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Dear Victor Vakaryuk:

Thank your for organizing the third round of review of our manuscript BH12397, “Thermal Conductivity Accumulation in Amorphous Silica and Amorphous Silicon.”

We are pleased that the Third Referee recommends publication. Our replies to the Third Referee’s comments are included in the attached rebuttal.

We look forward to your response.

Sincerely,

Alan McGaughey

Response to the First Report of the Third Referee

Modified text throughout the manuscript is highlighted in red.

1) The chosen density of the a-SiO2 models (2.35 g/cc) is relatively far from the standard experimental density of silica (~2.2 g/cc): this may have a consequence on the structure and most likely on the speed of sound. This choice should be justified and the structure of the a-SiO2 models should be described in more details, especially concerning the presence of coordination defects.

We added a comment to Section III A concerning the density of a-SiO_2:

"The simulated density of 2350 kg/m^3, which corresponds to zero pressure, is 7% larger than the experimental value of 2220 kg/m^3 [52]. At the experimental density, the BKS potential generates a structure in tension at a pressure of 2-3 GPa [47]. We use a density of 2350 kg/m^3 to be consistent with previous simulations."

A comment concerning the effect of the density on the sound speed is added to Section IV C:

"The transverse sound speed predicted for our model of a-SiO2 is 85% of that predicted by the other methods (Table I) and that measured by experiment [63,67,70], which is likely related to the smaller density of our samples (see Section III A)."

The radial distribution function [Fig. 3 (a)] describes the bonding environment of our model of a-SiO_2 and compares well with experiment. As for the presence of coordination defects, as reported by McGaughey and Kaviany for these structures [Table 2 in Ref. 47] at least 99.5% of the atomic coordinations are at the expected values (4 for silicon and 2 for oxygen). A comment is added to Section III A:

"At least 99.5% of the atomic coordinations are at the expected values (4 for silicon and 2 for oxygen) [47]."

2) I have a problem with Fig. 3: The calculation of the dispersion curves of a-SiO2 does not seem to make much sense beyond 0.5 wavevector, as the S(omega,k) curves are very broad.

We agree that the "goodness of fit" (coefficient of determination) for the a-SiO_2 structure factors above a normalized wave vector of 0.2 is low. We commented directly about this point in Section IV B:

“For a-SiO2, the coefficients of determination are greater than 0.8 for |κ|/κmax ≤ 0.2 and less than 0.7 for |κ|/κmax > 0.2, where the structure factors peaks are less than an order of magnitude larger than the background.”

We report the errors explicitly in the text and represent them graphically in Fig. 3 so that the reader is not misled. We feel that it is useful to plot the (pseudo) dispersion curve for a-SiO_2 above a wave vector of 0.2 to compare with previous experimental [18,26,28,63,67] and numerical [41,42,64] results, as well as to compare with our a-Si data.

3) The choice of using the speed of sound obtained from the DOS looks rather arbitrary, as it does not properly take into account longitudinal modes as propagating modes. This choice probably affects the estimate of the thermal conductivity of a-Si (where propagating modes count most), and may explain why the agreement between eq. (1) and Green Kubo calculations (Fig. 6b) is rather poor.

We agree that there is some arbitrariness in the choice of the sound speed and believe that we addressed this comment honestly in the last paragraph of Section IV C. We argued that using a single polarization with mode properties dominated by transverse values will maximize the contribution to thermal conductivity from propagating modes. Because the predicted k_{GK} for a-Si is lower than k_{vib} (but still within the uncertainty), adjusting the propagating mode properties towards longitudinal values would only make the agreement with k_{GK} worse. As such, we do not feel that the use of Eq. (2) with a single polarization is the cause for the difference between k_{vib} and k_{GK}.

4) In the last paragraph of the discussion about lifetimes a comparison is made with the results in Ref. 9. It is shown that discrepancies between the present and the former results do not come from the use of different empirical potentials. It should also be mentioned that samples are generated in different ways. In this work WWW a-Si models are used, while in Ref. 9 models were generated by quenching from liquid Si, and therefore may display significant structural differences. It is worth pointing out that also the converged bulk value obtained in this work using MD and the Green Kubo method (2.1 W/m/K) differs significantly from the one in Ref. 9 (3 W/m/K), probably due to large differences between the models.

We thank the referee for these good points. We added these comments to the end of Section IV D:

There are several issues to consider when comparing our results to those of He et al.[9] The a-Si models in Ref. 9 were prepared using a melt-quench technique that may lead to structural differences. When we applied the Tersoff potential (as used by He et al.) to the WWW a-Si models, we predict similar lifetimes

to those from the SW potential. Furthermore, in Ref. 9

the NMD analysis was performed in the time domain, where the effects of metastability can be more strongly pronounced. Finally, we note that the a-Si bulk thermal conductivity predicted by He et al. using the Green-Kubo method is 40% larger than our prediction (3 W/m-K vs. 2.1 W.m-K).

5) I do not see the point of defining a "diffusion mean free path" as in eq. 22. It is not physically meaningful, as the physical interpretation of "diffusions" does not imply the concept of a propagating wave.

The mean free path definition is only physically meaningful for low-frequency diffusons, which may be “marginally propagating” (Refs. 4 and 6). By using Eq. (22), which has been used in previous studies (Refs. 4 and 6) to estimate MFPs of diffusons and to demonstrate marginal propagation [4,6], we find that the lowest frequency modes in our models have diffuson MFPs between the lattice constant and the supercell size. This finding supports the notion that the lowest-frequency modes in our systems are marginally propagating.

6) In page 6, Voigt should be written without "h"!

Thanks for pointing out this error, which has been corrected.