Experimental Platform for Thermal Conductivity Measurements

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Abstract

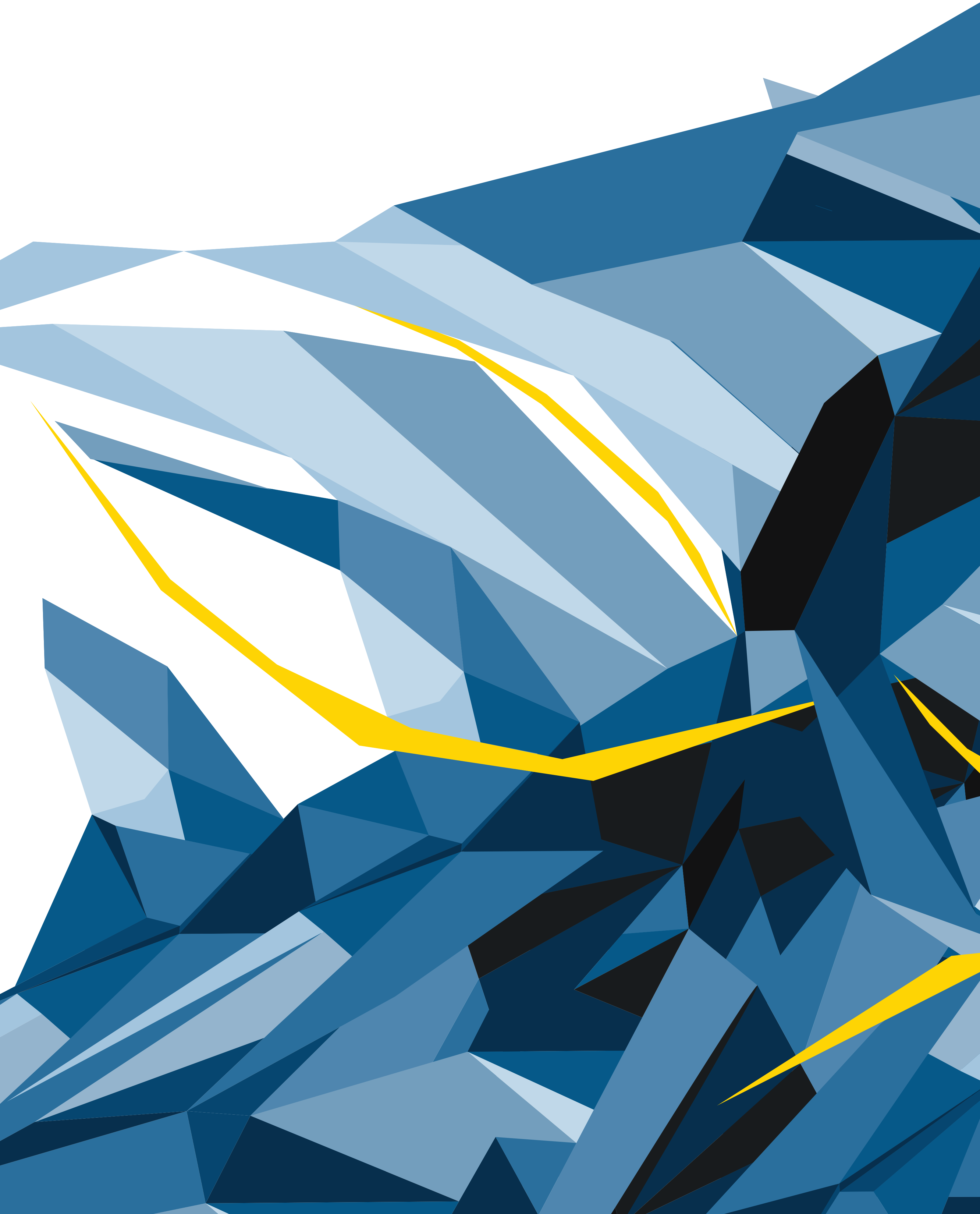
Thermal conductivity measurement techniques are extremely well established at room temperature, but become more challenging at cryogenic temperatures. The development of a dedicated thin film measurement platform for sub-Kelvin temperatures is explored here. Delicately suspended Si-N platforms ensure thermal isolation of the sample, while lithographically patterned Joule heaters provide a controllable temperature gradient across the platforms. A carbon-platinum (C-Pt) composite, fabricated using focused ion beam (FIB) assisted deposition, was patterned on the platforms as a local, resistive thermometer. These C-Pt thermometers are highly sensitive below 1K, and comparable to commercially available cryogenic thermometers. While suspended platforms have been used for thermal conductivity measurements before, the integration of highly sensitive, locally deposited cryogenic thermometers allows for more precise measurements over a wider temperature range.

Keywords: Thermal conductivity, cryogenic, thin film

Introduction

The measurement of thermal conductivity at cryogenic temperatures is critical to the study of a variety of cutting edge fields including quantum thermodynamics [1] and quantum thermoelectrics [2, 3], as well as fundamental physics like the Wiedemann-Franz law [4, 5]. Techniques for the measurement of thermal conductivity, κ, of various materials at room temperature are extremely well-established for both bulk samples and thin films [6, 7, 8], due to its importance in the development of micro- and nano-scale electronic devices [9]. However, thermal conductivity measurements at cryogenic temperatures, where many quantum phenomena are studied, are much more challenging than measurements at room temperature. Electron-phonon scattering, a common thermal transport mechanism, is temperature dependent [10]. Thus, the sensitivity of standard measurement techniques decreases with decreasing temperature, making low temperature measurements difficult.

Thermal conductivity at low temperatures is theoretically well-understood, and can be extracted fairly easily from electrical conductivity using the Wiedemann–Franz law [10]. The Wiedemann-Franz law quantifies the proportional relationship between electrical conductivity and thermal conductivity for a material. This law holds at both high and low temperatures [10, 11], and for bulk samples, thin films, and nanostructures [5, 12, 13, 14, 15]. However, experimentally determined values can vary greatly depending on material purity, defects, boundary resistance, and growth or deposition techniques, particularly for thin films and nanostructures [16, 17, 14, 11, 5]. Therefore, a versatile, repeatable technique for measuring thermal conductivity at low temperatures is critical to minimizing method dependent variation. This is especially important for thin films and nanostructures, as measurements are already difficult due to the limited material. There have been some studies done on the low temperature thermal conductivity of thin films and nanostructures, from 77-1 K [18, 19, 20, 15, 14, 21, 17, 11, 5, 12]. However, the development of a standardized, flexible measurement platform to has yet to be established, particularly below 1 K.



Precise thermal conductivity measurements at cryogenic temperatures require thermal isolation of the sample, sensitive local thermometry, and a controllable temperature gradient. Thermal isolation is a nontrivial problem at low temperatures, because the background thermal conductivity from the sample substrate creates measurement noise, decreasing the accuracy of the measurements. In order to remove these background effects, we use a Si-N suspended platform that has been uniquely designed for low temperature thermal conductivity measurements. Joule heaters are easily patterned on to the platforms during the fabrication process to create a measurable temperature gradient across the Si-N bridge. Low temperature thermometry is often done with resistive thermometers; however, traditional fabrication processes can easily destroy the delicate suspended platforms. Here, we use a focused ion beam deposited carbon-platinum composite (FIB C-Pt) as a local, highly sensitive low temperature thermometer, since it can be deposited at any point during the fabrication process without damaging the suspended platforms. In this study, we describe the development of a dedicated thermal conductivity measurement chip using a unique suspended platform design that directly addresses low temperature measurement challenges.

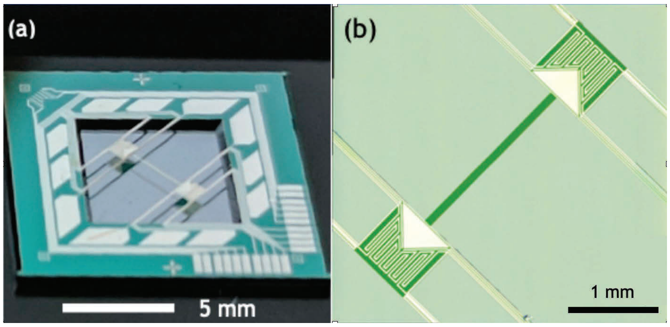


Figure 1: a) Optical microscope image of the measurement platform chip design, showing the Si-N platforms and suspended bridge between them. b) SEM image of the modified lithography pattern on the suspended platforms and Joule heaters. Both images were provided by collaborators at the University of Denver.

Methods

The suspended platforms and Si-N bridge were fabricated at the University of Denver by the Zink research group. The fabrication process is outlined in [13], and modified as follows for this particular application. Both sides of an Si wafer were coated with 500 nm of Si-N via low-pressure chemical vapor deposition (LPCVD). A 10 nm Cr adhesion layer followed by a 40 nm layer of Pt were deposited using electron beam pressure vapor deposition (E-beam PVD) on the polished side of the wafer, and lithographically patterned into heaters and electrical leads to measure across the bridge (Figure 1b). Plasma etching (CF_4) is used to make windows into the Si-N film, and the Si substrate underneath the suspended platforms and Si-N bridge is removed via a chemical etch (see Figure 1a).

After the fabrication of the suspended platforms, C-Pt thermometers were deposited across two of the leads connected to the Si-N bridge via FIB deposition (Figure 2). The C-Pt precursor gas, $(CH_3)_3(CH_3C_5H_4)Pt$, was heated to 45-46°C. Each wire was deposited via a 290-300 pC/μm dose, 30 kV ion voltage, 200 ns dwell time, 150% overlap, and a current of 24 pA. Once the thermometers are deposited, the suspended platform was bonded to a sample stage using gold wire, and then cooled in a dilution refrigerator to 0.01 K. The resistance of the FIB thermometers was measured as a function of the dilution refrigerator temperature (from a standard RuO_2 thermometer on the mixing chamber of the fridge) using standard lock-in techniques. For this particular experiment, heating effects become relevant when the excitation current of the lock-in measurements produce a power of ~1 nW. All temperature measurements were taken with a much lower excitation current of 100 nA, where heating effects were negligible.

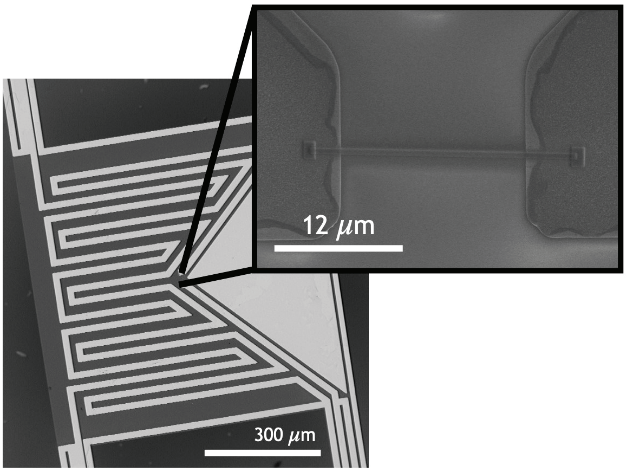


Figure 2: SEM close-up image of the patterned joule heaters on one of the suspended platforms. Zoom in: SEM image of the FIB C-Pt thermometer between two patterned leads at the point of the triangle, to measure the resistivity in a pseudo-four probe configuration.

Results

Heat is primarily conducted by electrons and phonons moving through a material. The number of phonons decreases with decreasing temperature [10], which in turn decreases electron-phonon and phonon-phonon interactions (primary thermal transport mechanisms). Pathways for thermal transport at low temperatures are therefore limited, and heat conduction becomes dependent upon boundary and defect scattering effects [10]. This is especially true for thin films and nanostructures, where fabrication techniques can influence sample morphology and composition [16, 17, 14, 11, 5]. This can lead to variation in measured values for thermal conductivity, especially at low temperatures.

At cryogenic temperatures, the thermal conductivity of the sample is often of order with the substrate. Unfortunately, directly measuring the effects of a substrate on thermal conductivity are difficult and prone to error [22]. This makes it difficult to differentiate between the thermal conductivity of the sample and the background value of the substrate. It is especially challenging when measuring thin films and nanostructures, as decreasing dimensions correlates with decreasing thermal conductivity. Thus, it is critical to minimize background contributions from the substrate and surroundings to ensure more accurate measurements. One method to decrease background thermal conductivity is to use a substrate with an extremely low thermal conductivity, such that the background is significantly smaller in magnitude than the thermal conductivity of the sample. Here, we use silicon nitride (Si-N) as the substrate, since it is a well-known electrical insulator with low thermal conductivity [21]. This makes it an ideal substrate for low temperature thermal measurements. By using suspended Si-N thin film platforms as the substrate, the substrate effects are additionally reduced. The smaller size of the thin film minimizes its thermal conductivity, and suspending the sample maximizes its thermal isolation (a standard practice at low temperatures) [23, 12, 5, 13].

Thermal isolation is critical, as all external connections to a sample could become heat sources. Any temperature difference between the sample and external connections becomes more significant at cryogenic temperatures. As the measured temperature gradient across a sample is inherently a measure of its thermal conductivity, it is important to ensure that the generated ΔT is controlled. Thermometric methods are standard for creating a ΔT due to their easy implementation, and have been shown to work reliably even at low temperatures [24, 11]. Two common thermometric methods are the 3ω technique and steady-state joule heating. The 3ω technique uses an applied AC input signal to the heaters, and uses a lock-in amplifier at a particular frequency to extract the voltage or current measurement [25, 26, 27], while steady-state joule heating uses a DC excitation current to measure the change in temperature [28, 24]. Thermal isolation ensures that any thermometrically generated ΔT is directed across the suspended Si-N bridge through the sample, avoiding radiative heat loss to the surroundings. Additionally, it prevents a larger ΔT being generated by external connections to the sample acting as heat sources. Collaborators at the University of Denver have demonstrated that the unique geometry of the suspended platforms minimizes radiative heat loss and maximizes the thermal isolation of the Si-N bridge from any external heat sources [13]. Thus, a generated ΔT across the suspended platforms will only be conducted through the bridge and the sample.

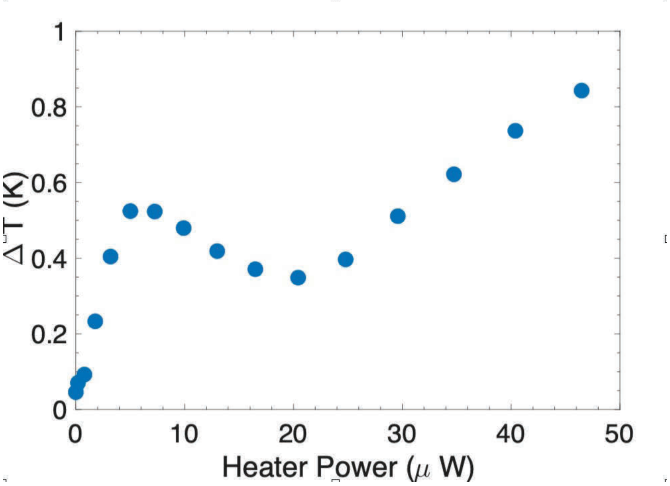


Figure 3: Change in temperature as a function of applied heater power to a bulk SiO_2 substrate at 10 mK.

Nanoscale patterned heaters on the suspended platforms can reliably create a temperature gradient at low temperatures. Given a bulk SiO_2 substrate at 10 mK, this method can reliably create a ΔT of almost 1 K (Figure 3). Standard literature values for the thermal conductivity of bulk SiO_2 range between 1.1 and 1.4 W/m-K, while thin film Si-N has a thermal conductivity of 3.6 W/m-K [13]. Because the thermal conductivities for SiO_2 and Si-N are close and of order, we would expect a similar ΔT. The additional unique geometry of the Si-N platform is expected to contribute to a comparable or greater ΔT than shown in Figure 3. Furthermore, the low thermal conductivity of Si-N indicates that lower heater power could generate an equivalent or greater ΔT across the suspended bridge. A greater ΔT is important because it is a larger measurable signal compared to the background thermal noise. Improving the sample to background ratio means that materials with lower thermal conductivities can be measured with more confidence.

Figure 3 demonstrates the functionality of steady-state joule heating at cryogenic temperatures, though there are additional benefits to the joule heaters being suspended. The nonlinearity of the plot is a result of the dilution refrigerator itself; at low temperatures, the fridge cooling power is nonlinear as the sample is heated, giving a nonlinear ΔT. The heater on the SiO_2 sample is thermally connected to the fridge, so at higher input power, the heater begins to overcome the cooling power of the fridge. The heater becomes a global heater instead of a local one, increasing the fridge temperature as well as that of the sample. The thermal isolation of the SiN bridge and the joule heaters via suspension ensures that the heaters are not heat sunk to the fridge, making it much more difficult to achieve global heating. The fact that lower heater powers could generate a comparable or greater ΔT across the Si-N bridge also safeguards against global heating.

At low temperatures, temperature gradients are often measured using resistive thermometry. However, traditional cryogenic resistive thermometry methods are inaccessible for this particular experimental chip design. Standard methods for resistive thermometry require a mask for deposition and patterning, which can damage the delicate suspended platforms. Experimentally developed focused ion beam deposited C-Pt thermometers are ideal for this platform design, as they use a maskless deposition process with nanometer resolution [29]. While both platinum and carbon are sensitive at low temperatures, neither are independently suited for local, cryogenic thermometry. The sensitivity of a pure Pt thermometer decreases below 10 K as phonon modes freeze out and change its resistivity [10]. Carbon resistors, though highly sensitive at cryogenic temperatures, are too large to be used for local, microscale thermometry. However, the FIB deposited C-Pt composite has unique conduction characteristics that are distinct from both pure Pt and carbon at low temperatures, due to Pt nanoparticles integrated in an amorphous carbon matrix [29] (Figure 4a). Due to their varied composition and morphology, each thermometer has a unique percent change in resistance and requires individual calibration (Figure 4b). However, their high sensitivity at low temperatures and ease of fabrication make these thermometers uniquely suited for local, cryogenic thermometry on suspended platforms. These thermometers have a sensitivity comparable to other leading commercially available resistive thermometers like RuO_2 (plotted in Figure 4b for comparison). Precise, local thermometry ensures more accurate measurements of the generated temperature gradient across the suspended bridge. This allows for more accurate measurements of materials with low thermal conductivities, and broadens the range of possible materials to measure at low temperatures.

Future Applications

Thermal conductivity is a fundamental material property that is critical to many different fields in physics. This is especially true at low temperatures, where measurements become difficult as thermal transport pathways freeze out. The experimental platform described in this study addresses the primary challenges of cryogenic measurements and allows for the direct measurement of the thermal conductivity of a thin film. Using Si-N suspended platforms with a unique geometry minimizes background thermal effects and maximizes thermal isolation of the sample and joule heaters. Applying power to one of the heaters creates a hot bath on one side of the bridge, such that the heat is conducted across the bridge and not lost to the surroundings. The local temperature on either side of the bridge is measured using recently developed, highly sensitive FIB C-Pt thermometers, which gives a ΔT. Knowing the geometry of the platforms, the thermal conductivity can then be calculated using equation 1.

$$K = \frac{Q \cdot d}{A \cdot \Delta T}$$

Equation 1

In equation 1, Q is the amount of heat transferred through the material in Watts (the applied power to create the hot bath), d is the distance between the two measurements points (bridge length) and A is the area of the surface through which the heat is passing (bridge width×thickness). ΔT is the measured change in temperature between the two sides of the bridge.

Since we know the dimensions of the Si-N bridge, the measured ΔT across the bridge, and the applied power to the system, we can calculate the thermal conductivity of the bridge. Standard thermal evaporation or sputtering techniques can be used to deposit thin film materials upon the Si-N bridge without damaging it [4, 30, 13, 18]. The thermal conductivity of the deposited thin film can then be measured, subtracting out the background thermal conductivity of the Si-N bridge to isolate the contribution of the thin film. This technique is straightforward and easily repeatable, as well as versatile for use with many different materials. A wide range of thin films and nanostructures can be measured using this platform, even those with very low thermal conductivities, due to the low background thermal contributions, the ability to generate a larger ΔT across the bridge, and sensitive, local thermometry.

There is an increasing need for low temperature thermal conductivity measurements for thin films of various materials, as electronic devices get smaller and thermal effects become more predominant [12]. Many cutting edge quantum phenomena can only be studied at cryogenic temperatures, where measurement devices need to be precise [1, 3, 2]. This requires knowledge of how these devices will behave at low temperatures, which is intrinsically linked to their thermal and electrical conductivities. Additionally, it is possible to analyze how theories like the Wiedemann-Franz law work at extremely low temperatures – if and why they break down, for what materials, etc. There is evidence both for and against the breakdown of the Wiedemann-Franz law at low temperatures [11, 5, 12, 4, 31], but it can be difficult to compare since the materials and measurement methods differ. Having a standardized low temperature measurement method for thermal conductivity would simplify the evaluation of both emergent quantum phenomena and traditionally accepted theories. The wide range of measurable materials, in addition to the repeatability of this technique, makes this experimental platform uniquely suited for low temperature thermal conductivity measurements, for future use in a variety of classical and quantum applications.

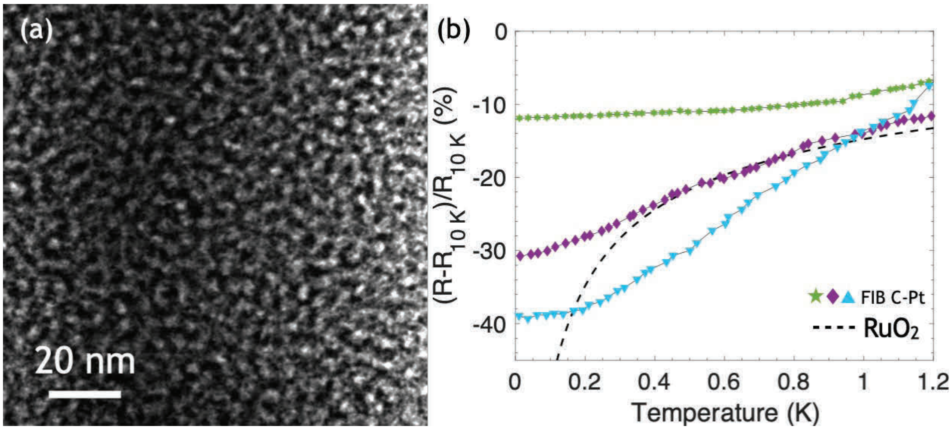


Figure 4: a) A transmission electron microscope (TEM) image of a thermometer deposited on a carbon grid. The grains of platinum (dark spots) are clearly embedded in a carbon matrix (lighter background). b) Percent change in resistance (R – R10K/R10K) of C-Pt thermometers as a function of temperature. Plot shows ultra low temperature resistance of three different thermometers used for calibration, as well as that for RuO2. Both figures taken from [29].

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