A New Regime of Nanoscale Thermal Transport: Collective Diffusion Counteracts Dissipation Inefficiency

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Abstract We uncover a new regime of nanoscale thermal transport that dominates when the separation between heat sources is small compared with the substrate's dominant phonon mean free paths. Surprisingly, the interplay between neighboring heat sources can facilitate efficient, diffusive-like heat dissipation.

1 Introduction

Understanding thermal transport from nanoscale heat sources is important for a fundamental description of energy flow in materials, as well as for thermal management in many technological applications including nanoelectronics, thermoelectric devices, nano-enhanced photovoltaics and nanoparticle-mediated thermal therapies. Recent work has shown the rate of heat dissipation from a heat source is reduced significantly below that predicted by Fourier's law for diffusive heat transport when the characteristic dimension of the source is smaller than the mean free path (MFP) of the dominant heat carriers (phonons in dielectric/semiconductor materials) [1, 2]. However, a complete fundamental description of nanoscale thermal transport is still elusive and current theory is limited by a lack of experimental validation.

Diffusive heat transport requires many collisions between heat carriers to establish a local thermal equilibrium and a continuous temperature gradient along which energy dissipates. However, when the dimension of a heat source is smaller

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than the phonon MFP, the diffusion equation is intrinsically invalid as phonons move ballistically without collisions, and the rate of nanoscale heat dissipation is significantly lower than the diffusive prediction. Furthermore, heat-carrying phonons in real materials have a wide distribution of MFPs, from several nanometers to hundreds of microns. For a given heat source size, phonons with MFPs shorter than the hot spot dimension remain fully diffusive and contribute to efficient heat dissipation and a high thermal conductivity (or equivalently, a low thermal resistivity). In contrast, phonons with long MFPs travel far from the heat source before scattering, with an effective thermal resistivity far larger than the diffusive prediction. Phonons with intermediate MFPs fall in between: heat transport is quasi-ballistic with varying degrees of reduced contributions to the conduction of heat away from the nanoscale source.

Most work to date explored the reduction in heat transport from functionally isolated micro- and nanoscale heat sources [1, 2]. Indeed, characterizing heat transport from nanostructures with varying size can be used to experimentally measure cumulative phonon MFP spectra of materials, with the proof-of-principle demonstrated for long-MFP (>1 μm) phonons in silicon [2].

2 A New Regime of Nanoscale Thermal Transport

In this work, we use tabletop extreme ultraviolet (EUV) high harmonic beams to investigate the different regimes of nanoscale thermal transport—purely diffusive and quasi-ballistic—and surprisingly uncover a new *collectively-diffusive* regime that occurs when the *separation* between nanoscale heat sources is smaller than the average phonon MFP. Quasi-ballistic transport dominates when the size of isolated nanoscale heat sources is smaller than dominant phonon MFPs as the long-MFP contributions to heat dissipation are suppressed relative to diffusive predictions. In the new collectively-diffusive regime, the separation between heat sources is small enough that long-MFP phonons can interact with phonons originating from a neighboring heat source as they would if both originated from the same, larger heat source, reintroducing their diffusive-like contributions to thermal transport. This can counteract the reduction in nanoscale heat dissipation to such an extent that heat transport recovers toward the diffusive limit, as shown in Fig. 1. In the limiting case, the spacing between heat sources vanishes and this regime approaches heat dissipation from a uniformly heated layer.

This work has two important implications. First, both size *and spacing* of heat sources are important for determining nanoscale heat dissipation, offering new ideas to mitigate scaling problems for thermal management in nanoelectronics [3]. Second, this new transport regime contains clear signatures of a material's phonon MFP spectrum, enabling detailed characterization of MFP-dependent thermal conductivity.

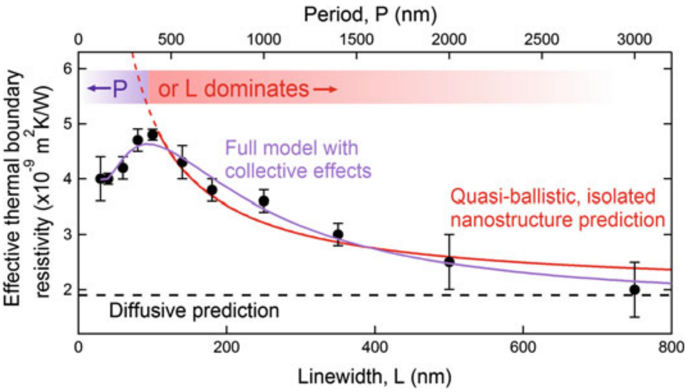

Fig. 1 Effective thermal boundary resistivity extracted from dynamic EUV diffraction. For each heat source linewidth L, on sapphire substrate, the resistivity increases with decreasing linewidths due to the suppression of the contribution to thermal conductivity of phonon modes with MFP larger than L. Decreasing the period P = 4L can reactivate modes with MFP larger than P, returning the effective resistivity towards the diffusive limit (*black dashed line*)

3 The Experiment

Arrays of nickel nanowires were fabricated by e-beam lithography to form periodic gratings on the surface of sapphire substrates. The linewidths L range from 750 to 30 nm, with period P = 4L and a rectangular profile height of ≈13.5 nm. The nanowires are heated by a 25 fs pump pulse centered at a wavelength of 800 nm. Laser excitation creates an array of nanoscale hot spots (lines) on the surface of a cold, transparent substrate. All nanowires are fabricated on the same substrate at the same time, for a constant intrinsic thermal boundary resistivity across all samples: any variation in heat dissipation efficiency as the hot spot size or spacing is varied can thus be attributed to different regimes of thermal transport. The thermal expansion and subsequent cooling of the gratings is probed using coherent EUV light centered at a wavelength of 29 nm, created by high harmonic up-conversion of an 800 nm Ti:Sapphire laser [4]. As EUV light diffracts from the periodic grating, the thermal expansion and relaxation of the nanowires can be extracted from the changes in the diffraction efficiency [5].

4 Characterizing Phonon Transport in Materials

We use this new phenomenon to extract the contribution to thermal transport from specific regions of the phonon MFP spectrum, opening up a new approach for thermal transport metrology and mean free path spectroscopy. This is because by varying both nanostructure size and separation, an effective phonon filter is introduced that suppresses specific MFP contributions to thermal conductivity, resulting in the trends pictured in Fig. 1. We compare our extracted phonon MFP spectra with predictions from first-principles calculations and find excellent agreement between experiment and theory.

This unique new capability for characterizing phonon transport in materials will enable for the first time the experimental characterization of MFP-dependent phonon thermal conductivity spectra down to MFP ≈ 10 nm for more complex nanostructured or metamaterials, where theoretical predictions are not yet possible.

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