Thermal and Electrical Properties of Quasi-1D van der Waals Nanowires

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Primary CNF Tools Used: Heidelberg mask writer - DWL2000, Autostep i-line stepper, LPCVD nitride - B4, GSI PECVD, AJA sputter deposition, AJA ion mill

Abstract:

Using the microdevices fabricated at CNF, we conducted extensive measurements of thermal transport through quasi-one-dimensional (quasi-1D) van der Waals (vdW) crystal $NbSe_3$ nanowires. Dimensional transition from 3D to 1D was demonstrated when the nanowire diameter scales down below 26 nm. Importantly, we found that one-dimensional (1D) phonons led to superdiffusive transport with the nanowire thermal conductivity becoming divergent with the wire length following a 1/3 power law extending over an unprecedented 42.5 μm. This important result has been published in *Nature Nanotechnology*.

Summary of Research:

Because of the pandemic, no on-site nanofabrication at CNF was conducted during the reporting period. However, using the previously fabricated microdevices, we conducted extensive measurements of thermophysical properties of quasi-1D vdW crystal $NbSe_3$ of different diameters and lengths. The schematic diagram of the measurement scheme, an SEM micrograph of an $NbSe_3$ wire placed on the measurement device, and a TEM micrograph of an ultra-thin nanowire were shown in Figure 1.

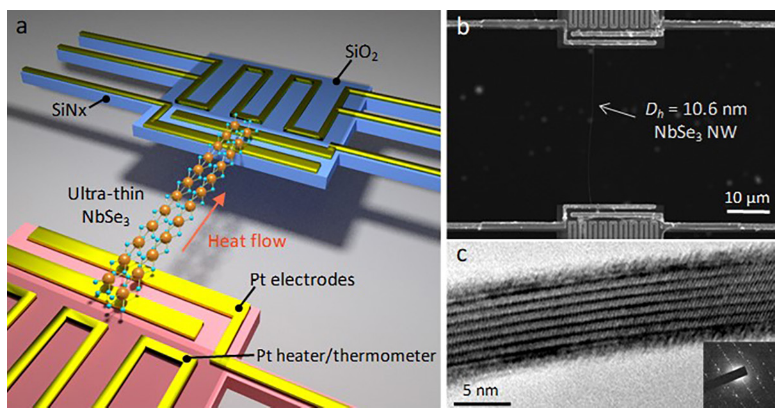

Figure 1: (a) Schematic illustration of the measurement scheme. (b) An SEM micrograph showing a nanowire on the device with a suspended length of 28.2 µm. (c) An HRTEM image of an ultra-thin $NbSe_3$ nanowire showing the crystalline structure.

These measurements have led to a major breakthrough with the first experimental observation of superdiffusive transport of 1D phonons, which has been published in *Nature Nanotechnology* [1].

In 1955, Fermi, Pasta, Ulam, and Tsingou reported their “shocking little discovery” that an excited vibration mode in single atomic chains did not dissipate into heat over a long period of time, which attracted tremendous attention due to its broad implications. A direct consequence of this discovery is that the thermal conductivity of 1D lattices becomes divergent with the chain length, suggesting a type of thermal superconductors of ever-increasing thermal conductivity with the sample length.

The concept remains purely conceptual and is regarded as of academic interest only as single atomic chains of sufficient length remain experimentally unattainable.

Our study shows that the thermal conductivity of ultra-thin $NbSe_3$ nanowires increases with the wire length beyond a record level of 42.5 μm following a 1/3 power law, providing the first experimental evidence for superdiffusive transport.

Figure 2a plots the measured room temperature thermal conductivity (κ) versus the hydraulic diameter (D_h) of the nanowires, with all nanowires of ~ 15 μm long. Interestingly, the data indicate a clear transition at D_h = 26 nm. For thicker wires, κ decreases as D_h reduces due to phonon-boundary scattering; however, as D_h further drops, κ demonstrates an unexpected steep upward trend with ~ 25 fold increase for a 6.8 nm wire.

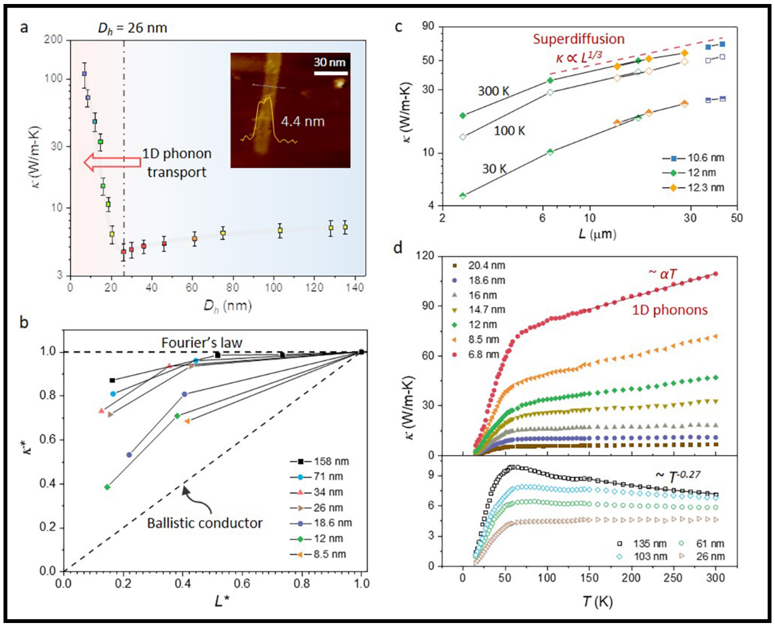

Figure 2: Divergent and superdiffusive transport of 1D phonons. (a) Measured room temperature thermal conductivity (κ) versus the hydraulic diameter (D_h). Inset: AFM scanning profile of the nanowire with D_h = 6.8 nm. (b) Normalized room temperature κ versus the normalized suspended length, which indicates a convergence-divergence transition as D_h decreases. (c) Measured κ versus suspended length at different temperatures. (d) Temperature dependence of κ for different diameter wires.

Next we examine the length dependence of κ, and Figure 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 (~ 15 μm), for seven wires of different D_h. Interestingly, for wires with larger D_h, κ first increases with L from ~ 2 to ~ 6 μm, and then converges to a saturated value. However, as D_h reduces to below 26 nm, κ exhibits a much stronger length dependence even for $L > 6\,\mu$m, suggesting a transition from convergence to divergence.

To further explore the length dependence, we measured more samples with much longer suspended length. Figure 2c indicates that for nanowires with D_h in the range of 10 to 12 nm, the length dependence extends beyond 42.5 μm, much larger than the previously reported values for different wires. Interestingly, in the length range of > 6.5 μm, the measured κ follows a trend of $\kappa \propto L^{1/3}$, consistent with the theoretical prediction of superdiffusive phonon transport in 1D lattices.

To further confirm the superdiffusive transport, we also plot the measured κ at 100 K, which again follows the trend of $\kappa \propto L^{1/3}$ in the same length range. This consistent trend over very different temperatures of 100 and 300 K strongly suggests that the length dependence in the range of > 6.5 μm is due to the super-diffusive behavior of 1D phonons, instead of partially ballistic transport.

This intriguing transport is due to excitation of 1D phonons in the ultra-thin nanowires, as shown in Figure 2d with a linear increasing trend for thinner wires in the temperature range of 50-300 K (upper panel) in contrast to the decreasing trend of thicker wire dure to Umklapp scattering (lower panel).

In addition to this important discovery, we have also done other measurements with the microdevices and published two papers [2,3].

References:

[1] L. Yang, Y. Tao, Y. Zhu, M. Akter, K. Wang, Z. Pan, Y. Zhao, Q. Zhang, Y. Xu, R. Chen, T. T. Xu, Y. Chen, Z. Mao, and D. Li, "Observation of Superdiffusive Phonon Transport in Aligned Atomic Chains," Nature Nanotechnology, published online, DOI: 10.1038/s41565-021-00884-6 (2021).

[2] Z. Pan, L. Yang, Y. Tao, Y. Zhu, Y.-Q. Xu, Z. Mao, and D. Li, "Net Negative Contributions of Free Electrons to the Thermal Conductivity of $NbSe_3$ Nanowires," Phys. Chem. Chem. Phys. 22, 21131-21138 (2020).

[3] Y. Zhao, M. L. Fitzgerald, Y. Tao, Z. Pan, G. Sauti, D. Xu, Y.-Q. Xu, and D. Li, "Electrical and Thermal Transport through Silver Nanowires and their Contacts: Effects of Elastic Stiffening," Nano Lett. 20, 7389-7396 (2020).