ABSTRACT

Heat is one of the most fundamental forms of energy, and the ability to control heat plays a critical role in most current and future energy applications. Recently, interface engineering between heterogeneous solids has provided new approaches to manipulate heat transport at the scales of the energy carriers in solids, *i.e.* phonons which are quantized lattice vibrations. For example, nanocrystalline materials, which are polycrystalline materials with nanoscale grain sizes, are promising thermoelectric (TE) materials that have achieved substantially improved figure of merits compared to their bulk counterparts. This enhancement is typically attributed to a reduction in lattice thermal conductivity by phonon scattering at grain boundaries. On the other hand, inefficient heat dissipation across interfaces has been a long-standing problem that shortens the lifetime of electronics such as light-emitting diodes.

Despite the importance of interfaces, we still lack a comprehensive understanding of interfacial thermal phonon transport. For instance, the Fresnel coefficients enable the straightforward mathematical description of light as it moves between media of differing dielectric constants. Similarly, interfacial phonon transport can also be characterized by transmission coefficients that vary over the broad phonon spectrum in an analogous manner to Fresnel coefficients for light. However, despite decades of work, the spectral profile of these coefficients and how the profile is influenced by the atomic structure of actual interfaces remains unclear. As a result, the basic phenomenon of interfacial heat transport remains among the most poorly understood transport processes.

To elucidate this process, in this thesis we investigate interfacial thermal phonon transport using both modeling and experiment. The first portion of the thesis examines the impact of frequency-dependent grain boundary scattering in nanocrystalline silicon and silicon-germanium alloys using a novel computational method. We find that the grain boundary may not be as effective as commonly considered in scattering certain phonons, with a substantial amount of heat being carried by low frequency phonons with mean free paths longer than the grain size. Our result will help guide the design of more efficient TEs.

The second part of the thesis focuses on studying heat conduction using the Boltzmann transport equation (BTE), which is the governing equation of energy transport at length scales comparable to phonon mean free paths. The BTE is an integro-differential equation of time, real space, and phase space. Due to its high dimensionality, it is extremely challenging to solve. Here, we develop analytical methods to solve the frequency-dependent BTE, which allow us to obtain simple, closed-form solutions to complex multidimensional problems that have previously been possible to solve only with computationally expensive numerical simulations. We demonstrate that the solution leads to a more accurate measurement of phonon MFP spectra in thermal transient grating experiments.

Finally, we report the first measurements of thermal phonon transmission coefficients at a metal-semiconductor interface using ab-initio phonon transport modeling based on the BTE we develop in the second part and a thermal characterization technique, time-domain thermoreflectance. With our approach, we are able to directly link the atomic structure of an interface to the spectral content of the heat crossing it for the first time. Our work realizes the long-standing goal of directly measuring thermal phonon transmission coefficients and demonstrates a general route to study microscopic processes governing interfacial heat conduction.

PUBLISHED CONTENT AND CONTRIBUTIONS

(*1*) Chengyun Hua and Austin J Minnich. “Importance of frequency-dependent grain boundary scattering in nanocrystalline silicon and silicon–germanium thermoelectrics”. In: *Semiconductor Science and Technology* 29.12 (2014), p. 124004. URL: `http://stacks.iop.org/0268-1242/29/i=12/a=124004`.
Contributions: Conducted Monte Carlo simulations in nanocrystalline structrues, analyzed the data, and wrote the manuscript.

(*2*) Chengyun Hua and Austin J. Minnich. “Analytical Green’s function of the multidimensional frequency-dependent phonon Boltzmann equation”. In: *Phys. Rev. B* 90 (21 2014), p. 214306. DOI: `10.1103/PhysRevB.90.214306`. URL: `http://link.aps.org/doi/10.1103/PhysRevB.90.214306`.
Contributions: Generated the idea, solved and analyzed the equation, and wrote the manuscript.

(*3*) Chengyun Hua and Austin J. Minnich. “Transport regimes in quasiballistic heat conduction”. In: *Phys. Rev. B* 89 (9 2014), p. 094302. DOI: `10.1103/PhysRevB.89.094302`. URL: `http://link.aps.org/doi/10.1103/PhysRevB.89.094302`.
Contributions: Generated the idea, solved and analyzed the equation, and wrote the manuscript.

(*4*) Chengyun Hua and Austin J. Minnich. “Semi-analytical solution to the frequency-dependent Boltzmann transport equation for cross-plane heat conduction in thin films”. In: *Journal of Applied Physics* 117.17, 175306 (2015). DOI: `http://dx.doi.org/10.1063/1.4919432`. URL: `http://scitation.aip.org/content/aip/journal/jap/117/17/10.1063/1.4919432`.
Contributions: Generated the idea, solved and analyzed the equation, and wrote the manuscript.

(*5*) Chengyun Hua et al. “Fresnel transmission coefficients for thermal phonons at solid interfaces”. In: *In review process* (2016).
Contributions: Developed the analytical model, analyzed the experimental data, and wrote the manuscript.

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