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Dear *Physical Review B* Editor:

We are submitting the manuscript titled “Thermal Conductivity Accumulation in Amorphous Materials” by Jason M. Larkin and Alan J. H. McGaughey for consideration for publication in *Physical Review B*.

The objective of this study is to predict the thermal conductivity accumulation functions for amorphous silica (a-SiO_2) and amorphous silicon (a-Si). The accumulation function describes the contribution of vibrational modes with different mean free paths to the total thermal conductivity. For amorphous materials, it provides critical information about the relative contributions of propagating and non-propagating modes, an area of active debate for more than twenty years [*PRB* **42** (1990) 3, *PRB* **48** (1993) 17, *PRB* **50** (1994) 9, *PRB* **59** (1999) 5, *PRB* **77** (2008) 21, *PRL* **102** (2009) 3, *PRB* **81** (2010) 10].

Our work is timely because the development of the frequency-domain thermoreflectance technique by Regner et al. [*Nat. Comm.* **4** (2013) 1640] has provided an unprecedented resolution for the measurement of accumulation functions, with excellent agreement found for crystalline silicon. An intriguing result from Regner et al. is the measurement of accumulation functions for a-SiO_2 and a-Si. While a-SIO_2 has a flat accumulation at the bulk value, indicating an absence of propagating modes, a-Si shows a large accumulation up to mean free paths of hundreds of nanometers, indicating the presence of phonon-like modes. Even though a-SiO2 and a-Si are widely studied, data is not currently available to build their accumulation functions to allow for comparison to Regner et al.’s data and an elucidation of the underlying thermal transport physics.

Our study combines molecular dynamics simulations, lattice dynamics calculations, and theoretical modeling to predict the accumulation functions for a-SiO_2 and a-Si. For a-SiO2, we find good agreement in terms of magnitude and trends with the data of Regner et al. For a-Si, the predicted and measured thermal conductivity magnitudes are comparable, but the range of the mean free paths over which the accumulation occurs is different. This discrepancy points to important questions as to how to interpret the experimental accumulation functions and the need for

further studies of a-Si, which has processing dependent structure and thermal conductivity.

We suggest the following reviewers:

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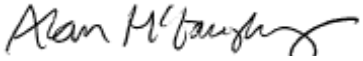
We look forward to your response.

Sincerely,



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