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.

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1. Introduction

Understanding thermal transport from nanoscale heat sources is important for a fundamental description of energy flow in materials, as well as for many technological applications including thermal management in 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 heat source is smaller than the mean free path (MFP) of the dominant heat carriers (phonons in dielectric and semiconductor materials) [1,2]. However, a complete fundamental description of nanoscale thermal transport is still elusive, and current theoretical efforts are limited by a lack of experimental validation.

Diffusive heat transfer 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 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 nanoscale 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 ballistically 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; here heat transfer 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 transfer from functionally isolated micro- and nanoscale heat sources [1,2]. Indeed, characterizing heat transfer from nanostructures with varying size can be used to experimentally measure cumulative phonon mean free path spectra of materials, with the proof-of-principle demonstrated for long-wavelength (> 1 μm) MFP 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 uncover a new regime of thermal transport that occurs when the *separation* between nanoscale heat sources is smaller than the average phonon MFP. Surprisingly, the interaction of phonons from neighboring heat sources can counteract the reduction in nanoscale heat dissipation, to such an extent that collective effects increase heat transfer to near the diffusive limit. This work has two important implications. First, mitigating nanoscale heat transport may not be as challenging as once thought [3]. Second, the size of the heat source is not the only important scale that determines nanoscale heat dissipation.

Figure 1 illustrates the differences between the three regimes of thermal transport from nanoscale heat sources – purely diffusive (top left), quasi-ballistic (middle left) and collectively-diffusive (bottom left). Quasi-ballistic transport dominates when the size of isolated nanoscale heat sources is smaller than dominant phonon MFPs. In the new collectively-diffusive regime we uncovered, the separation between heat sources is small enough that long-MFP phonons, whose contribution to heat dissipation would normally be limited by the small size of nano-heat sources, can interact with phonons originating from a neighboring heat source, thus creating an effectively larger heat source size. In the limiting case, the spacing between heat sources vanishes and this regime approaches heat dissipation from a uniformly heated layer. Most importantly, the appearance of this new collectively-diffusive regime mitigates scaling problems for thermal management in nanoelectronics, which may not be as serious as projected [3].

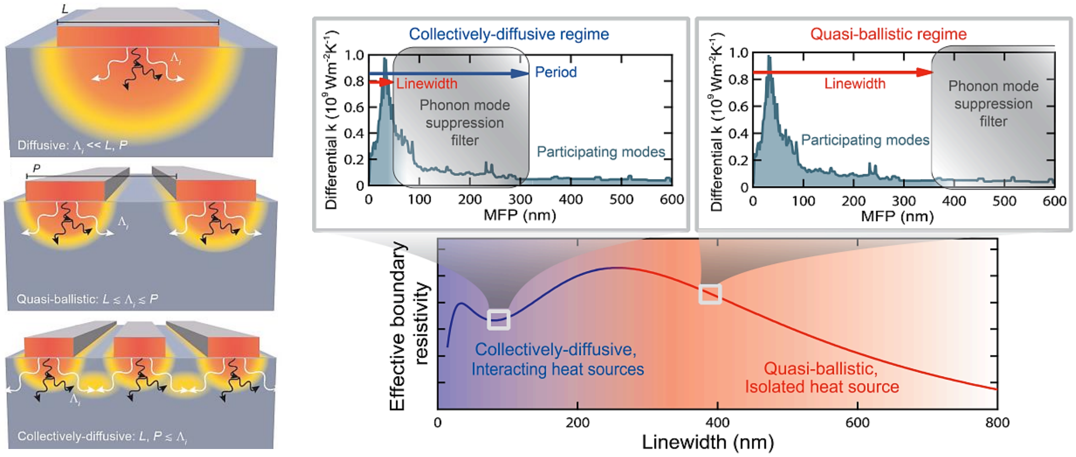

Fig. 1. When heat sources are large compared to dominant phonon MFPs, heat flow is fully diffusive with all phonon modes participating (top left). For smaller heat sources (middle left), long MFPs travel non-diffusively and their contribution to thermal conductivity is suppressed (top right). This causes a rise in the effective thermal boundary resistivity (bottom right). However, when heat sources are closely spaced (bottom left), a single, large effective heat source is created, and more phonon modes are reintroduced (top middle), allowing thermal boundary resistivity to decrease once again.

3. The experiment

Arrays of nickel nanowires were fabricated by e-beam lithography to form periodic gratings on the surface of sapphire and silicon substrates. The nanowire linewidths L range from 750 nm down to 30 nm, with period $P = 4L$ and a rectangular profile height of $\approx$ 13.5 nm. The use of nano-patterned structures rather than optical absorption allows us to explore heat sources much smaller than the diffraction limit of visible light.

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 substrate (sapphire and silicon are transparent or semi-transparent at the pump wavelength). 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, expansion and cooling of the nanowires changes the diffraction efficiency, and this signal can be used to directly extract the thermal expansion and relaxation of each individual nanowire [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, as pictured in Fig. 1 (right). We compare our extracted phonon MFP spectra with predictions from first-principles calculations and find excellent agreement between experiment and theory.

Looking forward, 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 for more complex nanostructured or metamaterials, where theoretical predictions are not yet possible.

5. References

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