Comment on “Theoretical Paradigm for Thermal Rectification via Phonon Filtering and Spectral Confinement”

A. A. Maznev and Bai Song

Phys. Rev. Lett. **128**, 129601 — Published 21 March 2022
DOI: 10.1103/PhysRevLett.128.129601

Comment on "Theoretical Paradigm for Thermal Rectification via Phonon Filtering and Spectral Confinement"

In a recently published Letter [1], Donovan and Warzoha suggest that a layered structure consisting of diamond layers of different thicknesses yields a significant "thermal rectification" effect (i.e., the magnitude of the heat flux changes when the temperature difference across the structure switches sign). The proposed mechanism based on filtering out long-wavelength acoustic phonons by a thin layer goes against an established notion that thermal rectification requires both a spatial asymmetry and a nonlinearity [2]. We believe that the results of Ref. [1] are incorrect and, moreover, that "phonon filtering" in principle cannot result in thermal rectification.

If no anharmonic interactions occur, as assumed in Ref. [1], the analysis of thermal transport must be done separately for each phonon frequency (then the total heat flux can be calculated by adding up frequency-specific contributions). If a given phonon frequency is blocked by one of the layers, this frequency cannot contribute to the heat transport across the entire structure, no matter what the sign of the temperature difference is. The phonons that actually carry heat across the structure will not be affected by the blocked frequency because there is no interaction between phonons of different frequencies. Thus, blocking part of the phonon spectrum cannot possibly cause any asymmetry in the heat flux when the sign of the temperature difference is flipped.

The structures proposed in Ref. [1] belong to a broader class of structures for which the absence of thermal rectification has already been shown [3]. If anharmonic interactions are absent, a phonon of a given frequency emitted from one side of the structure will be either eventually transmitted and absorbed at the opposite side or backscattered and reabsorbed at the same side; heat transport across the structure can then be described in terms of phonon transmission (similarly to the Landauer approach in electronic transport). Within this framework, Ref. [3] has shown that as long as the second law of thermodynamics holds, the thermal rectification effect cannot arise.

The incorrect result of Ref. [1] stems from several methodological mistakes. (i) Thermal conductivity is treated as a local quantity as if the Fourier law were still locally valid. However, when anharmonic phonon interactions are absent, local thermal equilibrium cannot be established, and local thermal conductivity can no longer be defined. The Fourier law also breaks down when the layer thickness becomes comparable to or smaller than the phonon mean free path, which is certainly the case for a 1-nm-thick or even 1-μm-thick diamond film [4, 5]. When the Fourier law breaks down, one has to consider the *conductance* of the entire structure, i.e., the ratio of the heat current to the temperature difference between the thermal reservoirs placed on either side of the structure. The "effective" conductivity is then obtained by multiplying the conductance per unit area by the film thickness [4].

(ii) Ref. [1] does not define the thermal boundary conditions at the top and bottom surfaces of the structure. It is mentioned that the structure is a "freestanding membrane," but thermal reservoirs are necessary to close the problem of the cross-plane conductance. (For example, "blackbody" walls are often used in the analysis of the cross-plane thermal conductivity of thin films [4-6].)

(iii) The assumption that a thin layer of thickness d blocks phonons with wavelengths exceeding $2d$ is incorrect. Long wavelengths not only are not blocked by a thin layer, but are transmitted more easily: the transmission of an acoustic wave through a solid layer approaches 100% when the wavelength approaches infinity [7]. It is true that in a suspended membrane the normal component of the phonon wavevector is quantized and its lowest value is π/d [8]. However, this is no longer true when a thin layer is part of a bigger structure (consider, for example, phonons in a superlattice [9]).

(iv) The analysis of Ref. [1] ignores the phonon transmission and reflection conditions at interfaces and as such does not pass a limiting-case test: when the transmission approaches 100%, a bilayer becomes equivalent to a single layer as far as thermal transport is concerned and any effects related to the individual layer thicknesses must disappear.

The contribution of A.A.M. was supported by the U.S. Department of Energy Award DE-SC0019126. B.S. appreciates support by the National Natural Science Foundation of China (Grant No. 52076002), and the XPLORER PRIZE from the Tencent Foundation.

A. A. Maznev[1*] and B. Song[2**]

[1]Department of Chemistry, Massachusetts Institute of Technology, Cambridge MA 02139, USA

[2]College of Engineering, Peking University, Beijing 100871, China

[*]alexei.maznev@gmail.com

[**]songbai@pku.edu.cn

References

1. B. F. Donovan and R. J. Warzoha, Phys. Rev. Lett. **124**, 075903 (2020).

2. G. Wehmeyer, T. Yabuki, C. Monachon, J. Wu, and C. Dames, Appl. Phys. Rev. **4**, 041304 (2017).

3. A. A. Maznev, A. G. Every, and O. B. Wright, Wave Motion **50**, 776 (2013).

4. A. Majumdar, J. Heat Transfer **115**, 7 (1993).

5. G. Chen, *Nanoscale energy transport and conversion: a parallel treatment of electrons, molecules, phonons, and photons* (Oxford University Press, Oxford, 2005).

6. B. Vermeersch, J. Carrete, and N. Mingo, Appl. Phys. Lett. **108**, 193104 (2016).

7. L. M. Brekhovskikh and O. A. Godin, *Acoustics of Layered Media I: Plane And Quasi-Plane Waves* (Springer, Berlin, 2012).

8. Note, however, that the phonon confinement effect in a suspended membrane does not amount to imposing a wavevector cut-off on the bulk phonon spectrum and modifying the bulk phonon mean free path by a boundary scattering term, see, e.g., B. Fu, K. D. Parrish, H.-Y. Kim, G. Tang, and A. J. H. McGaughey, Phys. Rev. B **101**, 045417 (2020).

9. S. Tamura, D. C. Hurley, and J. P. Wolfe, Phys. Rev. B **38**, 1427 (1988).