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Dear *Journal of Applied Physics* Editor:

We are submitting the manuscript titled “Predicting alloy vibrational mode properties using lattice dynamics calculations, molecular dynamics simulations, and the virtual crystal approximation” by Jason M. Larkin and Alan J. H. McGaughey for consideration for publication in *Journal of Applied Physics*.

Due to their low thermal conductivities, alloys are currently an active area of research, notably in the thermoelectric energy conversion field. The ability to predict alloy thermal conductivity is critical in narrowing down a large materials design space. Recent papers [e.g., *PRL* **106**, 045901 (2011), *PRL* **109**, 095901 (2012), *PRB* **85**, 184303 (2012)] have used anharmonic lattice dynamics and the virtual crystal approximation (an approach we call VC-ALD) to make such predictions.

In VC-ALD, the disorder in the alloy is treated as a perturbation. The limits of this approach have yet to be determined. In our work, we use computationally-inexpensive empirical potentials to assess the virtual crystal approximation by self-consistently treating the disorder explicitly and as a perturbation. We are not aware of any such previous study. Our results indicate that while VC-ALD is generally an accurate method, care must be taken when modeling alloys with low thermal conductivities, where significant underprediction of thermal conductivity is likely.

We suggest the following reviewers:

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We look forward to your response.

Sincerely,

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