論 文 の 内 容 の 要 旨

論文題目　　Structure dependent thermal transport properties of single walled carbon nanotubes
（単層カーボンナノチューブの構造依存熱輸送特性）

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Since the discovery of carbon nanotubes in 1991 by Iijima, many potential applications have been suggested and examined in mechanics, optics, electronics and thermotics. Due to the quasi-one-dimensional atomic structure, individual carbon nanotube possesses significant thermal conductivity along the axial direction, thus its probable prospects in the field of thermal managements has attracted many researchers and engineers, especially as a thermal interface material to enhance the heat dissipation in electronics that limits the further miniaturization. Besides, the newly developed methods have fabricated high-quality free-standing carbon nanotube thin films, and they inherit some of the extraordinary characteristics and manifest great optical, mechanical, electrical and thermal properties as well.

Waste heat utilization has attracted worldwide attention due to the pressing energy crisis, hence thermoelectric materials owe to its simple and direct conversion from low-grade waste heat into high-grade electric power stand out as a promising candidate to ease the severe problem of this century. Of the current commercially available TE materials, the high cost, toxicity, inflexibility, and heavy weight are the obstacles for their broad applications. Therefore, extensive researches have been invested in organic TE materials that are cheaper, non-toxic, flexible and lighter, among which carbon nanotube thin film shows its great potential thanks to its abundance in nature and superb mechanical properties. Although the thermal conductance of individual carbon nanotubes is significantly high, the weak van der Waals interactions between nanotubes and the finite length of carbon nanotubes in network are hugely suppressing the thermal conductance of the thin mats or films, while its electrical properties has not been affected much. Contrast to the easiness of obtaining the electrical properties of carbon nanotube thin films with well-studied theories and experiments, the thermal properties on the other hand are short of fairly verified methods to account for. Therefore, this thesis proposes a new method for studying the in-plane thermal conductance of thin film with infrared thermography. Infrared thermography is a quick method to obtain the temperature profiles of the targets, along with some ad hoc designing to calculate the heat flux, it can develop into a direct and handy technique for contactless thermal conductance determination. In this experiment, the single walled carbon nanotube thin films are free-standing in between two cantilevered silicon thin plates which work as the reference to calculate the heat flux through SWNT thin film. The temperature profile along the silicon-SWNT-silicon bridge is recorded with the infrared camera. Besides, another unloaded parallel silicon pair is set in order to offset the noise from the camera. A control experiment without film is also conducted to account for background conductance originating from thermal radiation or residual convection. Since the thickness of single walled carbon nanotube thin film in this study is hard to determine and it is changeable under different conditions, sheet thermal conductance is proposed to evaluate the capability of heat transfer of single walled carbon nanotube thin films. The results indicate the sheet thermal conductance are 15613.4±2672.5nW/K, 9925.4±1417.1nW/K, 5454.8±594.9nW/K and 3405.7±773.8nW/K at room temperature for single walled carbon nanotube thin film with transparency of 60%, 70%, 80% and 90%, respectively. Furthermore, for the 50nm single walled carbon nanotube thin film of 90% transparency, its thermal conductivity is around $68.1 Wm^{-1}K^{-1}$, which is benefitting from the composition of the very long high-quality single walled carbon nanotubes by aerosol chemical vapor deposition synthesis method. The sensitivity of This method is on the order of 10^{-6} W/K and can be applied to any other films (especially low thermal conductance films) that can be transferred to free-standing in between the cantilevered silicon pair plates. Besides, in comparison with the Raman measurements performed on the same thin films, the non-equilibrium among different phonon polarizations in Raman measurements might have contributed to the lower sheet thermal conductance by a factor of 1.9 to 2.5 in the condition of this study.

There has been large amount of experiments conducted concerning the axial thermal conductivity of individual carbon nanotubes. However, the various sample preparation methods as well as different measurement methods make the results ranging hugely and hardly comparable with each other. The hidden reason behind this challenge is the difficulty to identify the precise nanoscale structure of the carbon nanotubes being studied. Furthermore, the chirality-specific SWNT growth has been realized, which have excited the application in electronics, then the thermal behaviors of individual single walled carbon nanotubes with specific atomic structure are more urgently needed to be addressed. Besides, the length of SWNT is a critical parameter that determines the number of phonons in it, but the length dependency of phonon conductance in single walled carbon nanotubes is still a controversial debate in theoretical investigations, and due to the challenges of sample preparations, there are scarce experimental studies to clarify the disagreement. The unsettlement and insufficiency of the structure dependency of thermal transport properties in SWNTs motivate us to systematically study the relationship of the thermal conductance and its nanoscale structure with the high-quality defect-free horizontally aligned SWNTs. Tackling the structure dependency of thermal transport in individual SWNTs would help us better understand and manipulate its extraordinary thermal properties to serve for future applications.

The micro-thermometer compatible with transmission electron microscope has been fabricated in this thesis to investigate the axial thermal conductance of individual carbon nanotubes and their nanoscale structure. The transmission electron microscope images show that the transferred horizontally aligned carbon nanotubes across the micro-thermometers are very clean and straight. Through comparison of the SEM and TEM images, the number of SWNTs in a bundle can be determined by counting the extended SWNTs dispersedly distributed on the supporting membranes. The thermal conductance of four samples, including three bundles with different sizes as well as three isolated single carbon nanotubes, have been studied. Thanks to the very long extended carbon nanotubes on the membranes and the annealing process during carbon nanotube transfer, the contact strength and contact area is immensely enhanced that the thermal boundary resistance can be effectively ignored in this investigation. The measured background conductance is much higher than the theoretical prediction, makes the deduction of background conductance compulsory for the precision of measurements. The effective thermal conductivity of the four samples under investigation are increasing with the temperature first and saturates around room temperature. The thermal conductivity of the isolated carbon nanotubes reaches around 5000$Wm^{-1}K^{-1}$ around 300K, fold larger than that of the bundles. Furthermore, the larger the size of the bundles, the lower its effective thermal conductivity. The logarithmical decrease of the thermal conductivity with the bundle size is the first-time quantitative study. This experiment proves the serious degeneration of thermal transport properties by the interactions between carbon nanotubes, and in consistent with the IR measurements of higher thermal conductivity of thin films composed of smaller size bundles.

In macro-scale, a quick and handy method is presented here to measure the sheet thermal conductance of carbon nanotube thin films; in nanoscale, the correlation between the thermal transport properties of individual carbon nanotube and its nanoscale structure is investigated to provide scientific guides to carbon nanotube electronics architecture. Therefore, the clear relationship of the thermal conductivity of carbon nanotube and its nanoscale structure can help to design the thermal conductance of carbon nanotube thin films through control growth, tuning it to be higher for thermal managements or lower for insulator or thermoelectric applications in the future.