SIMULATING THERMAL TRANSPORT IN NANOSTRUCTURES FROM THE BALLISTIC TO THE DIFFUSIVE REGIME

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FIGURE 1 (TOP RIGHT): Non-equilibrium molecular dynamics simulations of heat transport in two-dimensional graphene carried out to large length and time scales have revealed a transition from ballistic to transitional transport. The deviation of the thermal conductivity from the logarithmic dependence demonstrates the transition clearly.

FIGURE 2 (BOTTOM RIGHT): We analyzed the competition between disorder and low-dimensionality in the materials known as "diamond nanothreads," which are the (CONT.) newest member of the carbon material family. The ordered system (blue line) shows the expected linear variation of the thermal conductivity on the length. When a small distribution of topological defects (Stone-Wales defects) is introduced (red line), the thermal conductivity quickly converges to a finite value.

EXECUTIVE SUMMARY:

With the power of the Blue Waters supercomputer, we have been carrying out large-scale molecular dynamics simulations to reveal the unusual physics that underlies thermal transport in nanostructured and low-dimensional materials. In these newly emerging materials, the physics of thermal transport remains not well understood. Thanks to Blue Waters, our work in this area has spanned simulations ranging from the ballistic to the diffusive transport regime and has enabled key developments to enhance understanding of how phonons, the primary heat carriers in solids, interact with structure and disorder at these scales. Our simulations have allowed us to answer several outstanding questions relating to (i) the possible divergence of the thermal conductivity in low-dimensional crystals as the sample size increases [1], (ii) how disorder and defects scatter phonons in low-dimensional materials [2], (iii) anomalous effects of strain on thermal transport in low-dimensions [3], and (iv) how low-dimensional superlattices can give rise to coherent phonon transport, enabling the design of nanostructured systems that serve as phonon waveguides and filters.

INTRODUCTION

Understanding heat transport is critical across a variety of applications: optimizing heat shields for rocket and jet engines, solid-state refrigeration via thermoelectrics, and enabling next-generation cooling technologies for powerful supercomputers. Heat transport at the macroscale is governed by Fourier's law and is comparatively well understood. Phonons are quantized lattice vibrations that are responsible for transporting heat through a solid. Fourier's law applies when the system size is large, relative to the mean free paths, which are typically a few hundred nanometers. If not, interesting physics arises. For example, when structure at the nanoscale is introduced, several phenomena emerge that have no analog at larger scales. This arises because the characteristic lengths are comparable to the mean free paths of the primary heat carrying phonons. At the nanoscale, phonons interact with the structure. In addition, the emergence of flexural phonon modes in low-dimensions (out of plane vibrations) introduces heat carriers with anomalous dispersion, which gives rise to unusual temperature-dependence of the thermal conductivity. The role of these carriers (scatterers of phonons, or contributors to conduction?) is now widely debated in the research community.

METHODS & RESULTS

The method we use to carry out our analysis is large-scale molecular dynamics simulations. We use highly parallelizable molecular dynamics frameworks on Blue Waters to access computational domains and simulation sizes that would otherwise be too computationally costly. In the molecular dynamics approach, the motion of each atom in the computational domain is tracked over time, and ensemble averages over the sampled configurations give access to properties of interest. In particular, we use two methods within molecular dynamics. The first is equilibrium molecular dynamics, in which a system is simulated under equilibrium conditions (without net heat current). The equilibrium fluctuations in the system contain information about the subtleties of heat transport. For instance, phonon mean free paths can be extracted from the heat flux autocorrelation function and the thermal conductivity can be determined. The second approach is non-equilibrium molecular dynamics, in which a temperature profile is established across the computational domain. The phonon flow in the system can be statistically analyzed, and linear response characteristics via the fluctuation dissipation theorem once again enable us to extract features of the thermal conductivity.

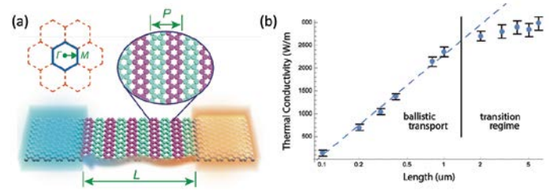
Some of the most exciting highlights of our work from the previous year are:

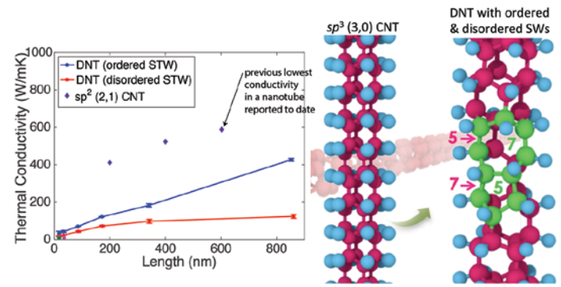
• Simulation of transport in two dimensional materials from the ballistic to the diffusive regime. In low dimensions, heat transport is governed by two competing factors. The first is the long-ranged, ballistic phonon carriers that give rise to increasing thermal conductivity with growing sample size; the second is the presence of flexural modes that may scatter these carriers. Figure 1 clearly demonstrates a transition from ballistic to transitional flow in two-dimensional graphene. In addition to demonstrating the transition, we obtain new physical insights into why the divergence of the thermal conductivity is suppressed: the flexural modes.

• Competition between disorder and low-dimensionality: Which one wins? For diamond nanothreads (1D materials), we were able to demonstrate the dramatic effects of disorder in 1D materials. On one hand, the thermal conductivity of a 1D material is expected to diverge linearly with increasing system length. On the other hand, disorder effects are greatest in low dimensions. How these two effects play out in a disordered, one-dimensional system is currently not well understood. Our simulations, as shown in Figure 2, show a dramatic effect: Even at the smallest degrees of disorder sampled, the divergence of the thermal conductivity becomes quickly suppressed in one dimension.

• Superlattices in two-dimensional materials: observation of coherent phonon transport. We have shown, in work published in 2014 in *Physical Review B*, that the introduction of a superlattice structure in two-dimensions allows tuning of the thermal conductivity through wave interference effects. These superlattice structures effectively demonstrate the design of thermal circuit components, such as the phonon equivalent of optical waveguides or filters.

• Anomalous strain effects in low-dimensional materials. Conventional wisdom tells us that applying strain (in 3D solids) tends to reduce the thermal conductivity of a material. We have demonstrated that in 2D, the thermal conductivity of a material exhibits an anomalous, non-monotonic dependence on the applied strain. We are able to offer physical insights for the underlying mechanisms. This work was published in 2015 in *Physical Review B*.





hundreds of thousands of atoms. Accessing these size scales is critical since diffusive transport in low dimensions does not emerge until system sizes approach ~10 μm or larger. Other research approaches are not capable of accessing these scales without phenomenological models or assumptions about governing transport laws. For example, we were able to demonstrate a transition in the nature of the flow from the ballistic to the transitional flow regime in two-dimensional graphene (Figure 1). This required simulations of sample size ranging from 2 to 6 μm, the largest simulations of low-dimensional heat transport carried out to date within the non-equilibrium molecular dynamics framework.

PUBLICATIONS

Zhu, T. and Ertekin, E. Phonon transport on two-dimensional graphene/boron-nitride superlattices. *Physical Review B* 90 (2014), 195209.

Zhu, T. and Ertekin, E. Resolving anomalous strain effects on two-dimensional phonon flows: the cases of graphene, boron nitride and planar superlattices. *Physical Review B* 91 (2015), 205421.

WHY BLUE WATERS

The Blue Waters supercomputer has enabled us to carry out simulations of systems of several