Influence of the sample mounting on thermal conductance measurements using PPMS TTO option

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Abstract. We discuss the performance of the automated heat conductivity measurement system manufactured by the Quantum Design, Inc. The Thermal Transport Option implemented into the Physical Properties Measurement System (PPMS) measures the thermal transport properties of materials (thermal conductivity, Seebeck coefficient and electrical resistivity simultaneously) in the temperature range 1.8 – 395 K and in magnetic fields generated by the installed superconducting solenoid. Recently, discrepancies up to 30% in measured quantities at 390 K have been reported. We critically analyze the experimental method used to measure the above mentioned quantities and show possible sources of problems.

1. Introduction
The thermal conductivity belongs to the most significant characteristics of materials. The experimental method of its measuring is based on the definition of the heat conductance as an amount of heat $Q$ driven through the material by unit temperature gradient $\Delta T$, $k = Q / \Delta T$. This simple definition, however, meets technical obstacles when exactly determining the individual quantities, namely the heat flow. In real experiments, the sample under study together with the sensors is not ideally thermally isolated from its surroundings and the heat flow is thus not exactly defined. Various methods to measure the thermal conductivity have been published; the most common cases are (i) one-heater-two-thermometers configuration [1] and (ii) two-heaters - three thermometers [2] configuration. For its Physical Properties Measuring System, Quantum Design, Inc., developed the Thermal Transport Option (TTO) that uses the one-heater-two thermometers configuration.

Recently Rudajevova et al [3] published the thermal conductivity data of various materials obtained using the TTO. Based on the SRM 8420 from National Institute of Standards and Technology the difference between the measured data and those cited by NIST reached about 30% at 390 K. The contribution presents a result of series of experiments in order to find weak points of this commercial device.

2. Thermal transport measurements
The QD TTO for the PPMS enables a measurement of thermal conductance $k$, thermopower (Seebeck coefficient) $\alpha$ and electrical resistance $\rho$, for sample materials over the entire temperature and magnetic field range of the PPMS. TTO measures thermal conductance by monitoring the temperature drops $\Delta T$ along the sample as a known amount of heat passes through the sample. While the
measurements taken with the TTO system are quite elementary in principle, to get the data was typically very error prone, time consuming, and laborious, due - for example - to problems in controlling heat flow and accurately measuring small temperature differentials in a convenient manner. The TTO system has solved or greatly reduced many of these experimental complications.

There are two modes of measurement - single and continuous. In the single mode the system reaches a steady state in both the heater "off" and "on" states. To determine the thermal conductance \( k \), the stable time independent data for \( \Delta T = T_{\text{hot}} - T_{\text{cold}} \) are used. The second one, continuous, is more complicated for evaluation of the data.

Having established the stable temperature gradient over the sample the thermal conductance \( k \) is calculated from \( k = P/\Delta T \) where \( P \) is the heat flowing through the sample. Since the heat flux cannot be measured directly, the net conducted heat through the sample is estimated as the power dissipated in the heater resistor, minus losses due to radiation (\( P_{\text{rad}} \)) and thermal conduction down the leads from the shoe assemblies (\( K_{\text{shoe}} \)). The conductance is calculated from the equation (1)

\[
k = \frac{RI^2 - P_{\text{rad}}}{\Delta T} \quad \Delta T = T_{\text{hot}} - T_{\text{cold}}.
\]

where the parameters are estimated as

\[
K_{\text{shoe}} = aT + bT^2 + cT^3
\]

\[
P_{\text{rad}} = \sigma_T (S/2) \varepsilon (T_{\text{hot}}^4 - T_{\text{cold}}^4)
\]

Here \( I^2R \) is the Joule heat produced in the heater of resistance \( R \) by the current \( I \); \( a, b \) and \( c \) are constants given by QD, Stephan-Boltzmann constant \( \sigma_T = 5.7 \times 10^{-8} \text{ W/K}^4\text{m}^2 \), \( S \) is sample surface area and \( \varepsilon \) its emissivity. The equation (1) is derived from well-known general heat equation for the one-dimensional case and the simplest boundary conditions. The procedure implicitly assumes that the temperature of any part of the assembly is close to the system temperature; the sample properties are changing with temperature slowly. The strongest boundary condition is that the temperature gradient exists just only on the sample part between the thermometers.

3. Experiment

We used two samples for measurement of the thermal conductance. The nickel standard, which is a part of the TTO for testing, is made of thin metal sheet 0.25 mm thick and 1.25 mm wide. The central part of the sample, of the length 8.3 mm, is used for the experiment; the legs for connecting thermometers, heater and basement (\( T_{\text{system}} \)) are about of the same length as the central part. Nickel is of 201 grade [4]; our experiment gives low value of RRR = 5.5, which suggests a low purity. As a next sample, Ni-foil, was prepared with a special distribution of contact pads. It is a strip of a 0.2 mm thick nickel foil 1.7 mm wide and about 22 mm long. The pads made of 0.4 mm dia Cu wire were soft soldered to both ends and at a distance 2 mm and 10 mm from one side of the strip. The experiment has been performed with two positions of the sample with respect to the basement - for clarity marked "down" and "up", see the inset of figure 2. The thermal conductance is always measured over the same (8 mm long) part of the sample between thermometers.

We have chosen the single mode experiment that waits until the temperature in the "heater off" period is stable with a given precision (\( \Delta T/T < 0.1\% \) in our case) and then it switches "heater on" and waits for stability again. The output of the software represents a full set of quantities evaluated by the fitting procedure of measured \( T_{\text{hot}}, T_{\text{cold}} \) and \( T_{\text{system}} \).

In the experiment we gradually lowered \( P \) with decreasing temperature (12 mW above 100 K, 2 mW in the range 30 K < \( T < 100 \) K, 0.5 mW for 10 K < \( T < 35 \) K and 0.1 mW below 10 K).

4. Results and Discussion

Figure 1 presents the data of our nickel grade 201 compared with the QD nickel standard data available on the web page of Quantum Design, Inc. The measured heat conductance deviates from the published values for a \( T^3 \) term. The discrepancy of the measured and standard data are about 12 % at
350 K, which is less than published by Rudajevova. It could be caused by a different way of making the thermal contacts. Rudajevova used an epoxy bond, which impairs the thermal contact efficiency.

In attempt to find possible sources of systematic errors we shall concentrate on two main problems – a) thermal contacts between the base and the sample and b) thermal difference between the hot thermometer and the heater. The TTO software calculates the conductivity using equation (1). It can be rewritten in form of energy balance

\[ F R = k \Delta T + P_{\text{rad}} + K_{\text{shoe}} (T_{\text{hot}} - T_{\text{system}}) + K_{\text{shoe}} (T_{\text{system}} - T_{\text{cold}}). \]  

Here we stress that the heat flows between temperatures explicitly shown in the parentheses on the RHS. The energy flux from the heater (LHS of the equation (4)) consists of the heat flow through the sample, radiation losses from the sample, heat flow through the upper and the lower shoes. In the real experiment there is a non-ideal thermal contact of the sample to the base. Because \( T_{\text{system}} \) is the lowest temperature, the last term in the RHS is negative! and the resulting conductance is therefore high.

In the case b) the hot thermometer and the heater are at different temperatures and no additional correction is done for it. In both cases the procedure brings systematic errors. The experiment with our Ni-foil sample should test the sensitivity of the procedure to the different boundary conditions. We had expected different thermal losses for each position, resulting in different measured thermal conductance. An example of the time record of the temperatures during of the one measured point at 270 K is given in figure 2. The influence of an additional thermal resistance of the sink namely on the \( T_{\text{cold}} \) is evident.
Results in the deduced thermal conductances for both sample positions are displayed in figure 3. The values $P_{rad}$ and $K_{shoe}$ are really different in both cases but their overall influence is surprisingly small.

We could try to take into account the fact that $T_{cold} > T_{system}$ by using

$$k = k^{QD} - K_{shoe} \left( \frac{T_{rise}}{\Delta T} - 1 \right)$$

(5)

where $k$ is the conductance corrected for $T_{cold} > T_{system}$, $k^{QD}$ is the value given by the TTO procedure and $T_{rise} = T_{hot}^{ON} - T_{hot}^{OFF}$ the increase of $T_{hot}$ by the heater pulse.

The corrections are also plotted in figure 3. It stands to reason that the corrections are not sufficient. The correction for the upper side of the sample, $T_{heater} > T_{hot}$, is still missing due to a lack of knowledge of $T_{heater}$. The next attempt for correction could be highly speculative.

![Figure 3. Thermal conductivities of the nickel foil for both positions (see text) and their corrections.](image)

5. Conclusions

The Thermal Transport Option installed in the PPMS utilizes the simplest approach to the thermal conductance measurement. In real experiments, however, one often meets situations beyond limited parameter space suitable for this approach that appreciably affect results of the experiment. According to our experience, doubts about correctness of the results exist in the temperature range above 100 K. There is no simple proof that would exactly tell whether or not the experiment is set up correctly, however, following the advices below one can try to minimize the errors:

a) Choose sample dimensions such that you can use maximal available heater power. It minimizes the needed corrections.

b) Any temperature gradient along the sample not corresponding to the space distribution of the thermometer contacts serves as an evidence of a bad thermal contact to the base. Remake contacts to the base and to the heater (enlarge the surface area, solder it instead of gluing, etc).

c) Two-probe lead configuration could be another compromise.

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References

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