The Rigaku Journal
Vol. 4/ No. 1/ 2 / 1987

CONTRIBUTED PAPERS

TRANSIENT HOT STRIP TECHNIQUES FOR MEASURING THERMAL CONDUCTIVITY AND THERMAL DIFFUSIVITY

SILAS E. GUSTAFSSON

Department of Physics, Chalmers University of Technology, S 412 96 Gothenburg, Sweden

ABSTRACT: A review is given of the recent developments in the use of transient hot strips for measuring thermal transport properties of materials. Depending on the particular material and the desired probing depth in a particular measurement the length of the current pulses of the transient recordings has been vaired from 10 ns to 100 s.

In order to extract information on both thermal conductivity and thermal diffusivity from the recordings it is necessary to vary the strip sizes from 0.01 * 20 * 500 cu mm^3 to 0.00001 * 0.005 * 0.5 cu mm^3.

The microstrips are made by vapour deposition on the surface of insulating materials and used for the so called PTHS (Pulse Transient Hot Strip) technique, primarily designed for studies of deposited layers of thicknesses down to half a micrometer.

1. Introduction

Information on thermal transport properties has become increasingly important in a wide range of engineering fields. It is also important in understanding the very basic properties of materials at different temperatures and pressures.

Thermal conductivities vary extensively depending on the structure, density, porosity etc. of different materials. In some cases also very large temperature dependencies have been recorded. Extreme situations that can be mentioned are the thermal conductivity of loose powders with low interstitial gas pressure which typically is of the order of 0.01 W/mK (Saxena *et al.*, 1986) while the thermal conductivity of dense single crystals at temperatures around 10 K may be as high as 10000 W/mK or higher.

Because of the large variation of the thermal conductivities (and also the thermal diffusivities) a number of different experimental techniques have been used for different materials in different experimental conditions. The introduction of the transient hot strip (THS) techniques was made with the specific intention of covering as large ranges of thermal conductivities as possible. This has been achieved by the use of strip sizes which range from vapour deposited microstrips to cm-wide strips made of metal foils as well as widely varying recording or characteristic times of the experiments.

Maximum applicability as well as convenience is achieved with this experimental method by the use of the hot strip both as a heater and as a temperature sensor. By recording the voltage increase over the strip while it is exposed to one or several current pulses it is possible to get information on the resistance increase of the hot strip, which is made of a material with as high TCR (temperature coefficient of the resistivity) as possible.

In typical situations the TCR is of the order of 0.001 (1/K), which means that a mean temperature increase of about one degree in the hot strip is sufficient for precise recordings of both the thermal conductivity and the thermal diffusivity.

So far the THS- (or single shot) method has been applied to studies of a large number of different materials with thermal conductivities ranging from 0.01 to 100 W/mK. The PTHS-method on the other hand has made it possible to study thermal transport properties of materials that have not been accessible to any thermal conductivity measurements before. By the introduction of vapour deposited microstrips with the approximate dimensions: length 0.5 mm, width 0.005 mm and thickness 20nm and by limiting the duration of the current pulses through the sensor to a submicrosecond range it is possible to study the thermal properties of dielectric coatings with thicknesses down to 0.5 micrometers if the thermal diffusivities are in the range of 1 mm sq/s.

With the PTHS-technique there are also obvious possibilities of measuring thermal conductivities which are orders of magnitude higher than what conveniently can be achieved with the single shot THS-method. This makes the PTHS-techniques a very interesting candidate for thermal conductivity measurements of pure single crystals at cryogenic temperatures.

2. Theory

2a. Single current pulse

The theory needed to describe the temperature increase in a hot strip during a transient recording is dependent on both the electrical circuit and how the recordings are most conveniently made.

The first situation to be discussed here is when the strip is exposed to a single current pulse (single shot) and during this pulse a number of recordings of the strip resistance or the mean temperature increase of the strip are being made.

The temperature increase in an infinite solid due to a quantity of heat, *Qpc*, instantaneously generated at the time $t = t'$ at a point (x', y', z'), is, for an anisotropic material with principal conductivities Λ_1, Λ_2, and Λ_3 in the directions of the x-, y-, and z-axis, respectively, given by

$$\frac{Q}{8\left(\pi^2\kappa_1\kappa_2\kappa_3\right)^{1/2}(t-t')^{3/2}} \quad (3)$$
$$-\exp\left\{-\left[\frac{(x-x')^2}{4\kappa_1(t-t')}+\frac{(y-y')^2}{4\kappa_2(t-t')}+\frac{(z-z')^2}{4\kappa_3(t-t')}\right]\right\}$$

where $\kappa_i = \Lambda_i/pc$ and pc is the specific heat capacity per unit volume (Carslaw and Jaeger, 1959).

Assuming that the point sources are distributed over an area in the form of an infinite strip of the *yz* plane and that the power supplied to the strip is practically constant over the entire area and varies very little as a function of time, we can write an expression of the temperature distribution in the strip accordingly

$$T(y,t) - T_0 = \frac{P_0}{16\pi d h\left(\Lambda_1\Lambda_2\right)^{1/2}} \quad (2)$$
$$\cdot\int_0^t dt'\left(t-t'\right)^{-1}\int_{-d}^{d} dy' \exp\left[-\frac{\left(y-y'\right)^2}{4\kappa_2\left(t-t'\right)}\right]$$

where 2d is the width of the strip, P_0 is the total output of power in the strip and its actual length is 2h.

The resistance variation (R) of the strip due to the temperature increase given by eq. (2) can be expressed as

$$R_0/R = \left(2d\right)^{-1}\int_{-d}^{d} dy\left\{1+\alpha\left[T\left(y,t\right)-T_0\right]\right\}^{-1} \quad (3)$$

where α is the temperature coefficient of the resistivity (TCR) of the strip and R_0 is its resistivity at the temperature T_0 before any heating has taken place.

Neglecting second and higher orders of α and remembering that the temperature increase of the strip is kept around one Kelvin equations (2) and (3) can be combined to give

$$R/R_0 = 1 + \frac{P_0\alpha}{4h\left(\pi\Lambda_1\Lambda_2\right)^{1/2}} f\left(\tau_2\right) \quad (4)$$

where $\tau_2 = (t/\theta_2)^{1/2}$, with the characteristic time $\theta_2 = d^2/\kappa_2$ and

$$f\left(\tau\right) = \tau\, erf\,\tau^{-1} - \left(2\pi\right)^{-1/2}\tau^2\left[1-\exp\left(-\tau^{-2}\right)\right] \quad (5)$$
$$-\left(2\pi\right)^{-1/2} Ei\left(-\tau^{-2}\right)$$

The following definitions of the integral functions apply:

$$\mathrm{erf}\,u = 2\pi^{-1/2}\int_0^u \exp\left(-v^2\right)dv \quad (6)$$

$$-Ei\left(-u\right) = \int_u^\infty v^{-1}\exp\left(-v\right)dv \quad (7)$$

The derivation of eq. (4) is made with the assumption that the strip is located inside an infinite medium. When the strip is evapourated on or in some other way is attached to the surface of a semi-infinite substrate and the heat loss of the surroundings, other than that to the substrate, can be neglected, the theory shows that P_0 in eq. (4) should be replaced by $2P_0$.

In a single shot experiment recordings of the voltage or resistance increase are made at a number of times measured from the start at the current pulse.

The procedure use for obtaining the dual information on thermal conductivity and thermal diffusivity from one such transient event, was to make a computational plot of R vs. $f(\tau_2)$, using an arbitrary θ_2-value. Since this plot should be a straight line according to eq. (4), the maximization of the correlation coefficient has been used in the iteration procedure to find the best value of θ_2 or

$$\kappa_2 = d^2/\theta_2 \quad (8)$$

The intercept of the final R vs. $f(\tau_2)$ plot gives R_0 and the slope of the line is proportional to $(\Lambda_1\Lambda_2)^{1/2}$ with the constant of proportionality known.

By making three independent measurements with the orientation of the strip along the principal directions of the crystal we get the following information (see Fig. 1)

$\Lambda_1\Lambda_2 = a_A \qquad \kappa_1 = b_A \qquad (9a)$

$\Lambda_2\Lambda_3 = a_B \qquad \kappa_2 = b_B \qquad (9b)$

$\Lambda_3\Lambda_1 = a_C \qquad \kappa_3 = b_C \qquad (9c)$

Using the general relation

$\Lambda i = \kappa_i pc \quad$ with $\quad i = 1, 2, 3$

we get

$\Lambda_1 = (a_A a_C/a_B)^{1/2} \qquad (10a)$

$\Lambda_2 = (a_B a_A/a_C)^{1/2} \qquad (10b)$

$\Lambda_3 = (a_C a_B/a_A)^{1/2} \qquad (10c)$

$pc = (a_A a_C/a_B)^{1/2}/b_A$

$\quad = (a_B a_A/a_C)^{1/2}/b_B$

$\quad = (a_C a_B/a_A)^{1/2}/b_C \qquad (11)$

Eq. (11) is giving three different expressions of pc, which can serve as an excellent check of the internal consistency of the measurements.

In case of isotropic materials

$\Lambda_1 = \Lambda_2 = \Lambda_3 = \Lambda \qquad (12)$

and the equations are simplified accordingly. This means that it is possible to extract information on both the thermal conductivity and the thermal diffusivity of an isotropic material from one single transient recording.

The possibility to extract this dual information is actually based on the choice of a recording time which is of the same order of magnitude as the characteristic time. This can be seen if we look a little more closely at the $f(\tau)$-function, which in principle is valid for all τ-values.

For very small τ-values (infinite plane source) we see that

$f(\tau) = \tau \qquad (13)$

and for very large T-values (hot wire solution) we get

$f(\tau) = (4\pi)^{-1/2}(3 - \gamma + 2 \ln \tau) \qquad (13a)$

where $\gamma = 0.5772$ ...

Using eq. (13) the resistivity as expressed with eq. (4) will vary as $t^{1/2}$ and eq. (13a) will result in an increase proportional ln t. In none of these limiting cases it is possible to obtain both the transport coefficients. To be able to do that one must work in the "strip source" range (Gustafsson et al., 198 1) with observation times of the same order of magnitude as the characteristic time

$\theta = d^2/\kappa$

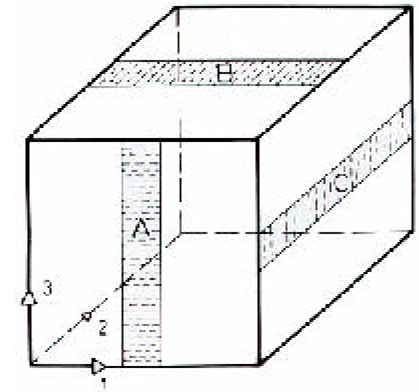

Fig. 1. Orientations of the hot strip in relation to the principle directions of a single crystal.

An approximation of considerable practical importance is

$f(\tau) = \tau - (4\pi)^{-1/2}t^2 \qquad (14)$

which is valid with good accuracy for τ-values less than 0.7.

Using this approximation it is possible to write down the resistance variation as a function of time as

$R = a_1 + a_2 t^{1/2} + a_3 t \qquad (14a)$

A second order fit of R vs. $t^{1/2}$ will give the three coefficients a_1, a_2 and a_3 by which the thermal transport coefficients can be calculated accordingly (Gustafsson et al., 1982)

$\Lambda = \alpha I_0^2(-a_1^2 a3/a_2^2)/2h \qquad (14b)$

$\kappa = 4\pi d^2(a_3/a_2)^2 \qquad (14c)$

2b. Train of current pulses

In experiments with the THS method it is possible to design experiments with a wide range of characteristic times (θ_2) simply by varying the strip width (2d). It is obvious that as the observation times of the experiment are restricted to less than say $k\theta_2$, where k is of the order of unity, only a part of the specimen will be heated during the transient event and only the properties of this part of the specimen will determine the value of the measured thermal conductivity and thermal diffusivity.

In order to estimate the thickness (Δ) of the heated region along the x-axis (this thickness will be referred to as the probing depth) we assume that by placing a perfect thermal insulator or perfect conductor at a distance Δ from the strip and parallel with the plane of the strip the temperature of the strip will only be influenced to a degree, which is well below the expected experimental precision (Gustafsson, 1982).

Based on these assumptions one obtains the following expression of the probing depth

$$\Delta = 1.4(\kappa_1 k \theta_2)^{1/2} \qquad (15)$$

where it is assumed that experimental recordings of the resistance variation have been made for times up to but not beyond $k\theta_2$. Eq. (15) can be rewritten to read

$$\Delta = 1.4d(k\kappa_1/k_2)^{1/2} \qquad (16)$$

From this equation we see that, if k=2 and the diffusivities in the directions perpendicular and parallel with the surface of the strip are not too different, the probing depth will be almost exactly the same as the strip width.

The concept of a probing depth in transient thermal conductivity measurements is very important since it opens up the possibilities that by varying recording times and the strip widths experiments can be designed to probe materials at different depths.

Based on this concept a transient method for measuring thermal conductivity and thermal diffusivity of solid surface layers with low electrical conductivity has recently been described. The technique is based on the THS-method and by reducing the strip width to between 5 and 10 micrometers it is possible to limit the probing depth so that direct measurements of the thermal transport properties of vacuum deposited insulating layers can be made. These kind of studies are presently not possible to perform with any other experimental technique.

To perform experiments with these microstrips in a single shot mode would require that the current pulse would be limited to the microsecond or submicro-second ranges.

The actual mapping of the voltage-versus-time trace over this time period, while possible, would require quite expensive electronic equipment. To reduce this problem of time resolution we have used pulse non-linearity measurements, an elegant technique developed and used for studies of thermal time constants of non-linear resistors (Hebard *et al.*, 1978). The technique is based on a procedure by which a string of pulses is applied to an AC-coupled circuit containing the hot strip actually acting as a non-linear resistance.

If a circuit is constructed which contains a large blocking capacitor in series with the hot strip, its RC time constant will be long compared to the pulse width and the strip will be exposed to a train of square pulses.

Because of the AC coupling there will appear a base-line shift of the instantaneous voltage over the strip. This is a consequence of the fact that the current is changing direction twice during one period. By working with duty cycles in the range from 0.02 to 0.05 the reverse current during the time between the positive pulses can be made very small with the essential heating of the strip during the positive excursions of the current.

Because of the non-linear part of the strip resistance, an average voltage, rocorded over a number of periods across the strip, will develop and theory shows that it is inversely proportional to the thermal conductivity of the substrate on top of which the thin film resistor has been deposited.

The theoretical description of the heat dissipation from the strip also shows that by making measure-

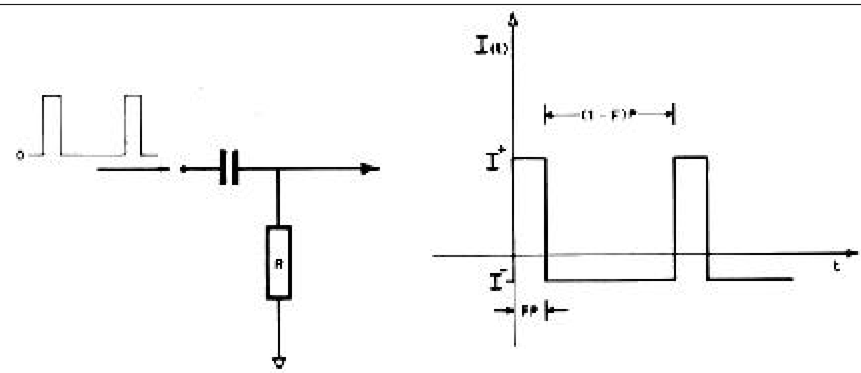

Fig. 2 a) The electrical circuit with a large blocking capacitor and the hot strip as the non-linear resistor R
b) Base-line shift introduced by the blocking capacitor

ments at more than one pulse frequency it is possible to derive information on the thermal diffusivity of the substrate.

If an AC coupled network consisting of linear components is exposed to a train of square pulses, the time average of the output voltage over a number of whole periods will be zero. However, if we include a non-linear resistance in the circuit, the zero net charge condition which prevails will give rise to a similar baseline shift, but the average output voltage will not be zero. The average voltage will as a matter of fact adjust itself to a value which reflects very directly the nonlinear part of the resistance.

The circuit used is shown in Fig. 2. The generator imposes the pulse train on the THS probe through the large blocking capacitor. If the rectangular current pulses have a magnitude I and a duty cycle F, we have

$$I = I^+ + I^- \tag{17}$$

where I^+ and I^- are the positive and negative excursions of the current, respectively.

From the zero net charge condition it is required that

$$FI^+ = (1 - F)I^- \tag{18}$$

As we will see below the most interesting arrangement is to use a small duty cycle, which means that the probe (hot strip) will be exposed to a strong current pulse during the first part of the period while the reverse current during the second part of the period will be rather small. The fact that there is a reverse current during the second part of the period will, however, mean that the temperature of the strip will not relax back to its initial temperature but will after each full period have experienced a small but finite temperature increase.

Considering a series of pulses from the generator, the average voltage, $\overline{U}_n$, over *n* periods may be expressed as

$$\overline{U}_n = (nP)^{-1}\int_0^{nP} u(t)dt \tag{19}$$

where P is the pulse period and u(t) is the instantaneous voltage over the THS probe. Because of the complexity of $\overline{U}_n$, we may write

$$\overline{U}_n = n^{-1}\sum_{k=1}^{n}\overline{U}_k \tag{20}$$

where $\overline{U}_k$ is the average voltage over the period number *k*, or more explicitly

$$\overline{U}_k = P^{-1}\left\{ I^+\int_{(k-1)P}^{(k-1+F)P} R_k^+(t)dt - I^-\int_{(k-1+F)P}^{kP} R_k^-(t)dt \right\} \tag{21}$$

where $R_k^+(t)$ and $R_k^-(t)$ are the time-dependent resistances of the strip during the positive and negative excursions of the current, respectively. These resistances are directly dependent on the two different heat dissipations, Q^+ and Q^-, which with good accuracy can be given by

$$Q^+ = R_0(I^+)^2 \tag{22}$$

and

$$Q^- = R_0(I^-)^2 \tag{23}$$

From eq. (18) we have

$$Q^-/Q^+ = r^2 \tag{24}$$

where R_0 is the initial resistance of the hot strip and

$$r = F/(1 - F) \tag{25}$$

If we assume that a long, resistive strip of negligible heat capacity on the surface of a solid substrate is electrically heated so that the output of power, Q, is constant, the time-dependent resistance, R(t), can be given (Gustafsson et al., 1984) as

$$R(t) = R_0\left\{1 + \frac{\alpha Q}{16\pi d^2 h(\Lambda_1\Lambda_2)^{1/2}}\int_{t1}^{t2}(t-t')dt' \cdot \int_{-d}^{d}dy\int_{-d}^{d}dy'\exp\left[-\frac{(y-y')^2}{4\kappa_2(t-t')}\right]\right\} \tag{26}$$

This equation is general in the sense that it is assumed that the output of power in the strip only lasts from the time t_1 to t_2.

Starting with these equations it is possible although somewhat tedious to derive a recussion formula for $\overline{U}_k$ accordingly

$$\overline{U}_k = C\sigma^{-1}\Big\langle (1+r)(1+r^2)\Big\{\sum_{i=0}^{k-2}\left[J_{(i+F)\sigma} + J_{(i+1-F)\sigma}\right] - 2\sum_{i=0}^{k-3}J_{(i+1)\sigma}\Big\} - \left[1+(1+2r)(1-r^2)\right]J_{(k-1)\sigma} + (1+r)J_{(k-1+F)\sigma} + r(1-r^2)J_{(k-F)\sigma} - rJ_{k\sigma}\Big\rangle \tag{27}$$

where k = 1, 2, ..., n. It should be noted that whenever the upper limit of the summation in this equation is less than zero the sum should be con-sidered identically zero.

Further more

$$C = \alpha(1-F)^3 U^3_{in}(24\pi h R_0)^{-1}(\Lambda_1\Lambda_2)^{-1/2} \quad (28)$$

and U_{in} is the total pulse height given by

$$U_{in}=R_0(I^+ + I^-) \text{ and } I^+=U_{in}(1-F)/R_0$$

We have further that

$$\sigma = P/\theta,$$

$$J_x = 8\pi^{1/2}\tau_x^3 \operatorname{erf} \tau_x^{-1}-\tau_x^2 \exp(-\tau_x^{-2})$$
$$- 3\tau_x^4[1 - \exp(-\tau_x^{-2})] -(1 +6\tau_x^2) \operatorname{Ei}(-\tau_x^{-2}) \quad (29)$$

and

$$\tau_x = (x/\theta)^{1/2} \quad (30)$$

These derivations lead to an average voltage over a specified number of periods, which can be expressed as

$$\overline{U}_n = C\Phi(n, F, \sigma) \quad (31)$$

Experimentally it is observed that $\overline{U}_n$ tends to stabilize very quickly and become very little dependent on the number of periods particularly for small F-values. This fact can also be verified from eq. (27), which shows that it is possible to design experiments with very small contributions to the average voltage from all but the very first period.

Because of this fact a convenient way of performing measurements with the PTHS-method is to fix the duty cycle F and then measure the average voltage as a function of the length of the period, P. Making then a computational plot of $\overline{U}_n$ vs. $\Phi(\sigma)$ it is possible to find by iteration a characteristic time θ_2 such that the plot represents a straight line through the origin with a slope equal to C according to eq. (31).

An important aspect of these measurements is that short current pulses are being used and that the probing depth may be orders of magnitude smaller than in all previous measurements.

For a signal shot measurement the probing depth has been defined according to eq. (15). However, in PTHS measurements it seems reasonable to assume that the total time in this equation corresponds to say 2FP. The probing depth of a pulse measurement (Δ_p) would then be

$$\Delta_p = 2(\kappa FP)^{1/2} \quad (32)$$

An interesting experimental observation from pulse measurements is that one can work with FP values which are appreciably smaller than θ_2 and still get thermal diffusivities with good precision. This is very likely due to the AC coupling of the electrical circuit.

3. Mathematical Model and Experimental Arrangements

The mathematical model given above presup-poses that the strip sensor/heat source is ideal in the sense that: a) it has no heat capacity, b) the electrical contacts do not disturb the measurement, c) all energy is transported by diffusion and not by radiation, d) there is no thermal contact resistance between the heater and the substrate and 3) the electrical power delivered to the strip is constant throughout the current pulse(s).

The present section will deal with these devia-tions from the mathematical model, and it will be shown that in some cases it is possible to neglect the effects by a proper design of the experiments and in other cases it is necessary to slightly modify the equations used for evaluating the transport coef-ficients.

3a. Heat capacity of the sensor

A basic assumption in the theory above is that the hot strip does not have any heat capacity. The actual influence from this heat capacity can be estimated by calculating the power (P_s) needed to raise the tem-perature of the strip itself accordingly

$$P_s = 8dhv\rho_s c_s \frac{\partial\left(\overline{\Delta T}\right)}{\partial t} \quad (33)$$

where 2v is the thickness of the strip, ρc_s is the specific heat per unit volume of the strip material and

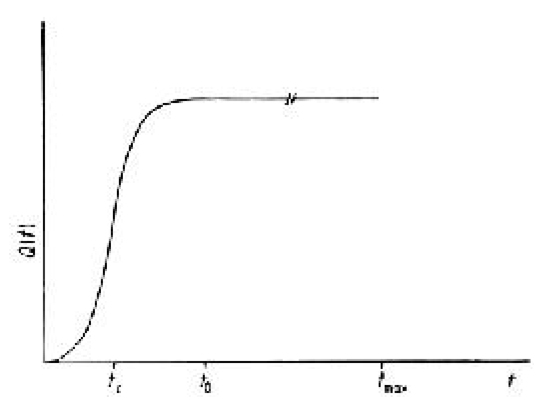

Fig. 3 Assumed time dependence of the power deli-vered to the sample. After a time t_0 the power is assumed to be constant, that is the processes consuming energy at the beginning of the transient have died out and cannot be resolved experimentally.

$$\overline{\Delta T} = \frac{P_0}{4h(\pi \Lambda_1 \Lambda_2)^{1/2}} f(\tau_2) \quad (34)$$

cf. eq. (4).

It is obvious from eq. (33) as well as from a more complete treatment of this effect (Gustafsson, 1985) that the heat capacity of the strip is influencing the recording only at the beginning of the current pulse. The detailed theory also shows that the disturbance increases if the ratio of the heat capacity of the strip to that of the sample material is large.

The situation with a loss of power delivered to the sample can be depicted as in Fig. 3, where the power of Q(t), delivered to the sample is assumed to have reached a constant value, P_0, only after a certain time, to. The word constant is here taken to mean that any variation of the power input to the substrate after the time t_0 cannot be resolved experimentally.

If the length of the current pulse is t_{max}, we can for any time less than t_{max} give the temperature increase, $T(y, t) - T_0$, of the hot strip (Gustafsson, 1967) as

$$\begin{aligned} T(y,t) - T_0 &= (16\pi d h)^{-1} (\Lambda_1 \Lambda_2) \\ &\cdot \int_0^t dt' Q(t') (t - t')^{-1} \\ &\cdot \int_{-d}^{d} dy' \exp\left\{ -(y - y')^2 / [4\kappa_2 (t - t')] \right\} \end{aligned} \quad (35)$$

where a strip shaped heat source with the same dimensions as the hot strip, but without heat capacity, is assumed to be placed on and in perfect thermal contact with the surfaces of two semi-infinite samples.

With these assumptions the time varying resistance of the strip can then be given (Gustafsson *et al.*, 1979) by

$$R = R_0 \left[\begin{aligned} &1 + \alpha (32\pi d^2 h)^{-1} (\Lambda_1 \Lambda_2)^{-1/2} \\ &\cdot \int_0^t dt' Q(t') \Psi(t, t') \end{aligned} \right] \quad (36)$$

where

$$\begin{aligned} \Psi(t, t') &= (t - t')^{-1} \int_{-d}^{d} dy \\ &\cdot \int_{-d}^{d} dy' \exp\left\{ -(y - y')^2 / [4\kappa_2 (t - t')] \right\} \end{aligned} \quad (37)$$

This theoretical approach means that the actual strip with a certain heat capacity and with inevitable difficulties of defining a precise start of the current pulse is being replaced by ideal strips on the sample surfaces but having a variable output of power, which, however, in its entirety is delivered to the sample.

It is then assumed that whatever disturbances at the beginning of the current pulse will die out rather quickly so that Q(t) will become constant, P_0 after the time t_0, we can, using the mean-value theorem for integrals, find a time, t_c, such that

$$\int_0^t dt Q(t') \Psi(t, t') = P_0 \int_{t_c}^{t} dt' \Psi(t, t') \quad (38)$$

and consequently

$$R = R_0[1 + \alpha P_0 (4h)^{-1} (\pi \Lambda_1 \Lambda_2)^{-1/2} f(\tau_c)] \quad (39)$$

where the $f(\tau_c)$-function has been defined earlier and

$$\tau_c = [(t - t_c)/\theta_2]^{1/2} \quad (40)$$

Eq. (38) does not require Q(t) to behave in any particular manner other than that the mean value theorem can be applied. The time correction, t_c, can be both positive and negative depending on the actual time history of Q(t).

When applying this theory it is necessary that the equipment is designed so that the time correction is much smaller than the characteristic time, θ_2, of the transient experiment in order not to reduce appreciably the time available for data collection. it should be remembered that θ_2 can be increased by making the strip wider.

It should be noted that any process, that would cause a short time variation of the power delivered from the strip to the sample at the beginning of the transient event, is covered by this theory.

3b. Thermal radiation

The heat loss per unit area of the strip due to radiation into a "black" enclosure can be given by

$$Qr = \sigma_s(\varepsilon T_1^4 - \alpha_{12} T_2^4) \quad (41)$$

where T_1 is the strip temperature and T_2 is the temperature of the enclosure, σ_s is the Stefan-Boltzman constant and the emissivity $\varepsilon \approx \alpha_{12}$.

Since the temperature increase of the strip in room temperature measurements is typically around one Kelvin, we have

$$Qr = \sigma_s \varepsilon T^3 \overline{\Delta T}) \quad (42)$$

Here $\overline{\Delta T}$ can be taken from eq. (34).

A rather simple analysis of this equation indicates that the power dissipated as radiation is indeed very small and the reason for this fact is that only a rather small temperature increase is necessary for transient recordings of the thermal transport properties.

However, at temperatures approaching 1000 K one might have to consider radiation effects particulary when looking at materials that are transparent to such radiation.

3c. Finite length of the strip

The theory developed so far is based on the assumption of an infinitely long hot strip. To cover a more realistic situation one can solve eq. (1) for a strip of finite length (Chohan, 1987). However, there is also the additional complication with the sometimes heavy electrical contacts that have to be attached to the strip. This problem has been discussed elsewhere (Gustafsson *et al.*, 1979).

The theoretical work as well as experimental results indicate that the length should be about thirty times the width of the strip in order to keep the accuracy of thermal transport coefficients around one percent.

The deviations from the simple mathematical model due to the end effects as well as from thermal radiation tend to increase as the temperature of the strip (heater/sensor) is raised, which means that a disturbance if any would appear at the end of the current pulse.

3d. Thermal contact resistance

When evapourating a thin metal strip on the surface of an insulating material to be studied, all indications are that the thermal contact between the sensor/heater and the material is very good. However, there are situations when a thermal contact resistance cannot be avoided. A case in point is when electrically conducting materials are to be studied and there is a need to electrically insulate the hot strip.

When describing this situation we refer to the theoretical model given under section 3a with an "ideal" strip on and in perfect thermal contact with the metal surface but with a variable output of power at the beginning of the current pulse. The time correction so introduced will take care of the heat capacity of the strip as well as the insulating material and other effects at the beginning of the current pulse which prevent the output of power to develop instantaneously.

However, in addition to the rise time problem we must also consider the fact that the combined heater temperature sensor is thermally displaced from the ideal strip. Let us now assume that the thickness (δ) of the insulating layer is much less than the width (2d) of the hot strip and that after a certain time, t_0, we can assume that all the power produced in the hot strip will be delivered to and released from the insulating layer into the metal. This means that after a certain time we can assume the temperature difference between the two surfaces of the insulating layer to become constant although the mean temperature of the layer will keep increasing throughout the transient event. This theoretical picture is a direct consequence of the assumption that after a certain short interval of time at the beginning of the transient the influence from the heat capacity of the insulating layer can be neglected.

According to Tyrrel (1961), the time it takes to establish a linear temperature distribution across a slab with the boundaries kept at constant but different temperatures is less than δ^2/κ_i, where κ_i is the thermal diffusivity of the insulating material.

This means that provided δ^2/κ_i is less than t_0, we can express the resistance of the hot strip outside the insulator as

$$R = R_0[1 + \alpha\Delta T_i + Cf(\tau_c)] \qquad (43)$$

If $P_0^*=R^*I_0^2$ is the power output of the hot strip after the time t_0 then

$$\Delta Ti=P_0^*\delta(8dh\Lambda_i)^{-1} \qquad (44)$$

Λ_i is the thermal conductivity of the insulating layer. The hot strip is assumed to be placed between two samples of the same material.

Since it would be inconvenient to measure this constant temperature difference, ΔT_i, across the insulating layer, we can rearrange eq. (10) accordingly.

$$R = R^*[1 + Cf(\tau_c)] \qquad (45)$$

where

$$C=\alpha R_0 I_0^2(4h)^{-1}(\pi\Lambda_1\Lambda_2)^{-1/2} \qquad (46)$$

and

$$R^* = R_0[1 +\alpha\Delta R_0 I_0^2(8dh\Lambda_i)^{-1}]. \qquad (47)$$

I_0 is the constant current through the hot strip during the transient event and in deriving eq. (47) we have used the fact that R* is only slightly larger than R_0.

It should be noted that both C and R* are constants during a transient recording for times larger than t_0. This means that if the insulating layer is thin

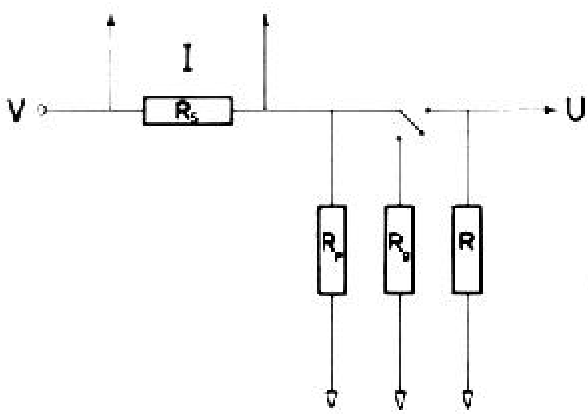

Fig. 4 Circuit to obtain constant power in the non-linear resistor, R, (hot strip). R_B is introduced to achieve stabilization of the driving voltage and the series resistance.

and its thermal conductivity not too small, R* will be very nearly equal to R_0 and eq. (45) can be used directly for evaluating the results. To achieve a higher accuracy it is necessary to make at least two transient recordings at the same initial temperature but with different currents. A plot of R* versus I_0^2 should then, according to eq. (47), give a straight line with an intercept equal to R_0, which then may be used for calculating the thermal conductivity from eqs. (45) and (46).

Note that the determination of the thermal diffusivity is completely independent of the temperature difference across the insulating layer.

It is also important to note that the experimental information is derived from eq. (45) which is in its form identical to the equation for studying insulators. The only difference being that we are not getting the true initial resistance R_0 but rather the apparent initial resistance R*, which is a function of the current, I_0, through the strip.

3e. Current variation and electrical matching

In all the discussions above we have assumed an electrical circuit such that the current after a short initial period is essentially constant. However, this assumption would in some cases lead to very inconvenient voltages and extremely poor matching of the pulse generator to the rest of the electrical circuit in all PTHS measurements.

The circuit that has been most commonly used for transient recordings is depicted in Fig. 4.

If the initial resistance R_0 of the strip is of the same order of magnitude as R_s (assuming for the moment that R_p and R_B in the circuit are infinitely large) the increase of the resistance in the nonlinear component (heating strip) will cause the current to decrease and by that seriously affect the resistance recordings during the transient event.

The traditional way of avoiding this difficulty is to arrange for R_s to be about one hundred times larger than R_0 and connect the circuit to a constant driving voltage (constant current source). In this way one assures that the resistance increase of the nonlinear component is so small compared with the total circuit resistance that the current may be considered constant. However, if the resistance of the strip is around 50 ohms and an output of power in the strip of 0. 5 W is needed, then we would require a series resistance, R_s, of around 5 kohm and a driving voltage in excess of 500 V which is rather inconvenient.

In order to deal with this general problem with a time varying resistance in a circuit, we write eq. (4), eq. (26) or eq. (45) as

$$R=R_0 + \partial R \qquad (48)$$

where ∂R is the time varying part of the strip resistance.

We can similary write

$$I=I_0 + \partial I \qquad (49)$$

for the current through the circuit.

The voltage U measured over the strip can, with these variables, be expressed as

$$U = U_0\left[1+\frac{\partial R}{R_0}+\frac{\partial I}{I_0}+\left(\frac{\partial R}{R_0}\right)\left(\frac{\partial I}{I_0}\right)\right] \qquad (50)$$

and we see that two additional terms appear within the brackets.

Introducing the constant driving voltage V, the general expression of the current becomes

$$I_0 + \partial I= V/(R_s + R_0 + \partial R) \qquad (51)$$

where R_s is assumed to be constant. From this equation one gets the most general expression of the current variation and its relation to the resistance change accordingly

$$\frac{\partial I}{I_0} = -\frac{\partial R}{R_s + R_0} \qquad (52)$$

Introducing this expression in eq. (50) we get

$$U = U_0\left[1+\left(\frac{R_s}{R_s+R_0}\right)\frac{\partial R}{R_0}-\left(\frac{R_s}{R_s+R_0}\right)\left(\frac{\partial R}{R_0}\right)^2\right] \quad (53)$$

With R_0 and R_s of the same order of magnitude and $\partial R/R_0$) typically of the order 0.001, the third term within the brackets can be neglected, which yields

$$U = U_0\left[1+\left(\frac{R_s}{R_s+R_0}\right)\frac{\partial R}{R_0}\right] \quad (54)$$

and the constant in eqs. (4), (26) and (45) in front of the time varying function should all be multiplied by the factor $R_s/(R_s + R_0)$.

Since this factor appears in eq. (26) it should also be included in eq. (31).

It can also be shown (Gustafsson et al., 1984) that by choosing $R_0 = R_s$ the output of power in the strip can be kept extremely constant throughout the transient event beyond $t = t_0$.

In certain situations where one cannot arrange to meet the condition expressed by $R_0 = R_s$, it is necessary to arrange a parallel resistor R_p as indicated in Fig. 4. With such a resistor in place the factor $R_s/(R_s + R_0)$ should be replaced by

$$R_0^{-1}/(R_0^{-1}+R_s^{-1}+R_p^{-1}) \quad (55)$$

To minimize the power variation the condition

$$R_0^{-1}=R_s^{-1}+R_p^{-1} \quad (56)$$

applies.

The facility to work with a time varying current makes it possible to arrange for necessary matching of the circuit to the power supply/pulse generator which is particularly important for PTHS measurements when working with submicrosecond pulses.

4. Experimental

In addition to what has been said under section 3 above there are a few rather essential things that must be remembered when arranging transient measurements of thermal conductivity.

The sample preparation is rather straight forward for liquids and powders, which normally require a container with a suspented metal strip (foil). It is also possible to arrange a hot strip as a probe by double folding a long metal strip and glue it to the outer sides of an extended piece of insulating material (e.g. mica).

When studying solid samples the first step in the sample preparation is to obtain one or two pieces of the material with a flat surface. The question "what is a flat surface" is not possible to specify for all situations since work has been done with large strips (600 x 20 mm^2) as well as microstrips (0.5 x 0.01 mm^2). An obvious requirement is that the surface roughness or the granularity of the material is much less than the strip width (2d).

When evaporating a thin film strip on the surface of a piece of electrically insulating material, the actual measurements have to be performed in vacuum to avoid heat losses to the surrounding gas. For most solid materials the heat loss by thermal conduction through the gas is negligible, however, thermal convection does seriously affect the recordings. Measurements with evaporated strips are normally very easy and convenient to make because it is easy to arrange a convenient resistance (R_0) of the strip. The only important thing to remember is that at least two recordings at different temperatures must be made. The reason being that for thin films it is normally not possible to get a stable ot value (TCR) when cycling the temperature unless the film is very well annealed. However, by arranging at least two measurements and making the first one at a higher temperature than the subsequent ones, very stable values of $\Delta R/\Delta T$ or $R_0\alpha$ can be calculated from the R_0-values obtained from the recordings at the different temperatures.

The evaporated strips are obviously more ideal both as heaters and sensors than any other arrangement, because a higher R_0-value means a lower current pulse and less "strain" on other circuit components (see below). At the same time there is excellent thermal contact between the strip and the sample surface.

Other experimental arrangements for studying solid samples are to a) clamp a strip, made of a thin (0.01 to 0.02 mm) metal foil, between two pieces of the same material or b) press a metal foil strip against a flat surface of the sample with the help of a piece of material with a thermal conductivity much less than that of the test sample. In most of these cases the electrical leads have been attached with some kind of pressure contacts, however soldered contact have also been used. The electrical contacts at the ends of the strip must be made with a certain amount of care in order to avoid any disturbances during the transient recording. At the same time the strip must have a well defined length.

The electrical contacts to thin film strips have been fixed by the use of pressure contacts, with or without indium on the contact pads or some other standard technique developed by the electrical industry.

When designing an electrical circuit for transient recordings, it is very important to make sure that those components (active or passive) which are assumed to behave in a particular way, do not change their properties during the transients (e.g. resistors, electrical leads etc.). One particularly important component is the power/voltage supply, which during a short period of time sometimes is exposed to a heavy load. The constancy required can be expressed by $\Delta V R_0/(R_0 + R_s)$ which must be of the same order of magnitude as the least count of the voltage recording instrument. ΔV in this expression is a possible variation of the output voltage from the power supply. A way to sometimes avoid this problem is to arrange a loading of the voltage source (see figure) prior to the initiation/triggering of the current pulse.

The electrical circuits used so far for THS- and PTHS-measurements have been simple DC circuits and the only addition to Fig. 4 is a stable offset voltage introduced to approximately match the initial voltage, U_0, and by that making the voltage recordings more convenient. If the driving voltage and the constant resistances in the circuit are not known, it is necessary to introduce some kind of arrangement for measuring the initial current, I_0. A simple way to do that is to introduce a small standard resistance in series with the strip and read the voltage across.

The most elegant way of avoiding the comparatively large initial voltage jump is to include the strip in one arm of a simple bridge as indicated in Fig. 5.

When working with this bridge, it would first be balanced by passing a current small and short enough not to heat the strip. This can be achieved by selecting a convenient value of the resistance R_A. After balancing the main switch would be thrown from the preloading of the power supply to the position which initiates the transient recording. This means that the bridge is then actually working off balance and is essentially used only as an offset facility.

When working in this mode it is necessary to analyse how the voltage variation recorded by the digital voltmeter is related to the resistance variation of the hot strip.

This is most readily done in the same way as indicated under section 3*e* above, taking into consideration all the resistances in the circuit. It turns out that the voltage variation, $U - U_0$ can, irrespective of the combinations of resistors, be expressed as

$$U - U_0 = [R_s/(R_s + R_0)]I_0 \partial R \quad (57)$$

where I_0 is the initial current through the strip, R_s is the resistance of one main part of the bridge, R_0 is the initial resistance of the strip and ∂R is defined by eqs. (4) and (48).

When analysing the bridge a little further one finds that the most sensitive arrangement, as far as voltage recordings are concerned, is to select R_s to approximately match the initial resistance of the hot strip.

An additional advantage with the bridge is that it gives a precise value of $R^* - R_0$ when working with electrically conducting materials and applying the theory for weaker thermal contact between the strip and the surface of the substrate.

An important aspect of the implementation of the THS- and the PTHS-methods is the way the experimental recordings are treated and the thermal transport coefficients are calculated. There are obviously many different ways that can be used and we will here only indicate the ways we have found most convenient.

Referring for instance to eq. (4), it is obvious that an iteration procedure in which the characteristic time, θ_2, is varied so that a straight line plot of R- or U-values versus $f(\tau_2)$ is achieved, would automatically give both the thermal conductivity and the thermal diffusivity. The maximization of the correlation coefficient can be used as a measure of the best fit of a straight line.

When applying eq. (45) a similar procedure can be used (provided the time correction, t_c, has been estimated in advance).

t_c can be estimated by the use of eqs. (14) and (40) and experimental points with times larger than t_0 or say 2 t_c but less than $\theta_2/2$. For these points a second order least squares fit can be made of resistance or voltage recordings as a function of $(t-t_c)^{1/2}$ and by iteration determine t_c so that the sum of the squared deviations assumes a minimum.

When evaluating a PTHS-experiment it should be noted that the average voltages being recorded are strictly zero for vanishing pulse periods, which means that a line through the origin with minimization of the sum of the squared deviations will give the best

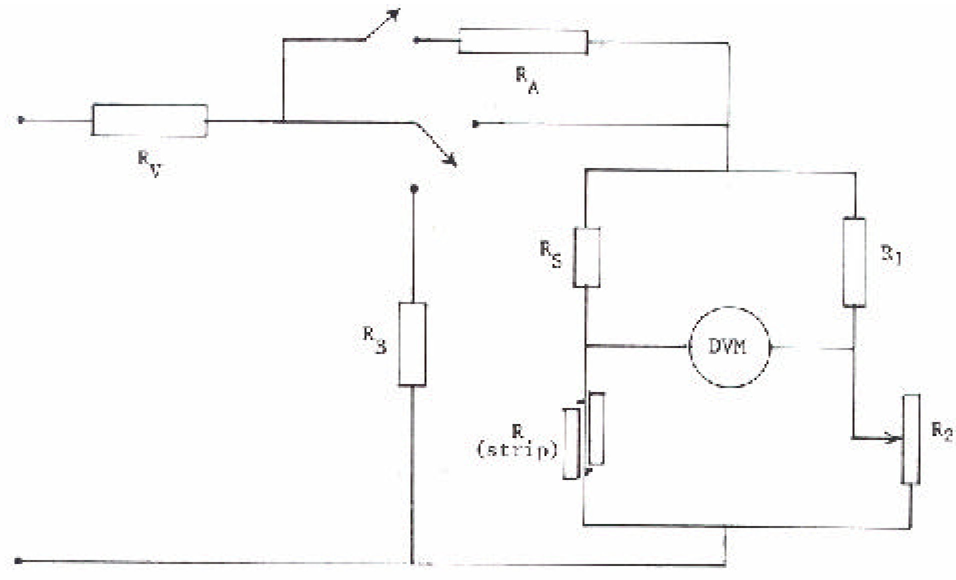

Fig. 5 Bridge circuit to offset the initial voltage over the hot strip.

characteristic time/thermal diffusivity and subsequently the thermal conductivity.

As a guide for designing experiments it may be mentioned that strips have so far been made of the following materials:

Strips made of metal foils:

- Platinum
- Silver
- Nickel
- Aluminium
- Tantalum
- Brass
- Stainless steel

Strips made by vacuum deposition:

- Gold (Fig. 6)
- Silver
- Aluminium

When there is a need to electrically insulate the strip one should of course use a material as thin and with as high thermal conductivity as possible in order to reduce the difference between R* and R_0 and achieve as good a thermal contact between the strip and the surface of the substrate as possible.

Conclusions

The THS-method as described above has been successfully used for measurements on a number of different materials. The follwing can be mentioned:

- Glasses
- Ceramics
- Minerals
- Building materials
- Metals
- Alloys
- Powders
- Liquids
- Wood
- Fabrics

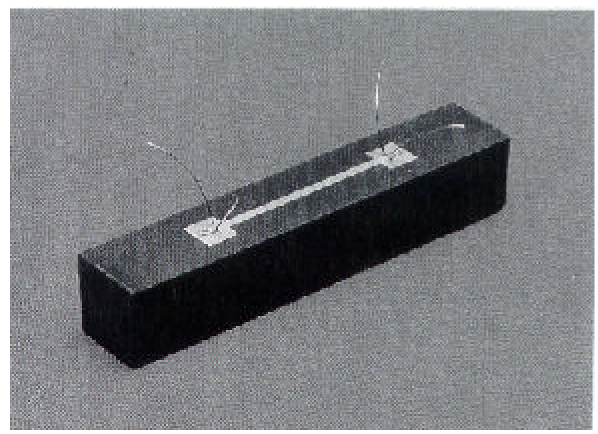

Fig. 6 Evaporated thin film strip on the surface of a piece of insulating material together with attached electrical leads.

The PTHS-method has been used for measurements on:

- Silicon dioxide (fused)
- Silicon dioxide (produced by a PVD technique)
- Gallium Arsenide

- Liquid crystals

The successful use of this particular experimental technology has been made possible by the use of sensors of widely different sizes and experimental recording times that has been varied over nine orders of magnitude.

The PTHS-method in particular has opened up fields of studies of thermal transport properties which have not been accessible before. This is obvious from the fact that the probing depth, in studies of materials with typical thermal diffusivities, has been reduced by more than two orders of magnitude as compared with any other experimental technique and has even made vacuum deposited insulating materials accessible for measurements.

This new method has opened up possibilities of interesting transient measurements of thermal conductivities also at cryogenic temperatures particulary in crystals with a minimum of defects.

As has been elaborated above the restriction of the length of the heating pulses guaranties that only the solid immediately under the probe (hot strip) will determine its temperature increase. The strip only "sees" its immediate surroundings and behaves as if placed on the surface of an infinite solid.

By increasing the probing depth and by that the width of the strip by say two orders of magnitude and still keep the pulse length in the microsecond or submicrosecond range it should be possible at least in principle to study materials with thermal diffusivities approximately four orders of magnitude higher than typical values at room temperature.

The particular interest in these kind of studies, which are in the process of being started, is connected with the possibility of making measurements without any boundary or Casimir scattering of the phonons at appreciably lower temperatures than with other experimental techniques.

In spite of the possibilities outlined here the question still remains whether or not we in the future will be able to measure the "thermal superconductivity" that should exist in infinitely large and perfect crystals. The answer to this question can only be sought in the further development of transient techniques, which in these lower temperature ranges might give us a quite different picture of the thermal conductivity than what we have at present.

References

N. S. Saxena, M. A. Chohan and S. E. Gustafsson (1987), *Jap. J. Appl. Phys.*, **26,** 51.

H. S. Carslaw and J. C. Jaeger (1959), Conduction of Heat in Solids (Oxford).

S. E. Gustafsson E. Karawacki and M. N. Khan (1981), *J. Appl. Phys.*, **52**, 2596.

S. E. Gustafsson (1982), Proc. 8th Symp. Thermophys. Prop. II., ed. Sengers, Y. V. (Book No 100 152), Am. Soc. Mech. Eng., New York.

A. F. Hebard and W. M. Steverson (1978), *J. Appl. Phys.*, **49,** 5250.

S. E. Gustafsson M. A. Chohan, K. Ahmed, and A. Maqsood, (1984), *J. Appl. Phys.*, **55**, 3348.

S. E. Gustafsson, K. Ahmed, A. J. Hamdani, and A. Maqsood (1982), *J. Appl. Phys.*, **53**, 6064.

S. E. Gustafsson (1985), Thermal conductivity 18, Ed. Ashworth, T. and Smith, D. R., Plenum.

S. E. Gustafsson (1967), *Z. Naturforsch*, **22a**, 1005.

S. E. Gustafsson, E. Karawacki, and M. N. Khan (1979), *J. Phys. D: Appl. Phys.,* **12**, 1411.

M. A. Chohan (1987), Thesis.

S. E. Gustafsson, E. Karawacki and M. A. Chohan (1984), *Rev. Sci. Instrum.*, **55**, 610.