Measurement and Modulation of Thermal Transport in One-dimensional Materials with Scanning Electron Microscopy

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論 文 内 容 の 要 旨 Thesis summary

The accurate measurement of the thermal transport properties of nanomaterials is crucial for uncovering the microscopic heat transfer mechanisms and guiding the modulation of material performance for applications. Despite the impressive advancements in nanoscale thermal metrology over the past decades, there are still some problems and challenges, including (1) how to eliminate the thermal contact resistance (TCR) error between the sensor and the sample for the resistive contact methods, (2) how to directly evaluate TCR at a single junction between nanostructures, and (3) how to efficiently establish the structure-property relationship for thermal transport at the nanoscale. Besides, the modulation of thermal transport at the nanoscale has long remained challenging. This dissertation aims to address the above issues with scanning electron microscopy (SEM) and focuses on the measurement and modulation of thermal transport in one-dimensional (1D) materials.

First, a method combining the electron beam (EB) heating in SEM with two suspended line-shaped heat flux sensors was developed to eliminate the TCR error at the sample-sensor junction in the thermal conductivity measurement of 1D materials. The two ends of the sample are anchored to the two sensors respectively and the focused EB is utilized as a mobile heating source to heat the 1D sample locally with a high spatial resolution, while the two sensors simultaneously measure the heat fluxes induced by the EB heating and further the thermal resistance ratio of the two sample segments separated by the EB spot is extracted. The spatially resolved thermal resistance distribution can be obtained by moving the nanosized EB heating spot along the sample. The *in situ* thermal resistance mapping along a single cupstacked carbon nanotube (CNT) was performed using this method, from which the thermal conductivity of the CNT without the TCR error was extracted to be around 40 W/m·K and the TCR at the sample-sensor junction was measured to be 1.35 MK/W.

Second, we developed a method for the in situ and real-time measurement of TCR at a single junction

between 1D samples in SEM. In this method, the two 1D samples are anchored respectively to a manipulator-based tungsten probe serving as a movable heat sink and a suspended line-shaped sensor. A thermal path from the sensor to the probe is formed and the total thermal resistance of the two samples in contact can be extracted. Afterward, we separate the two samples and measure their respective thermal resistance in the SEM. Then the TCR is extracted by subtracting the thermal resistance of each sample from the total thermal resistance. Since the movement of two samples can be separately controlled inside the SEM chamber, the contact condition can be dynamically and flexibly adjusted and the high-resolution images of the contact conditions can be *in situ* captured by SEM. Using this method, we measured the TCR between two multi-walled CNTs for 11 contact conditions. The TCR results were on the order of 10⁷ K/W and were studied in combination with the SEM-captured images of the contact morphology.

Third, we developed a method to simultaneously measure the thermal conductivity and observe the internal structure of a 1D sample using scanning transmission electron microscopy (STEM) in the SEM for the structure-property correlation. The two ends of the sample are anchored to a tungsten probe and a suspended line-shaped sensor, respectively. The sensor serves simultaneously as a heater and a resistance thermometer, ensuring highly sensitive thermal measurements. The sensor is fabricated on the edge of the silicon wafer so that the EB can transmit through the 1D sample and that the inner nanostructure of the sample can be observed in real-time. Using this method, we *in situ* measured the thermal conductivity of two cup-stacked CNTs and concurrently observed the internal non-uniform structures. The results indicated that the sample with more structural disorders had a lower thermal conductivity.

In addition, we successfully modulated the thermal transport in cup-stacked CNTs by filling liquid into the samples. We prepared CNTs infused with pure water and LiCl solutions and measured their thermal conductivities with our *in-situ* STEM-in-SEM setup. The internal structures of three waterinfused CNTs were also observed in real-time. We also measured the temperature dependence of thermal conductivity for a water-infused and a LiCl-solution-infused CNT. The liquid-infused cup-stacked CNTs exhibited higher thermal conductivity (60-360 W/m·K) than the dry samples (about 40 W/m·K). This thermal conductivity enhancement can be attributed to the strengthened interfacial coupling between graphene layers due to the penetration of water molecules or ions into the gaps between graphene cups. which is supported by the molecular dynamics simulation results in the literature.