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Dear *Journal of Physics: Condensed Matter* Editor,

Please find attached the manuscript “Comparison and evaluation of spectral energy methods for predicting phonon properties” by Larkin et al., which we are submitting for review for *Journal of Physics: Condensed Matter*.

This manuscript presents a detailed derivation, comparison, and evaluation of two methods for extracting phonon properties, notably lifetimes, from molecular dynamics simulations. Phonon properties are needed to predict the thermal conductivities of bulk and nanostructured materials, a property critical to evaluating the performance of new materials for use in electronic, opto-electronic, and energy related systems. While lattice dynamics-based methods can be used to access low temperatures, molecular dynamics-based methods are needed at high temperatures where higher orders of anharmonicity are important.

The significant contributions of our manuscript are:

1. The formal derivation of the Φ method for predicting phonon properties (a frequency-domain approach that is rigorous and correct). This new formulation has not been published and has advantages over a similar time-domain approach described previously.
2. The demonstration that the Φ’ method for predicting phonon lifetimes (which was previously proposed but never comprehensively validated) is incorrect, while it has been (and continues to be) used in research articles. The Φ’ method can, however, be used to predict phonon frequencies in systems that are perturbed from a perfect periodicity.

Please let me know if you require any more information.

Regards,

Jason M. Larkin

Joseph E. Turney

Alex D. Massicotte

Cristina H. Amon

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