Improved Method for Measuring Thermal Diffusivity of Bulk Samples and Films

Honors Research Thesis

Presented in Partial Fulfillment of the Requirements for graduation "with Honors Research Distinction in Physics" in the undergraduate colleges of The Ohio State University

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ABSTRACT

We have developed a technique to measure the thermal diffusivity of bulk samples and films. It builds off of the widely used laser flash method. Basically, we measure the time interval between a laser created heat pulse and its arrival at a detector some distance away. In parallel with the laboratory set-up we use computer modeling to simulate our detector's response and account for losses due to air convection. One advantage to our method is that we are able to accurately determine thermal diffusivity without needing to enclose the apparatus in a vacuum as is required with most other set-ups. Experimentally, we shine a laser pulse on a free-floating end of a long sample, the other end of the sample is secured to a heat sink, and measure, via a thermistor, the heat pulse as it travels a well defined distance. In our testing with a silver wire, we found our measurement to be in agreement with the accepted value. As an important application, this method determines the thermal diffusivity for thin films that would otherwise be difficult to measure. As an example, we measured the thermal diffusivity of graphene films upon ceramic substrates. By combining computer modeling and a simple experimental procedure, we are able to efficiently and accurately determine thermal conductivities for a wide variety of samples.

ACKNOWLEDGEMENTS

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INTRODUCTION

Thermal diffusivity tells us how quickly a material will transmit or conduct heat across its geometry. Being able to measure this quantity accurately is then essential to find materials best suited for thermally sensitive technologies like heat sinks for CPUs or semi-conductors, blast furnace linings, or new thermal greases.

Many methods to measure thermal conductivity exist and we shall cover three of these techniques [3][5][14]. The most important for us is the flash method where an impulse of heat is created within a sample and the temporal temperature profile is measured at another position. The thermal conductivity can then be derived. Another is the 3ω method which uses AC current to generate a voltage signal in the third harmonic from which thermal oscillations can be determined and from that the thermal properties. The third method was designed for a single atom layer of graphene and takes advantage of the strong temperature dependence in the raman shift to measure the thermal profile.

The method forming the basis of this thesis is a modification on the standard laser flash method. Our apparatus does not require a vacuum like most other instances of the flash method and greatly simplifies the experimental approach from those other techniques. In our system we have a laser create a pulse of heat, typically 1W for 10-100ms or 0.01 to 0.1 joules of energy. This pulse travels along the sample to a thermistor at an accurately measured distance from the target location, which allows us to record the temperature

profile over time. To allow for optimum absorption of the laser power, a spot of graphite is painted onto the location we wish to send the flash. The thermistor's resistance depends on its temperature. We measure the resistance of the thermistor and convert that to temperature via LabVIEW with the DAQ computer instrument attachment. The temperature data is then compared to predictions from computer modeling designed for our system from whence we can correct for the convection losses and find the thermal conductivity.

An important experimental check on our method is to measure established values for known materials. We measured the thermal conductivity of a silver sample with our system and found it to be in close agreement with the established value. Fig 16 shows the results of this important check and is one of the principal results of this thesis. The theory and the experiment for silver are shown to be in close agreement.

The purpose behind developing this technique was to measure the thermal conductivity of graphene. Using the raman scattering method on a single atom layer of graphene, Baladin et. al. found an upper value of 5300 ± 480 W/mK which is the highest of all known materials [3]. Silver, by comparison, is 429 W/mK [17]. It is then of great importance to see if this extraordinary conductivity holds for multi-layered graphene since a single layer is inadequate to use for many purposes. Graphene tends to be difficult to layer in a sufficient thickness for use as a sample by itself. Instead our samples are created by the L. James Lee group using a novel technique developed by them. With a 600nm layer on a 3.1mm ceramic strip we were able to measure a thermal conductivity of ~1200W/mK

which is roughly that of in-plane graphite. Fig 17 shows the experimental data and the related computer modeling [12]. This is the other principal experimental result of this thesis.

BACKGROUND INFORMATION: THERMAL DIFFUSIVITY

To understand how a material transfers heat and the rate of that transfer, one must know its thermal conductivity or thermal diffusivity. These quantities are closely linked and are directly proportional to each other. Their relation is defined as:

$$\alpha = \frac{\kappa}{c \rho}$$

where α = thermal diffusivity (m²/s), κ = thermal conductivity (W/mK), c = heat capacity (J/K), and ρ = density (kg/m³) [2]. This relation is defined in terms of the fundamental heat equation.

To derive the heat equation, we shall consider a flux of heat traveling along a rectangular box [6].

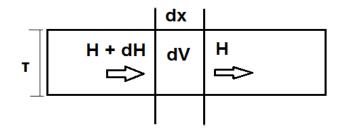


Figure 1: Simple model of 1-D heat flow.

We take a region of that box of thickness dx and volume dV into which heat H+dH is flowing in and heat H is flowing out. The box has a height of τ and a width of z which makes up the cross sectional area A. The first relation we need to consider is the heat flux.

$$\delta H \ A \ \delta t = c \ \rho \ dV \ \delta T = c \ \rho \ z \ \tau \ \delta x \ \delta T$$
$$\frac{\delta H}{\delta x} \ A = c \ \rho \ z \ \tau \frac{\delta T}{\delta t}$$

Next we need to consider the conductivity of the sample.

$$H A = \kappa \frac{\delta T}{\delta x} \tau z$$
$$A \frac{\delta H}{\delta x} = \kappa \frac{\delta^2 T}{\delta x^2} \tau z$$

If we relate these two, we find the heat equation and the definition of the thermal diffusivity.

$$\frac{\delta H}{\delta x} A = c \rho z \tau \frac{\delta T}{\delta t} = A \frac{\delta H}{\delta x} = \kappa \frac{\delta^2 T}{\delta x^2} \tau z$$

$$c \rho z \tau \frac{\delta T}{\delta t} = \kappa \frac{\delta^2 T}{\delta x^2} \tau z$$

$$c \rho \frac{\delta T}{\delta t} = \kappa \frac{\delta^2 T}{\delta x^2}$$

$$\frac{\delta T}{\delta t} - \frac{\kappa}{c \rho} \frac{\delta^2 T}{\delta x^2} = 0$$

$$\frac{\delta T}{\delta t} - \alpha \frac{\delta^2 T}{\delta x^2} = 0$$

Since the main motivation for this work is to measure the thermal properties of a thin film on a much thicker substrate to the extent that the total thickness of the sample is much thinner than that of the length we are measuring along we will need to calculate for such a system. For this layered substrate, our system looks slightly different.

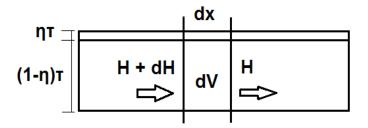


Figure 2: Model for 1-D heat transfer on layered substrate.

This will cause some differences in our heat flux equations.

$$\frac{\delta H}{\delta x} A \,\delta t = c_c \rho_c \, dV \,\delta T + c_s \rho_s \, dV \,\delta T = (c_c \rho_c \eta \,\tau + c_s \rho_s (1-\eta) \,\tau) z \,\delta x \,\delta T$$
$$\frac{\delta H}{\delta x} A = (c_c \rho_c \eta \,\tau + c_s \rho_s (1-\eta) \,\tau) z \frac{\delta T}{\delta t}$$

Where subscript c indicates values for the coating and subscript s indicates those for the substrate. For the conductivity relation:

$$H A = H_{c} A_{c} + H_{s} A_{s} = \kappa_{c} \frac{\delta T}{\delta x} \eta \tau z + \kappa_{s} \frac{\delta T}{\delta x} (1 - \eta) \tau z = (\kappa_{c} \eta + \kappa_{s} (1 - \eta)) \tau z \frac{\delta T}{\delta x}$$
$$A \frac{\delta H}{\delta x} = (\kappa_{c} \eta + \kappa_{s} (1 - \eta)) \frac{\delta^{2} T}{\delta x^{2}} \tau z$$

Combining these together to get our heat equation and our new effective diffusivity.

$$(c_{c}\rho_{c}\eta\tau+c_{s}\rho_{s}(1-\eta)\tau)z\frac{\delta T}{\delta t} = (\kappa_{c}\eta+\kappa_{s}(1-\eta))\frac{\delta^{2}T}{\delta x^{2}}\tau z$$

$$\frac{\delta T}{\delta t} - \frac{\kappa_{c}\eta+\kappa_{s}(1-\eta)}{c_{c}\rho_{c}\eta+c_{s}\rho_{s}(1-\eta)}\frac{\delta^{2}T}{\delta x^{2}} = 0$$

$$\frac{\delta T}{\delta t} - \alpha_{eff}\frac{\delta^{2}T}{\delta x^{2}} = 0$$

$$\alpha_{eff} = \frac{\kappa_{c}\eta+\kappa_{s}(1-\eta)}{c_{c}\rho_{c}\eta+c_{s}\rho_{s}(1-\eta)}$$

If we make the assumptions that the thickness of the coating is much less than the total thickness ($\eta \ll 1$) and that the conductivity of the substrate is much less than that of the

coating ($\kappa_s \ll \kappa_c$), then we can simplify the effective diffusivity to:

$$\alpha_{eff} \approx \frac{\kappa_c \eta + \kappa_s}{c_s \rho_s}$$

In many cases the terms from the substrate can be ignored and the diffusivity is essentially equal to the conductivity. This makes converting these two quantities trivial.

METHODS OF MEASURE

The Flash Method

The flash method was first described in 1961 by W.J. Parker et. Al [14]. The basic premise behind it is that you create a pulse or flash of heat, typically with a laser, to a sample then you measure the change in temperature over time at another point on the sample. From that temperature profile you can derive the thermal diffusivity and there-fore the conductivity. In most instances of this method the sample must be placed in a vacuum and machined to a specific required shape, a disk. The flash is shone on one side of the disk and the temperature is measured on the opposing side measuring the conduct-ivity perpendicular to the plane of the sample.

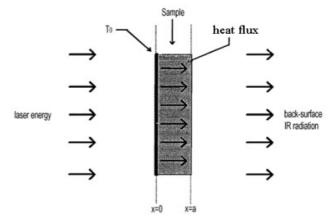


Figure 3: Diagram of basic laser flash operation [6].

Because samples must be prepared to a disk-type shape and the properties are measured across the thickness of the sample, materials to be measured have to be able to be prepared to a thickness typically on the scale of millimeters. In addition most experimental setups must be equipped with a vacuum system to enclose the sample to eliminate any heat loss due to air-convection. This requirement for a vacuum leads to a greater experimental complexity and therefore more chances for human error. Commercial systems can ease difficulties in constructing an apparatus but are still often bulky and complex. To measure a sample of less than a micrometer in thickness is impossible with the older standard flash method and slower thermal sensors.

3w Method

The 3ω method was developed in 1912 by O.M. Corbino initially as a method to measure thermal diffusivity of metal filaments for use in light bulbs [4]. In 1987, Cahill and Pohl re-purposed it for use in measuring the thermal diffusivity of solids [5]. It functions by sending an AC current across a metal line deposited onto the sample. This current excites the metal line and creates oscillations in the 2ω harmonic in the electrical resistance. This in turn generates a 3ω harmonic in the voltage signal which are used to derive the magnitude of the thermal oscillations. From this the thermal properties can be found.

For preparation the sample needs to have metal deposited onto its surface to act as both the heater and thermometer as shown in fig 4.

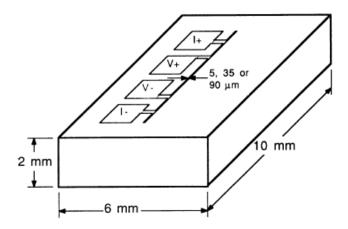


Figure 4: Diagram of sample prepared for 30 measurements [4].

Cahill and Pohl mention several ways to deposit this metal layer onto the sample ranging between 5-90µm in thickness of the metal layer. A requirement is then imposed that the sample needs to be at least five times the width of the metal line. Our graphene sample with a thickness of 600nm is far too thin to be measured with this method.

Raman Scattering

As we have seen, our desired material of multi-layered graphene is simply too small to be measured with some of the standard measuring techniques. When it was first measured as a single-atom layer this was even more so of an issue. Balandin et. al. instead utilized the strong temperature dependence in the raman shift for graphene [3]. Similar to the flash method, they shone a laser at a point on the mono-layer and used raman scattering to detect the temperature profile and the heat spread across the atoms. This temperature profile could then be used to derive the desired thermal properties. This method only works for

single-atom layers due to fast heat loss in thicker samples. Rather than being too thin, our sample is now too thick.

OUR METHOD: MODIFIED FLASH METHOD

Our method is a modified form of the flash method that can measure thin films coated on substrates without requiring the complexity of a vacuum enclosure of the standard flash method. We also measure along the plane of the sample rather than through the sample. Since the sample is not contained within a vacuum when taking the measurement, heat loss due to convection would ordinarily be an issue. Computer modeling is used to account and correct for this heat loss.

Apparatus

Our apparatus is significantly easier, simpler, and more economic than previous flash method techniques. As shown in fig 5, a laser (LASEVER LSR1064NL 1200mW 1064nm DPSS Infrared Laser) sends out a pulse of light between 1ms and 1s to a suspended sample [13]. The laser has a 30cm focusing lens attached so that the sample can be place in a position such that the laser spot is the desired size. The sample is contained in a simple Plexiglas barrier to remove any large scale heat losses due to the surrounding environment. To operate the laser we send a voltage step function to the laser. Five volts will cause the laser to operate at full power and 1.5V was found to be the minimum voltage at which we could get an output [13]. Most data sets were taken with a short pulse at full power though in some cases smaller voltages were used. When lining up the laser to hit the desired spot on a sample the minimum power voltage was used to prevent

burning over the longer exposure.

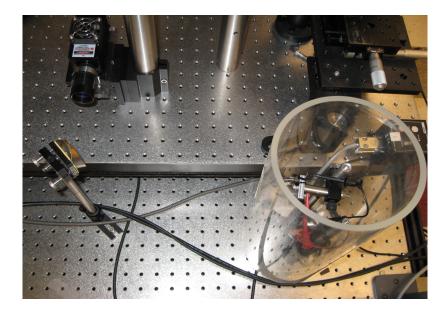


Figure 5: Experimental apparatus.

Sample Preparation

A sample consists of the material to be tested, usually shaped as a long thin strip. On this strip a thermistor is attached and acts as the detector or thermometer. A well-defined distance below the detector a graphite spot is placed to act as a standard target for the laser and to allow better absorption of the heat energy. This is then attached to a heat sink on one end so that we can establish a well-defined arrangement for later modeling treatment while the other is left free. The graphite spot is often placed at the bottom edge of the sample so that we get the full pulse of heat traveling across the detector. Having the full pulse allows a better reading than only half a pulse that would result if the target was not at the end. Though at the same time a closer target also allows for a better signal. The

pulse we send will depend on these factor as we want as short a pulse as possible but at the same time need to have a detectable signal at the farthest distance possible to reduce our error in the distance. An error of ± 1 mm in the position is much less significant for a 3cm distance than for a 1cm distance.

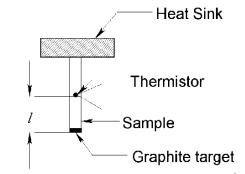


Figure 6: Diagram of typical sample setup.

Fig 8 shows an actual sample that we measured, a silver wire, prepared and suspended in the Plexiglas barrier with and without the laser shining for alignment. The wire is held fast to a aluminum block which acts as a heat sink. Melcor TG-002 Thermal Grease is used to allow better thermal contact between the sample and the heat sink. The thermistor we use is of the micro-bead variant (VECO 42A29) and is attached via a minimal amount of thermal epoxy (Arctic Alumina Thermal Adhesive). This thermistor is extremely small and its wire leads are even smaller yet as is shown in fig 7. Without taking care the thermistor leads can be easily broken through simple handling with tweezers. Attaching the thermistor with the minimum amount of epoxy is another challenge all its own. The final complication due to the detector was soldering the leads to wires without either breaking the sample, the leads, or detaching the thermistor. After the sample has its thermistor attached but before it is attached to the heat sink and the wires are soldered, the graphite targets are painted at certain distances away from the detector with AquaDag E.

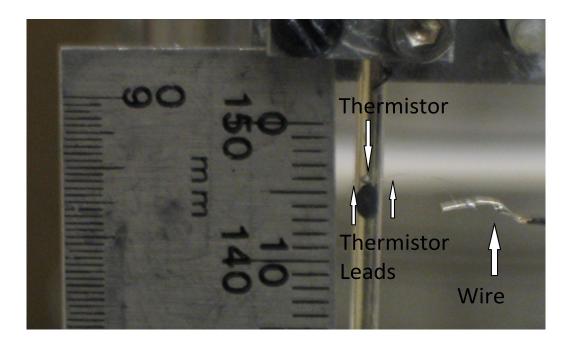


Figure 7: Thermistor attached to silver wire with graphite target.

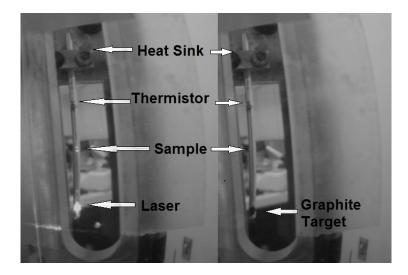


Figure 8: Silver suspended from heat sink with and without laser hitting 3cm target.

Several representative samples are shown in fig 9. From left to right, a silver wire with graphite targets placed at 1mm, 1cm, 2cm, and 3cm distances, an aluminum strip with a graphene coating, a thin quartz strip with a gold coating, and a ceramic strip similar to that used as the substrate to the final graphene tests.

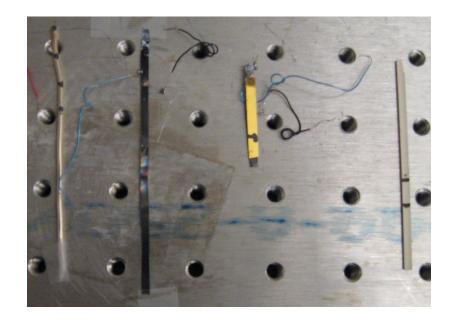


Figure 9: Representative samples used in our apparatus.

On more recent tests we have eliminated the use of a heat sink to simplify the boundary conditions when solving the heat equation. This results in heat reflecting back along the length of the sample but that complication is readily treated in the modeling. As we test for the accuracy of our system in and out of a vacuum we now use a sample suspended solely by the wires attached to the thermistor as shown in fig 10.

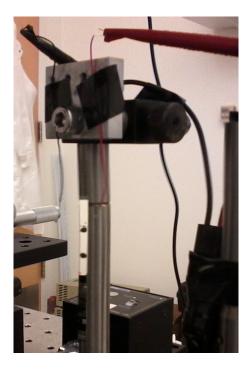


Figure 10: Sample of a silver coating on a quartz substrate isolated from physical contact.

A main trick we utilize is the use of coating layers of a desired material on a known substrate with a low conductivity as shown in the above theory sections. Whether it be graphene on a ceramic substrate or silver on a quartz substrate, the assumptions that the conductivity of the coating be much larger than that of the substrate, the thickness of the coating be much less than the total thickness, and the total thickness being much less than that of the measurement length generally hold true. A degree of sacrifice must be made in requiring a much thinner coating than the substrate since we need to have a detectable difference between the uncoated and coated samples.

Circuit

Fig 11 shows the circuit for our apparatus. A nine volt battery acts as the voltage source. The thermistor is in series with a $20k\Omega$ ballast resistor to divide the voltage since the thermistor is approximately $20k\Omega$ at room temperature. It can be shown that the maximum detected change in voltage due to the change in resistance of the thermistor is when $R_{ballast}$ approximately equals R_{therm} . The voltage across the thermistor is read by a DAQ board on the computer which is processed by a LabVIEW program (See Appendix I & II). Noise from the signal is reduced by adding in a capacitor to eliminate fast oscillations in the signal. But this noise reduction comes at the cost of a slight delay as is shown in fig



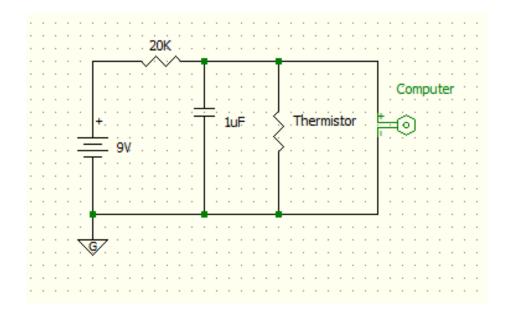


Figure 11: Circuit used by apparatus.

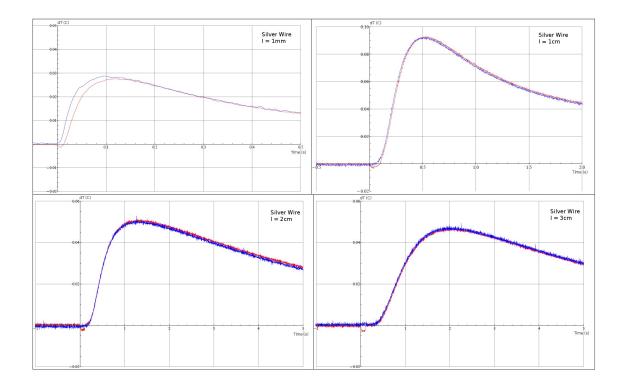


Figure 12: With and without capacitor data sets for various distances for a silver wire sample. Data shown is averaged over 20 consecutive trials. A slight delay due to the capacitors is only evident in the 1mm data and barely perceptible in the 1cm data. This delay then is not a concern for data above 1cm. This goes along with desiring as long a distance as possible for our measurements.

Thermistor Characterization

In order to find temperatures from the voltages we measure from the circuit we convert them to resistances and then use the Steinhart & Hart equation

$$\frac{1}{T} = A + B \ln(R) + C \ln(R)^3$$

to convert from resistance (R) to temperature (T) [16]. The three coefficients A, B, and C

are either given by the manufacturer or can be derived by solving a set of three simultaneous equations with three sets of known temperatures per resistance. Our coefficients were: A = 8.163E-4, B = 2.734E-4, and C = 2.53E-8. To find the resistance in the first place we input the room temperature resistance measured with a multimeter into the program and then the program takes the first voltage read and calculates a general first order current via Ohm's Law. We then use this current to convert the remainder of our voltages to resistance which is plugged into the Steinhart & Hart equation.

The LabVIEW program outputs the temperature difference from the first read in temperature along with the laser voltage output and timing. Based on how many trials you specify, the program will repeat the measurement that many times with a several second delay between each trial to allow for additional time for the system to return to equilibrium. These columns of data can then be input into Mathematica or any program of your choosing and plotted or compared to computer simulations. Appendix III shows the Mathematica program used to create plots.

When we are taking data, an important consideration is the time response of the detector itself. The thermal mass of the thermistor and the epoxy it is attached with affect that time response. We measure this by taking data at 1mm below the thermistor, fig 13 shows the 1mm data from the silver wire shown in fig 12. This allows us to eliminate the effects from the thermal mass of the thermistor and epoxy and to find the time response of a particular experiment. The thermistor response is modeled by the following differential equation [15]:

$$\frac{\delta T_{thermistor}}{\delta t} = \frac{(T_{sample} - T_{thermistor})\kappa}{\mu_{thermal}}$$
$$\frac{\delta T_{thermistor}}{\delta t} - \alpha T_{thermistor} = \alpha T_{sample}$$

where $\mu_{thermal}$ is the thermal mass, κ is the thermal conductance from the sample to the thermistor, $T_{thermistor}$ is the temperature of the thermistor, T_{sample} is the temperature of the sample. If we take

$$T_{thermistor} = e^{-\alpha t} \tau$$

then we can substitute this back into the differential equation to get

$$-\alpha e^{-\alpha t} \tau + e^{-\alpha t} \frac{\delta \tau}{\delta t} + \alpha e^{-\alpha t} \tau = \alpha T_{sample}$$

which simplifies out to

$$\frac{\delta \tau}{\delta t} = \alpha e^{\alpha t} T_{sample}$$

and finally integrating this gives us

$$\tau(t) = \alpha \int_{0}^{t} e^{\alpha t'} T_{sample}(t') dt'$$

where we can plug in the result of the computer modeling into T_{sample} and find the difference between the predicted value at the sample and the measured value at the thermistor.

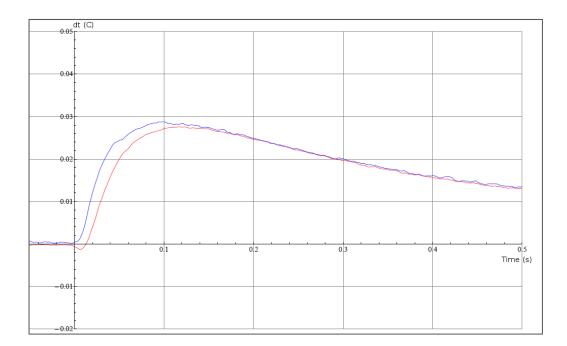


Figure 13: Blow up of 1mm silver wire measurement from fig 12. The heat pulse was 10ms long and the peak arrives about 100ms after the start of the pulse.

Computer Modeling

As has been mentioned previously, convection heat losses need to be considered since our system is not isolated from the air in a vacuum. A simple way to model this heat loss is by expanding the heat (H) to the first order

$$H = h(T - T_a)$$

where h is the heat transfer coefficient, T is the temperature of the sample, and T_a is the temperature of the surrounding air [9].

To model our experimental arrangement Dr. Wei-Ching Liao creates a mesh of the sample in COMSOL based on the properties we supply from the experiment including the pulse size, duration and geometry. From this and the convection heat loss the boundary conditions in fig 14 are applied to the system. As in the physical experiment we then choose a position along the mesh to sample the temperature profile.

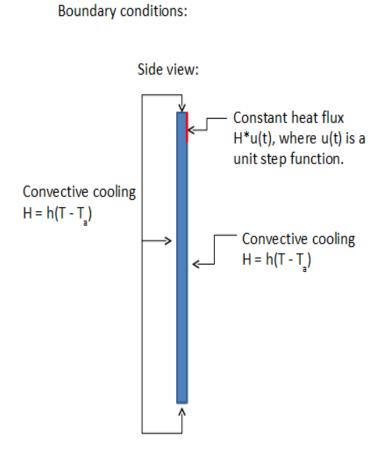


Figure 14: Example boundary conditions applied to computer simulation. On all surfaces we assume convective cooling and at one point we apply a unit step function of heat.

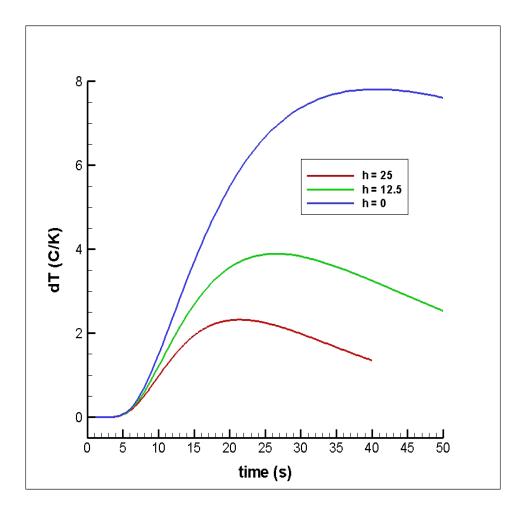


Figure 15: Temperature curves for three different value of the heat transfer coefficient. Blue represents the curve with no convection while the other two show two different values off by a factor of two.

Since the heat transfer coefficient is a function of the surface, we account for convection by either creating models with a range of coefficients and seeing which one fits best or by comparing a sample with a known thermal conductivity with an unknown that has a similar geometry. In the latter we compare the experimental data with the model and use that difference to get our heat transfer coefficient.

RESULTS

Using a silver sample similar to those shown previously we plotted the experimental raw data along side predictions for the established thermal diffusivity of 165mm²/s with additional curves at 10% either way.

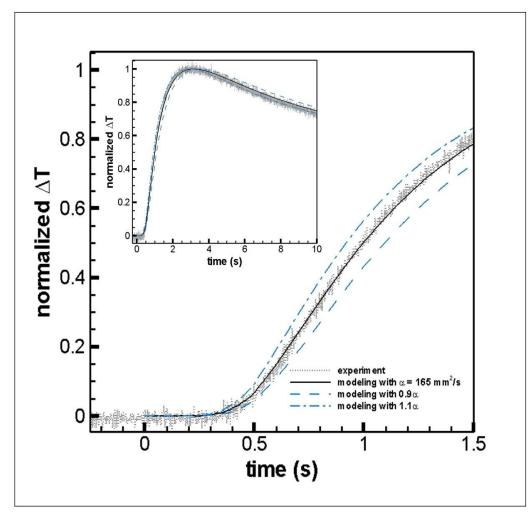


Figure 16: Silver experimental data with simulated curve overlay.

We can clearly see from fig 15 that our apparatus is in very close agreement with established values. Having established that the method is indeed valid we then proceeded with the testing of a graphene layer upon a ceramic substrate. The graphene layer was approximately 600nm thick upon 3.1mm of ceramic material showing that the $\eta \ll 1$ assumption is valid. Since graphene was previously measured in its mono-layer form to have one of the highest thermal conductivities known and ceramic is low compared to the traditional high conductivity materials the $\kappa_c \gg \kappa_s$ assumption also holds valid. Fig 16 shows data for the sample with and without the ceramic substrate and predictions from the simulation for several values of the thermal conductivity for the graphene layer.

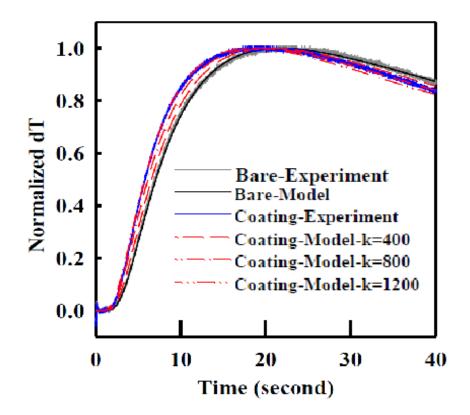


Figure 17: Graphene thermal response tests showing the data and model for the uncoated ceramic along with the data for the graphene coated ceramic with three curves corresponding to k = 400W/mK, k = 800W/mK, and k = 1200W/mK [12].

We can see that the 1200W/mK curve fits the closest to the experimental data allowing us to determine the graphene has a thermal conductivity of approximately 1200W/mK. Slightly higher than that of in-plane graphite which has a conductivity of approximately 1000W/mK [1].

CONCLUSION

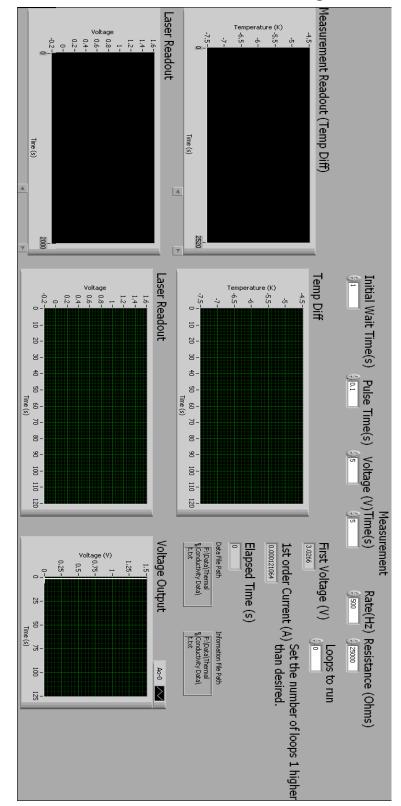
We have shown that the laser method combined with computer modeling can accurately measure thermal properties without the use of a vacuum or the cumbersome apparatii of older techniques. A final end-all test of the system with a sample measured inside and outside a vacuum chamber is currently in progress. The graphene measurements appear to agree with other measurements done on few-layer graphene samples. Specifically the measurement of ~1300W/mK on a four layer sample by Ghosh et. Al [10].

There are ways in which the apparatus could be improved. For instance, we could replace the physical contact detector with an IR sensor to measure the temperature profile without needing to account for the thermistor response. Without having to go through the difficulty of attaching the thermistor to the sample, preparation would become significantly easier, but at the same time new complications would arise from the more complex tool. In the end we present a viable method for measuring thermal properties that is easy to construct, simple to operate, and more economic than most alternatives.

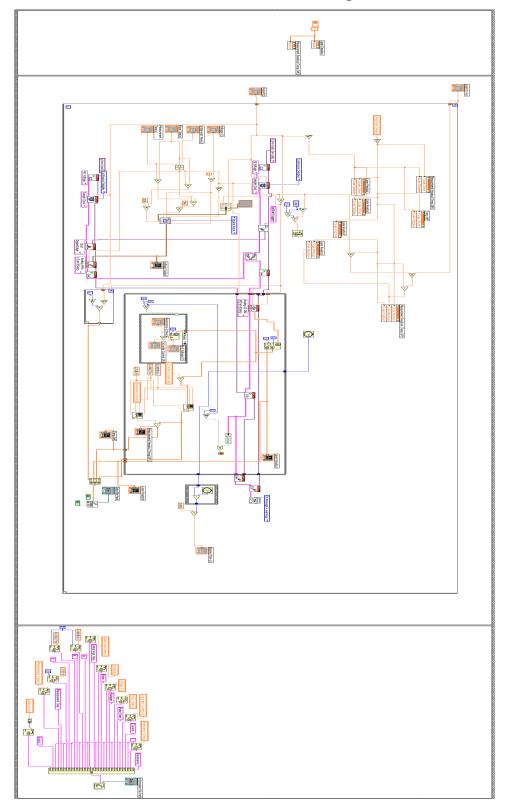
REFERENCES

- 1) Alofi, A., and G. P. Srivastava. "Thermal conductivity of graphene and graphite." *Physical Review B*. no. 11 (2013).
- 2) Ash, J., J. Baldwin, J. Hirt, and A. Lance. "Heat Conductions." presentation, Viginia Tech University, http://www.math.vt.edu/people/gao/class_home/Presentations/heatconduction.ppt
- 3) Balandin, A. A., S. Ghosh, W. Bao, I. Calizo, D. Teweldebrhan, F. Miao, and C. N. Lau. "Superior Thermal Conductivity of Single-Layer Graphene." *Nano Letters*. no. 3 (2008): 902-907.
- 4) Corbino, O. M. "Measurement of specific heats of metals at high temperatures." *Atti della Reale Accademia Nazionale dei Lincei*. (1912): 181-188.
- 5) Cahill, D. G., and R. O. Pohl. "Thermal conductivity of amorphous solids above the plateau." *Physical Review B*. no. 8 (1987): 4067-4073.
- 6) Cahill, David G. "Measurement of thermal conductivity." lecture. University of Illinois at Urbana-Champaign, http://users.mrl.uiuc.edu/cahill/thermal_school09.pdf.
- 7) Center for Advanced Life Cycle Engineering, "Details about Property Measurements." http://www.calce.umd.edu/TSFA/laser_flash/details.htm.
- 8) de Koninck, David. *Thermal conductivity measurements using the 3-omega technique: application to power harvesting microsystems*. Masters thesis, McGill University, 2008.
- 9) Engineers Edge, "Convection Review Heat Transfer." http://www.engineersedge.com/heat_transfer/convection.htm.
- 10) Ghosh, S., et. al. "Extremely high thermal conductivity of graphene: Prospects for thermal management applications in nanoelectronic circuits al transport in few-layer graphene." *Applied Physics Letters*. no. 15 (2008).
- Ghosh, S., W. Bao, D. L. Nika, S. Subrina, E. P. Pokatilov, C. N. Lau, and A. A. Balandin. "Dimensional crossover of thermal transport in few-layer graphene." *Nature*. (2010): 555-558.
- 12) Huang, Wenyi, et al. "Carbide-Bonded Graphene Coating." Sumitted for publication. .
- 13) Lasever, "LSR1064-NL 100~1200mW IR lasers." http://www.lasever.com/productitle.asp-id=1381.htm.

- 14) Parker, W. J., R. J. Jenkins, C. P. Butler, and G. L. Abbott. "Flash method of determining thermal diffusivity, heat capacity, and thermal conductivity." *Journal of Applied Physics*. no. 9 (1961): 1679.
- 15) Personal Communication with Dr. Gregory Lafyatis.
- 16) Steinhart, J. S., and S. R. Hart. "Calibration curves for thermistors." *Deep-Sea Research*. no. 4 (1968): 497-503.
- 17) Stewart, Dr. Doug. Chemicool, "Silver Element Facts." Last modified 2013. http://www.chemicool.com/elements/silver.html.
- 18) Vozar, L., and W. Hohenauer. "Flash method of measuring the thermal diffusivity. A review." *High Temperatures High Pressures*. no. 3 (2003/2007): 253-264.



APPENDIX I: LabVIEW Front Page



APPENDIX II: LabVIEW Block Diagram

APPENDIX III: Mathematica Program

(*Data Set 1*) (* Get the all the information stored within the info file*) ininfo = ReadList["F:/Data/Thermal Conductivity Data/Silver Sample 1um, 3cm with Cap .1s Pulse, 5V 4-1-13 info.txt", Word, RecordLists - True, WordSeparators - {" "}]; Resistance = ToExpression[ininfo[[1]][[2]]]; Current = ToExpression[ininfo[[1]][[4]]]; PulseTime = ToExpression[ininfo[[1]][[7]]]; Voltage = ToExpression[ininfo[[1]][[9]]]; Rate = ToExpression[ininfo[[1]][[11]]]; InitialWaitTime = ToExpression[ininfo[[1]][[15]]]; a = ToExpression[ininfo[[1]][[17]]]; b = ToExpression[ininfo[[1]][[19]]]; c = ToExpression[ininfo[[1]][[21]]]; MeasurementTime = ToExpression[ininfo[[1]][[24]]]; If[Length[ininfo[[1]]] > 24, Loops = ToExpression[ininfo[[1]][[26]]], Loops = 20]; OneRunLength = Rate * (MeasurementTime + InitialWaitTime + PulseTime) LaserStartPosition = {Rate * InitialWaitTime}; (*Get the data from the data file for plotting manipulation*) intable1 = ReadList["F:/Data/Thermal Conductivity Data/Silver Sample 1um, 3cm with Cap .1s Pulse, 5V 4-1-13.txt", Word, RecordLists → True, WordSeparators → {"\t"}]; Ttable1 = Transpose[intablea1]; AbsoluteTempList = ToExpression[Ttable1[[1]]]; TempDiffList = ToExpression[Ttable1[[2]]]; LaserOutList = ToExpression[Ttable1[[3]]]; TimeList = ToExpression[Ttable1[[4]]]; AbsoluteTempLists = Partition[AbsoluteTempList, Length[AbsoluteTempList] / Loops]; TempDiffLists = Partition[TempDiffList, Length[TempDiffList] / Loops] LaserOutLists = Partition[LaserOutList, Length[LaserOutList] / Loops]; TimeLists = Partition[TimeList, Length[TimeList] / Loops]; For[i = 1, i <= Loops, i++, AppendTo[LaserStartPosition, 1+Length[TakeWhile[LaserOutLists[[i]], # < Voltage - 1 6]]]]
For[i = 1, i <= Loops, i++, TimeLists[[i]] = TimeLists[[i]] - TimeLists[[i]][LaserStartPosition[[i+1]]]];</pre> For[i = 1, i <= Loops, i++, TempDiffLists[[i]] = TempDiffLists[[i]] - TempDiffLists[[i]][[LaserStartPosition[[i+1]]]]; ListOfLists1 = {Transpose[{TimeLists[[2]], TempDiffLists[[2]]}]; ListOfLaserLists1 = {Transpose[{TimeLists[[2]], LaserOutLists[[2]]}}; For[i = 3, i <= Loops, i++, AppendTo[ListOfLists1, Transpose[{TimeLists[[i]], TempDiffLists[[i]]}]]; For[i = 3, i <= Loops, i++, AppendTo[ListOfLaserLists1, Transpose[{TimeLists[[i]], LaserOutLists[[i]]}]];</pre> AverageList = TempDiffLists[[2]]; For[i = 3, i <= Loops, i++, AverageList = AverageList + TempDiffLists[[i]]];</pre> AverageList = AverageList / Loops; AveragedPlot1 = Transpose[{TimeLists[[1]], AverageList}]; TempDiffMax = Max[AverageList]; NormalizedList = AverageList / TempDiffMax; NormalizedPlot1 = Transpose[{TimeLists[[1]], NormalizedList}]; (*Data Set 2*) (* Get the all the information stored within the info file*) ininfo = ReadList["F:/Data/Thermal Conductivity Data/Silver Sample 1um, 3cm no Cap .1s Pulse, 5V 4-1-13 info.txt", Word, RecordLists -> True, WordSeparators -> {" "}]; Resistance = ToExpression[ininfo[[1]][[2]]]; Current = ToExpression[ininfo[[1]][[4]]]. PulseTime = ToExpression[ininfo[[1]][[7]]]; Voltage = ToExpression[ininfo[[1]][[9]]]; Rate = ToExpression[ininfo[[1]][[11]]]; InitialWaitTime = ToExpression[ininfo[[1]][[15]]]; a = ToExpression[ininfo[[1]][[17]]]; b = ToExpression[ininfo[[1]][[19]]]; c = ToExpression[ininfo[[1]][[21]]]; MeasurementTime = ToExpression[ininfo[[1]][[24]]]; If[Length[ininfo[[1]]] > 24, Loops = ToExpression[ininfo[[1]][[26]]], Loops = 20]; OneRunLength = Rate * (MeasurementTime + InitialWaitTime + PulseTime); LaserStartPosition = {Rate * InitialWaitTime}; (*Get the data from the data file for plotting manipulation*)
intablea1 = ReadList["F:/Data/Thermal Conductivity Data/Silver Sample 1um, 3cm no Cap .1s Pulse, 5V 4-1-13.txt", Word, RecordLists → True, WordSeparators → {"\t"}]; Ttable1 = Transpose[intablea1]; AbsoluteTempList = ToExpression[Ttable1[[1]]]; TempDiffList = ToExpression[Ttable1[[2]]]; LaserOutList = ToExpression[Ttable1[[3]]]; TimeList = ToExpression[Ttable1[[4]]]; AbsoluteTempLists = Partition(AbsoluteTempList, Length[AbsoluteTempList] / Loops]; TempDiffLists = Partition(TempDiffList, Length[TempDiffList] / Loops]; LaserOutLists = Partition[LaserOutList, Length[LaserOutList] / Loops]; TimeLists = Partition[TimeList, Length[TimeList] / Loops]; For[i = 1, i <= Loops, i++, AppendTo[LaserStartPosition, 1 + Length[TakeWhile[LaserOutLists[[i]], # < Voltage - 1 &]]]]</pre> For[i = 1, i <= Loops, i++, TimeLists[[i]] = TimeLists[[i]] - TimeLists[[i]][LaserStartPosition[[i+1]]]];
For[i = 1, i <= Loops, i++, TempDiffLists[[i]] = TempDiffLists[[i]] - TempDiffLists[[i]][LaserStartPosition[[i+1]]]];</pre> ListOfLists2 = {Transpose[{TimeLists[[2]], TempDiffLists[[2]]}); ListOfLaserLists2 = {Transpose[{TimeLists[[2]], LaserOutLists[[2]]}]; For[i = 3, i <= Loops, i++, AppendTo[ListOfLists2, Transpose[{TimeLists[[i]], TempDiffLists[[i]]}]];
For[i = 3, i <= Loops, i++, AppendTo[ListOfLaserLists2, Transpose[{TimeLists[[i]], LaserOutLists[[i]]}]];</pre> AverageList = TempDiffLists[[1]]; For[i = 2, i <= Loops, i++, AverageList = AverageList + TempDiffLists[[i]]];</pre> AverageList = AverageList / Loops AveragedPlot2 = Transpose[{TimeLists[[1]], AverageList}]; TempDiffMax = Max[AverageList]; NormalizedList = AverageList / TempDiffMax; NormalizedPlot2 = Transpose[{TimeLists[[1]], NormalizedList}];

(*Plot the Data*) (*Raw Data*)

Show[ListPlt[ListOfLists2, Joined + True, AxesLabel + {Time, dT}, PlotRange + {{-1, 20}, {-.02, .1}}, LabelStyle + Directive[FontSize + 15], GridLines + Automatic], Show[ListPict[ListOfLists2, Joined + True, AxesLabel + {Time, dT}, PlotRange + {(-1, 20), {-.02, .1}}, LabelStyle + Directive[FontSize + 15], OridLines + Automatic]; ListPlot[ListOfLists1, Joined + True, AxesLabel + {Time, dT}, PlotRange + {(-1, 20), {-.02, .1}}, LabelStyle + Directive[FontSize + 15], OridLines + Automatic] ListPlot[ListOfLists1, Joined + True, AxesLabel + {Time, dT}, PlotRange + {(-1, 20), {-.02, .1}}, LabelStyle + Directive[FontSize + 15], OridLines + Automatic] ListPlot[ListOfLists2, Joined + True, AxesLabel + {Time, dT}, PlotRange + {(-1, 20), {-.02, .1}}, LabelStyle + Directive[FontSize + 15], OridLines + Automatic] ListPlot[ListOfLists1, Joined + True, AxesLabel + {Time, dT}, PlotRange + {(-1, 00, {-.02, .1}), LabelStyle + Directive[FontSize + 15], OridLines + Automatic] ListPlot[ListOfLists2, Joined + True, AxesLabel + {Time, "Voltage"}, PlotRange + {(-0, 05, .05), {-.02, 5.1}}, LabelStyle + Directive[FontSize + 15], OridLines + Automatic]

dified Data*)

ListPlot[{AveragedPlot1, AveragedPlot2}, AxesLabel + {Time, dT}, PlotRange + {{-1, 20}, {-.02, .1}}, PlotStyle + {Red, Blue, Green}, LabelStyle + Directive[FontSize + 15], Joined \rightarrow True, GridLines \rightarrow Automatic] ListPlot[NevingAverage[AveragedPlot], 5], NovingAverage[AveragedPlot2, 5]], AxesLabel → {Time, dT}, PlotRange → {{-1, 20}, {-.02, .1}}, PlotStyle → {Red, Blue, Green},

ListPlot([MovingAverage[AveragedPlot], 5], MovingAverage[AveragedPlot2, 5]), AxesLabel + (Time, dT), PlotRange + {(-1, 20}, (-.02, .1)), Pl LabelStyle + Directive[TontSize + 15], Joined + True, Gridlines + Automatic] ListPlot[[MovingAverage[AveragedPlot], 10], MovingAverage[AveragedPlot2, 10]], AxesLabel + (Time, dT), PlotRange + {(-1, 20), (-.02, .1)}, PlotStyle + [Red, Blue, Green], LabelStyle + Directive[TontSize + 15], Joined + True, GridLines + Automatic] ListPlot[[MormalizedPlot], NormalizedPlot2], AxesLabel + (Time, dT), PlotRange + {(-1, 20), (-.05, 1)}, PlotStyle + {Red, Blue, Green}, LabelStyle + Directive[TontSize + 15], Joined + True, GridLines + Automatic]

(soutput the average and moving average to a files)

Export["F:/Data/Thermal Conductivity Data/Silver Sample 1um 4-1, 3cm wCap 0.1s Loop Average.txt", AveragedPlot1]

Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam WCap 0.1s Loop Average.txt", AveragedPlot1]
Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam KCap 0.1s Hoving Average.txt", NovingAverage[AveragedPlot1, 5]]
Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam KCap 0.1s Hoving Average.txt", AveragedPlot2]
Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam KCap 0.1s Hoving Average.txt", AveragedPlot2]
Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam KCap 0.1s Hoving Average.txt", AveragedPlot2
Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam KCap 0.1s Hoving Average.txt", AveragedPlot2
Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam KCap 0.1s Hoving Average.txt", AveragedPlot2, 5]]
Export["F:/Data/Thermal Conductivity Data/Silver Sample lum 4-1, Sam KCap 0.1s Hoving Average.txt", AveragedPlot2, 5]]