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A practical cryogenic resistive sensor for thermal conductivity measurements

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Abstract

The line heat-source method is one among many classes of transient methods for measuring thermal properties, such as thermal conductivity, diffusivity and specific heat, which are directly related to heat conduction. Belonging to this category is the transient plane source (TPS) technique for simultaneous measurements of thermal conductivity and diffusivity of solids. The technique uses a 'resistive element' made of nickel foil in the form of a bifilar spiral (TPS element/sensor) covered on both sides with an insulating layer of Kapton. For the first time, the calibration procedures of such a sensor have been extended down to 35 K. By fitting the calibration data to a fourth-order polynomial, a reliable equation for the measured resistance of the TPS element covering the whole temperature range of interest (35–300 K) was obtained. A moderate decrease in the sensitivity of this sensor was observed around 60 K; it is attributed to a nearly temperature-independent resistivity coefficient (TCR) behaviour in this temperature range. The sensitivity decline is linked to a design factor that is related to the resistance behaviour of the material used in the construction of the sensor. To improve the practicality of using the sensor in this temperature range, a comparison was made between the behaviour of the TCR values of nickel and silver. It is found that a sensor made of silver foil will improve the accuracy and extend the measurements to lower temperatures.

Keywords: Cryogenic; Transient methods; Sensors; Thermal conductivity

1. Introduction

The advancement of technology requires the development of new materials having physical properties suitable for use in severe conditions, for instance, the materials used in the construction of the high-pressure vessels in chemical distillation plants or in the construction of large furnaces. Furthermore, a fundamental understanding of the physical processes that are related to the transport of thermal energy in some of these new materials is also essential. Therefore, the development of experimental methods to measure these properties under various environmental conditions should follow a line parallel with this progress. The reliability of a specific method is decided by several factors, such as the speed of operation, the required accuracy and performance under various environmental conditions, the physical nature of material, and the geometry of the available sample. However, in most methods the main concern is to obtain a controlled heat flow in a prescribed direction, such that the actual boundary conditions in the experiment agree with those assumed in the theory.

A class of methods for measurements of thermal properties is the transient methods. In these methods, the sample is initially at thermal equilibrium with the surroundings. Then one end of the sample is subjected to a short heating pulse. The change in temperature is monitored at one or more points during the time of measurement. The thermal diffusivity is then evaluated by correlating the experimental temperature measurements with the theoretical relationship obtained from the solution of the differential heat equation. Solutions for different experimental arrangements and various boundary conditions are given by Carslaw and Jaeger [1].

The line heat-source method is a specific transient method in which a line heat source (wire or strip) is embedded in the specimen initially at uniform temperature. With this method, it is possible to measure both the heat input and the temperature changes, and then the thermal conductivity and diffusivity are simultaneously determined.

The transient hot wire (THW) method based on the empirical work of Stålhane and Pyk [2], and the transient hot-strip (THS) method initially introduced by Gustafsson [3] for the study of transparent liquids belong to this specific class. It is based on a transient temperature rise of the hot strip (thin metal foil) at constant energy input. The strip is used as both a constant heat source and temperature sensor. The duration

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of the current pulse (time of measurement) is usually of the order of a few seconds. Measurements are simply performed by recording the voltage (resistance/temperature) variations across the strip, which is supplied with pulsed electrical current. Recently, an extension of the THS method has led to the development of a method called the transient plane source (TPS) method that uses a 'resistive element' (TPS element/sensor) in the same way as the strip is used in the THS method [4]. In this work, we are investigating important design remarks related to the fabrication of this resistive sensor used for thermal conductivity measurements at low temperature.

2. Experimental

2.1. Design requirements

One of the major developments of the THS method leads to the TPS method. This development was first meant to ease the strip handling in the THS method, and to match several design requirements, some of which are the following:

1. To make a strip which can be protected against any mechanical damage that could happen while preparing the experiment.
2. To use the same strip for measurements on different samples.
3. The circuit design improves if the strip has a larger resistance (see discussion below).
4. To find a practical way to retain the same properties (type and thickness) of the insulating layer for specific measurements.
5. To improve the way of connecting the electrical leads to the strip.

These requirements, and others [5], finally led to the design of the TPS element/sensor [6].

This TPS element/sensor is used in the TPS method both as heat source and temperature sensor. There are a variety of geometric configurations [7] for the sensors, such as disks, squares, etc. Which geometric configuration is most convenient depends on many factors such as material type, sample size, kind of leads used, etc. The sensor used in this work is made of nickel foil in the form of a bifilar spiral covered on both sides with an insulating layer of Kapton, Fig. 1(a). The thicknesses of the foil and the Kapton layer are 10 and 25 μm, respectively, the effective diameter of the bifilar spiral is 13 mm and the diameter of the Kapton layer is 20 mm. The dimensions of this sensor are chosen to be smaller than the dimensions of the previously used sensors [6,7] so that it has higher resistance. A higher-resistance sensor is preferable to compensate the decrease in the absolute value of the resistance as the temperature is lowered and as a result the error in measuring $\Delta R(T)/R(T)$ is minimized. Details about the specific characteristics and the design requirements of these sensors are given by Gustafsson [7].

In the experimental arrangement, see Fig. 1(b) and (c), the sensor is pressed between the sample halves, which usually are two identical cylinder-shaped pieces, each having a diameter of 18 mm and a thickness of 8 mm. It should be noted that the size of the sample halves must satisfy the condition of an infinite medium, that is, the probing depth, which indicates how far the heating pulse has propagated into the sample during the transient time, is less than the distance from the heater to the nearest boundary of the sample.

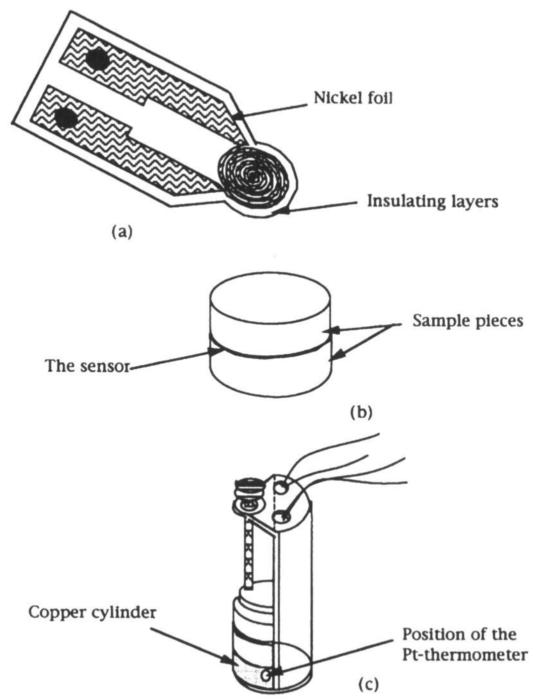

Fig. 1. The TPS element (a), the shape of the sample (b), and the sample arrangement in the sample holder (c).

2.2. Theoretical principle

The theory of the method considers a three-dimensional heat flow inside the sample, which can be regarded as an infinite medium, provided that the measurement time of the thermal transient is ended before the thermal wave reaches the boundaries of the sample. The experiment is performed by recording the voltage variations over the TPS element while its temperature is slightly raised (≈ 1 K) by a pulsed electrical current (≈ 300 mA). The time-dependent resistance of the TPS element, during the transient recording, can be expressed in a first approximation, as

$$R(t) = R_0[1 + \alpha \Delta T(t)] \quad (1)$$

where R_0 ($\approx 5\ \Omega$ at room temperature) is the resistance of the TPS element before the transient recording has been initiated, α is the temperature coefficient of resistance (TCR) (for the TPS element $\alpha \approx 5.0 \times 10^{-3}\ \mathrm{K}^{-1}$ at room temperature) and $\Delta T(t)$ is the temperature increase of the TPS element. A typical plot of temperature (voltage) versus time is shown in Fig. 2. Depending on the temperature range of interest, the TCR values for the TPS element are determined within that particular temperature range from separate cali-

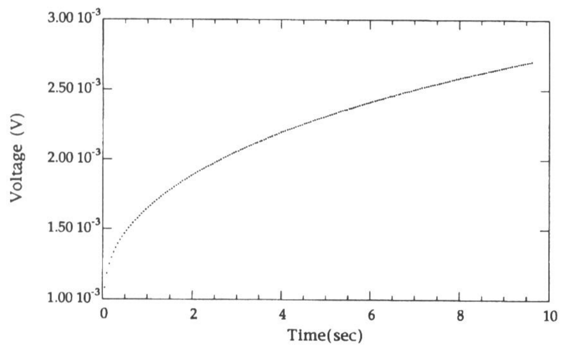

Fig. 2. A typical temperature (voltage or resistance) recording vs. time.

bration procedures by means of different thermometers such as a Pt thermometer.

The evaluation of $\Delta T(t)$ in the heater depends on the power output in the TPS element, the design parameters of the sensor and the thermal transport properties of the surrounding sample. For the disk-shaped sensor used in this work, $\Delta T(t)$ is given by the following equation [Appendix I in Ref. [5]], from which the thermal conductivity and diffusivity can be obtained:

$$\Delta T(\tau) = P_o(\pi^{3/2}a\lambda)^{-1}D(\tau) \qquad (2)$$

where

$$D(\tau) = [m(m+1)]^{-2}\int_0^{\tau} \mathrm{d}\sigma\sigma^{-2}\left[\sum_{l=1}^{m} l \sum_{k=1}^{m} k \times \exp\left[\frac{-(1^2+k^2)}{4m^2\sigma^2}\right] I_0\left(\frac{lk}{2m^2\sigma^2}\right)\right] \qquad (3)$$

P_o is the total output power, λ is the thermal conductivity of the sample, and a is the radius of the sensor.

$D(\tau)$ is the theoretical expression of the time-dependent temperature increase [7], which describes the conducting pattern of the disk-shaped sensor, assuming that the disk consists of a number m of concentric ring sources, see Fig. 1(a). For convenience the mean temperature change of the sensor is defined in terms of the non-dimensional variable τ, where $\tau = (\kappa t/a^2)^{1/2}$ or $\tau = (t/\theta)^{1/2}$, where t is the time measured from the start of the transient heating, $\theta = a^2/\kappa$ is the characteristic time and κ is the thermal diffusivity of the sample.

To match theory with experiment, the design parameters of the experiment should be chosen, and the data-reduction procedures should be carried out in such a way that the deviations from the ideal conditions can be justified. In other words, there are possible deviations due to experimental factors, and there are possible deviations due to improper data handling. Most of these factors including precision/accuracy and reproducibility of the measured λ, have been discussed fully elsewhere [5]. However, this is the first time the TPS method has been used down to 35 K and we have met difficulty around 60 K due to a nearly temperature-independent TCR of the sensor. In this temperature range, the difficulty is linked to a design factor that is related to the resistance behaviour of the material used in the construction of the sensor.

3. Results and discussion

Concerning the temperature dependence of resistivity for metals in general, it is easy to have a good fit to a theoretical equation for the measured resistivity as a function of temperature. In fact, for our case (nickel) we have obtained a good fit of the resistivity for the temperature range in which we are interested (35–300 K). In the calibration procedures of this sensor, both a Pt thermometer and copper–constantan thermocouple were used to monitor the temperature. The TCR values are obtained by evaluating the derivative of the resistivity equation as a function of temperature at the particular temperature of interest.

To obtain a good representation for the measured resistance of the TPS element in the whole temperature range of interest (35–300 K), the measured data were fitted to the following fourth-order polynomial:

$$R(T) = A - BT + CT^2 - DT^3 + ET^4 \qquad (4)$$

where the values of the constants are $A = 0.666$, $B = 5.119\times10^{-3}$, $C = 1.598\times10^{-4}$, $D = 5.057\times10^{-7}$ and $E = 6.750\times10^{-10}$. The experimental data and the fitting polynomial are shown in Fig. 3. The reliability of the fitting has been judged by the obtained value of the regression coefficient, $r = 0.9999$.

The best TCR value estimation can be obtained from

$$\alpha(T) = \frac{1}{R(T)}\frac{\mathrm{d}R(T)}{\mathrm{d}T} = \frac{-B+2CT-3DT^2+4ET^3}{A-BT+CT^2-DT^3+ET^4} \qquad (5)$$

Table 1 shows typical values of $R(T)$ and $\alpha(T)$ as obtained by using Eqs. (4) and (5). Looking at the values of α (see Table 1 and Fig. 4 (the dashed line)), we observe a nearly temperature-independent resistivity coefficient for nickel in the temperature range around 60 K. This implies the following:

(1) The variations of $R(T)$ in this temperature range are small, particularly if we are referring to the slighter variation in $R(T)$ that could be induced by the small temperature

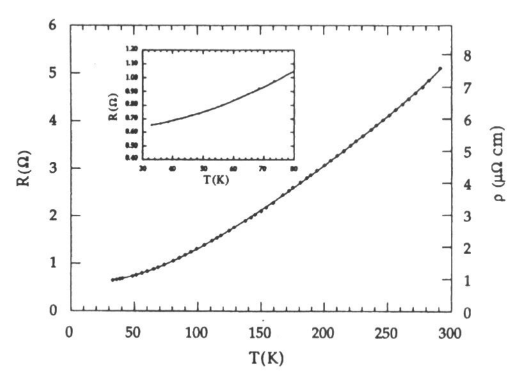

Fig. 3. The resistance of the TPS element as a function of temperature fitted to Eq. (4). The inset shows the behaviour of $R(T)$ around 60 K.

Table 1
Typical values of resistance and TCR for the TPS element

T [K]	R [Ω]	α [K^{-1}]
50	0.751	0.009924
55	0.790	0.010582
60	0.834	0.011054
65	0.882	0.011367
70	0.934	0.011545
75	0.995	0.011614
80	1.049	0.011593
90	1.176	0.011357
100	1.315	0.010952
125	1.701	0.009656
150	2.129	0.008365
175	2.588	0.007278
200	3.069	0.006428
225	3.575	0.005807
250	4.110	0.005394
275	4.687	0.005166
300	5.313	0.005098

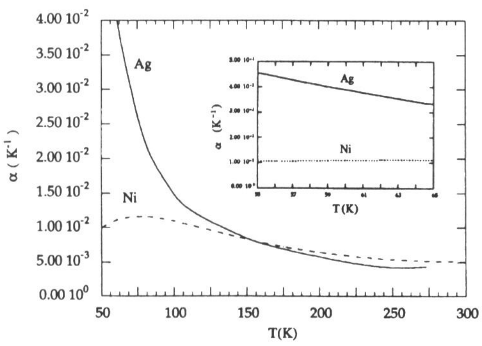

Fig. 4. The variations of the temperature coefficient of resistivity (α) around 60 K for Ni (our data) and Ag (from Ref. [13]).

increase, of the order of one degree Kelvin, which is the expected temperature rise of the sensor during the delivery of the constant current pulse. This will lead to a smaller sensitivity and to large errors from 5 to 8% in the values of λ around these temperatures, see for example, values of λ for samples $YBa_2Cu_{3-x}Co_xO_{7-\delta}$ ($x=0.0$, 0.1) in Fig. 5 from Ref. [8].

(2) The relative error in the value of $\Delta R(T)/R(T)$ becomes larger as the temperature is lowered.

(3) The resistivity values calculated from $R(T)$ and the dimensions of the sensor [7] depicted in Fig. 3 are comparable to the resistivity values given for nickel wires [9–12] in this temperature range. In other words, the size effects in resistivity are negligible since the mean free path of electrons for nickel films [9] at 77 K is ≈700 Å, which is much smaller than the thickness of the foil used in this work (10 μm). Therefore, nickel foils and possibly nickel in general have resistance variations that do not satisfy the required sensitivity in this temperature range.

(4) It is obvious from the behaviour of the TCR of silver [13] also shown (solid line) in Fig. 4 that a sensor made out of silver will certainly improve the accuracy of the measurements since its α values have higher slope and magnitude than the corresponding values of nickel.

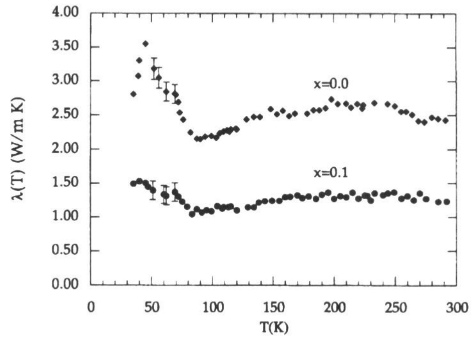

Fig. 5. The thermal conductivity data of $YBa_2Cu_3O_{7-\delta}$ (♦) and $YBa_2Cu_{3-x}Co_xO_{7-\delta}$ (●) against temperature. The error bars around 60 K for both samples are due to a nearly temperature-independent resistivity coefficient for the nickel sensor in this temperature range (taken from Ref. [8]).

4. Conclusions

The development of the present Ni foil sensors fulfilled some of the design criteria that are required for the measurements of thermal transport properties of various samples under altered experimental conditions. Some of these requirements are a protection against mechanical damage, larger resistance, which implies lower errors in $\Delta R(T)/R(T)$, efficient electrical contacts, and practical durability for using the same sensor/element for several times. Furthermore, fabrication of these sensors from materials other than nickel will extend the temperature range under which this method could be used. It turns out from our analysis that the sensor behaviour versus temperature follows a fourth-order polynomial with a sensitivity (TCR) value within the range $(0.5\text{–}1.1)\times10^{-2}$ in the temperature interval 300–60 K. Furthermore, it seems that silver sensors appear as a good alternative to extend the measurement field to lower temperatures, providing that the dimensions of the silver sensors are chosen so as to obtain an optimum ratio of $\Delta R(T)/R(T)$.

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References

[1] H.S. Carslow and J.C. Jaeger, *Conduction of Heat in Solids*, Oxford University Press, London, 2nd edn., 1959.
[2] B. Stålhane and S. Pyk, Ny metod för bestämning av värmelednings koefficienter, *Tekn. Tidskr. 61* (28) (1931) 389–393.
[3] S.E. Gustafsson, A non-steady-state method of measuring the thermal conductivity of transparent liquids, *Z. Naturf.*, *22a* (1967) 1005–1010.

[4] S.E. Gustafsson, E. Karawacki and M.N. Khan, Transient hot-strip method for simultaneously measuring thermal conductivity and thermal diffusivity of solids and fluids, *J. Phys. D: Appl. Phys.*, *12* (1979) 1411–1421.

[5] B.M. Suleiman, The transient plane source technique for measurement of thermal properties of polycrystalline ceramics including high-T_c superconductors, *Ph.D. Thesis*, Physics Department, Chalmers University of Technology, Gothenburg, Sweden, 1994.

[6] S.E. Gustafsson, *Int. Patent Appl. No. PCT/SE89/00137.*

[7] S.E. Gustafsson, Transient plane source technique for thermal conductivity and thermal diffusivity measurements of solid materials, *Rev. Sci. Instrum.*, *62* (1991) 797–804.

[8] B.M. Suleiman, L. Börjesson and P. Berastegui, The effect of cobalt doping on thermal conductivity of $YBa_2Cu_3O_{7-\delta}$ superconductor, *Phys. Rev. B, 53* (1996) 5901–5906.

[9] K.-H. Hellwege and J.L. Olsen (eds.), *Metals: Electronic Transport Phenomena, Landolt–Börnstein, Numerical Data and Functional Relationships in Science and Technology*, Springer, Berlin, Vol. 15, Subvolume a, 1982, pp. 139–150.

[10] D. Grieg and J.P. Harrison, The low temperature electrical transport properties of nickel and dilute nickel–copper alloys, *Philos. Mag.*, *12* (1965) 71–79.

[11] S.J. Tzeng, Y.D. Yao and T.H. Chuang, Electrical resistivity of nickel-rich nickel–indium alloys between 10 and 800 K, *Phys. Status Solidi A*, *117* (1990) K47.

[12] M.J. Aus, B. Szpunar, U. Erb, A.M. El-Sherik, G. Palumbo and K.T. Aust, Electrical resistivity of bulk nanocrystalline nickel, *J. Appl. Phys.*, *75* (1994) 3632–3634.

[13] R.A. Matula, Electrical resistivity of copper, gold palladium and silver, *Phys. Chem. Ref. Data*, *8* (1979) 1147–1296.

Biographies

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Silas E. Gustafsson, born in 1935, received a Ph.D. in physics from Gothenburg University in 1969 and since then he has been the leader of the thermal transport properties group at the Physics Department, where he is an associate professor. From 1969 until 1972 he worked as the director of studies at the Physics Department, then from 1972 until 1993 he was the head of the council of the Science Faculty in Gothenburg University. Since 1994 he has been the academic secretary and the head of the rector's secretarial office at Gothenburg University.

Lars Börjesson, born in 1957, received a Ph.D. degree from the Department of Physics, Chalmers University of Technology in 1987. He became professor and chair in solid-state physics at the Royal Institute of Technology, Stockholm, Sweden in 1993, and in condensed matter physics at Chalmers University in 1995.