (e.g. a connected domain of icosahedra is as long as the diagonal length of our simulation box).”

Furthermore, we provide the citation Ref. 35, which explicitly shows that the icosahedron network in supercooled Cu64Zr36 percolates the system near 800K.

*(continued) Don’t they percolate well before Tg? Shouldn’t the connected icosahedra already percolate through the box at ~TD, and then further permeate the entire system approaching Tg? For example, in Ref. 33 at the same Cu64Zr36 composition, even for the demanding connection scheme of interpenetrating icosahedra, their percolation was reported to onset at certain icosahedra population threshold: when the fraction of Cu in icosahedra goes above 26%.*

Supercooled Cu64Zr36 only exhibits a percolated icosahedron network just above Tg and below. Ref. 33 actually conducts its percolation analysis at 800K, which is significantly below TD and is about 50K above Tg. They show that interpenetrating icosahedra percolate the system when 26% of the system’s Cu atoms comprise the **center atoms** of the icosahedron network (last sentence of page 349 of Ref. 33). This indicates that far more than 26% of Cu will be involved in the overall network as vertex atoms of the icosahedra and that a tremendous amount of icosahedral ordering has developed in the system - far more than what we see at TD.

*5) Also, Ref. 33 reports that face-sharing icosahedra increase at the expense of edge sharing ones, along with cooling. This was related to a reduction in configuration potential energy (CPE). The current paper seems to lump all connection schemes together. Would the specific connection types, in addition to “connected or not”, play a role in dynamics? Some comments may also be useful to guide the next step in research.*

The roles played by the different types of connections between icosahedra do seem to be important. Indeed, in Ref. 35, we pay special attention to non-interpenetrating connections, whereas, seemingly out of convenience, many other studies only consider interpenetrating connections. While these considerations are important, they do take us out of the scope of this paper and towards the mechanical properties of a glass. We have added a sentence to point our readers to further reading on this topic:

“Not discussed in this paper are the roles played by the various types of connections that can form between icosahedra. For detailed discussions of this important aspect of icosahedral ordering, we refer the reader to early studies of this subject [33, 35].”

*6) I like their approach of analyzing how the dynamical slowdown is affected by the connection of SRO at longer lengthscales. But perhaps they could better summarize for us (or make a comment on) the increase of the dynamic and structural correlation lengths with undercooling, comparing one with the other (would the two correspond to one another, e.g., they are on similar length scales)?*

The relationship between dynamical and structural length scales is an important point that we failed to make sufficiently explicit in the previous draft of our paper. We modified the following discussion in the last paragraph of page 13:

“Y. Zhang et al. recently showed quite explicitly that diffusing atoms strongly avoid regions of icosahedral ordering, and that they instead reside in liquidlike regions of the system [47]. They demonstrate that underlying the development of medium-ranged icosahedral ordering and glassy dynamics is an important correspondence between dynamical and structural correlation length scales in the liquid.”