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Four-Probe Thermal Transport Measurements of Mesoscale Structures

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Four-Probe Thermal Transport Measurements of Mesoscale Structures

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Abstract

Four-Probe Thermal Transport Measurements of Mesoscale Structures

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A number of thermal transport measurement methods have been reported in the past few decades for bulk materials and nano-structures. However, challenges have remained for measuring the thermal conductivity of mesoscale structures with the dimension in the range between 1 μm and 1 mm. A recent reported four-probe thermal transport measurement method for nanostructures can separately obtain the intrinsic thermal resistance and the contact thermal resistance. This thesis reports an investigation of the applicability of this method for mesoscale samples. Mesoscale four-probe measurement devices have been designed, fabricated and calibrated with a SiN_x beam sample. Multiple error sources have been evaluated. The measured thermal conductivity of SiN_x , 3.7- 3.8 $\text{W m}^{-1} \text{K}^{-1}$ at room temperature, is in agreement with the literature values. In addition, consistent thermal conductivity values of the SiN_x layer have also been obtained based on the measured thermal and electrical conductances of the Pd/Cr/SiN_x beam of the measurement device and the Wiedemann-Franz law. The mesoscale four probe devices have also been used to perform thermal transport measurements of three polyethylene ribbon samples with different dimensions. The

measured thermal conductivity of polyethylene ranges between $4 \text{ W m}^{-1} \text{ K}^{-1}$ and $6 \text{ W m}^{-1} \text{ K}^{-1}$ at the temperature range between 150 K and 350 K, with the peak value appearing at 275K. The decreasing thermal conductivity with increasing temperature reveals the effect of lattice anharmonicity at temperatures higher than 275 K. These results demonstrate that the mesoscale four-probe thermal transport measurement device can obtain both the contact thermal resistance and intrinsic thermal resistance of mesoscale structures with acceptable errors, which are mainly caused by increasing radiation heat loss from the thermometer lines and the sample with increasing lengths. Improvement in the measurement devices can be made by reducing the length and width of the thermometer lines to reduce the radiation loss without reducing the conduction thermal resistance. Hence, this mesoscale four-probe thermal transport measurement method can fill the gap between thermal transport measurements of bulk materials and nano-structures and provide experimental data of mesoscale structures.

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Chapter 1 Introduction

A number of different thermal transport measurement methods have been developed in the past few decades for measuring thermal properties of bulk materials, thin films and nanostructures. Each of these techniques has its own advantages and limitations and is suitable for a limited range of materials. For bulk materials, steady-state techniques, transient hot-wire methods, laser flash diffusivity measurement approaches, and transient plane source set ups are commonly used.¹ The steady-state methods are based on the measurements of the rate of heat conduction and associated temperature drops in the samples subjected to a time-invariant heat flow rate. The transient methods require the measurements of a time dependent thermal response signal of the sample in either time domain or frequency domain.

Among various methods, the guarded hot plate method is a standard test method for steady-state heat flux measurements and thermal transmission properties.² The principle of the guarded hot plate method is to generate a known unidirectional heat flux through the sample. In a basic implementation, the sample is placed between a heater plate and a parallel cold plate, and is surrounded by an annular guard acting as a thermal barrier. Electrical current is supplied in the annular guard to compensate for lateral heat loss and maintain one-dimensional heat flow perpendicular to the heater plate and the cold plate. With the known heat flux applied in the heater plate, measured temperature drop, and sample thickness, the thermal conductivity can be obtained. Salmon has reported a guarded hot plate apparatus, which can measure the thermal conductivity of

low density insulating materials with uncertainties in the range 1.3–2.4% for specimens with a thickness between 50 mm and 250 mm.³ Over the past few decades, the accuracy achieved in the guarded hot plate method has improved considerably with numerical modeling to minimize edge heat losses, improved analysis systems, and advanced temperature controllers. However, challenges have remained in guarded hot plate measurements of thermal conductivity at high temperatures, where radiation loss becomes significant especially for materials.

Besides the standard guarded hot plate method, a steady-state longitudinal heat flow method has been used to measure thermal conductivity of a number of materials. In one implementation,⁴ a heat flux flows through an unknown sample and a known sample as a reference. The respective thermal gradient is inversely proportional to their thermal conductivities. Samples measured are usually cylindrical bars with typical size of $0.5 \times 0.5 \times (2\sim3)$ mm³. For a low thermal conductivity sample, the inevitable surface heat losses are of concern. For a sample with high thermal conductivity, the sample needs to be very long to establish a reasonably high temperature gradient that can then be accurately measured. In addition, the contact thermal resistance between the sample and thermal sensors can be large and cause errors in the temperature measurements.

The laser flash method, which is a time domain transient technique, can obtain the thermal diffusivity of bulk materials.⁵ Optical heating is applied as an instantaneous heating source, along with a thermographic technique for rapid non-contact temperature sensing, which avoids the error from contact thermal resistance. However, the heat capacity and density of the sample need to be determined from other measurements,

which may contain large errors, in order to obtain the thermal conductivity of the sample measured by the laser flash technique.

For thin film thermal conductivity measurements, the 3ω method and time-domain thermoreflectance technique are widely employed. For the 3ω method first introduced by Cahill,⁶ a narrow metal line serving as both an electrical heater and a temperature sensor is patterned on the thin film sample. A sinusoidal current at frequency 1ω is applied to the metal line, causing a temperature oscillation ΔT of the metal line at 2ω frequency. The electrical resistance of the metal line changes with temperature and contains an oscillating component at 2ω frequency. A lock-in amplifier can be used to measure the 3ω component in the voltage drop as a result of the 2ω electrical resistance modulation, which contains the information of thermal transport property of the thin film. In these electrical resistance thermometry based measurements, the temperature coefficient of resistance of the metal line needs to be sufficiently large and constant in the measurement temperature range.

The optical non-contact time-domain thermoreflectance (TDTR) method technique can be used to measure both bulk and thin film materials.⁷ The sample is usually coated with a metal thin film transducer. During the measurements, the laser pulse is used to heat the sample, while a probe laser is used to measure the change in the temperature-dependent reflectivity of the metal film. In most cases, analysis of TDTR experiments requires comparisons between the data and a model of the heat transport in the system with some unknown thermal properties treated as free parameters and adjusted

to minimize the differences between the model and the data.⁸ In addition, the sample surface needs to be smooth to avoid diffuse scattering of the probe laser beam.

When the characteristic dimension of a material is reduced to the scale of the scattering mean free path or even the wavelength of the charge or heat carriers, which are usually in the nanometer scale, the thermal properties of the nanostructures can be very different from the bulk counterparts. This feature has motivated the development of several methods for thermal transport measurements of nanostructures as discussed in a recent review,⁹ including suspended resistance thermometer devices, micro-Raman spectroscopy, thermal reflectance, transient electrothermal techniques, and bimaterial cantilevers. Both the 3ω and TDTR methods discussed above for thin film thermal transport measurements have been further developed for nanostructures measurements.

A suspended platinum resistance thermometer (PRT) device was fabricated for measuring the thermal conductivity of nanostructures.¹⁰ The device consists of two adjacent thermally isolated SiN_x membranes each supported by three long suspended SiN_x beams with built-in serpentine Pt resistance thermometers. Two Pt electrodes are patterned on each membrane to measure the thermovoltage drop along the nanostructure segment bridging the membrane, which can be used to determine the temperature drop at the contacts between the sample and the membranes, and the contact thermal resistance. This method is limited to electrically conducting nanostructure samples with a sufficiently large and uniform Seebeck coefficient along the entire length. Hence, it is unable to measure a thin nanostructure with a spatially varying Seebeck coefficient that depends sensitively on the surface charges.

A different four probe thermal resistance measurement method for determining both the contact thermal resistances and the intrinsic thermal conductivity of a nanostructured sample has been reported recently.¹¹ The device consists of four suspended metal/SiN_x lines that serve as heater and thermometers. The nanostructure sample bridges the four lines. A different line is used as a heater line in each of four different heating configurations. The obtained sixteen sets of temperature and heat flow measurement data can be used to obtain nine of the thermal resistances in the measurement device and the nanostructure sample, including the intrinsic thermal resistance and the two contact thermal resistances.

Micro-Raman spectroscopy has been employed measure thermal transport in different nanostructures. During the measurements,¹² the sample is heated with a laser beam and the sample temperature within the laser spot can be determined either with the Stokes and anti-Stokes intensity ratio or the Raman peak shift. However, both thermometry approaches have limitations. For the first method, the intensity ratio depends only on the zone center or zone boundary optical phonon populations, which can be used to determine the temperature of only the Raman-active modes. In addition, the sample needs to be heated to a high temperature to observe the anti- Stokes peak. For the second method, charged impurities and strain can contribute to the Raman peak shift, which can cause large errors. The temperature sensitivity of these Raman methods is usually inferior compared to electro-thermal sensors.

Besides steady state electro-thermal or micro-Raman measurements of thermal transport in nanostructures, transient electrothermal techniques (TET) have been

employed to obtain the thermal conductivity and thermal diffusivity of nanowires.¹³ In the TET technique, a nanostructure is suspended between two copper electrodes. A step current is applied to the sample as the heating source. The electrical resistance change of the sample can be used to determine the temperature rise, which can then be used to obtain the thermal diffusivity. For a non-conductive sample, a thin film of metal can be coated on the surface and used as the heater and resistance thermometer. However, the parasitic heat conduction in the metal coating needs to be accounted for, and the radial temperature gradient needs to be ignored.

Besides the electro-thermal and Raman based thermometry techniques that have been used for nanoscale thermal transport measurements, a bimaterial cantilever thermal sensor has been used to measure the thermal conductivity of polyethylene nanofibers.¹⁴ The nanofiber is drawn from a heated polymer gel by a tipless bimaterial atomic force microscopy cantilever, and was mechanically cut into 300 μm long samples. The free end of the nanofiber is attached to a microthermocouple and heated by an electrical heater. A laser is focused on the tip of cantilever to measure the thermally induced deflection of the cantilever, which is then used to obtain the heat flow from the fiber into the cantilever tip.

Although a number of experimental methods have been developed for probing the thermal properties of bulk materials, thin films and nanostructures, these methods are often not suitable for mesoscale samples with the dimension in the range between 1 μm and 1 mm. The motivation for this work is to develop a thermal transport measurement method for mesoscale samples. The recently reported four-probe measurement method for nanostructures is scaled up for thermal transport measurement of mesoscale samples.

The method is calibrated with silicon nitride samples, and used to measure thermal conductivity of polyethylene thin films. The errors in the mesoscale thermal transport measurements are analyzed.

Chapter 2 A Four-Probe Thermal Transport Measurement Method for Mesoscale Structures

A four-probe thermal transport measurement method for micro- and nano-scale rod or ribbon shape samples has been reported recently.¹¹ This chapter describes an investigation of the applicability of this method for mesoscale samples with a characteristic dimension in the range between 1 μm and 1 mm. Mesoscale four-probe measurement devices are designed, fabricated, and calibrated with a silicon nitride (SiN_x) sample. Various error sources in the measurements are evaluated.

2.1 Principle of Four-Probe Thermal Transport Measurement Method

The four-probe thermal transport measurement method reported recently can determine both the contact thermal resistances and the intrinsic thermal resistance of a rod or ribbon sample.¹¹ The measurement device consists of four suspended Pd/Cr/ SiN_x resistance thermometer lines, as shown in **Figure 2.1(a)**. The sample is suspended on top of the four lines. **Figure 2.1(b)** shows the thermal resistance circuit of the measurement device. During the measurement at each sample stage temperature, one of the four suspended thermometer lines is used as the heater line. The electrical resistance change of each line is measured to obtain the average temperature rise of each line. Based on the thermal resistance circuit, the following three sets of equations can be derived to solve for $R_{c,2}$, R_2 , $R_{c,3}$, $R_1 + R_{c,1}$, $R_3 + R_{c,4}$, where R_j is the thermal resistance of a suspended segment of the sample, and $R_{c,j}$ is the contact thermal resistances at one contact to the sample, as defined in the caption of **Figure 2.1(b)**.

$$\theta_{c,2,i} - \theta_{c,3,i} = Q_{2,i}R_{c,2} + (Q_{1,i} + Q_{2,i})R_2 - Q_{3,i}R_{c,3}, \text{ for } i = 1,2,3,4. \quad (2.1)$$

$$\theta_{c,1,i} - \theta_{c,2,i} = Q_{1,i}(R_1 + R_{c,1}) - Q_{2,i}R_{c,2}, \quad \text{for } i = 1,2,3,4. \quad (2.2)$$

$$\theta_{c,4,i} - \theta_{c,3,i} = Q_{4,i}(R_3 + R_{c,4}) - Q_{3,i}R_{c,3}, \quad \text{for } i = 1,2,3,4. \quad (2.3)$$

Figure 2.1. (a) Optical image of a four-probe measurement device with four 800-μm-long, 10-μm-wide suspended Pd/Cr/SiN_x thermometer lines. A SiN_x beam sample is suspended on top of the four thermometer lines. The black particle contaminates were deposited on the sample and device after the thermal measurement is completed and during the process to remove the sample from the chip carrier. (b) Thermal resistance circuit of the measurement device when the first thermometer line is electrically heated at a rate of $(IV)_1$. R_1 , R_2 , and R_3 are the thermal resistances of the left, middle, and right suspended segments of the suspended sample, respectively. $R_{c,j}$ is the contact thermal resistance between the j^{th} Pd/Cr/SiN_x line and the sample. $R_{b,j}$ is the thermal resistance of the j^{th} Pd/Cr/SiN_x resistance thermometer line. $\theta_{c,j,i}$ is the j^{th} Pd/Cr/SiN_x line temperature rise at the contact to the sample when the i^{th} line is used as the heater line. The temperature rise θ_0 at the two ends of each of the suspended Pd/Cr/SiN_x lines is assumed to be negligible.

Among the first set of four equations, the contact temperature rise $\theta_{c,j,i}$ and heat flow rate $Q_{j,i}$ into the sample from three lines must be known in order to solve for the three unknown thermal resistances $R_{c,2}$, R_2 and $R_{c,3}$. In the second and third sets of equations, one of the four equations can be used to obtain $R_1 + R_{c,1}$ and $R_3 + R_{c,4}$ based on the obtained $R_{c,2}$ and $R_{c,3}$ and the known contact temperature and heat flow rate in the four

lines.

The temperature rise $\theta_{c,j,i}$ of the thermometer line at the contact point can be obtained from the measured average temperature rise $\bar{\theta}_{j,i}$ when the i^{th} line is electrically heated. Similar to the work of Smith et al.,¹⁵ a finite contact width is considered in the following analysis, which neglects surface heat loss due to radiation, convection or conduction to residual gas molecules in the high-vacuum sample space. The radiation loss is considered in a subsequent section. For purely heat conduction along a thermometer line without the heating current, the temperature distribution is linear, as illustrated in **Figure 2.2(a)**, where the thermometer temperature within the contact width is assumed to be uniform. Thus, the contact point temperature ($\theta_{c,j,i}$) and the average temperature ($\bar{\theta}_{j,i}$) of the j^{th} line without the heating current are related according to

$$\theta_{c,j,i} = \frac{2L_j}{L_j + w_j} \bar{\theta}_{j,i} \quad , \quad \text{for } j \neq i \quad (2.4)$$

where L_j and w_j is the half-length and half-width of the j^{th} line, respectively.

$R_{b,j}$ is the thermal resistance of each thermometer line given as

$$R_{b,j} = \left(\frac{\kappa_j A_j}{L_j - w_j - d_j} + \frac{\kappa_j A_j}{L_j - w_j + d_j} \right)^{-1} = \frac{(L_j - w_j)^2 - d_j^2}{2\kappa_j A_j (L_j - w_j)} \quad (2.5)$$

where κ_j , A_j , and d_j are the effective thermal conductivity, cross-section area, and deviation of the center of the contact from the center of the thermometer line. For the heater line, the average temperature rise can be calculated as

$$\bar{\theta}_{i,i} = \frac{1}{2L_i} \int_{-L_i}^{L_i} \theta_{i,i}(x) dx \quad (2.6)$$

$$\bar{\theta}_{i,i} = \frac{1}{2L_i} \left(\int_{-L_i}^{d_i - w_i} \theta_{i,i,A}(x) dx + \int_{d_i - w_i}^{d_i + w_i} \theta_{c,i,i} dx + \int_{d_i + w_i}^{L_i} \theta_{i,i,B}(x) dx \right) \quad (2.7)$$

where $\theta_{i,i,A}$ and $\theta_{i,i,B}$ are the parabolic temperature distributions given below for each side of the suspended line:

$$\theta_{i,i,A}(x) = \frac{-(IV)_i}{4\kappa_i A_i L_i} x^2 + C_1 x + C_2 \quad (2.8)$$

$$C_1 = \frac{-(IV)_i}{4\kappa_i A_i L_i} [L_i - (d_i - w_i)] + \frac{\theta_{c,i,i}}{[L_i + (d_i - w_i)]} \quad (2.9)$$

$$C_2 = \frac{(IV)_i}{4\kappa_i A_i L_i} [L_i (d_i - w_i)] + \theta_{c,i,i} \frac{L_i}{[L_i + (d_i - w_i)]} \quad (2.10)$$

$$\theta_{i,i,B}(x) = \frac{-(IV)_i}{4\kappa_i A_i L_i} x^2 + D_1 x + D_2 \quad (2.11)$$

$$D_1 = \frac{(IV)_i}{4\kappa_i A_i L_i} [L_i + (d_i + w_i)] - \frac{\theta_{c,i,i}}{[L_i - (d_i + w_i)]} \quad (2.12)$$

$$D_2 = \frac{-(IV)_i}{4\kappa_i A_i L_i} [L_i (d_i + w_i)] + \theta_{c,i,i} \frac{L_i}{[L_i - (d_i + w_i)]} \quad (2.13)$$

Based on the temperature distribution given above, the contact temperature in the i^{th} heater line can be obtained as

$$\theta_{c,i,i} = \left(\frac{2L_i}{L_i + w_i} \right) \bar{\theta}_{i,i} - \frac{1}{6} (IV)_i R_{b,i} \frac{[(L_i - w_i)^2 (L_i^2 - 2L_i w_i + 3d_i^2 + w_i^2)]}{L_i (L_i + w_i) [(L_i - w_i)^2 - d_i^2]} \quad (2.14)$$

According to energy balance in the thermal circuit,

$$Q_{i,i} = - \sum_{\substack{j=1 \\ j \neq i}}^4 Q_{j,i} \quad (2.15)$$

$$Q_{j,i} = -\theta_{c,j,i} / R_{b,j} , \quad \text{for } j \neq i \quad (2.16)$$

These results can be combined to obtain

$$\sum_{\substack{j=1 \\ j \neq i}}^4 \frac{\theta_{c,j,i}}{R_{b,j}} = -\kappa_i A_i \frac{d\theta_{i,i,A}}{dx} \Big|_{x=d_i-w_i} + \kappa_i A_i \frac{d\theta_{i,i,B}}{dx} \Big|_{x=d_i+w_i} + (IV)_i \frac{w_i}{L_i} , \text{ for } i = 1, 2, 3, 4 \quad (2.17)$$

The temperature gradients in equation (2.17) can be obtained from equation (2.8)- (2.13) to yield

$$\sum_{j=1, j \neq i}^4 \frac{\theta_{c,j,i}}{R_{b,j}} = (IV)_i \frac{L_i + w_i}{2L_i} - \frac{\theta_{c,i,i}}{R_{b,i}} \quad (2.18)$$

Equation (2.4) and equation (2.18) can be combined to yield

$$\theta_{c,i,i} \equiv \theta_{i,i}(d_i - w_i < x < d_i + w_i) = R_{b,i}[(IV)_i \left(\frac{L_i + w_i}{2L_i}\right) - \sum_{j=1, j \neq i}^4 \left[\frac{\bar{\theta}_{j,i}}{R_{b,j}} \left(\frac{2L_j}{L_j + w_j}\right)\right]] \quad (2.19)$$

With equation (2.14) and (2.19), the $R_{b,j}$ can be obtained based on measured average temperature rise and dimensions of the suspended lines according to

$$\sum_{j=1}^4 \frac{\bar{\theta}_{j,i}/(IV)_i}{R_{b,j}} \frac{2L_j}{L_j + w_j} = \frac{2[(L_i^3 - w_i^3)(L_i - w_i) - 3L_i d_i^2 w_i]}{3L_i(L_i + w_i)[(L_i - w_i)^2 - d_i^2]}, \quad \text{for } i = 1, 2, 3, 4 \quad (2.20)$$

With the measured average temperature rise of the line and the calculated $R_{b,j}$, the contact temperature rise for the heater line ($\theta_{c,i,i}$) can be calculated from equation (2.19).

Figure 2.2. (a) Optical micrograph of a $0.44 \pm 0.01 \mu\text{m}$ thick, $9.5 \pm 0.1 \mu\text{m}$ wide SiN_x sample, assembled across four suspended Pd/Cr/SiN_x lines together with a schematic of the temperature profiles along the Pd/Cr/SiN_x heater line (i^{th} line) and one Pd/Cr/SiN_x resistance thermometer line (j^{th} line, $j \neq i$). The scale bar in the optical micrograph is $100 \mu\text{m}$. (b) A close-up view of the contact between the SiN_x sample with a Pd/Cr/SiN_x line. The scale bar is $20 \mu\text{m}$.

With equations (2.4 - 2.20), $\theta_{c,j,i}$ and $Q_{j,i}$, for $i, j = 1-4$ can be obtained from the measurement results. In the first set of four equations (2.1) of $R_{c,2}$, R_2 and $R_{c,3}$, there are four equations with three unknowns $R_{c,2}$, R_2 and $R_{c,3}$, and three equations are independent. Hence, the first set of equations of the thermal circuit can be solved. For this overdetermined system, the multiple linear regression analysis can be used to find the solution and to minimize the sum of the squares of the residuals made in the results of every single equation.¹⁶ The first set of four equations (2.1) can be represented in the matrix notation as follows:

$$Y = Xr \quad (2.21)$$

$$Y = \begin{bmatrix} y_1 \\ y_2 \\ y_3 \\ y_4 \end{bmatrix} \quad X = \begin{bmatrix} 1 & x_{1,1} & x_{1,2} \\ 1 & x_{2,1} & x_{2,2} \\ 1 & x_{3,1} & x_{3,2} \\ 1 & x_{4,1} & x_{4,2} \end{bmatrix} \quad r = \begin{bmatrix} R_2 \\ R_{c,2} \\ R_{c,3} \end{bmatrix} \quad (2.22)$$

where

$$y_i = \frac{\theta_{c,2,i} - \theta_{c,3,i}}{Q_{1,i} + Q_{2,i}} \quad (2.23)$$

$$x_{i,1} = \frac{Q_{2,i}}{Q_{1,i} + Q_{2,i}} \quad (2.24)$$

$$x_{i,2} = \frac{-Q_{3,i}}{Q_{1,i} + Q_{2,i}} \quad (2.25)$$

The solution is

$$r = \begin{bmatrix} R_2 \\ R_{c,2} \\ R_{c,3} \end{bmatrix} = (X'X)^{-1}X'Y \quad (2.26)$$

where X' represents the transpose of the matrix X , and $(X'X)^{-1}$ represents the matrix inverse of $X'X$. The results gives the best estimates for R_2 , $R_{c,2}$, $R_{c,3}$, based on four sets of measured $\theta_{c,j,i}$ and $Q_{j,i}$.

2.2 Design and Fabrication of Mesoscale Measurement Devices

In previous work of Kim et al. on four-probe measurements of micro- and nano structures, the separation between the two middle thermometer lines is smaller than 10 μm .¹¹ Consequently, these devices cannot be used to measure mesoscopic samples with a length considerably longer than 10 μm . New large-size four probe devices are designed and fabricated in this work for measuring mesoscopic samples. For measurements of different samples, the length of the suspended thermometer line is much longer than the contact width, and the length of the suspended segment of the sample is much larger than the sample width and thickness, so that one-dimensional heat conduction in the sample can be assumed.

According to an analysis similar to that reported in an earlier work,¹⁷ if the maximum applied temperature rise is about 5 K for measurements near room temperature, the largest sample thermal resistance or contact thermal resistance that can be measured would be about 100 times the thermal resistance of the suspended thermometer line, while the smallest sample or contact resistance that can be measured would be about 1% of the suspended line thermal resistance. For the devices fabricated in this work, the half-length of the suspended thermometer lines ranges from 100 μm to

400 μm , and the width of the line ranges from 6 μm to 10 μm . Consequently, the targeted thermal resistance of the thermometer line ranges from $5 \times 10^5 \text{ K W}^{-1}$ to $4 \times 10^6 \text{ K W}^{-1}$.

Hundreds of devices were fabricated together on a silicon wafers with a 500 nm thick low stress SiN_x film. As described in details in Ou's work,¹⁸ the fabrication process mainly includes two photolithography steps for metal deposition and SiN_x etching. In the metal deposition process, the suspended lines and metal pads for electrical connection consist of three layers. Ten nm thick Cr is deposited on 500 nm thick SiN_x followed by 60 nm thick Pd deposited on top. In the SiN_x etching process, windows are etched in the SiN_x layer, followed by wet etching of the silicon substrate to suspend the Pd/Cr/ SiN_x lines. After fabrication, the devices are examined with scanning electron microscopy (SEM) to ensure suspension of each line.

2.3 Device Calibration with SiN_x Beam Sample

For the measurements described in this thesis, the suspended thermometer lines of the four-probe devices are 600 – 800 μm long, and 10 μm wide. The spacing between the two middle lines is 150 μm or 200 μm . One of these devices has been calibrated with a long SiN_x beam sample.

The SiN_x beam sample was obtained from a suspended thermometer line of a mesoscale four-probe device, after the Pd layer and Cr layer were etched in diluted aqua regia and chromium etchant. The SiN_x sample was removed from the etched device and transferred to a new four-probe device with a micromanipulator. The measurement device was attached to a chip carrier with silver paint. Each metal pad of the suspended line was

wire-bonded to a pin in the chip carrier. The chip carrier was loaded into a high vacuum cryostat with two radiation shields to reduce the surface heat loss to gas molecules and radiation loss. The temperature inside the cryostat was stable before each measurement.

The thermal transport measurement was performed at a stage temperature of 350 K, 325 K and 300 K. The electrical resistance of the heater line was obtained from the voltage drop and current measured with a voltage amplifier and a current amplifier. For other three lines, the electrical resistance change was obtained with a sinusoidal sensing current and the measured voltage drop by a lock-in amplifier. The constant sinusoidal sensing current only changes the absolute temperature of the line by about 3×10^{-4} K, which has negligible effect on the temperature rise caused by the heating current in the heater line. Hence the sinusoidal sensing current does not influence the measurement result. After all the thermal measurements were completed, the sample was taken out from the cryostat for the dimension measurements.

2.3.1 Measurement Results of SiN_x

Optical microscopy and scanning electron microscopy (SEM) were used to measure the dimensions of the SiN_x sample at multiple locations. Parts of the measurement images are shown in **Figure 2.3**. The length and width of the middle suspended sample segment was found to be $160.9 \pm 0.9 \mu\text{m}$ and $9.5 \pm 0.1 \mu\text{m}$, respectively, based on the optic image. The thickness of the middle segment of the sample was found to be $0.44 \pm 0.01 \mu\text{m}$ based on SEM measurements at an 87° tilted

angle. With tilted SEM, it can be observed that all the Pd/Cr/SiN_x lines are suspended as shown in **Figure 2.3** (d).

Figure 2.3. Dimension measurements of SiN_x beam sample: (a) length measurements with optic microscopy (the scale bar is 100 μm), (b) width measurements with scanning electron microscopy, (c) thickness measurements with scanning electron microscopy, (d) tilted SEM measurement showing that the Pd/Cr/SiN_x lines are fully suspended and a particle attached to the end of the suspended SiN_x beam sample.

The electrical resistance rise of each line (ΔR_{ej}) in different heating current can be obtained by subtracting the electrical resistance measured at heating current $I = 0$ from

the measured electrical resistance data. **Figure 2.4** shows the measured electrical resistance rise ΔR_{ej} of four thermometer lines of SiN_x beam sample as a function of the heating current when Line 1 served as the heater line at stage temperature $T_0 = 350$ K.

Figure 2.4. Measured electrical resistance rises ΔR_{ej} of Line 1 ($j=1$, left Y axis), Line 2 ($j=2$, right Y axis), Line 3 ($j=3$, right Y axis), and Line 4 ($j=4$, right Y axis), as a function of the heating current I in Line 1 at sample stage temperature $T_0 = 350$ K for the SiN_x beam sample.

The measured electrical resistance at the sample stage temperature without electrical heating can be used to obtain the temperature (T) coefficient of resistance, which is needed to convert the measured electrical resistance rise at each different heating current to the temperature rise. In some previous works,^{11,19} a linear function of electrical resistance and temperature has often been used to fit the measurement results. However, according to the measurement data, linear fitting is inadequate to describe the curve R_{ej} versus T over the large temperature range of interest. Accordingly, assuming a

linear fit can lead to under-prediction of $dR_{e,j}/dT$ at low temperatures and over-prediction at high temperatures.

It is reported in some works that the Bloch-Gruneisen formula, which contains three adjustable parameters, is a much better description of the experimental curve of the experimental $R_{e,j}-T$ curve for many metals and their alloys.^{20,21} Based on the Bloch-Gruneisen formula, in this work $R_{e,j}$ vs T is fitted with a power law as equation (2.27), where a , n , and R_0 are three adjustable parameters, and γ is the relative error of the fitted data which can be obtained with equation (2.28).

$$R_{e,j} = aT^n + R_0, \quad \text{for } j=1,2,3,4 \quad (2.27)$$

$$\gamma = \frac{R_{e,j} - R_{e,fit}}{R_{e,j}} \quad (2.28)$$

Figure 2.5 shows the measured result of $R_{e,l}$ and the fitting curves using a linear relationship and a power law function, and relative errors in the fittings. In the small range of temperature from 300 K to 350 K, both fitting results contain errors smaller than 0.04%, while the linear fitting error is slightly larger than the power law fitting. In a wide range of temperature from 150 K to 350 K, which is shown in Chapter 3, power law fitting shows a smaller error than the linear in all the temperature of interest, which can better describe the temperature dependence of electrical resistance.

Based on the power law fitting of the measured electrical resistances of the four lines, the average temperature rise $\bar{\theta}_{j,i}$ of each suspended line can be obtained, as showed in **Figure 2.6** as a function of the heating current. The average temperature rise can be used to obtain the heat flow rate $Q_{j,i}$ from the line across the contact point to the sample,

shown in **Figure 2.7**, the thermal resistance $R_{b,j}$ of each line, shown in **Figure 2.8**, and temperature rise at the contact point.

Figure 2.5. Fitting of the electrical resistance $R_{e,l}$ of Line 1 in the SiN_x beam sample at $I = 0$. (a) Comparison between the measured electrical resistance $R_{e,l}$ (black circles) and the fits using a linear (red line) and power law (blue line) fitting. (b) Relative errors for the linear (red circles) and power law (blue squares) fits.

Figure 2.5 shows the measured result of $R_{e,l}$ and the fitting curves using a linear formula and a power law formula, and relative errors of the linear and power law fitting. In the small range of temperature from 300 K to 350 K, both fitting formulas have error lower than 0.04%, while the linear fitting error is slightly larger than the power law fitting. In a wide range of temperature from 150 K to 350 K, which is shown in Chapter 3, power law fitting has a smaller error in all the temperature of interest, which can better describe the temperature dependence of electrical resistance.

Based on the power law fitting of the measured electrical resistances of the four lines, the average temperature rise $\bar{\theta}_{j,i}$ of each suspended line can be obtained, as showed in **Figure 2.6** as a function of the heating current. The average temperature rise can be used to obtain the heat flow rate $Q_{j,i}$ from the line across the contact point to the sample,

shown in **Figure 2.7**, the thermal resistance R_{bj} of each line, shown in **Figure 2.8**, and temperature rise at the contact point.

Figure 2.6. Measured average temperature rise of Line 1 ($j=1$), Line 2 ($j=2$), Line 3 ($j=3$), and Line 4 ($j=4$) as a function of the heating current in Line1 when $T_0 = 350$ K for SiN_x beam sample.

Figure 2.7. Measured heat flow rate $Q_{j,1}$ into the sample from Line 1 ($j=1$), Line 2 ($j=2$), Line 3 ($j=3$), and Line 4 ($j=4$) as a function of heating current in Line 1 when $T_0=350$ K for SiN_x beam sample.

With 16 pairs of temperature rise and heat flow rate data, the thermal circuit can be solved to obtain $R_{c,2}$, R_2 , $R_{c,3}$ as shown in **Figure 2.8** together with the error bars obtained according to the uncertainty analysis method discussed in a subsequent section. The contact resistance at Line 2 and Line 3, ($R_{c,2}$ and $R_{c,3}$) are both smaller than the intrinsic thermal resistance of the SiN_x sample. The near zero value of $R_{c,2}$ indicates a good contact between Line 2 and the sample.

Figure 2.8. Measured intrinsic thermal resistances R_2 (blue circles), contact resistance $R_{c,2}$ (red squares), $R_{c,3}$ (yellow diamonds) and beam thermal resistance $R_{b,1}$ (white triangles), $R_{b,2}$ (white diamonds), $R_{b,3}$ (white circles), and $R_{b,4}$ (white squares) for SiN_x beam sample.

With the obtained intrinsic thermal resistance R_2 and the measured dimensions of the middle segment of each sample, the thermal conductivity values of three samples can be calculated and is shown in **Figure 2.9**. The measured thermal conductivity is

3.7- 3.8 W m⁻¹ K⁻¹ in the temperature range from 300 K to 350 K, which is comparable to the reported thermal conductivity values of SiN_x in other works.^{22, 23, 24}

The suspended thermometer lines of the device have three layers, 60 nm Pd deposited on 500 nm SiN_x and 10 nm Cr adhesion layer in between. The SiN_x layer thermal conductivity can be extracted based on the measured line thermal resistance R_{bj} and electrical resistance R_{ej} of Pd and Cr layer. According to Wiedemann-Franz law that relates the electrical resistivity and the electronic thermal conductivity of the metal layer, the thermal conductance of the metal layer can be obtained with

$$\text{Wiedemann-Franz law: } L_o = \frac{k_m}{\sigma T_m} \quad (2.29)$$

$$G_m = \frac{4}{R_e} L_o T_m \quad (2.30)$$

where L_o , k_m , σ , T_m , G_m , R_e is the Lorenz number, thermal conductivity of metal layer, electrical conductivity, temperature, thermal conductance of metal layer, measured electrical resistance, respectively. The factor of 4 in the second equation accounts for the fact that the electrical current for electrical resistance measurement flows in series and heat current flows in parallel in the two segments of the thermometer line. The Lorenz number has been reported to be about $2.7 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$ for Palladium near 350 K based on the measured electrical resistivity and the total thermal conductivity.²⁵ This experimental value is slightly higher than the Sommerfeld's theoretical result because the measured thermal conductivity includes an additional lattice contribution in addition to the electronic contribution described by the Sommerfeld value. The thermal conductance (G_{SiN_x}) and thermal conductivity (k_{SiN_x}) of the SiN_x layer can be found according to the

calculated thermal conductance of metal layer and the measured thermal resistance of the thermometer line, namely

$$G_{SiNx} = \frac{1}{R_b} - G_m \quad (2.31)$$

$$k_{SiNx} = \frac{lG_{SiNx}}{4A_{SiNx}} \quad (2.32)$$

where l and A_{SiNx} are the length of the suspended line and cross-section area of the SiN_x layer.

Figure 2.9. Measured thermal conductivity of SiN_x beam sample with the error bars smaller than the symbol size (blue circles). Shown in comparison is the extracted thermal conductivity of SiN_x layer of the four lines in the measurement device (squares).

Based on the measured line thermal resistance R_b , electrical resistance R_e , length, width of the SiN_x layer measured with optic microscopy and thickness measured with SEM, the SiN_x thermal conductivity is found to be in the range between 3.5 and

4.2 W m⁻¹ K⁻¹ for four different lines of the measurement device in the temperature range from 300 K to 350 K. As shown in **Figure 2.9**, the extracted thermal conductivity values of SiN_x are consistent with the measured thermal conductivity of the SiN_x sample in this work, and published data of SiN_x thermal conductivity.²²⁻²⁴ Some extracted thermal conductivity values of the SiN_x layer of the thermometer lines are slightly higher than the directly measured thermal conductivity of the SiN_x sample, likely due to the deviation of the actual Lorenz number from the literature Lorenz number used in the calculation.

2.3.2 Error Analysis

2.3.2.1 Background thermal conductance

In the aforementioned heat transfer analysis to determine the temperature distribution of the suspended beams, the background thermal conductance is not included. The background thermal conductance is due to radiation and conduction through the residual gas molecules in the cryostat, and more importantly, heat conduction in the substrate, which is not a perfect heat sink and can exhibit temperature rise at the junction to the heating line. This background thermal conductance of the device itself can lead to an overestimate of the thermal conductivity of the sample.

We have employed the four-probe measurement method to measure a blank device without a sample assembled on the device in order to evaluate the background thermal conductance. The blank device has the same dimensions as the device used to measure the polyethylene sample 3 discussed in Chapter 3. The obtained average temperature rise of each line in the blank device is compared with those in sample 3

device, as shown in **Figure 2.10**. The temperature rises of the heater lines are similar in the blank device and the sample 3 device. For the suspended lines not electrically heated, it is obvious that the temperature rises in the blank device are much smaller than those in sample 3, and are close to the measurement noise limited by the 10~50 mK temperature fluctuation of the cryostat sample stage and the measurement equipment, similar to an earlier measurement that used serpentine Pt resistance thermometer devices.^{19,26} This measurement result verifies that the background thermal conductance is negligible in these devices.

Figure 2.10. Measured average temperature rise of thermometer lines as a function of heating current applied in Line1 when $T_0 = 325$ K in the blank device ($j=1_b, j=2_b, j=3_b, j=4_b$) and in the polyethylene sample 3 device ($j=1_{s3}, j=2_{s3}, j=3_{s3}, j=4_{s3}$). Heater lines ($j=1_b, j=1_{s3}$) are with respect to left Y axis, other lines are with respect to right Y axis.

2.3.2.2 Spreading thermal resistance of the substrate

The temperature distribution of the suspended lines is calculated under the assumption that the silicon substrate temperature is uniform and the temperature rise at the junction between the line and silicon substrate is negligible. Here, the spreading thermal resistance can be calculated based on the conduction shape factor of a small area in contact with half of a semi-infinite medium as

$$R_{Si} = \frac{2}{Sk_{Si}} \quad (2.33)$$

where R_{Si} , S , k_{Si} is the spreading thermal resistance, shape factor, thermal conductivity of silicon substrate, respectively.²⁷ The shape factor S depends on the hydraulic diameter D of the small junction area between the Pd/Cr/SiN_x beam and the silicon substrate according to

$$S = 2D \quad (2.34)$$

The theoretical calculation results of the spreading thermal resistance in the silicon substrate is calculated to be about $2.88 \times 10^3 \text{ K W}^{-1}$, which is three orders of magnitude lower than measured thermal resistance $4 \times 10^6 \text{ K W}^{-1}$ of the suspended thermometer line. Thus, the temperature rise at the junction is negligible compared to the contact point temperature rise. Hence, it is reasonable to assume that the temperature distribution in the silicon substrate is uniform and temperature rise at the junction between the suspended line and silicon substrate is negligible.

2.3.2.3 Radiation loss of suspended thermometer lines and the sample

During the measurement, the heat is conducted through the suspended lines to the silicon substrate from the sample, as well as lost via radiation and conduction to residual

gas molecules from the surface of the long suspended beams. The measurement is performed in high vacuum and with two radiation shields to minimize the heat loss to the residual gas molecules and radiation loss. Although the radiation error has been found to be negligible for prior four-probe measurements of micro- and nano- scale samples, the radiation error is expected increase with the sample length for a fixed aspect ratio.²⁸ Hence, the radiation error of the new four-probe devices for mesoscale samples is examined here.

For a suspended line without the electrical heating current and with uniform cross-section and negligible temperature rise at junction to the silicon substrate, the temperature distribution can be calculated as

$$\frac{\theta_{j,i}(x)}{\theta_{c,j,i}} = \frac{(\theta(L_j)/\theta_{c,j,i}) \sinh mx + \sinh m(L_j-x)}{\sinh mL_j}, \quad \text{for } j \neq i \quad (2.35)$$

$$m = \sqrt{4\varepsilon_j \sigma T_0^3 P / k_j A_j} \quad (2.36)$$

where x is the distance from the contact point of the line and the sample, $\theta_{j,i}(x)$ is the difference between the local temperature of the beam and the sample stage temperature, and $\theta_{c,j,i}$ is the difference between the temperature at the contact point and the stage temperature.²⁷ In addition, k_j , L_j , P_j , A_j , ε_j and σ are the thermal conductivity, half length, perimeter of the cross section, and cross section area, surface emissivity of the j^{th} suspended line and the Stephan-Boltzmann constant. T_0 is the device stage temperature.

Based on the temperature distribution, the local fin heat conduction rate can be derived as

$$q_c(x) = k_j A_j \frac{\partial \theta_{j,i}(x)}{\partial x} = m k_j A_j \theta_{c,j,i} \frac{\cosh m(L_j - x)}{\sinh mL_j} \quad (2.37)$$

Hence, the heat conduction rates at $x = L_j$ and $x = 0$, which are the heat conduction from the sample to the suspended line and from the suspended line to the silicon substrate, respectively, can be calculated

$$q_c(L_j) = m k_j A_j \theta_{c,j,i} \frac{\cosh mL_j}{\sinh mL_j} \quad (2.38)$$

$$q_c(0) = m k_j A_j \theta_{c,j,i} \frac{1}{\sinh mL_j} \quad (2.39)$$

The ratio between the radiation loss from the surface of the suspended beam and the heat conduction into the substrate can be found to be

$$\beta_1 = \frac{q_c(L_j) - q_c(0)}{q_c(0)} = \cosh mL_j - 1 \quad (2.40)$$

In addition, with the surface radiation loss accounted for, the fin thermal resistance can be calculated as

$$R_f = (4\varepsilon_j \sigma T_0^3 P_j k_j A_j)^{-1/2} \tanh mL_j \quad (2.41)$$

In the limit of vanishing mL_j ,²⁸ corresponding to negligible radiation loss, the thermal resistance reduces to the expression for conduction thermal resistance as

$$R_{cond} = 4L_j / \pi k d^2 \quad (2.42)$$

The relative error in neglecting the radiation loss of the suspended beam is

$$\beta_2 = \frac{R_{cond} - R_f}{R_f} = mL_j \coth mL_j - 1 \quad (2.43)$$

In this work, the suspended lines of the measurement devices used in this work are 600 μm or 800 μm long, and 10 μm wide. For the suspended line consisting of 60 nm thick Palladium layer and 0.44 μm thick SiN_x layer,

$$k_j A_j = k_{j,Pd} A_{j,Pd} + k_{j,SiNx} A_{j,SiNx} \quad (2.44)$$

$$\varepsilon_j P_j = \varepsilon_{j,Pd} P_{j,Pd} + \varepsilon_{j,SiNx} P_{j,SiNx} \quad (2.45)$$

For the palladium layer, the emissivity is found to be 0.008 in the near infrared spectrum range close to $\lambda = 10 \mu\text{m}$.²⁹ According to Wiedemann-Franz law that relates the electrical resistivity and the electronic thermal conductivity of the metal layer, with measured electrical resistance and reported Lorenz number for palladium,²⁵ the $(k_{j,Pd} A_{j,Pd})$ can be obtained as

$$k_{j,Pd} A_{j,Pd} = \frac{G_m L_j}{4} = \frac{L_j}{R_e} L_o T_m \quad (2.46)$$

where L_o , T_m , G_m , and R_b are the Lorenz number, temperature, thermal conductance of metal layer, measured electrical resistance, respectively. As discussed in a preceding section, the factor of 4 in the equation accounts for the fact that the electrical current for electrical resistance measurement flows in series and heat current flows in parallel in the two segments of the thermometer line. The thickness of the Pd layer has not been measured accurately. According to the thickness reading in the metal evaporator, the thickness of the deposited Pd is about 60 nm. The width is measured to be 9.8 μm . Hence, the thermal conductivity of Pd is found to be around 53 $\text{W m}^{-1} \text{K}^{-1}$, which is smaller than the reported value of 72 $\text{W m}^{-1} \text{K}^{-1}$ for bulk Pd.²⁹ The suppressed thermal conductivity is due to the smaller grain size in the thin film of Pd than the bulk Pd.

The thermal conductivity of the SiN_x layer is 3.7 $\text{W m}^{-1} \text{K}^{-1}$ based on the measurement result of the SiN_x beam sample at 350 K in this work. According to Kirchhoff's law of thermal radiation, the emissivity can be assumed to be equal to the

absorptivity in thermodynamic equilibrium. For the semi-transparent thin film SiN_x layer, the optical properties can be calculated as,³⁰

$$\alpha = \frac{(1-\rho)(1-\tau)}{(1-\rho\tau)} \quad (2.47)$$

$$\rho = \left(\frac{n_2 - n_1}{n_2 + n_1} \right)^2 \quad (2.48)$$

$$\tau = \exp(-\kappa_\lambda t) \quad (2.49)$$

$$\kappa_\lambda = \frac{4\pi k_\lambda}{\lambda} \quad (2.50)$$

where α, ρ, τ and κ_λ are the absorptance, reflectance, transmissivity, absorption coefficient of SiN_x thin beam. In addition, n_1, n_2, k_λ are the refractive index of vacuum, the real part of the refractive index and the imaginary part or extinction index of SiN_x , respectively, at wavelength λ . In the near infrared spectrum range close to $\lambda = 10 \mu\text{m}$, it has been reported that $k_\lambda = 1.7$ and $n_2 = 1.1$ for SiN_x .³¹ Hence, the emissivity of the $0.44 \mu\text{m}$ thick SiN_x thin layer is calculated to be about 0.61.

According to equation (2.40), the ratio between radiation loss from the circumference of the suspended line and heat conduction into the substrate is found to be about 6.0% and 10.9% for the $600 \mu\text{m}$ and $800 \mu\text{m}$ long suspended lines, respectively. In addition, according to equation (2.43), the deviation of the conduction thermal resistance from the fin thermal resistance is 4.0% and 7.0% for the $600 \mu\text{m}$ and $800 \mu\text{m}$ long suspended lines, respectively.

The length of the mesoscale suspended lines of these devices is much longer than that used in previous works for nano- and micro- structures, leading to a larger radiation

loss error. **Figure 2.11** shows the relative error calculated with the same aspect ratio $L_j/w_j=100$ of the suspended lines. For the suspended lines with the same aspect ratio, the radiation loss ratios calculated from both methods increase with increasing L_j ,²⁸ while the thermal resistance is kept constant. If the length is reduced from 800 μm to 200 μm , and the width is reduced from 8 μm to 2 μm , the relative radiation error can be reduced from 11.0% to below 1.0% with equation (2.40) and from 7.0% to 0.5% with equation (2.43). Thus, the mesoscale measurement devices can be designed with shorter and narrower suspended lines to reduce the radiation loss while the method can still be used to measure a sample with the width below about 10 μm , which needs to be smaller than the length of the suspended line by a factor of 20 or more.

Figure 2.11. Relative error calculated with the same aspect ratio $L_j/w_j=100$ of the suspended lines, where $2L_j$ is the total length of the thermometer line. β_1 is the ratio between radiation loss from the circumference of the suspended line and heat conduction into the substrate. β_2 is the deviation of the conduction thermal resistance from the fin thermal resistance.

In addition, it is assumed in the aforementioned heat conduction analysis that the temperature distribution along a thermometer line without the heating current is linear. **Figure 2.12** shows the calculation result of the temperature distribution along the suspended lines in a dimensionless form based on equation (2.35) for a 400 μm half-length and 10 μm wide suspended line. The fin temperature profile can be fitted with a linear curve with a R-square value as high as 0.99981. Thus, it is reasonable to assume a linear temperature distribution along the beam.

Figure 2.12. Calculated temperature distribution along the suspended line.

Moreover, the spatially averaged temperatures rise along the suspended line without the applied electrical heating current can be calculated as,²⁸

$$\frac{\overline{\theta_{j,i}(x)}}{\theta_{c,j,i}} = \frac{1}{L} \int_0^{L_j} \frac{\theta(x)}{\theta_{c,j,i}} dx = \frac{1}{mL_j} \frac{\cosh mL_j - 1}{\sinh mL_j}, \quad \text{for } j \neq i \quad (2.51)$$

The ratio between the averaged temperature rise and contact temperature rise is calculated to be 0.491 with the calculated mL_j for a 400 μm half-length and 10 μm wide suspended line, which is close to 0.5 corresponding to a linear temperature distribution. Thus, it is reasonable to assume a linear temperature distribution along the beam.

In addition to heat loss from the suspended lines, radiation loss from the surface of the sample can result in an underestimate of the thermal conductance of the sample. For a low thermal conductance sample in which $\theta_L \ll \theta_0$, the relative error in the measured thermal conductance when neglecting radiation loss of the sample can be calculated with $\eta = mL \coth mL - 1$. With the obtained emissivity, measured thermal conductivity and dimensions of the SiN_x beam sample, the relative error in neglecting radiation loss from the SiN_x beam sample is found to be 4.0%, 5.2%, and 6.4% at temperature 300 K, 325 K, 350 K, respectively.

2.3.2.4 Uncertainty from dimension measurements and sample thermal resistance

To obtain the thermal conductivity of measured samples, measurements are made to obtain the sample length L , width w , thickness t , and thermal resistance R_2 . All the variables are related to the middle segment of the sample.

$$k = \frac{L}{wtR_2} \quad (2.52)$$

The error in the result δk is given by the error in the measured individual variable δL , δw , δt , δR_2 and sensitivity coefficient $\partial k / \partial L$, $\partial k / \partial w$, $\partial k / \partial t$, $\partial k / \partial R_2$.

$$\delta k = \sqrt{\left[\left(\frac{\partial k}{\partial L} \delta L\right)^2 + \left(\frac{\partial k}{\partial w} \delta w\right)^2 + \left(\frac{\partial k}{\partial t} \delta t\right)^2 + \left(\frac{\partial k}{\partial R_2} \delta R_2\right)^2\right]} \quad (2.53)$$

$$\frac{\partial k}{\partial L} = \frac{1}{wtR_2} \quad (2.54)$$

$$\frac{\partial k}{\partial w} = -\frac{L}{w^2 t R_2}$$

$$\frac{\partial k}{\partial t} = -\frac{L}{t^2 w R_2}$$

$$\frac{\partial k}{\partial R_2} = -\frac{L}{R_2^2 t w}$$

Precision errors of L , w and t in multiple measurements are estimated to be proportional to the standard deviation σP of N measurements results of a variable as equation (2.55), where S is the student t-distribution with 95% confidence and $(N-1)$ degree of freedom:

$$\delta P = \frac{\sigma P}{\sqrt{N}} S \quad (2.55)$$

In order to determine the error in R_2 , both the random error and radiation error should be considered. The least square regression method are used to find the approximate solution of the first set of four equations (2.1) of $R_{c,2}$, R_2 and $R_{c,3}$. In order to find the random uncertainties in the estimates of $R_{c,2}$, R_2 and $R_{c,3}$, the uncertainty in y_i need to be found as

$$\sigma_y^2 = \frac{Y'(I - X(X'X)^{-1}X')Y}{N-3} \quad (2.56)$$

where N is the number of measurements, and $(N-3)$ is degrees of freedom.¹⁶ With N measurements to find three unknowns, so we have $(N-3)$ degrees of freedom left. Here $N=4$ as we use four sets of measurement results of $\theta_{c,j,i}$ and $Q_{j,i}$.

The least square estimates are unbiased estimators of R_2 , $R_{c,2}$, and $R_{c,3}$, the variance-covariance matrix of the estimated solutions is obtained as follows:

$$C = \sigma_y^2 (X'X)^{-1} \quad (2.57)$$

$$\sigma_{r_{i,random}} = \sqrt{C_{ii}}, \quad \text{for } i=1,2,3 \quad (2.58)$$

$$r_1 = R_2 \quad r_2 = R_{c,2} \quad r_3 = R_{c,3}$$

where C is a symmetric matrix whose diagonal elements, C_{ii} , represent the uncertainty of the estimated R_2 , $R_{c,2}$ and $R_{c,3}$. With the calculated random error and radiation error, the error in R_2 can be obtained as

$$\delta R_2 = \sqrt{\sigma_{R_2,random}^2 + \sigma_{radiation}^2} \quad (2.59)$$

In the second and third sets of four equations of $R_l + R_{c,l}$ and $R_3 + R_{c,4}$, four values can be yielded with four equations based on the results of $R_{c,2}$ and $R_{c,3}$, respectively. Due to the presence of errors in the measurement, there is variation between the four values of each thermal resistance, which can be used to evaluate the random uncertainty of $R_l + R_{c,l}$ and $R_3 + R_{c,4}$, respectively.

To study the temperature dependence of thermal resistance, the temperature uncertainty is needed. The minimum temperature is the temperature of the cryostat without heating the suspended lines and the maximum temperature is the highest contact temperature of the middle segment of sample reached when heating one of the suspended lines.

$$T_{average} = T_{cryostat} + \frac{MAX[\theta_{c,j,i}]}{2} \quad (2.60)$$

$$T_{min} = T_{cryostat} \quad (2.61)$$

$$T_{max} = T_{cryostat} + MAX[\theta_{c,j,i}] \quad (2.62)$$

Chapter 3 Four-Probe Thermal Transport Measurements of Polyethylene

3.1 Sample Assembly

In this work, polyethylene film samples were measured with the mesoscale measurement devices. The polyethylene samples were provided by Dr. Yanfei Xu at Massachusetts Institute of Technology. The polyethylene came as a film shape with a micrometer scale thickness, width around 5 mm, and length around 30 cm. The drawn ratio of the polyethylene fiber is specified as 90. The sample is expected to have a higher thermal conductivity along the drawn direction. The melting temperature of the polyethylene film starts at 120°C, so higher temperatures must be avoided during sample preparation and measurement processes.

3.1.1 Sample Transfer Method

For transferring the polyethylene sample to a four-probe device, the polyethylene film was cut with a new blade perpendicularly to the length direction into short pieces. Two tweezers with sharp tips were then used to tear the short pieces into narrow ribbons along the length direction under the optical microscope, meanwhile the fiber was also cut with the sharp tweezers into short pieces to match the measurement devices size. A micromanipulator outfitted with a sharp probe was utilized to selectively pick up and transfer the thin fiber to the four-probe device. Because of the high stiffness of the fiber, the fiber was not tightly attached to the suspended lines, so that the contact thermal resistance between the sample and the suspend lines can be large. In order to address this

contact issue, once the ribbon was placed on the four lines, the contact points of the lines and the sample ribbon were pressed gently with the micromanipulator probe to improve the contact. In some samples, a small amount of poly(methyl methacrylate) (PMMA), which is sticky, was applied to the center point of the suspended lines with a sharp probe so as to attach the ribbon and the metal line together. Since the PMMA was applied only to the metal line and the influence of the PMMA was included in the contact resistance, this method did not influence the thermal conductivity measurement results of the suspended middle segment of the polyethylene sample.

3.1.2 Sample Characterization

Three polyethylene ribbon samples were measured in this work. Thermal measurement of each sample was performed in the temperature range from 150 K to 350 K, which is below the melting temperature of polyethylene. After thermal measurements, optical microscopy was used to measure the length and width of the middle segment of samples. For sample 1 and sample 2, the middle segment is twisted so the width of the sample was measured after the sample was transferred to a silicon substrate from the device. The width of sample 3 was measured on the device with the optic microscope. Atomic force microscopy (AFM) was used to measure the thickness of the sample after the sample was transferred to a silicon substrate for all three samples. The AFM measurement data can also give the width of the sample, which is consistent with the measured result from the optic microscopy. All the dimensions were measured more than 3 times. The average length, width and thickness of the middle segments of the

three samples are listed in **Table 3.1** together with their uncertainties. Only parts of the dimension measurements results are shown in **Figure 3.1-3.4**. Attempts were also made to use scanning electron microscopy (SEM) to visualize some other samples not included in this thesis. However, the polymer sample was easily destroyed by the electron beam and started to melt quickly upon exposure to the electron beam.

	Length (μm)	Width (μm)	Thickness (nm)
Sample 1	165.2 ± 1.2	3.98 ± 0.34	1084 ± 38
Sample 2	207.2 ± 7.3	3.74 ± 0.13	1053 ± 24
Sample 3	166.7 ± 4.5	3.02 ± 0.06	1052 ± 34

Table 3.1. Sample dimensions for measured polyethylene sample 1, 2, and 3

Figure 3.1. Top-down optical microscopy images of measured sample 1(a), sample 2 (b), and sample 3 (c). Scale bars are 100 μm for (a), (b), (c).

Figure 3.2. Optical microscopy results used to measure the length of sample 1 (a), sample 2 (b), and sample 3 (c). Scale bars are 50 μm for (a), (b), (c).

Figure 3.3. Optical microscopy results used to measure the width of sample 1 (a) on silicon substrate, sample 2 (b) on silicon substrate, and sample 3 (c) on the measurement device. Scale bars are 20 μm for (a) and (c), 10 μm for (b).

Figure 3.4. Atomic force microscopy data used to measure the thickness and width of sample 1 (a), sample 2 (b), and sample 3 (c) on silicon substrates.

3.2 Measurement Results

Figure 3.5 shows the measured electrical resistance rise ΔR_{ej} of four thermometer lines of sample 1 as a function of the heating current when Line 1 served as the heater line at stage temperature 350 K. The measured electrical resistance at the sample stage temperature without heating is used to obtain the temperature coefficient of resistance so as to convert the measured electrical resistance change to the temperature rise. **Figure 3.6** shows the measured R_{ej} of Line 1 in sample 1 and the fitting curves using a linear relationship and a power law function, and relative errors of the linear and power law fitting. The linear fitting has a larger error in the range of temperature from 150 K to 350 K than the power law fitting, which can better describe the temperature dependence

of the electrical resistance. The measured electrical resistance of each suspended line in each sample is fitted with different power law coefficients.

Figure 3.5. Measured electrical resistance rises $\Delta R_{e,j}$ of Line 1 ($j=1$, left Y axis), Line 2 ($j=2$), Line 3 ($j=3$, right Y axis), and Line 4 ($j=4$, right Y axis), as a function of the heating current I in Line 1 at sample stage temperature $T_0 = 350$ K for sample 1.

The average temperature rise $\bar{\theta}_{j,i}$ of each suspended line was obtained based on the temperature coefficient of resistance, as showed in **Figure 3.6** as a function of the heating current. The average temperature rise was used to obtain the heat flow rate $Q_{j,i}$ from the line across the contact point to the sample, shown in **Figure 3.7**, the thermal resistance $R_{b,j}$ of each line, shown in **Figure 3.8**, and temperature rise at the contact point.

Figure 3.6 Fitting of the electrical resistance $R_{e,l}$ of Line 1 in sample 1 when $I = 0$. (a) Comparison between the measured electrical resistance $R_{e,l}$ (black circles) and the fits using a linear (red line) and power law (blue line) formula. (b) Relative errors for the linear (red circles) and power law (blue squares) fits.

Figure 3.7. Measured average temperature rise of Line 1 ($j=1$, left Y axis), Line 2 ($j=2$, right Y axis), Line 3 ($j=3$, right Y axis), and Line 4 ($j=4$, right Y axis) as a function of the heating current in Line1 when $T_0 = 350$ K for sample 1.

Figure 3.8. Measured heat flow rate $Q_{j,1}$ into the sample from Line 1 ($j=1$), Line 2 ($j=2$), Line 3 ($j=3$), and Line 4 ($j=4$) as a function of heating current in Line 1 when $T_0=350$ K for sample 1.

The measurement results of $R_{c,2}$, R_2 , $R_{c,3}$ are shown in **Figure 3.9** together with the error bars obtained according to the uncertainty analysis method discussed in Chapter 2. These three samples have different thermal contact resistance ($R_{c,2}$ and $R_{c,3}$) values. For sample 1, both $R_{c,2}$ and $R_{c,3}$ are about half of the value of the intrinsic thermal resistance R_2 of the middle segment. For sample 2, $R_{c,2}$ and $R_{c,3}$ are much smaller than R_2 , and the value around zero indicates a good thermal contact between the sample and suspended lines. For sample 3, $R_{c,2}$ is much higher than $R_{c,3}$, which is comparable to R_2 . The variation of the contact resistance value from a contact point to another contact point due to the limited capability to control the contact condition during the sample transfer process. The applied PMMA on the contact points can reduce the contact resistance, but the process cannot be precisely controlled.

Figure 3.9. Measured intrinsic thermal resistances R_2 (blue circles), contact resistance $R_{c,2}$ (red squares), $R_{c,3}$ (yellow diamonds) and beam thermal resistance $R_{b,1}$ (white triangles), $R_{b,2}$ (white diamonds), $R_{b,3}$ (white circles), and $R_{b,4}$ (white squares) for sample 1 (a), sample 2 (b), and sample 3 (c).

With the obtained intrinsic thermal resistance R_2 and the measured dimensions of the middle segment of each sample, the thermal conductivity of three samples can be calculated and is shown in **Figure 3.10**. All three samples show thermal conductivity values in the range between $4 \text{ W m}^{-1} \text{ K}^{-1}$ and $6 \text{ W m}^{-1} \text{ K}^{-1}$ in the temperature range from 150 K to 350 K. The peaks of the thermal conductivity appear at around 275 K for all three samples. The decreasing thermal conductivity with increasing temperature reveals the effect of lattice anharmonicity higher than 275 K.³²

Figure 3.10. Measured thermal conductivity of Polyethylene sample 1 (red circles), sample 2 (green squares) and sample 3 (blue diamonds).

Radiation loss from the circumference of the polyethylene samples can result in an underestimate of the thermal conductance of the samples. The radiation loss of the polyethylene samples can be evaluated with fin heat transfer analysis similar as discussed in Chapter 2. Due to the very low emissivity reported for polyethylene (< 0.1),¹⁴ the relative error in neglecting radiation loss from the surface of polyethylene ribbon samples is found to be below 0.6% for all three samples at temperature 300 K, 325 K, 350 K, respectively.

Chapter 4 Summary and Outlook

New four probe devices are designed and fabricated in this work for thermal transport measurements of mesoscopic samples. The scaled up measured device was calibrated with a SiN_x beam sample. The measured SiN_x thermal conductivity results are in agreement with the literature values. In addition, consistent thermal conductivity values of the SiN_x layer have also been obtained based on the measured thermal and electrical conductance of the Pd/Cr/SiN_x beam and the Wiedemann-Franz law. A number of measurement errors are analyzed. First of all, the background thermal conductance of the measurement device is found to be negligible based on the measurement of a blank device without a sample bridging the thermometer lines. The spreading resistance of the substrate is found to be three orders of magnitude smaller than the thermal resistance of the thermometer lines, so that substrate temperature rise can be neglected. The relative errors due to radiation loss of the thermometer lines and mesoscale samples increases with the length for the same aspect ratio, and are within 10.9% for the 600-800 μm long suspended thermometer lines and 6.4% for the 160.9 μm long SiN_x calibration sample. The length and width of the thermometer lines of the measurement device can be reduced toward 200 μm and 2 μm to reduce the radiation loss error to below 1.0% for the line.

The mesoscale four probe devices have also been applied to perform thermal transport measurements of polyethylene thin film samples in the temperature range from 150 K to 350 K. Three polyethylene ribbon samples with different dimensions have been transferred with micromanipulator to the measurement devices. Optical microscopy and

atomic force microscopy have been used to determine the sample dimensions. In this work, a power law fitting method is used to fit the measured electrical resistances versus temperature data of the thermometer lines, in order to reduce the error compared to the linear fitting method. Due to the low emissivity of polyethylene, the radiation loss from the sample results in 0.6% error in the measured thermal resistance. The measurement result shows that the contact resistances in the contacts between suspended lines and three samples are not negligible and can vary considerably among the different contacts. The thermal conductivity values obtained from the measured intrinsic thermal resistances and sample dimensions ranges between $4 \text{ W m}^{-1} \text{ K}^{-1}$ and $6 \text{ W m}^{-1} \text{ K}^{-1}$ at the temperature range between 150K and 350K. The peak thermal conductivity appears at near 275K for all the three samples. The decreasing thermal conductivity with increasing temperature above 275 K reveals the effect of lattice anharmonicity.

These results show that the mesoscale four-probe thermal transport measurement method developed in this work can independently probe the contact thermal resistance and intrinsic thermal resistance of meso-scale structures. The measurement errors are mainly caused by radiation heat loss from the thermometer lines and the sample. The radiation loss from the thermometer lines can be reduced by designing thermometer lines with the length and width reduced toward $200 \text{ }\mu\text{m}$ and $2 \text{ }\mu\text{m}$, respectively. Thus, this mesoscale four-probe thermal transport measurement method can fill the gap between thermal transport measurements of bulk materials and nano-structures and provide experimental data of mesoscale structures.

References

- [1] Zhao, Dongliang, et al. "Measurement techniques for thermal conductivity and interfacial thermal conductance of bulk and thin film materials." *Journal of Electronic Packaging* 138.4 (2016): 040802.
- [2] Astm, C. "177-85. Standard test method for steady-state heat flux measurements and thermal transmission properties by means of the guarded hot plate apparatus." *Annual Book of ASTM Standards* 4.06 (1992): 21-42.
- [3] Salmon, David. "Thermal conductivity of insulations using guarded hot plates, including recent developments and sources of reference materials." *Measurement Science and Technology* 12.12 (2001): R89.
- [4] Yan, Jiaqiang. *A study of bond-length fluctuations in transition metal oxides*. Diss. 2004.
- [5] Min, S., J. Blumm, and A. Lindemann. "A new laser flash system for measurement of the thermophysical properties." *Thermochimica Acta* 455.1 (2007): 46-49.
- [6] Cahill, David G., et al. "Thermal conductivity of thin films: measurements and understanding." *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films* 7.3 (1989): 1259-1266.
- [7] Schmidt, Aaron J., Ramez Cheaito, and Matteo Chiesa. "A frequency-domain thermoreflectance method for the characterization of thermal properties." *Review of scientific instruments* 80.9 (2009): 094901.

- [8] Cahill, David G. "Analysis of heat flow in layered structures for time-domain thermoreflectance." *Review of scientific instruments* 75.12 (2004): 5119-5122.
- [9] Weathers, Annie, and Li Shi. "Thermal transport measurement techniques for nanowires and nanotubes." *Annual Review of Heat Transfer* 16.16 (2013).
- [10] Mavrokefalos, Anastassios, et al. "Four-probe measurements of the in-plane thermoelectric properties of nanofilms." *Review of scientific instruments* 78.3 (2007): 034901.
- [11] Kim, Jaehyun, et al. "A four-probe thermal transport measurement method for nanostructures." *Review of Scientific Instruments* 86.4 (2015): 044901.
- [12] Li, Qingwei, et al. "Measuring the thermal conductivity of individual carbon nanotubes by the Raman shift method." *Nanotechnology* 20.14 (2009): 145702.
- [13] Guo, Jiaqi, Xinwei Wang, and Tao Wang. "Thermal characterization of microscale conductive and nonconductive wires using transient electrothermal technique." *Journal of Applied Physics* 101.6 (2007): 063537.
- [14] Shen, Sheng, et al. "Polyethylene nanofibres with very high thermal conductivities." *Nature nanotechnology* 5.4 (2010): 251-255.
- [15] Smith, Brandon, et al. "Temperature and Thickness Dependences of the Anisotropic In-Plane Thermal Conductivity of Black Phosphorus." *Advanced Materials* 29.5 (2017).
- [16] Taylor, John. *Introduction to error analysis, the study of uncertainties in physical measurements*. 1997.

- [17] Shi, Li, et al. "Measuring thermal and thermoelectric properties of one-dimensional nanostructures using a microfabricated device." *Journal of heat transfer* 125.5 (2003): 881-888.
- [18] Ou, Eric. *Four-probe thermal measurement of a carbon nanotube sheet*. Diss. 2015.
- [19] Shi, Li, et al. "Measuring thermal and thermoelectric properties of one-dimensional nanostructures using a microfabricated device." *Journal of heat transfer* 125.5 (2003): 881-888.
- [20] Ziman, John M. *Electrons and phonons: the theory of transport phenomena in solids*. Oxford university press, 1960.
- [21] Chen, Z., et al. "Thermal contact resistance between graphene and silicon dioxide." *Applied Physics Letters* 95.16 (2009): 161910.
- [22] Mastrangelo, Carlos H., Yu-Chong Tai, and Richard S. Muller. "Thermophysical properties of low-residual stress, silicon-rich, LPCVD silicon nitride films." *Sensors and Actuators A: Physical* 23.1-3 (1990): 856-860.
- [23] Zink, B. L., and F. Hellman. "Specific heat and thermal conductivity of low-stress amorphous Si-N membranes." *Solid State Communications* 129.3 (2004): 199-204.
- [24] Sultan, Rubina, et al. "Thermal conductivity of micromachined low-stress silicon-nitride beams from 77 to 325 K." *Journal of Applied Physics* 105.4 (2009): 043501.
- [25] Powell, R. W., R. P. Tye, and J. Woodman Margaret. "Thermal conductivities and electrical resistivities of the platinum metals." *Platinum Metals Review* 6.4 (1962): 138-143.

- [26] Yu, Choongho, et al. "Thermal contact resistance and thermal conductivity of a carbon nanofiber." *Journal of heat transfer* 128.3 (2006): 234-239.
- [27] Bergman, Theodore L., and Frank P. Incropera. *Fundamentals of heat and mass transfer*. John Wiley & Sons, 2011.
- [28] Weathers, Annie, et al. "Reexamination of thermal transport measurements of a low-thermal conductance nanowire with a suspended micro-device." *Review of Scientific Instruments* 84.8 (2013): 084903.
- [29] Haynes, William M., ed. *CRC handbook of chemistry and physics*. CRC press, 2014.
- [30] Howell, John R., M. Pinar Menguc, and Robert Siegel. *Thermal radiation heat transfer*. CRC press, 2010.
- [31] Kischkat, Jan, et al. "Mid-infrared optical properties of thin films of aluminum oxide, titanium dioxide, silicon dioxide, aluminum nitride, and silicon nitride." *Applied optics* 51.28 (2012): 6789-6798.
- [32] Kittel, Charles. *Introduction to solid state physics*. Wiley, 2005.