Topical Whitepaper

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Towards All-Solid-State Heat Pipes

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Heat pipes, with an equivalent thermal conductivity ranging from 1,000 to 100,000 W/m-K, are critical in the thermal management of space vehicles. However, various limitations associated with the phase change and two-phase transport process in heat pipes render reliability issues, and tremendous efforts have been devoted to the design and operation of heat pipes in space.[1] Therefore, it would be extremely desirable to create an all-solid-state alternative, which might seem impossible given that the highest thermal conductivity of solids we have found so far only provides a maximum thermal conductivity of ~3,000 W/m-K at room temperature. However, it has been predicted that for one-dimensional (1D) lattices, their thermal conductivity (κ) could be divergent following a power law length dependence, i.e., $\kappa \sim L^{\beta}$, where L is the system length and β is a constant.[2] This phenomenon is called superdiffusive transport, and the concept has been thought to be of academic interest only because in reality, single atomic chains of sufficient length have remained to be unattainable.[2] However, a recent experimental demonstration of superdiffusive thermal transport in quasi-1D van der Waals (vdW) crystal $NbSe_3$ nanowires[3] reveal a real possibility of creating super heat conductors with thermal conductivity values higher than that of any known materials, which could be used to construct all-solid-state heat pipes that can revolutionize the thermal management of space vehicles.

This idea is built upon our recent observation that phonon transport in thin $NbSe_3$ nanowires becomes superdiffusive, with the thermal conductivity diverges with the wire length exceeding an unprecedented length of over 42.5 μm. As shown in Fig. 1a&b, a $NbSe_3$ nanowire is placed between a heat source and heat sink of a microdevice for thermal conductivity measurement using the microthermal bridge method. Fig. 1c displays a high resolution transmission electron microscopy micrograph showing the single crystalline nature of an ultra-thin $NbSe_3$ nanowire.

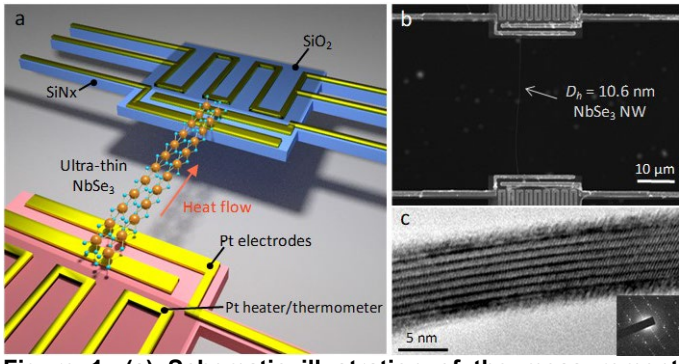

Figure 1: (a) Schematic illustration of the measurement device with a $NbSe_3$ nanowire placed between the heat source and heat sink. (b) An SEM micrograph showing a nanowire on the device with a suspended length of 28.2 um. (c) An HRTEM image of an ultra-thin $NbSe_3$ nanowire showing the crystalline structure.

Fig. 2a plots the measured room temperature thermal conductivity *versus* the hydraulic diameter of the wire (D_h), and all nanowires having suspended lengths of ~15 μm. Interestingly, the data indicate a clear transition at D_h = 26 nm. For thicker wires, κ decreases as D_h reduces, as a result of phonon-boundary scattering at nanowire surfaces; however, as D_h further drops, κ demonstrates a steep upward trend. In fact, compared to the moderate reduction from 7.1 to 4.3 W/m-K as D_h decreases from 135 to 26 nm, κ increases by ~25 fold and reaches 109 W/m-K for a 6.8 nm diameter wire.

The intriguing size-dependence prompts us to examine the length dependence of κ, through measuring the same nanowire with different suspended lengths. Fig. 2b plots the normalized room temperature thermal conductivity, κ^*, *versus* the normalized suspended length, L^*, both with respect to the values of the respective longest wires, for seven $NbSe_3$ nanowires of different D_h. For meaningful comparison, the maximum length of all samples in Fig. 2b is kept ~15 μm (14.2-16.3 μm). Interestingly, for wires with larger D_h, κ first increases significantly with L from ~2 to

~6 μm, and then only increases marginally as L further extends, indicating that κ converges to a saturated value as the wire length exceeds the phonon mean free path (MFP). However, as D_h reduces to below 26 nm, κ exhibits a much stronger length dependence even for L > 6 μm, suggesting a transition from convergence to divergence in the length dependence for κ.

To further explore the length dependence, we measured more samples with much longer suspended length. Fig. 2c illustrates the measured room temperature κ *versus* L, which indicates that for nanowires with D_h in the range of 10 to 12 nm, the length dependence extends beyond 42.5 μm, an unprecedented length dependence for nanowires, much larger than the previously reported 8.3 μm for SiGe wires,[4] ~10 μm for SWCNTs,[5] and ~15 μm for a 25 nm diameter GaP nanowire.[6]

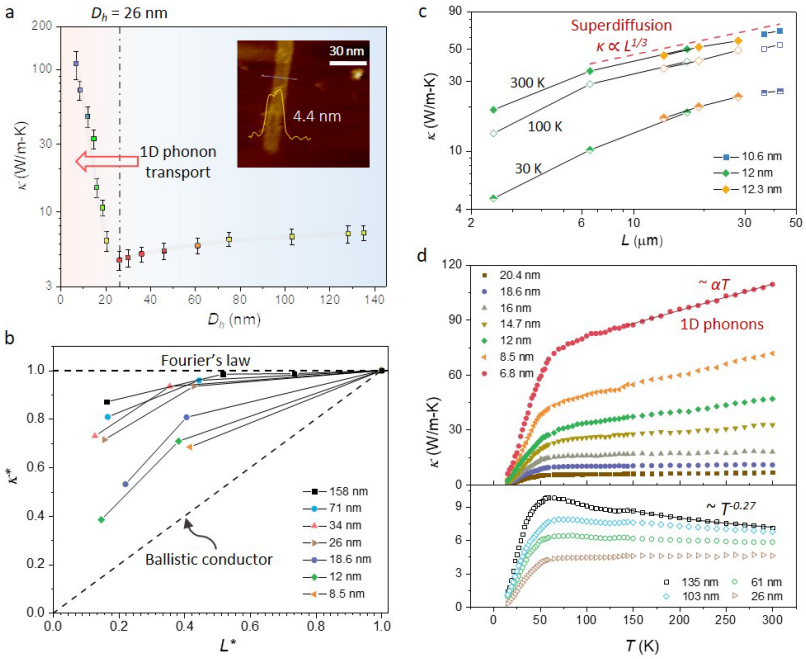

Figure 2. Divergent and Superdiffusive transport of 1D phonons. (a) Measured room temperature κ *versus* D_h. The gray solid line is guide to the eyes. Inset: AFM scanning profile of the nanowire with D_h = 6.8 nm. (b) Normalized room temperature κ *versus* the normalized suspended length, which indicates a convergence-divergence transition as the wire diameter decreases. (c) Measured κ *versus* suspended length at different temperatures. (d) Temperature dependence of κ for different diameter wires (the suspended lengths are ~ 15 μm for all samples).

Interestingly, in the length range of > 6.5 μm, the measured κ of the three nanowires follows a trend of $\kappa \sim L^{\beta}$, with a constant β of 1/3, consistent with the predicted thermal transport behavior of 1D lattices. To further confirm the superdiffusive transport, we also plot the measured κ at 100 K in Fig. 2c, which again follows the trend of $\kappa \sim L^{1/3}$ in the same length range. If partially ballistic phonon transport was responsible for the observed length dependence, a steeper slope would exist at 100 K as the phonon MFP is larger at lower temperature. Thus, the same power law dependence at the very different temperatures of 100 and 300 K strongly suggests that the length dependence in the range of > 6.5 μm is due to superdiffusive behavior of 1D phonons.

The excitation of 1D phonons in the ultra-thin nanowires is also supported by a transition in the temperature dependence of κ, as shown in Fig. 2d. For wires with D_h > 26 nm (lower panel), κ decreases as the temperature increases in the range of 50-300 K, a signature of Umklapp scattering. However, as D_h further reduces (upper panel), κ starts to display an increasing trend in the same temperature range, and a linear temperature dependence is observed for the measured κ of thin wires. This linear dependence is confirmed to be due to continuous excitation of 1D along chain phonons as the temperature escalates, which leads to a linearly increasing heat capacity (C), and hence a linearly increasing thermal conductivity according to $\kappa \sim Clv$, where v and l are the

phonon group velocity and MFP, respectively. Importantly, the length dependence at 30 K, where all three-dimensional phonons make important contributions to thermal transport, deviates from the 1/3 power law and the thermal conductivity tends to be converged for long wires, as also shown in Fig. 2c.

This experimental observation brings the once purely theoretical superdiffusive transport in nonlinear 1D lattices into reality and leads to the following three key scientific questions.

(1) What are the conditions for superdiffusive 1D thermal transport in vdW crystals? While all theoretical studies have been done on single atomic chains with only 1D phonons, our experimental observation indicates that superdiffusive behavior can emerge as long as 1D phonons dominate the transport process, even in arrays of covalently-bonded atomic chains with the perturbation of weak inter-chain vdW interactions. As such, the experimental results raise a fundamental question, that is, what is the threshold ratio of inter-chain to intra-chain interaction strengths under which the thermal conductivity of aligned vdW atomic chains becomes divergent with length. One more important question is whether superdiffusive transport is unique to $NbSe_3$ nanowires or it also occurs in other type of quasi-1D vdW crystals, such as $Ta_2Pd_3Se_8$ and $ZrSe_3$?

(2) What is the length limit of superdiffusive transport in quasi-1D vdW crystal nanowires and how can we extend the limit? Our experimental data show that the thermal conductivity of ultra-thin $NbSe_3$ nanowires increases with length following a 1/3 power law beyond 42.5 μm. It is of both fundamental interest and practical significance to explore the limit of this length dependence; and successful extension of the length limit could eventually lead to super heat conductors with ultra-high thermal conductivity values beyond that of any known materials.

(3) Whether it is possible to create novel bulk materials with super high thermal conductivities? Our characterization suggests that 1D phonons are excited as a result of a record level of elastic stiffening with a much enhanced Young's modulus in the along chain direction. Therefore, it is of great interest to explore whether we can alter the structure/composition of bulk vdW crystals through strain engineering to alter their elastic properties to achieve divergent thermal transport at a larger scale to achieve all-solid-state heat pipes that can transform the thermal management practice in aerospace engineering.

Conducting research to answer these scientific questions are of both fundamental significance and practical impacts. "What is the limit of heat conduction in solids" has been listed as one fundamental science question in a recent issue of *Science* and superdiffusive transport represents one most intriguing phenomenon in thermal sciences. The practical implication of all-solid-state heat pipes could produce immense impacts to thermal engineering including thermal management for space vehicles.

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