Measurements of the Thermal Conductivity

Measurement techniques:

- steady-state heat-flow technique
- comparative technique
- double comparative technique
- parallel thermal conductance technique
- radial flow method
- square wave a.c. drift method
- flash thermal diffusivity method — high temperatures
- 3ω technique — thin films

Systematic errors:

- heat loss via radiation
- thermal conduction through the lead wires

Measurements of the Thermal Conductivity

The Fourier–Biot equation:

$$\frac{\dot{Q}}{A} = -\varkappa \frac{dT}{dx} \,, \tag{1}$$

where $\dot{Q}$ is the rate of heat flow through area A with a temperature gradient, dT/dx.

The constant of proportionality $\varkappa$ — thermal conductivity.

The symbol λ is also often used.

Eq. (1) describes a diffusive heat transport process.

If T varies with time at a rate $\partial T/\partial t$ and if $\varkappa$ is T-independent, one can derive

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} \ . \qquad (2)$$

Here the constant of proportionality α is the thermal diffusivity.

The thermal diffusivity α can be determined from Eq. (2).

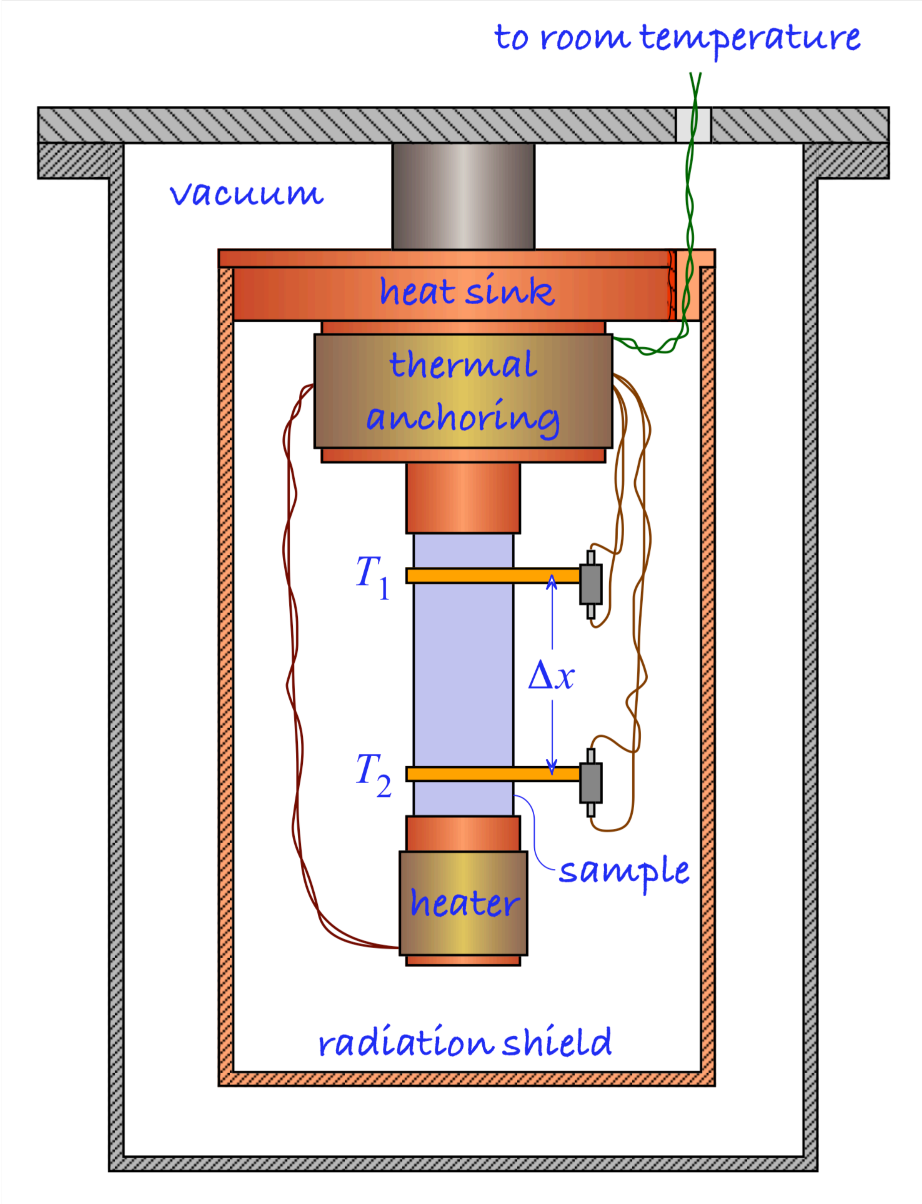
It relates to $\varkappa$ through the relation

$$\alpha = \frac{\varkappa}{\rho C} \ , \qquad (3)$$

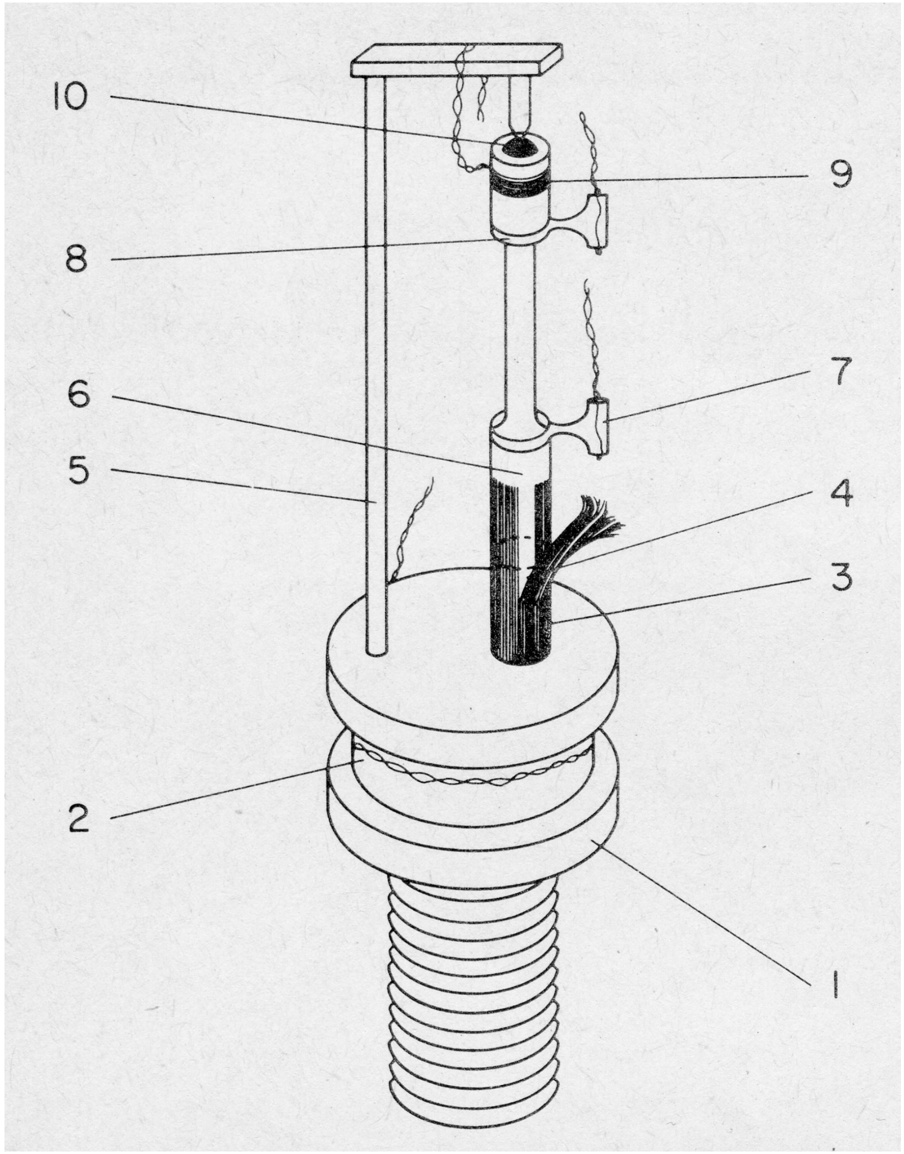
where ρ is mass density and C is the specific heat.

In general, $\varkappa(T) \rightarrow \alpha(T)$.

The Steady-State Heat-Flow Technique



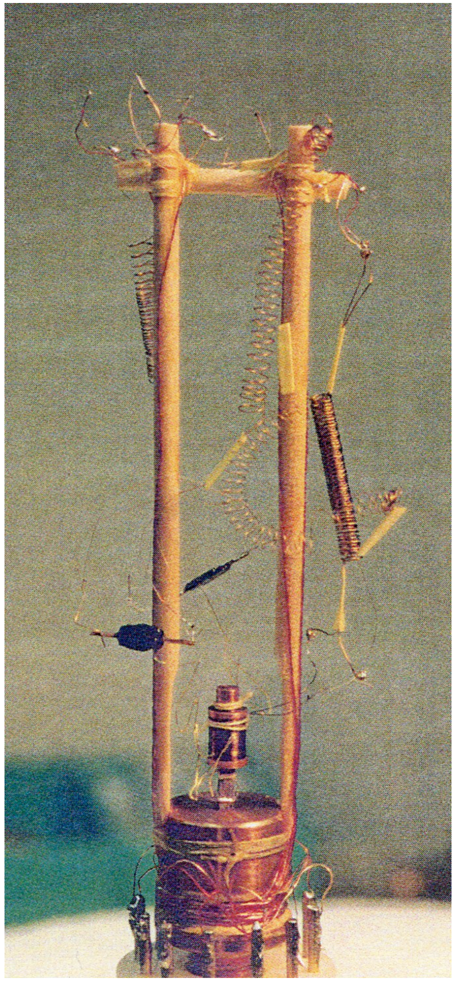
Experimental set-up

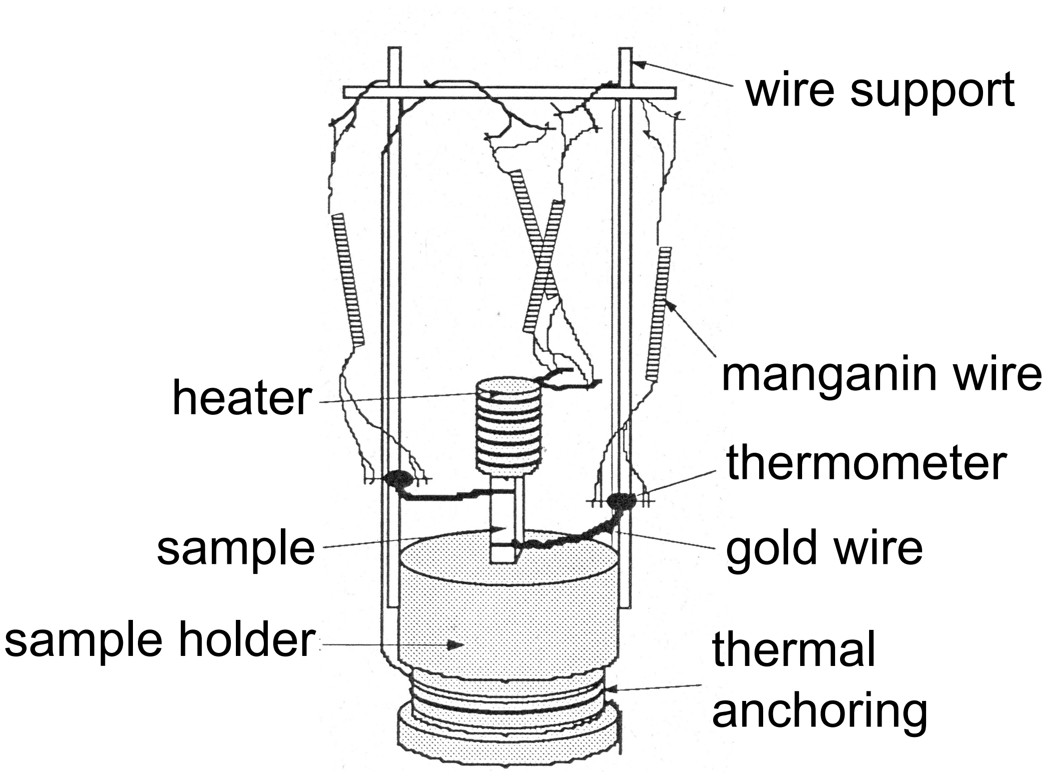


1 – copper holder
2 – thermal anchoring of the wires
3 – coil foil
4 – copper
5 – stainless-steel holder
6 – sample (Lead single crystal)
7 – carbon thermometer
8 – silver foil
9 – heater
10 – GE varnish; removed after mounting

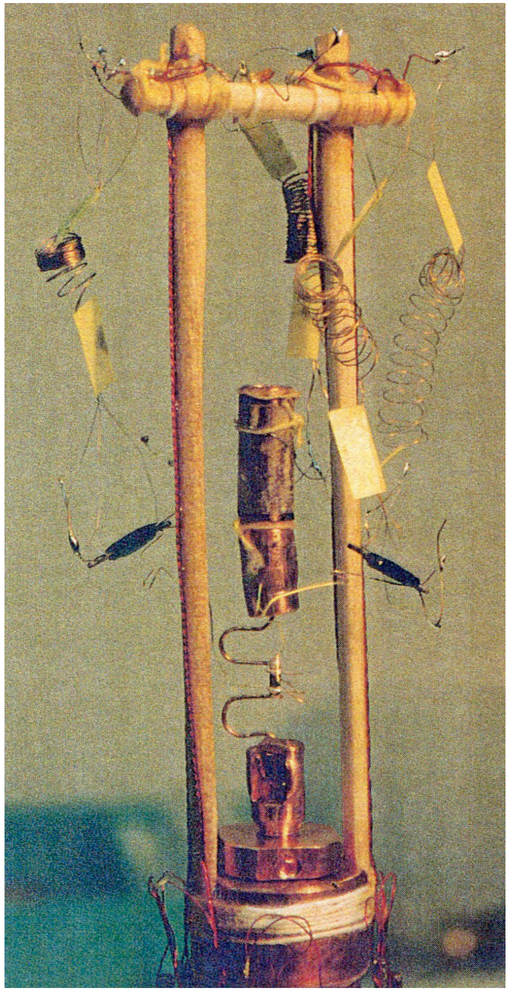
Experimental set-up: sample and sample holder

(from W. Odoni, P. Fuchs and H. R. Ott, Phys. Rev. B 28, 1314 (1983))



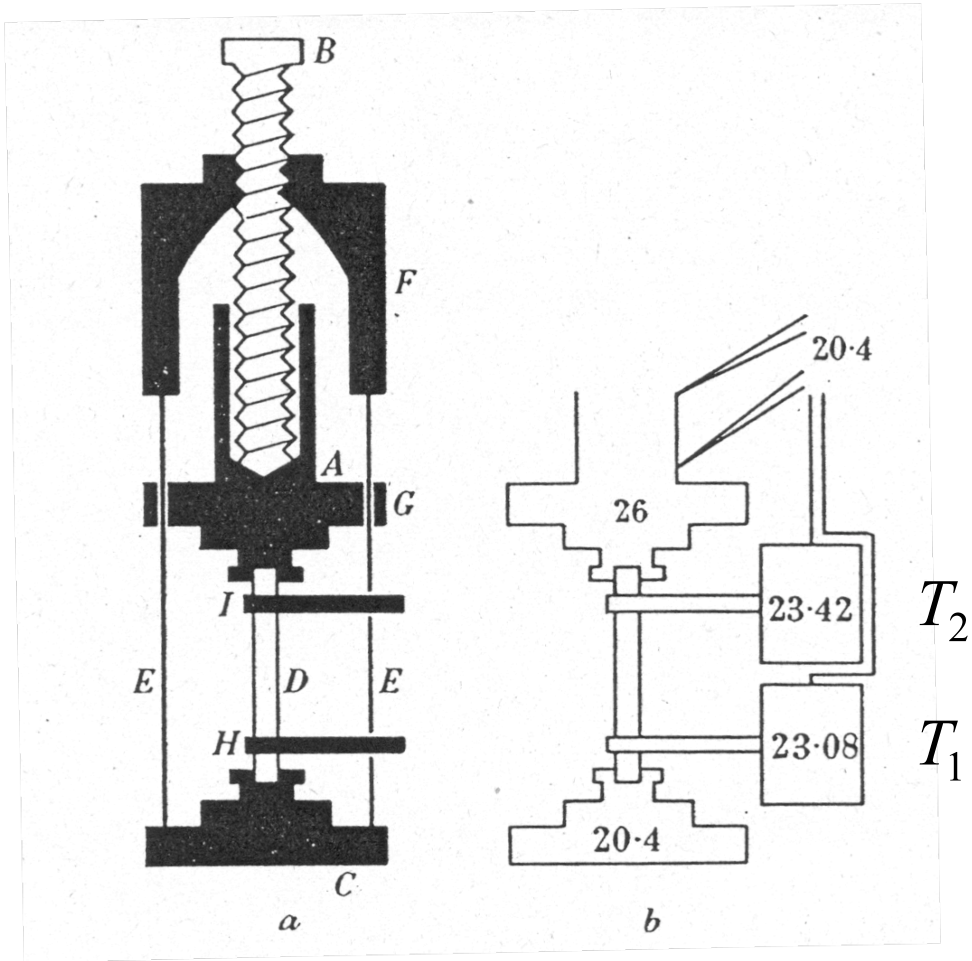


Experimental set-up; *T*-range between 0.065 and 100 K



Experimental set-up: small samples

What is the upper limit for ΔT?



$$\frac{\Delta T}{\bar{T}} \approx 0.015$$

Temperature distribution in a steady-state heat-flow set-up

(from R. Berman, F. E. Simon and J. M. Ziman, Proc. Roy. Soc. A **220**, 171 (1953))

Assume that the steady-state heat flow is established, and that the isothermal surfaces are planar and perpendicular to the sample between the thermometers T_1 and T_2. Integrating the Fourier – Biot equation from T_1 to T_2 yields

$$\int_{T_1}^{T_2} \varkappa(T)\, dT = -\frac{\dot{Q}\Delta x}{A}.$$

The mean value of $\varkappa$ between T_1 and T_2 is

$$\bar{\varkappa}(T) = -\frac{\dot{Q}\Delta x}{A\Delta T}.$$

Whether $\bar{\varkappa}(T)$ is a sufficiently accurate measure of $\varkappa(T)$ depends on the functional form of $\varkappa(T)$, the magnitude of ΔT, and how $\bar{T}$ is defined from the measured T_1 and T_2.

Usually, $\bar{T}$ is defined from T_1 and T_2 as $\frac{1}{2}\left(T_1+T_2\right)$.

Functional form of $\varkappa(T)$: replacing $\varkappa(T)$ by the 2nd order Taylor series approximation to $\varkappa(T)$ about $\bar{T}$ yields

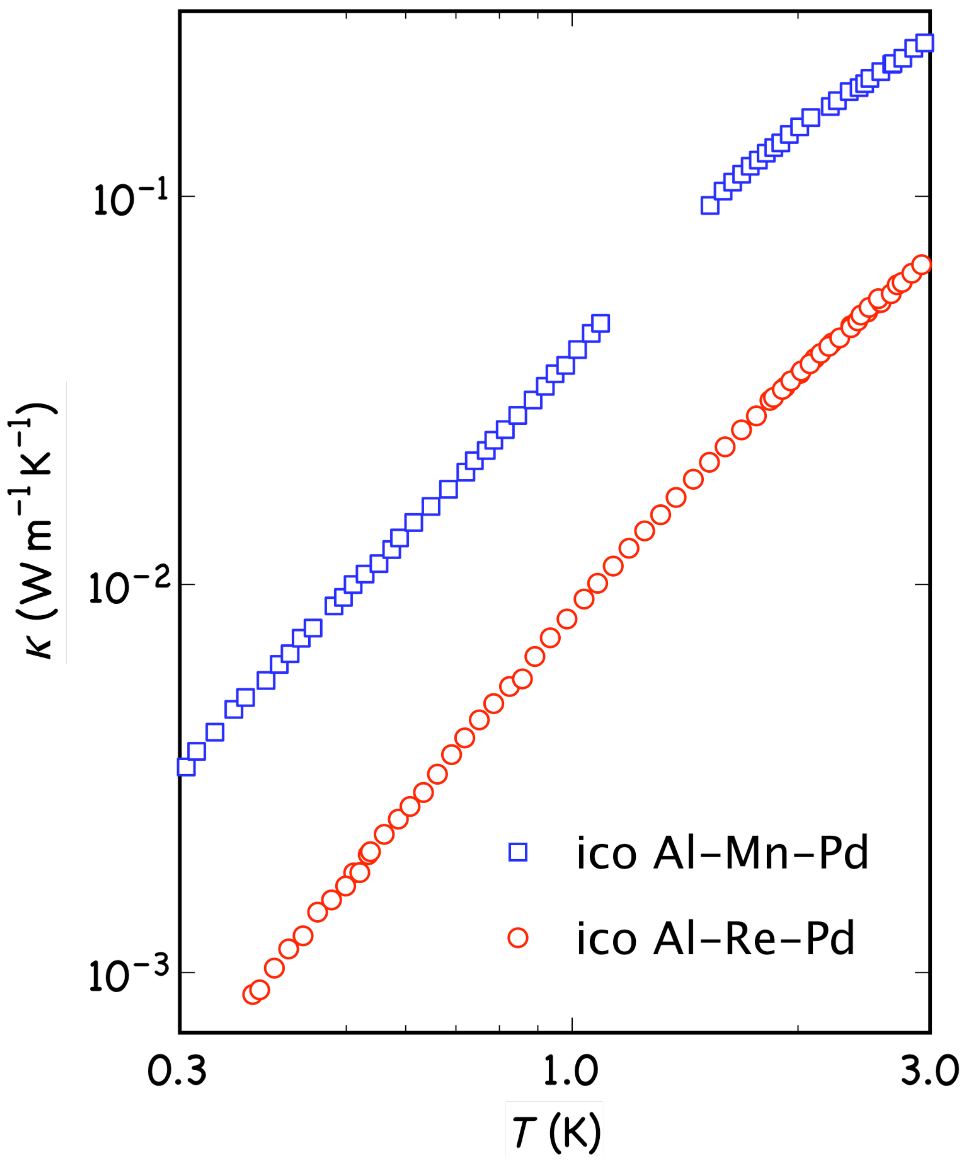
$$\bar{\varkappa}\left(\bar{T}\right)=\int_{T_1}^{T_2}\left(\varkappa\left(\bar{T}\right)+\varkappa'\left(\bar{T}\right)\left(T-\bar{T}\right)+\frac{1}{2}\varkappa''\left(\bar{T}\right)\left(T-\bar{T}\right)^2\right)dT=$$

$$\varkappa\left(\bar{T}\right)\Delta T+\frac{1}{24}\varkappa''\left(\bar{T}\right)\Delta T^2\Delta T$$

The relative error is $\dfrac{\delta\varkappa}{\varkappa}=\dfrac{\bar{\varkappa}\left(\bar{T}\right)-\varkappa\left(\bar{T}\right)}{\varkappa\left(\bar{T}\right)}=\dfrac{1}{24}\dfrac{\varkappa''\left(\bar{T}\right)}{\varkappa\left(\bar{T}\right)}\Delta T^2$

Therefore ΔT can often be quite large compared with $\bar{T}$.

If $\varkappa=\text{const}$ or $\varkappa\propto T$, $\bar{\varkappa}\left(\bar{T}\right)=\varkappa\left(\bar{T}\right)$ and ΔT can be any size.



Example:

for icosahedral phases

below ~ 1 K $\varkappa$ varies as T^2

the relative error in $\varkappa$ is

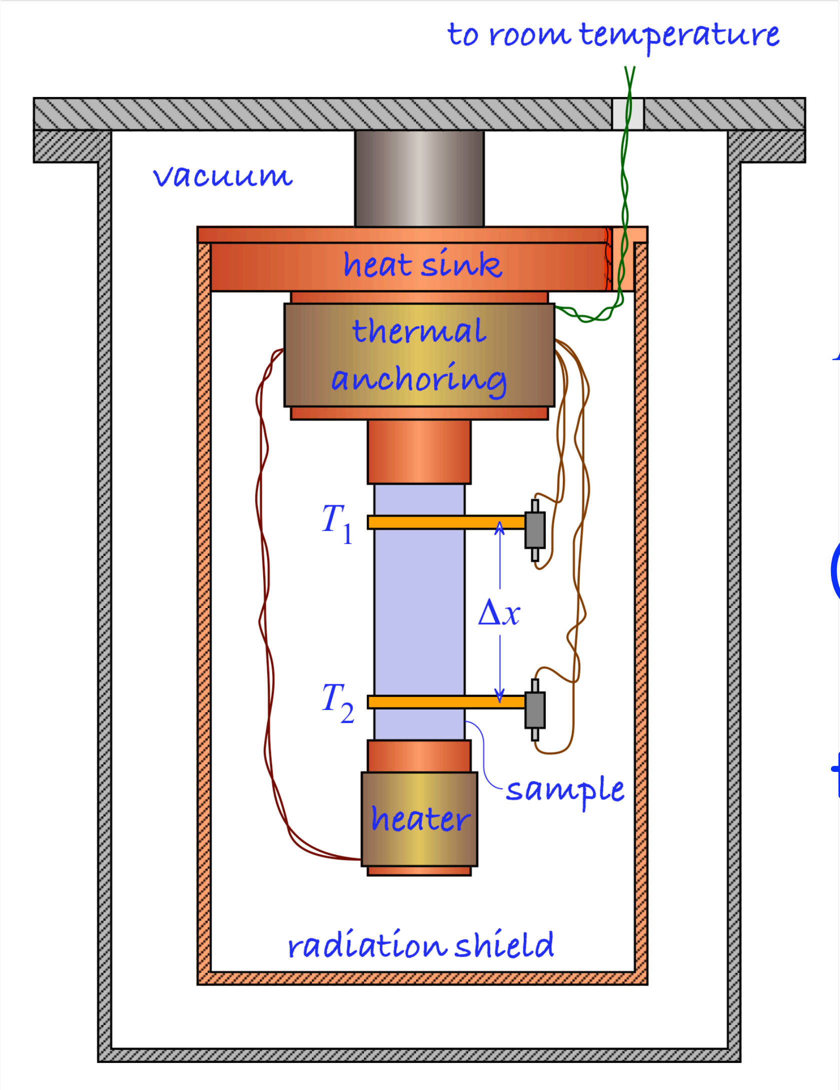
$$\frac{\Delta\varkappa}{\varkappa} = \frac{1}{24}\frac{\varkappa''(\bar{T})}{\varkappa(\bar{T})}\Delta T^2 = \frac{1}{12}\left(\frac{\Delta T}{\bar{T}}\right)^2$$

extremely small error!

Thermal conductivity of icosahedral phases at low temperatures

(from M. A. Chernikov, A. D. Bianchi, E. Felder, U. Gubler and H. R. Ott, Europhys. Lett. **35**, 431 (1996); M. A. Chernikov, A. D. Bianchi and H. R. Ott, Phys. Rev. B **51**, 153 (1995))
)

Heat Loss due to Radiation Effects



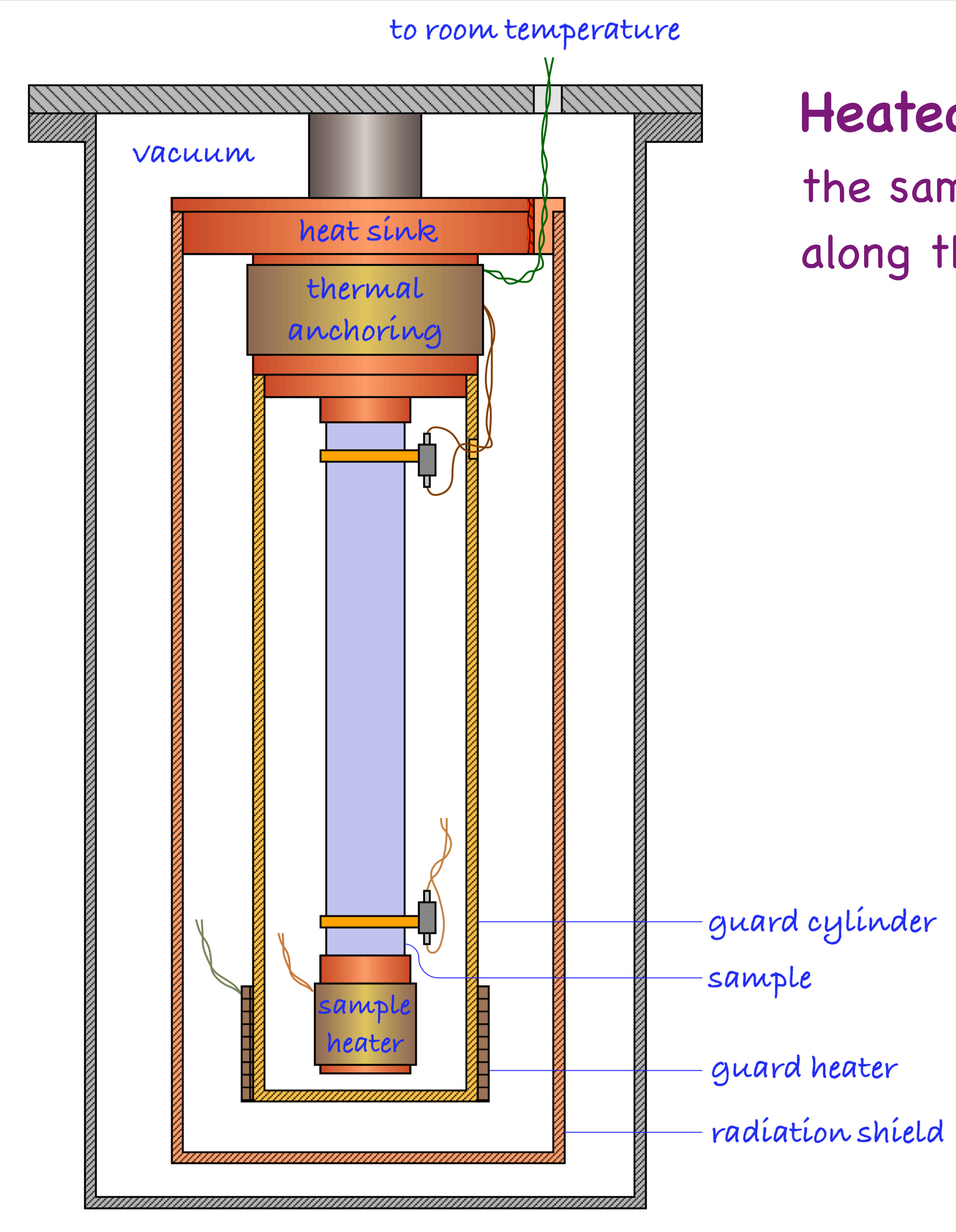
$$P = A\sigma\varepsilon\left(T_{\text{sample}}^4 - T_{\text{surrounding}}^4\right)$$

$$\sigma = 5.67\times10^{-8}\ \text{Wm}^{-2}\text{K}^{-4}$$

(the Stefan – Boltzmann constant)

translates to $\varkappa_{\text{error}} \propto T^3$

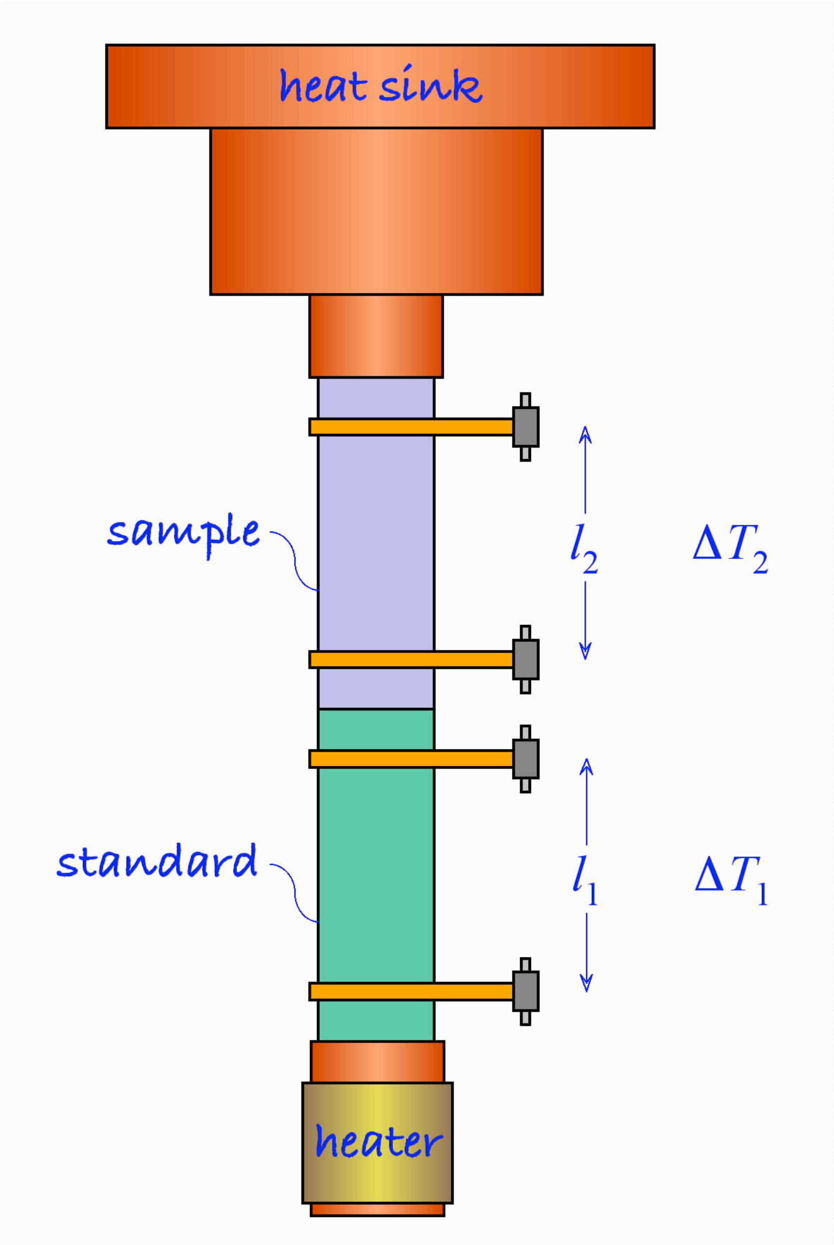
The maximum temperature reached in a conventional steady-state experiment is often limited by radiation losses



Heated guard set-up:

the same temperature distributions along the guard and the sample

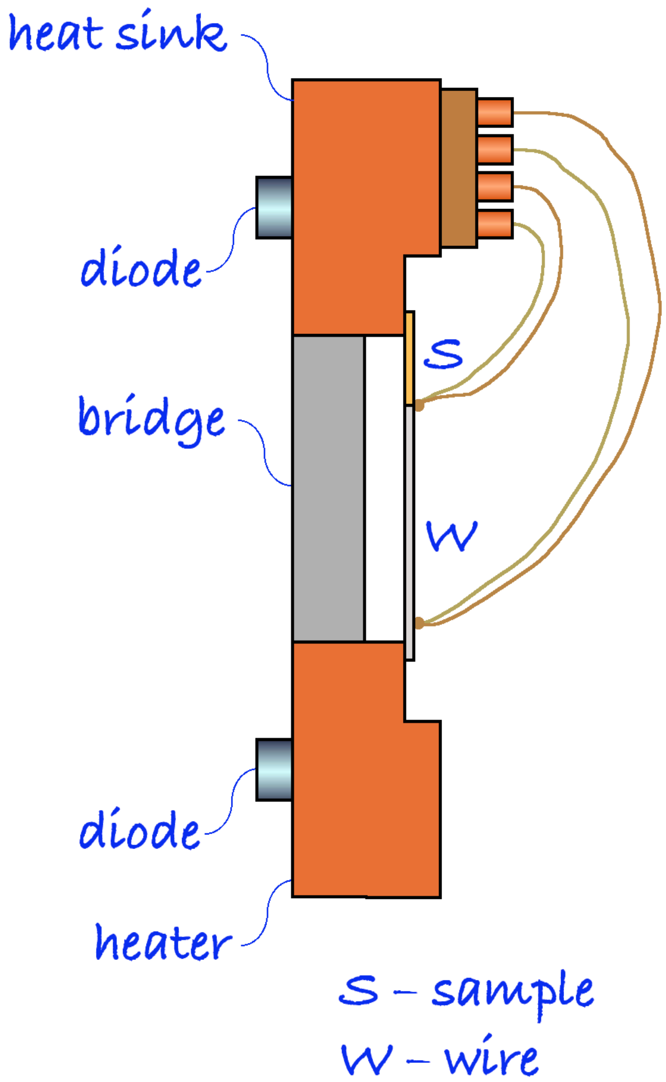
The Comparative Technique



$$P = \frac{\varkappa_1 \Delta T_1 A_1}{l_1} = \frac{\varkappa_2 \Delta T_2 A_2}{l_2}$$

$$\varkappa_2 = \varkappa_1 \frac{\Delta T_1}{\Delta T_2} \frac{A_1}{A_2} \frac{l_2}{l_1}$$

The Comparative Technique – Measuring Microgram Wiskers



Sample :

Pb – doped $Bi_2Sr_2Ca_1Cu_2O_x$

$800 \times 80 \times 4\ \mu m^3$

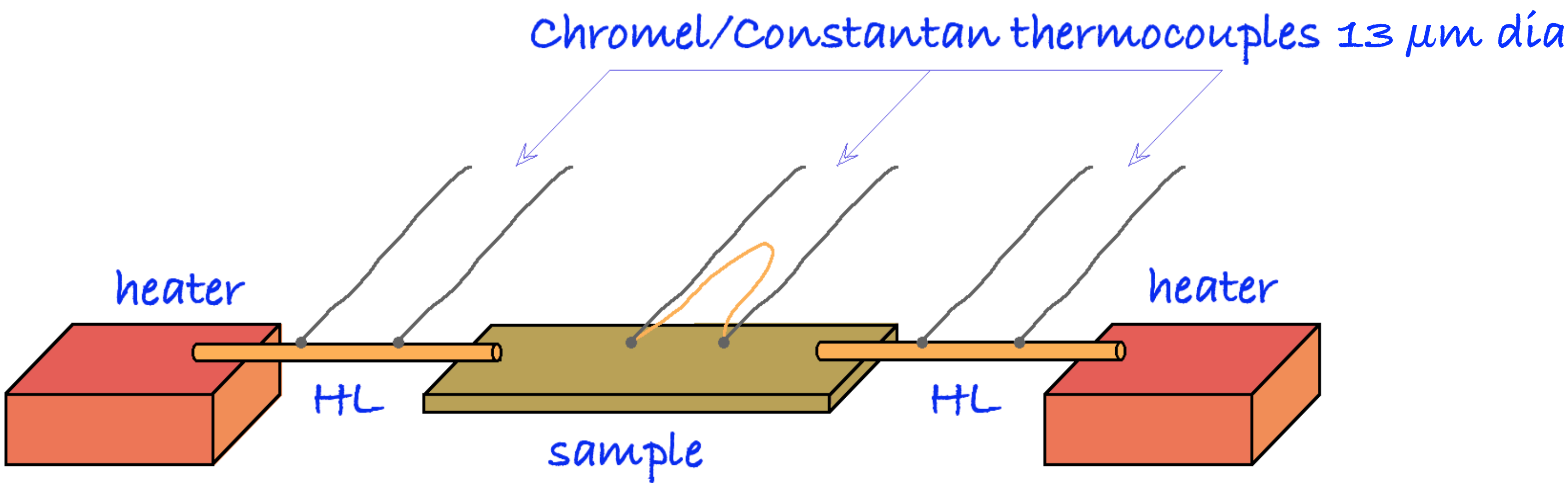
Thermocouples :

13 μm Chromel / Constantan

D. T. Verebelyi,

Rev. Sci. Instrum. **68**, 2494 (1997)

The Double Comparative Technique



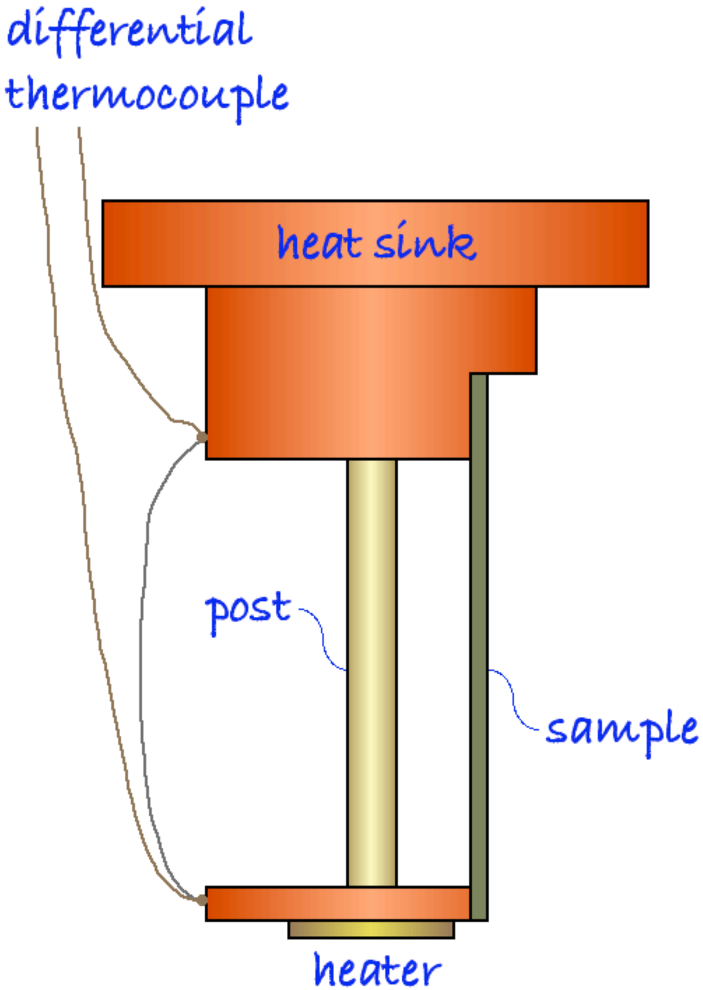
HL – heat link, Constantan 0,1 mm dia

sample – $Bi_2Sr_2YCu_2O_8$, $2 \times 1 \times 0.01$ mm^3

The experimental set-up: heat-flow reversal possible

(P. B. Allen, Xiaoqun Du, L. Mihaly and L. Forro, Phys. Rev. B **49**, 9073–9079 (1994))

The Parallel Thermal Conductance Method



Applications – structurally weak samples, which cannot support thermometers and heater.

Step 1 – a conventional steady-state set-up with a low-conductive post is assembled; thermal conductance is measured.

Step 2 – a sample is attached between the sink and the heater, in parallel with the post; the thermal conductance of the system is then again measured; parallel thermal conductance, which is determined by the sample and thermal contacts is calculated.

Error: blackbody radiation from the sample.

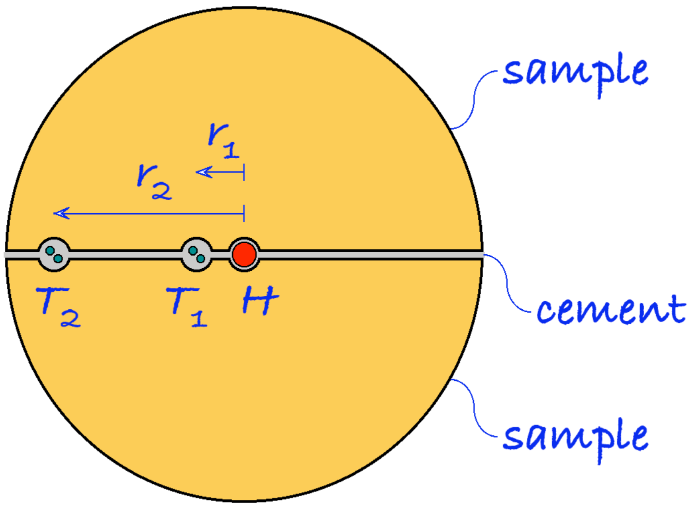
The Radial Flow Method

$$\varkappa = \frac{P}{L} \frac{\ln\left(r_2/r_1\right)}{2\pi\left(T_1 - T_2\right)}$$

L – sample length

The end-loss error

$\delta_{\text{end-loss}} < 0.5\%$ if $L > 4D$



H – heater

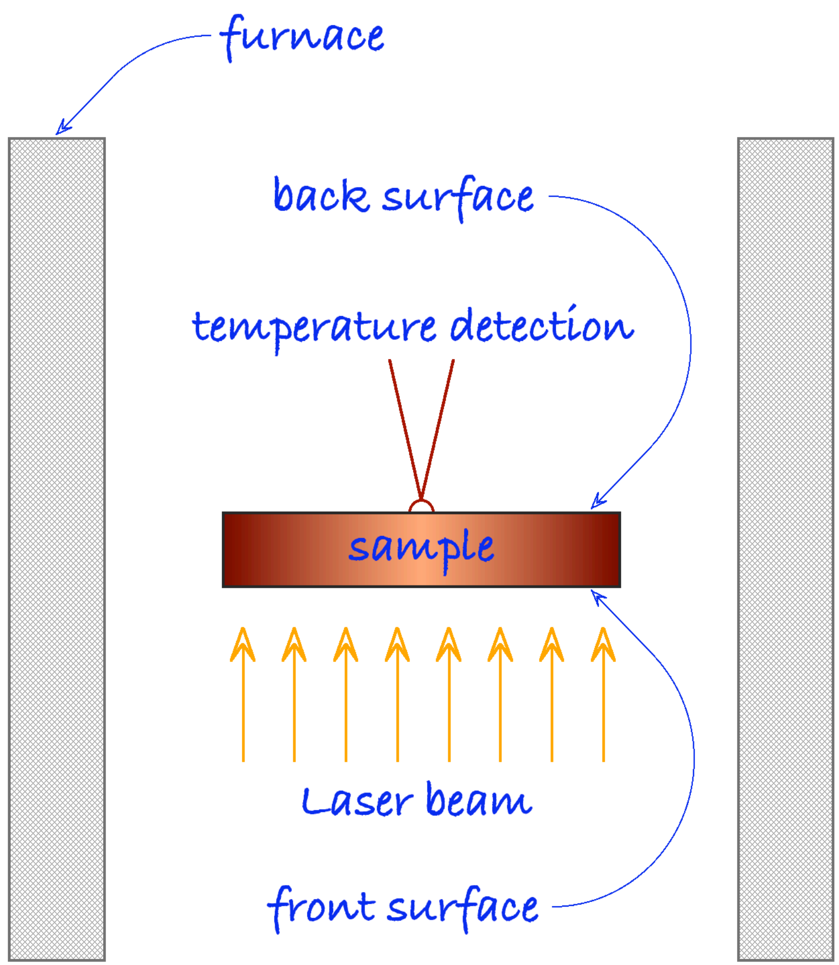
T_1, T_2 – thermocouples

Heat is applied internally – low radiation losses from the heater

(G. A. Slack and C. Glassbrenner, Phys. Rev. **120**, 782 (1960))

The Flash Method of Measuring Thermal Diffusivity

W. J. Parker, W. J. Jenkins, C. P. Butler and G. L. Abbott, J. App. Phys. **32**, 1679 (1961)



One face of a sample in the shape of a thin disk is irradiated by a short, of the order of ½ ms, light pulse.

The heat propagation through the sample results in a temperature rise on the other surface.

An infra-red detector monitors the temperature rise of the opposite side of the sample.

The thermal diffusivity is then calculated from the temperature versus time curve.

$$\frac{\partial T}{\partial t} = \alpha \frac{d^2 T}{dx^2} \qquad (1)$$

$$\Delta T(d,t) = \frac{Q}{\rho C_p d \pi r^2}\left[1 + 2\sum_{n=1}^{\infty}(-1)^n \exp\left(-\frac{n^2 \pi^2 \alpha t}{d^2}\right)\right] \qquad (2)$$

$$\alpha = \frac{1.37 d^2}{\pi^2 t_{1/2}} \qquad (3)$$

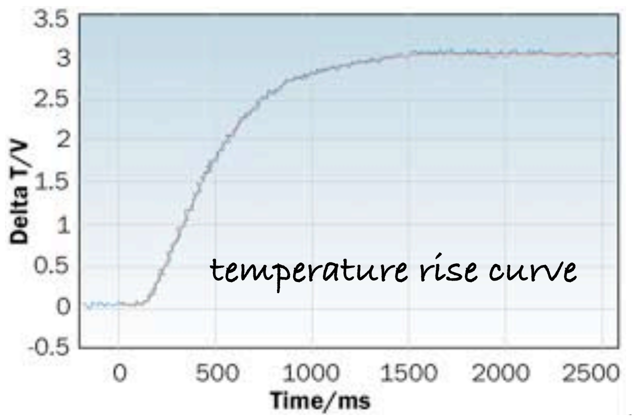
Here α is the thermal diffusivity, Q is the energy absorbed at the front surface, ρ is the mass density, C_p is the specific heat, d is the thickness of the sample, and r is the radius of the sample.

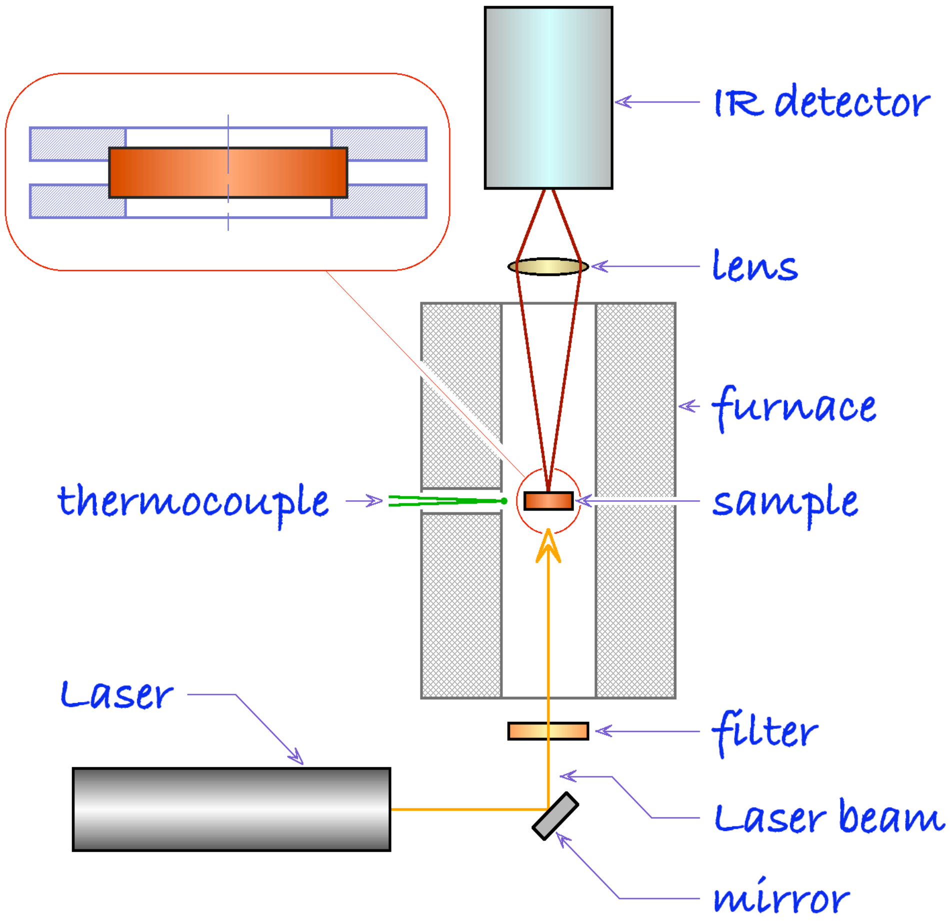
Eq. (2) – Carslaw and Jaeger's solution to one - dimensional heat flow

d^2/α – characteristic time of heat diffusion across the sample

The sample holder is specifically designed to shield the IR detector from direct laser or flash light.

The both sample surfaces must be highly emissive – often this requires a thin coating of graphite, which can lead to a significant error, if good adhesion is not achieved.

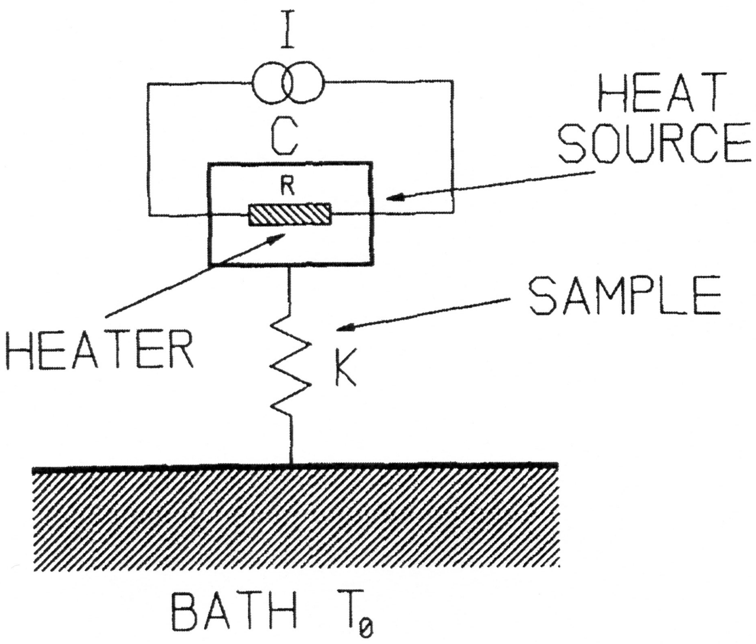




Flash diffusivity set-up: schematic

The sample temperature rise is kept fairly small, between about ½ to 2 °C. An InSb infra-red detector, cooled by liquid nitrogen, is a common choice. At very high temperatures up to 2800 °C pyrometers are used.

The Square Wave AC Drift Method



Square-wave modulation of the heater current creates temperature gradients

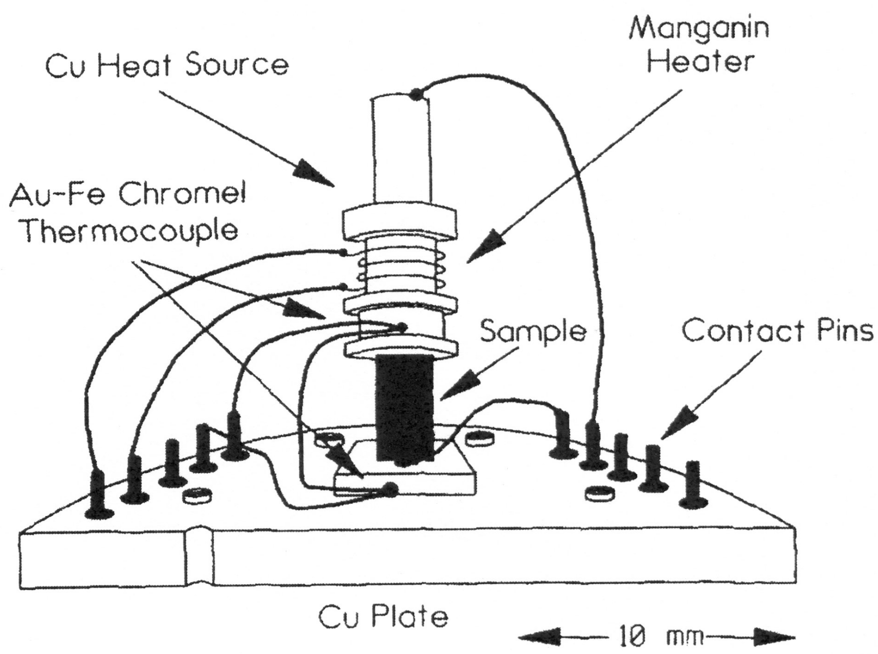
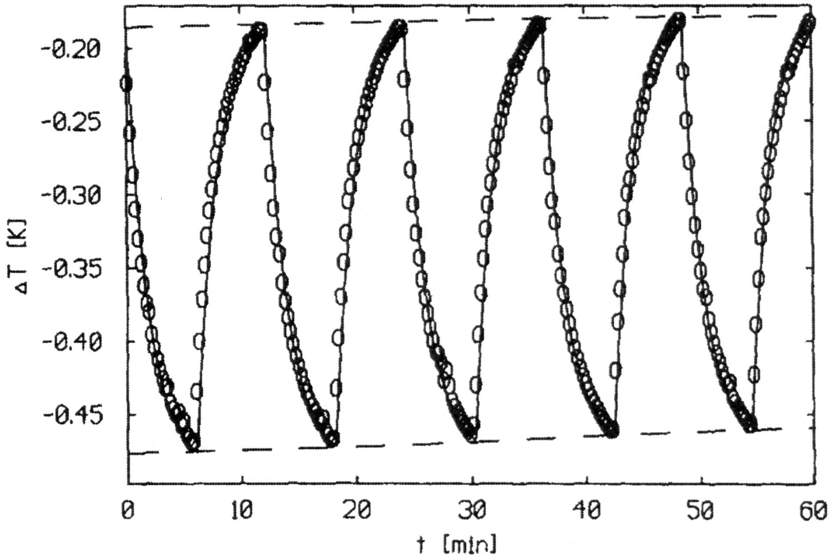


Diagram of the experimental set-up
(from O. Maldonado, Cryogenics **32**, 908 (1992))



Temperature vs. time variation

(from O. Maldonado, Cryogenics **32**, 908 (1992))

Assumption: the heater heat capacity, the heater resistance, and the thermal conductance of the sample are smooth functions of temperature.

The heat balance for the heater:

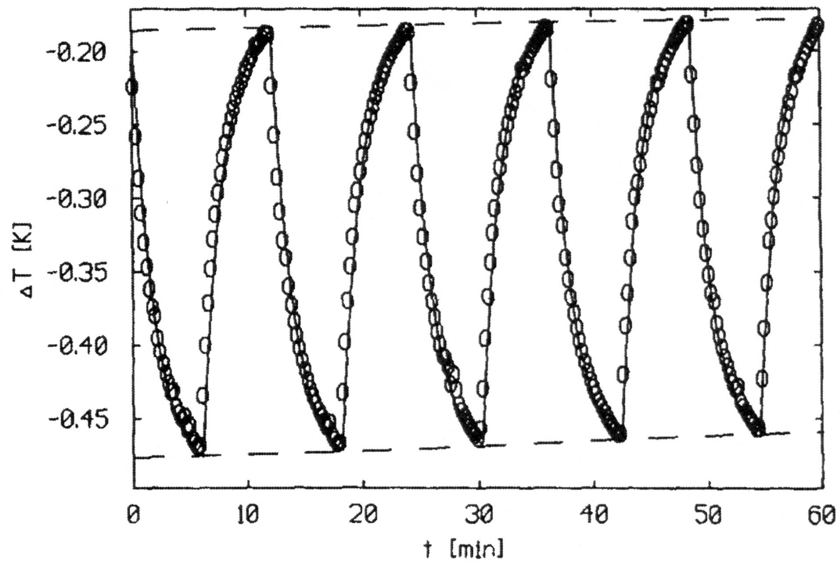
$$\frac{dQ}{dt} = C(T_1)\frac{dT_1}{dt} = R(T_1)I^2(t) - K(T_1 - T_0) \; .$$

Here dQ/dT is the rate of change of heat in the heater, T_1 is the heater temperature, T_0 is the bath temperature, C is the heater heat capacity, R is the heater resistance and K is the sample thermal conductance.

Assumptions:

(1) the heater heat capacity, the heater resistance, and the thermal conductance of the sample are smooth functions of temperature

(2) the temperature drift is slow compared to the periodic oscillations



$$K = \frac{RI_0^2}{\Delta T_{\text{peak-to-peak}}} \tanh\left(\frac{K\tau}{2C}\right)$$

Advantage – the sample temperature sweeps as the data is collected. The square wave a.c. method has been implemented in a commercial instrument (PPMS, Quantum Design).

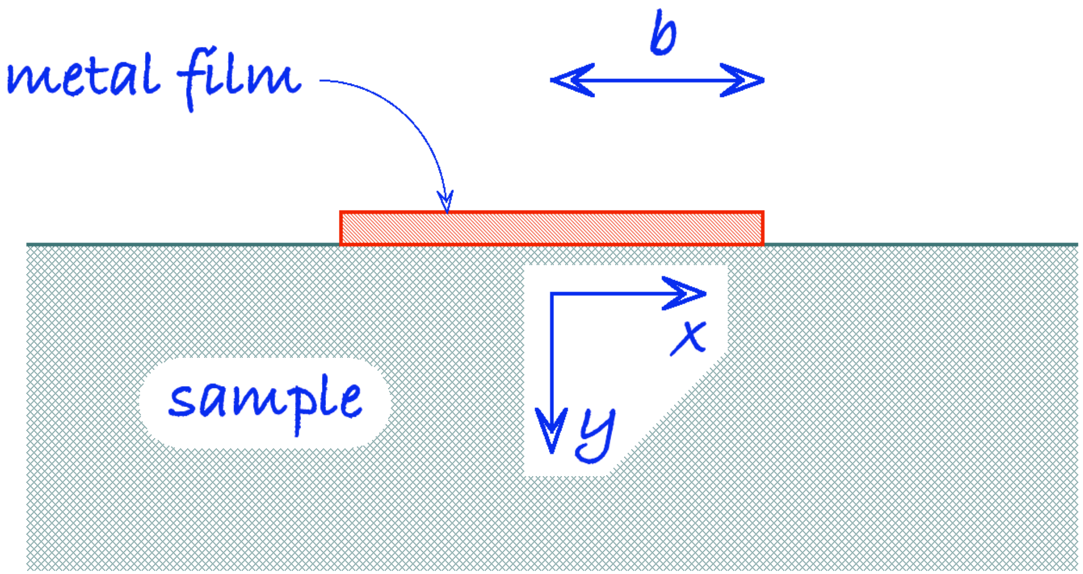
The 3ω Method

(D. G. Cahill, Rev. Sci. Instrum. **61**, 802 (1990))

Errors due to black-body radiation scale with a characteristic length of the experimental geometry.

The method is insensitive to these errors because the effective thickness of the sample is small, of the order of 100 μm.

The method uses a radial flow of heat from a single element that serves both as a heater and thermometer.

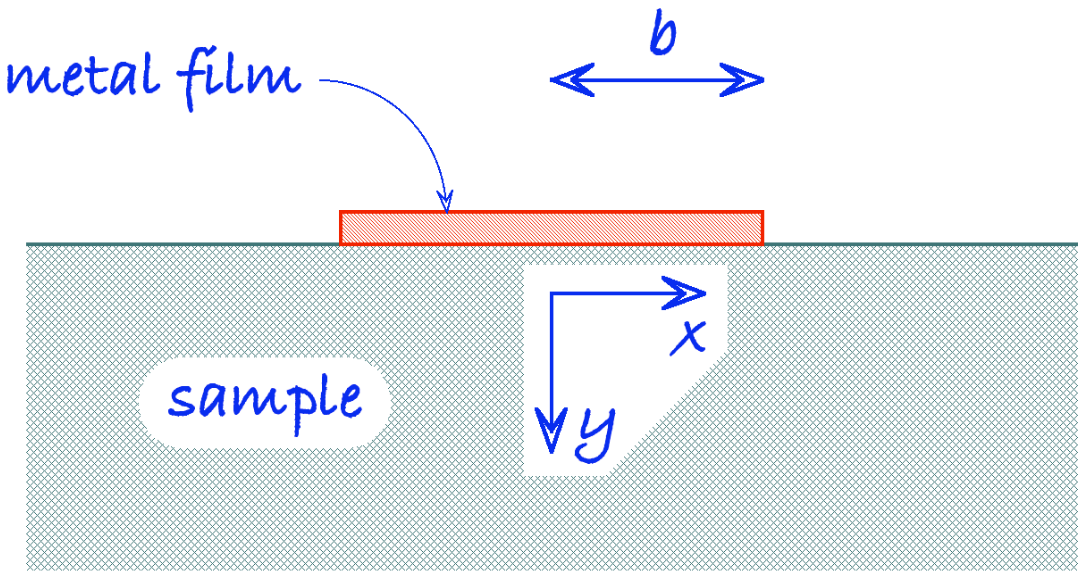


Temperature oscillations inside the sample at a distance $r = \sqrt{x^2 + y^2}$ from the line

$$\Delta T(r) = \frac{P}{l\pi\lambda} K_0\left(qr\right),$$

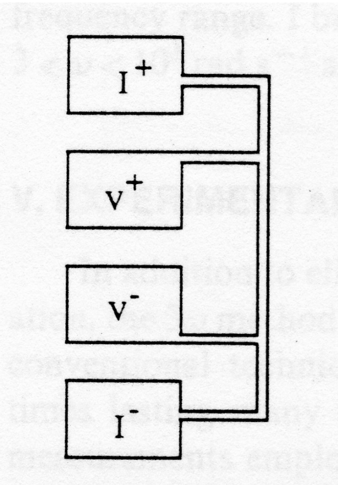
K_0 – the zeroth-order modified Bessel function,

$q^{-1} = \sqrt{\lambda/\left(2i\omega\rho C\right)}$ – the wavelength of the diffusive thermal wave.



The current through the line oscillates at a frequency ω and the power is generated at a frequency 2ω. The resistance of pure metals increases with increasing temperature. Therefore the resistance of the metal line has a small a.c. component that oscillates at 2ω. The resistance oscillation times the original driving current oscillating at ω results in an oscillation, with small amplitude, of the voltage across the line at 3ω.

Experimentally, measurements of the voltage across the line at 3ω are taken at selected fixed frequencies.



The thermal conductivity $\varkappa$ is determined from

$$\varkappa = \frac{V^3 \ln\left(f_2/f_1\right)}{4\pi l R^2 \left(V_{3,1} - V_{3,2}\right)} \frac{dR}{dT}.$$

Here V is the voltage across the metal line at ω,

R is the resistance of the metal line,

$V_{3,1}$ is the 3ω voltage at frequency f_1,

$V_{3,2}$ is the 3ω voltage at frequency f_2.