January 20, 2015

Re: BZ12352
Nonlocal thermal conductivity by nonequilibrium molecular dynamics
by Philip B. Allen and Yerong Li

Dear Dr. Allen,

The manuscript described here has been reviewed by two of our referees. Comments from the reports are enclosed.

To be publishable in the Physical Review, manuscripts must be technically correct, must contain significant new physics or understanding, be of high quality and scientific interest, and be recognized as an important contribution to the literature.

The comments of the referees suggest that the present manuscript does not meet the above criteria and thus is not suitable for publication in the Physical Review.

We would be willing to reconsider a paper revised along the lines suggested by the second referee.

Yours sincerely,

Anthony M. Begley
Managing Editor
Physical Review B
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Report of the First Referee -- BZ12352/Allen

The manuscript by B. Allen and Yerong Li with title “Nonlocal thermal conductivity by nonequilibrium molecular dynamics” proposes a new methodology for NEMD, using a sinusoidal pattern for extracting heat instead of the classical hot and cold thermostats. With this new method, the convergence time is getting shorter. The article is very technical and does not reveal new physics.

I regret not to be able recommend this article for publication in Physical Review B, as I believe that it would be much more appropriate in a computational journal.

Report of the Second Referee -- BZ12352/Allen

In this work, the authors discuss the calculation of thermal conductivities via Non-Equilibrium Molecular Dynamics (NEMD), i.e., by discussing a new variant of NEMD simulations. Here, the authors first discuss the typical NEMD approach that involves a constant temperature gradient across the cell induced by keeping two distant slabs of the supercell at a different, but constant temperature. Second, they introduce their own approach, which involves adding/subtracting heat in a sinusoidal pattern across the whole supercell. Third, the developed technique is applied to study thermal conductivity in the Lennard-Jones (LJ) liquid and crystal and fourth, an analytical formula based on the Boltzmann-Transport-Equation (BTE) is derived that facilitates the extrapolation to the bulk limit.

In principle, the work is interesting and the derivations are detailed and insightful. However, the work lacks some depth and truly novel aspects:

(a) Using the NEMD approach in classical MD simulations with and without periodic boundary conditions along the "long" part of the supercell has been done for decades – and many different variants of NEMD exist to aid and accelerate convergence with system size. None of these are discussed in the paper and compared to the proposed approach. To my knowledge, the introduced formulas for the Fourier representation of the thermal conductivity (here and in Ref. 3) are indeed new, although similar concepts are used in experimental measurements of the thermal conductivity (grating).

(b) The proposed NEMD variant using a sinusoidal heat pattern is indeed ingenious. I really appreciated the detailed derivation and the thoughtful description of the algorithm. However, the same senior author already published many crucial aspects and ideas that form the core of the proposed technique in Ref. 3. Again, I miss the link to existing literature, for instance Eur. Phys. J. B 87, 96 (2014).

(c) Although a nice proof of concept, the simulations of the LJ liquid and crystal at one single temperature do not provide enough insights to really judge the usefulness and accuracy of the proposed technique. As a matter of fact, LJ based NEMD simulations can be converged in time and size even by brute force on standard computational equipment. Here, it would be very useful to compare the performance of the proposed NEMD for more challenging systems, e.g., silicon [Phys. Chem. Chem. Phys. 14, 16209 (2012)] or even graphene [Ref. 23, 24, and Nat. Comm. 5, 3689 (2014)]. At least, one system that features also optical modes should be included as well.

(d) Again, I enjoyed the detailed and insightful derivation of the extrapolation technique derived in the last part of the work. The found divergence arising in anisotropic cells is very interesting and constitutes an important insight. However, the concepts of the presented formalism are again covered to a large extent already in Ref. 3 by the same senior author. Again, the model should at least be extended to include the influence of (slightly dispersed) optical modes.

Although pleasant to read and generally compelling, I cannot support a

publication of this manuscript in PRB at this stage, since it contains too few novel aspects. The core concepts of this work are already presented by the same senior author in Ref. 3. The numerical simulations performed to substantiate these core concepts are a nice proof of concept, but too few to allow for a detailed discussion and judgment of the presented algorithm. I suggest that the authors extend their formalism both analytically and numerically, i.e., for instance by including also optical modes in their considerations, by performing simulations for different, more challenging systems, and by systematically comparing both the accuracy and the performance of their algorithm with other NEMD (or even equilibrium MD) techniques. In that case, I would support publication in PRB.