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  
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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.  
  
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Report of the Second Referee -- BZ12352/Allen  
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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.