Electronic Supplementary Material (ESI) for Nanoscale.


**Electronic Supplementary Information (ESI)**

# Thermal Conductivity Measurements of High and Low Thermal Conductivity Films Using a Scanning Hot Probe Method in the 3ω Mode and Novel Calibration Strategies

Adam A. Wilson,[a*] Miguel Muñoz Rojo,[b*] Begoña Abad Mayor,[b] Jaime Andrés Perez,[b] Jon Maiz,[b] Jason Schomacker[a], Marisol Martín Gonzalez,[b] Diana Andra Borca-Tasciuc[a] and Theodorian Borca-Tasciuc[a†]

[a] Department of Mechanical, Aerospace and Nuclear Engineering, Rensselaer Polytechnic Institute, 110 8th Street, Troy, NY 12180 USA
[b] Instituto de Microelectronicas de Madrid, Consejo Superior de Investigaciones Científicas, Calle de Isaac Newton, 8, Tres Cantos, Madrid 28760, Spain

## 1. Probe Properties

To accurately use the Wollaston probe for measurements, the probe's properties must be known. The properties obtained and reported in Table 1 are the probe's length, diameter, thermal conductivity, probe-to-air heat transfer coefficient, temperature coefficient of resistance, and nominal electrical resistance. To find the probe's nominal electrical resistance, and the geometry of the probe, an SEM image was obtained to measure the heating element length and diameter. Figure S.1 contains this image.

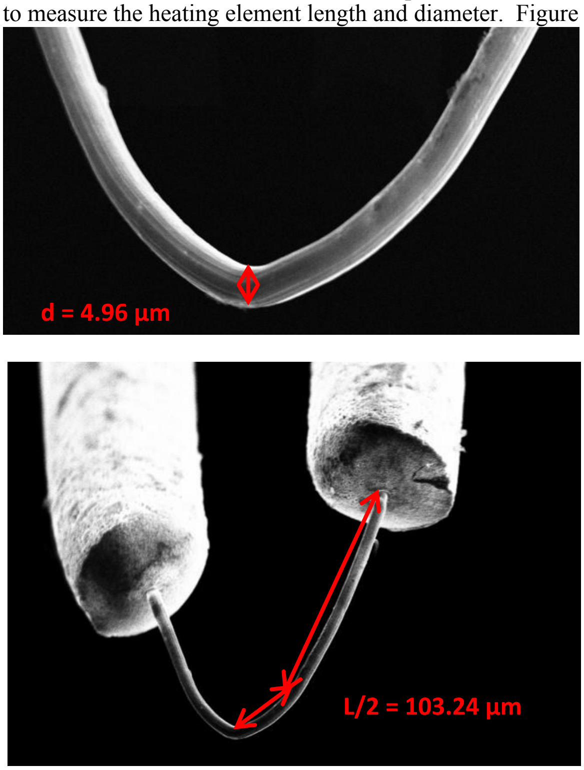



***Figure S.1*** *– SEM image of probe to obtain probe geometry for nominal resistance: a)(top) probe diameter; b)(bottom)half of probe lenth*

Note that the image for length was taken with the stage skewed at an angle such that the plane of the image was parallel to the right leg of the probe, giving it an accurate measure of length. The probe is known to be symmetric, so this length was doubled to obtain the total length.

Having the length and diameter of the circular wire, and taking the electrical resistivity value provided for Pt90/Rh10 (the material of which the heating element of the probe is made), we can readily calculate the nominal electric resistance of the probe by: $R_0 = \frac{4\rho L}{\pi d^2}$.

Thermal conductivity is assumed to be the manufacturer's specified value (the value provided for Pt90/Rh10 – 38 W/mK). To obtain the probe-to-air heat transfer coefficient, $h_{eff}$, it is necessary to find the thermal resistance of the probe in air, with no sample present. This represents the case in which the heat flux between probe and sample is zero. Once the thermal resistance is determined and the value introduced to a heat transfer model, the $h_{eff}$ parameter in the program is adjusted until the value of thermal resistance under no heat transfer to the sample matches what is observed experimentally. Another important consideration is how the electrical resistance changes with increasing temperature. This is the probe's temperature coefficient of resistance (TCR). Its value was obtained experimentally by placing the probe in a furnace and measuring the resistance with increasing furnace temperature. Figure S.2 shows this.

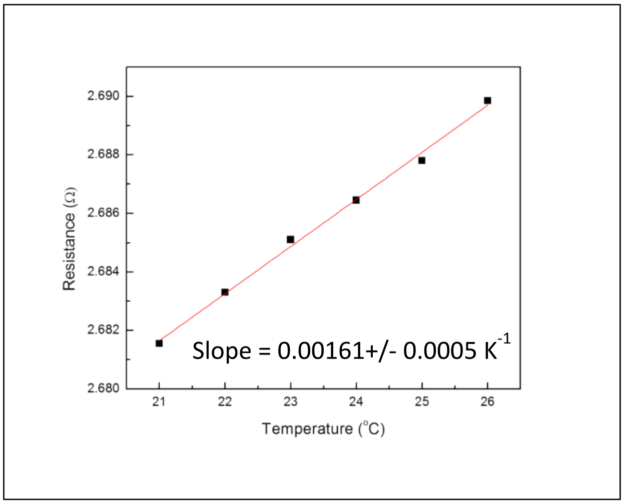



***Figure S.2** – Probe Resistance vs. Temperature to obtain probe's TCR*

## 2. Obtaining Seebeck Voltage/Seebeck Coefficient

In order to determine the sample's Seebeck coefficient, the sample is scanned at a fixed frequency in contact and the $V_{2\omega}$ voltage response is recorded. The average value of $V_{2\omega}$ is given from an analysis of the data collected in the scan, shown in image S.3. Note that the data needs to be scaled for a gain previously applied to the sample.

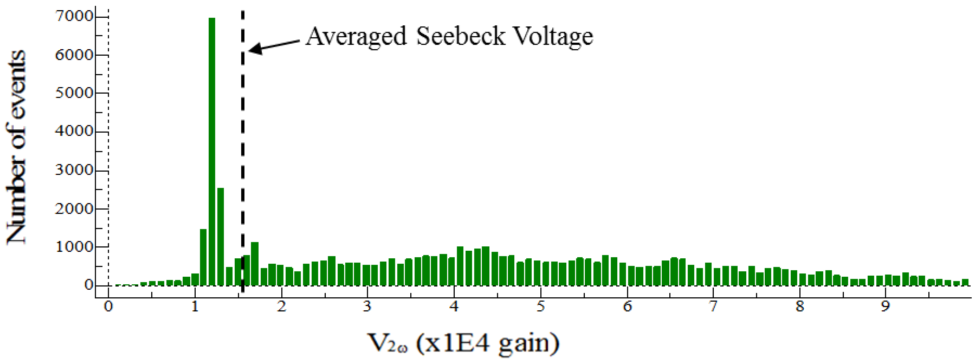
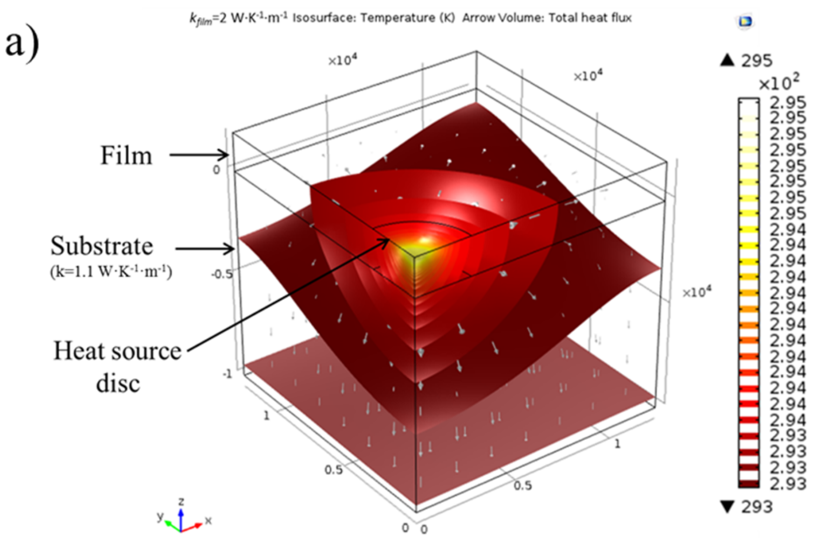



***Figure S.3** –Occurrence distribution of Seebeck voltage between probe and Sb-doped $Bi_2Te_3$; the mean voltage value is found from this graph and taken to be the Seebeck voltage for the sample*

Once this data is obtained, the Seebeck coefficient is calculated by dividing the Seebeck voltage by the sample temperature (which is obtained from scaling $\Delta T_p$ by $\Delta T_S$, given by the heat transfer model described in the text) and correcting for the contribution of the probe to the Seebeck voltage.

## 3. Finding thermal conductivity of films with two-dimensional heat spreading

In order to find the thermal conductivity of samples whose substrate contributes a non-negligible amount to the overall sample thermal resistance, a model which accounts for 2D heat conduction across multi-layered heat samples was developed based on heat propagation as induced by laser heating [2]. This model was cross-checked by analyzing the film thermal conductivity of the SiGe film, using COMSOL Multiphysics ®. For both models, the strategy to find the film's thermal conductivity is to assume a value for sample thermal conductivity (and assign a known value for the substrate thermal conductivity), and check the overall sample thermal resistance predicted by the program. The sample's thermal conductivity is then adjusted until the thermal resistance was found to match that obtained by experiment. Figure S.4 shows the thermal model developed in COMSOL, together with the means of finding the thermal conductivity of the sample by matching thermal resistance obtained from the model to that obtained from experiment. The thermal conductivity of the SiGe film was found by the analytical model to be 1.22±0.21 W/mK, compared with 1.23 W/mK obtained by the COMSOL model. The thermal conductivity of the thin-film gold on glass was found by the analytical model to be 104.2 W/mK. The governing equation and boundary conditions for the COMSOL model are the same as used in reference [1]



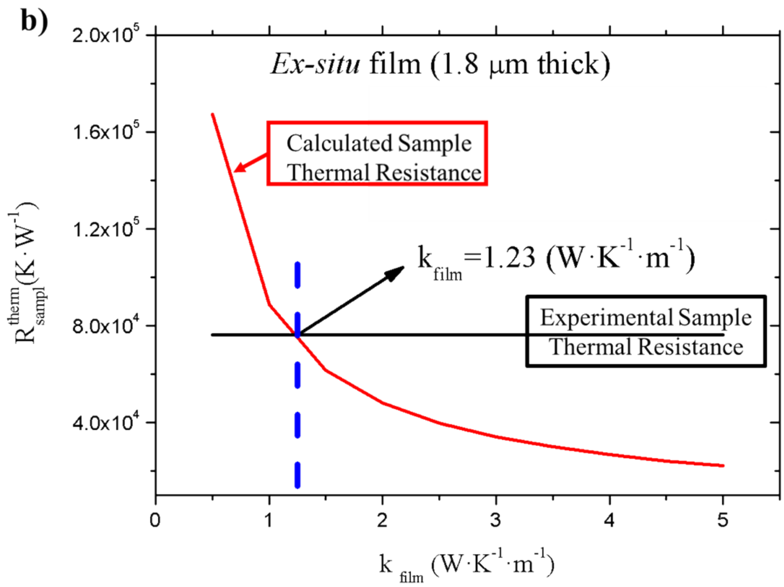



***Figure S.4 – a) Thermal model developed in COMSOL Multiphysics ®, used to find the thermal properties of film samples whose substrate contributs to the overall sample thermal resistance in agreement with predictions of the model employed by Son et al*[2] *and b) Finding the film thermal conductivity by fitting the experimental thermal resistance with a film on substrate model for ex-situ heat-treated SiGe thin film.***

## 4. Uncertainty analysis for thermal exchange parameters and thermal conductivity

To calculate the uncertainty in our measurements, the following procedure was used. Experimentally recorded data is voltage across probe, resistor, and entire circuit. These are recorded at a fixed applied AC voltage, varying the frequency from 5 Hz – 10 kHz. In the work represented here, the data considered was in the frequency independent range, around 10Hz. The experimental data at 10 points each above and below a frequency of 10 Hz was averaged, and the standard deviation of the data was found for the voltage values recorded. Propagating uncertainty from these measurements to the average probe temperature rise and applied power gives the uncertainty in the thermal resistance for this range.

The uncertainty in the values for $R^{th}_C$ and b obtained by intersection was calculated by measuring samples with known thermal conductivity and fitting $R^{th}_C$ for a range of values of b, and assuming the curves follow a linear trend near the point of intersection, the uncertainty in the point of intersection is obtained by first order propagation of variance[3], [4]. This method was employed for the two higher thermal conductivity calibration samples to yield the uncertainty in $R^{th}_C$ and b. From this, the maximum sample thermal resistance is obtained by inputting average probe thermal resistance plus uncertainty into equations 1-4 in the main text, with calibrated $R^{th}_C$ from intersection minus uncertainty and b from intersection plus uncertainty. Similarly, the minimum sample thermal resistance was found by taking the minimum probe thermal resistance, maximum $R^{th}_C$ and minimum b. The same procedure was carried out for all the low thermal conductivity calibration samples, and the locations of the intersection for each line was recorded. Finally, the standard deviation of the values of $R^{th}_C$ and b from those intersections were calculated to yield the uncertainty in $R^{th}_C$ and b. For the samples which were bulk, or which had bulk-like thickness, the uncertainty was propagated through equations 1-4 in the main text to obtain the uncertainty in the sample thermal conductivity. For the samples which were analyzed by the 2D heat transfer models, the film thermal conductivity is found by adjusting the film's thermal properties in the model until the sample thermal resistance matches that obtained after data reduction using the probe heat transfer model. Thus, uncertainty in thermal conductivity is found by adjusting the sample thermal properties at the upper and lower bounds of the thermal exchange radius, and matching to the upper and lower bounds of sample thermal resistance.

## References

[1] M. Muñoz Rojo, J. Martín, S. Grauby, T. Borca-Tasciuc, S. Dilhaire, and M. Martin-Gonzalez, "Decrease in thermal conductivity in polymeric P3HT nanowires by size-reduction induced by crystal orientation: new approaches towards thermal transport engineering of organic materials.," *Nanoscale*, vol. 6, no. 14, pp. 7858–65, Jul. 2014.

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