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Radiative heat losses in thermal conductivity measurements: a correction for linear temperature gradients

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ABSTRACT

A method for the quantification of the radiative heat losses in a steady-state thermal conductivity measurement set-up is developed based on the Stefan–Boltzmann radiation law with the assumption of a linear temperature distribution along the specimen. The resulting expression can be applied to any regular-shaped sample with well-defined side surfaces and cross-section. Owing to the quantification of the radiative heat losses the accuracy of the steady-state thermal conductivity measurement method is improved and the measurement range can be extended to higher temperatures. An exemplary application in a commercial device is presented. The results are in an excellent agreement with the independently measured high temperature thermal conductivity.

1. Introduction

Conductive heat transport is important for thermal management in building technology, vehicles as well as in production and conversion of energy. For instance, the efficiency of thermoelectric conversion depends on the thermal conductivity of thermoelectric materials. Therefore, in the mentioned areas a precise and reliable thermal conductivity measurement method is of great importance [1,2].

One of the simplest ways to measure the thermal conductivity is to exploit the one-dimensional Fourier heat transfer equation

$$\dot{Q}_{sam} = \kappa_x A\left(-\frac{dT}{dx}\right), \tag{1}$$

where $\dot{Q}_{sam}$ is the heat flow through the sample, κ_x is the thermal conductivity, A is the cross-section of the specimen and $\frac{dT}{dx}$ is the temperature gradient. Here and below the heat conduction process will be considered as one-dimensional flow along the x direction. In general, thermal conductivity depends on the orientation of the material in case the anisotropy results from the crystal structure or texturing. In the experimental arrangement discussed in this work only the component along x-axis is considered. For measuring κ in other directions the sample needs to be oriented adequately.

Heat flows from the high T to the low T region. In the steady-state arrangement a constant temperature gradient over the sample is established and the heat flow can be expressed as

$$\dot{Q}_{sam} = \kappa A\left(-\frac{\Delta T}{\Delta x}\right), \tag{2}$$

where ΔT is the temperature difference measured over the distance Δx. In this case it is possible to determine κ when $\dot{Q}_{sam}$, the sample cross-section and at least 2 temperature measurement points and their positions along the x-axis are known. However, at temperatures much higher than the absolute zero a precise estimation of $\dot{Q}_{sam}$ is challenging. If the source of the heat current is represented by P, the energy flow balance can be written as an equation

$$\dot{Q}_{sam} = P - \dot{Q}_{conv} - \dot{Q}_{cond} - P_{rad}, \tag{3}$$

schematically represented in Fig. 1, where $\dot{Q}_{conv}$ stands for convective heat losses, $\dot{Q}_{cond}$ for conductive heat losses and P_{rad} for radiative heat losses. The thermal conductivity κ can be now expressed as:

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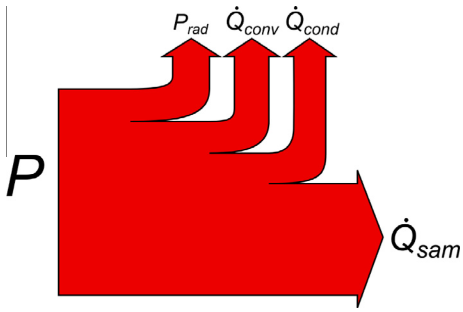

Fig. 1. A scheme of energy transfer in a thermal conductivity measurement.

$$\kappa = \frac{P - \dot{Q}_{conv} - \dot{Q}_{cond} - P_{rad}}{A}\left(-\frac{\Delta T}{\Delta x}\right)^{-1}. \quad (4)$$

In the presence of non-negligible heat losses the κ values obtained from Eq. (4) can be significantly different from the one obtained by assuming zero heat loss ($\dot{Q}_{sam} = P$). In this case omitting the relevant term ($\dot{Q}_{conv}, \dot{Q}_{cond}$ or P_{rad}) leads to overestimation of κ. The overview of experimental difficulties related to minimizing the loss terms and how to cope with them can be found in [3].

An introduction to the problem of heat losses in a thermal conductivity measurement can be found in [4]. If $\dot{Q}_{sam} \approx P - P_{rad}$ the radiative heat losses can be extracted analytically from the data, as described in [3]. The thermal conductivity of materials can be divided in two parts: the charge carrier thermal conductivity κ_{carr} and the lattice thermal conductivity κ_{latt}:

$$\kappa = \kappa_{carr} + \kappa_{latt}. \quad (5)$$

Using the Wiedemann–Franz relationship ($\kappa_{carr} = LT\sigma$) κ_{carr} can be calculated approximately from the electrical conductivity σ and the Lorenz number for metals $L = 2.44 \times 10^{-8}\ \mathrm{W\,\Omega\,K^{-2}}$. Subsequently, κ_{latt} can be obtained by a simple subtraction $\kappa - \kappa_{carr}$. In many cases it is justified to assume a certain dominant phonon scattering process resulting in a defined dependence of the lattice thermal conductivity on the temperature, for instance in case of phonon–phonon Umklapp scattering $\kappa_{latt}(T) \sim \frac{1}{T}$ [4]. Therefore, a deviation from the expected $\kappa_{latt}(T)$ behavior is often related to the neglected P_{rad} term, which starts to be visible at T higher than approximately 200 K. However, in cases when the Lorenz number is not constant or when the dominant phonon scattering mechanism is not clear the above described correction is not reliable. In the current work an estimation of the P_{rad} term is proposed without assumptions on the sample properties.

A previous attempt to estimate the contribution of the radiation term to the heat losses was done by measuring κ several times for the same material in similar conditions, but each time with different total surface of the sample S [5]. Since the radiation term depends linearly on the radiating surface size the subsequent extrapolation of the $\kappa(S)$ dependency to $S = 0$ allowed for elimination of the radiation term. However, this experimental procedure is relatively time-consuming, tedious and requires several samples of the same material.

Another approach represented the measurement system and the sample numerically and employed finite element modeling to describe the physical process of heat conduction and radiation [6]. The radiation was included in the model as a boundary condition on all free surfaces. It was demonstrated that including the heat losses from the elements of the experimental setup, especially from the heater, has a large impact on the final value of κ. However, the influence of the temperature distribution $T(x)$ along the heat conduction direction on the experimental results was not analyzed. The solution proposed in [6] is limited to materials with a thermal conductivity similar to electrolytic iron, which was used as a reference.

An advanced numerical solution of analytical equations representing the radiative heat exchange between a cylindrical sample within a cylindrical radiation shield in a thermal conductivity measurement apparatus was analyzed in [1]. The aim of that work was to quantify the radiative heat losses and to design an experimental setup according to the assumed geometry which would follow the elaborated equations and minimize the measurement error. The approach is found to produce reliable results, however limited to a certain geometrical arrangement and at exceptionally high computational expense. The model developed in the current work simplifies the geometry details and results in much simpler expression for κ, retaining good agreement with the reference, high-temperature data.

2. Mathematical model of the experiment

The model of a steady-state thermal conductivity measurement is schematically presented in Fig. 2. Usually, the heat flow is imposed by connecting the sides of the specimen to an external heat source and a heat sink. As aforementioned, it is necessary to know the heating power P of the heat source and estimate the heat losses due to convection $\dot{Q}_{conv}$, conduction $\dot{Q}_{cond}$, and radiative transfer P_{rad} in order to correctly determine the amount of heat transferred through the sample. Convective heat losses are most often negligible as the measurement chamber is typically evacuated to high vacuum. The conductive heat loss $\dot{Q}_{cond}$ depends on the measurement setup and includes all sources of heat leakage, for instance electrical connections to the sample thermometers or to the heater. In many cases $\dot{Q}_{cond}$ can be neglected. Otherwise it can be represented as:

$$\dot{Q}_{cond} = K_{cond}(T)\Delta T_{est}, \quad (6)$$

where $K_{cond}(T)$ is a temperature-dependent conductance comprising all conductive heat losses and ΔT_{est} is an estimation of the temperature difference driving them [7].

To estimate the radiative heat loss P_{rad} a model is developed assuming radiative heat transfer between the sample and the ambiance (c.f. Fig. 2, where the sketch of the measurement setup is presented). A linear temperature distribution between a heat source at c and a heat sink at $-w$ over the sample is assumed with

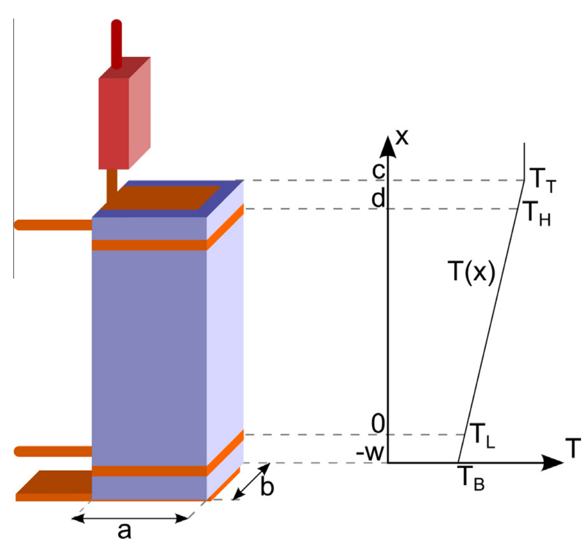

Fig. 2. A model of a steady-state thermal conductivity measurement.