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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**

*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-SiO2. The modified text is highlighted in red. As for the presence of coordination defects, …. The radial distribution function [Fig. 3 (a)] describes the bonding environment of our model of a-SiO2 and compares well with experiment.

*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-SiO2 structure factors above a normalized wavevector of 0.2 is low. We commented directly about this point in Section IV B. We report the errors explicitly in the text and represent them graphically in Fig. 3 so that the reader is not mislead. We feel that it is useful to plot the (pseudo) dispersion curve for a-SiO2 above a wavevector of 0.5 to compare with previous experimental [18,26,28,63,67] and numerical results [41,42,64], 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 kGK for a-Si is lower than kvib (but still within the uncertainty), adjusting the propagating mode properties towards longitudinal values would only make the agreement with kGK worse. As such, we do not feel that the use of Eq. (2) with a single polarization is the cause for the difference between kvib and kGK.

*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 comments to the end of Section IV D that reiterate all these points.

*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 definition is only physically meaningful for low-frequency diffusons, which may be “marginally propagating.” By using Eq. (22), we note 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”. Please note that Eq. (22) has been used in previous studies to estimate MFPs of diffusons to demonstrate marginal propagation [4,6].

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

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