Comparison and evaluation of spectral energy methods for predicting phonon properties

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The authors compare two formal definitions of the phonons' spectral function in the following way. They carry out numerical simulations (at relatively high temperatures, i.e., in the classical regime) of finite systems in which they solve the equations of motions (molecular dynamics). They then fit the results to the formal definitions, and find the frequencies and the life times of the vibrations that fit best the numerical data. Finally, they use those to compute the thermal conductivity of certain systems.

Whereas such a study may have a limited interest, I think that the paper in its present form is not appropriate for publication. First, the manuscript is far too long; it contains repetitions, for example, Eqs. (4-6) appear also in Appendix A) and it repeats basic textbooks expressions [Eqs. (1-3)]. Second, one wonders why the two formal expressions, denoted as Φ and Φ', are not compared theoretically, by introducing into the second the normal-mode expansions, for example; Third, the comparison is between two different sets (for each system) of numerical results obtained for small systems, and one wonders what are the computational errors in each of them, and how does the numerical protocol affect the comparison; and last but not least, the vast use of the term 'predict' leads the reader to believe that the predictive power of Φ and Φ' will be examined by comparing the results with those measured on real systems; however, it seems that the term predict serves as a synonym to values extracted by fitting.

In summary, it is not at all clear why do the authors prefer one definition over the other, and what is the actual difference of the two–from the theoretical point of view.