Journal of Thermal Analysis, Vol. 14 (1978) 151—156

THE USE OF A CONTINUOUS-FLOW HELIUM CRYOSTAT IN LOW-TEMPERATURE CALORIMETRY

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(Received September 13, 1977)

For experimental investigation of the temperature-dependences of specific heat and thermal conductivity in the range 4—300 K a continuous-flow helium cryostat has been developed. Its adaptation for low-temperature calorimetry and its use for measurement of the temperature-dependences of the specific heats of bulk samples of metals and insulators are described in this note. The phase transition from the normal to the superconductive state has been measured on NbTi and its critical temperature determined. Two methods of determination of the temperature-dependences of the specific heats of metals and insulators have been developed. The inaccuracy of specific heat determination did not exceed 2 % with metal materials and 5 % with insulating materials.

To reach and maintain any temperature in the range from room temperature to 4 K continuous-flow helium cryostat [1] has been developed, which operates with liquid or gaseous helium without a nitrogen bath, and which is capable of operation either under usual laboratory conditions or after being inserted into a bath He cryostat with a superconductive solenoid for measurements in high magnetic fields.

The principle of the measuring method, which is the most suitable for the characteristics of the continuous-flow cryostat, is the determination of the thermal capacity of a sample localized in the calorimeter with isothermal shield in the operating mode of low thermal impulses. The heat capacity is then derived from the well-known relation between the amount of heat supplied by the heating impulses and the increase of temperature of the sample at constant pressure.

Experimental

To make possible calorimetric measurements in the high magnetic field of a superconductive magnet, the outer diameter of the cryostat nowhere exceeds 50 mm and its length reaches 1630 mm. Figure 1 shows the function of the cryostat operated as a calorimeter for the investigation of specific heats of bulk samples.

The temperature of the isothermal shield, which is thermally connected through an indium joint with first heat exchanger 6, can be adjusted either by the control of coolant flow by needle valve 7 or by electric heating 8 controlled by the PID controller, which in turn is controlled by the CLTS thermal sensing device. The long-term temperature stability of the lower heat exchanger and of the shield during 30 min is about 0.001 K.

The lowest temperature of the sample separated quasi-adiabatically from the surrounding parts is reached by filling with gaseous helium at a partial pressure of

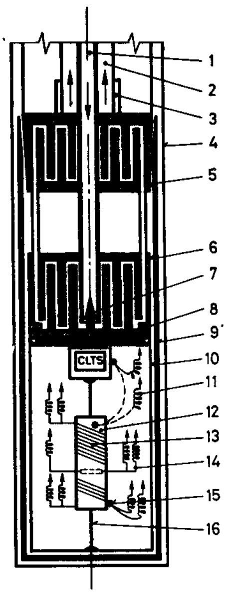

Fig. 1. Schematic section through lower part of cryostat arranged for measurement of specific heat of superconductive material; 1 — inlet of cryogenic medium, 2 — outgassing pipe, 3 — thermal anchoring, 4 — vacuum container, 5 — second heat exchanger, 6 — first heat exchanger, 7 — inlet needle valve, 8 — controlling heating, 9 — radiation shield, 10 — isothermal shield, 11 — differential thermocouple, 12 — sample, 13 — heating of sample, 14 — Allen-Bradley resistor, 15 — thermocouple, 16 — nylon fibre

approx. 1 Pa. After the required temperature is reached, the space with the sample is pumped off for a long period, until the steady state (from the point of view of helium desorption from the sample surface) is stabilized, while the cryostat temperature is maintained at exactly the same value as the temperature of the sample.

The temperature of the sample was measured with the use of a Au + 0.03 at. % Fe vs. chromel thermocouple, which has a sufficient sensitivity and stability in the range 4–300 K and has very low heat capacity. The thermoelectric power of the sensor, the reference junction of which was maintained at the temperature of the water triple point, was measured by the compensation method, using the TETTEX measuring set with an accuracy of the order of 10^{-2} μV. Calibration of the thermocouple was performed using platinum (Rosemount 162 D – calibrated at NBS U.S.A.) and germanium (Scientific Instruments N1L) substandard with an accurancy of 2×10^{-2} K. In the cases when the phase transition was measured on the superconductor, an Allen-Bradley resistor was used in the temperature range 4–20 K instead of the thermocouple, because the thermocouple sensitivity at low ΔT in this range was not sufficient. Temperature measurement was performed on a Cryobridge S 72 Řež measuring bridge in a three-point-connection of the Allen-Bradley resistor.

The accuracy of specific heat determination is mainly affected by the intensities of parasitic heat flows and by the heat capacities. The first effect was substantially reduced by evaluation of the temperature increase of the sample by extrapolation of the time-dependence of the sample temperature close to the thermal impulse. The second effect was lowered to 1 % of the heat capacity of the complete sample (using a suitable choice of sample heating winding, adhesive, temperature sensor, electrical inlets and mechanical gripping), and thus the error in their determination did not substantially influence the measurement.

To test the method, measurement on a sample of very pure Fe (99.995 %) was performed. The results are in full agreement with the representative measurements of the literature [2].

The metal samples were bulk cylinders. Insulant samples were thin-walled hollow cylinders (diameter 17 mm, wall thickness approximately 1 mm), which had on their outer surface symmetrically displaced heating windings, and on their inner surface a thermally anchored thermocouple (Fig. 2). Thus, the relaxation

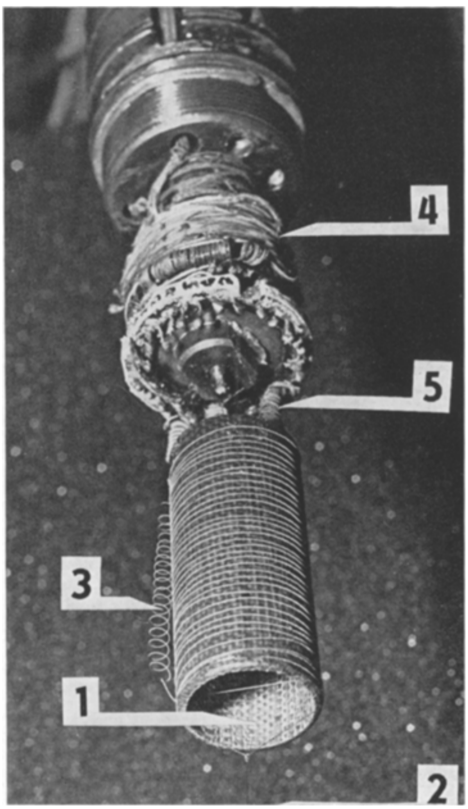

Fig. 2. Arrangement for determination of specific heat of insulators; 1 — sample, 2 — nylon fibre, 3 — inlets wound into self-supporting spirals, 4 — thermal anchoring of inlets, 5 — inlets of thermocouples

times of temperature equilibrium establishment were shortened to an acceptable level.

For the fast determination of the location of the phase transition in the temperature scale and for preliminary determination of the thermal dependence of the specific heat, a DTA method was modified and the Au + 0.03 at.% Fe vs. chromel thermocouple was used in differential connection. One of the thermocouple junction points was thermally anchored to the sample, while the second junction was located on the thermally well-stabilized isothermal shield. The difference of thermoelectric power for slow continuous growing or impulse heating of the sample was reinforced by a TETTEX photoelectric amplifier and recorded.

Results and discussion

More than one hundred experiments have shown that cooling down of the cryostat to 5 K lasts 3 min and the consumption of liquid helium (LHe) without any precooling is 0.8 l. When operation with the mentioned temperature stability is controlled at the temperature of 5 K, the coolant consumption of the cryostat is

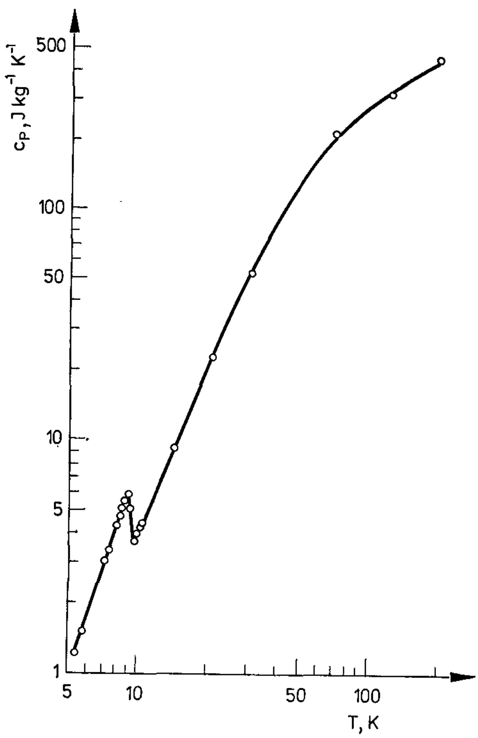

Fig. 3. Temperature-dependence of specific heat of superconductive material NbTi50

4 l LHe/h, at 10 K 3 l LHe/h, at 20 K 1.5 l/h, at 50 K 0.5 l/h, and at $T > 100$ K the consumption rate is not greater than 0.2 l/h. The upper heat exchanger covers thermal losses of about 600 mW, and the lower heat exchanger 40 mW.

Parasitic heat flows to the sample were minimized by proper choice of the material and by arrangement of the inlets to maintain the time course of the temperature of the sample almost constant, when the temperature increase of the sample ΔT was always smaller than $10^{-2}\,T$.

In the quasi-adiabatic calorimeter with isothermal shield the temperature-dependences of the specific heats of several metal materials (Czechoslovak stainless steels, permalloy), as well as of insulators (epoxy resin insulators using textile or glass laminate as basic structure) were measured. The dependence $c(T)$ for the superconductive material 50% Nb + 50% Ti (Fig. 3) was also measured and the critical temperature for transition from the normal into the superconductive state was determined at zero external magnetic field as $T_c = 9.2$ K.

For metal materials the relative error in measurement did not reach more than 2% and for insulators 5%. The results of measurement on alloys were compared with measurements on equivalent foreign-made materials (for example on AISI steels) and a good agreement was found.

As the first verification experiments show, both methods may be used successfully for experimental investigation of phase transitions of developed superconductive materials, of intermetallic rare earth compounds, and of metal hydrides.

References

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Résumé — Un cryostat à circulation continue d'hélium, destiné à la détermination expérimentale de la chaleur spécifique et de la conductivité thermique en fonction de la température entre 4 et 300 K, a été mis au point. Son utilisation comme calorimètre à basses températures est décrite ainsi que son application à la mesure de la chaleur spécifique en fonction de la température d'échantillons massifs de métaux et isolants. La transition de phase de l'état normal à l'état supraconducteur a été étudiée sur un échantillon de NbTi et la température critique a été déterminée. Deux méthodes ont été mises au point pour déterminer la variation de la chaleur spécifique en fonction de la température des métaux et isolants. L'imprécision de la détermination de la chaleur spécifique est inférieure à 2% pour les métaux et 5% pour les isolants.

Zusammenfassung — Ein Heliumkryostat mit kontinuierlichem Strom wurde für Versuchszwecke zur Untersuchung der Temperaturabhängigkeit der spezifischen Wärme und der Wärmeleitfähigkeit im Bereich von 4 bis 300 K entwickelt. Sein Einsatz als Tieftemperaturkalorimeter sowie seine Anwendung zur Messung der Temperaturabhängigkeit der spezifischen Wärme einer Anzahl von Metall- und Isoliermaterialproben werden beschrieben. Der Phasenübergang vom normalen in den Supraleitungszustand wurde an NbTi gemessen und seine kritische Temperatur bestimmt. Zwei Methoden wurden zur Bestimmung der Temperaturabhängigkeit der spezifischen Wärme von Metallen und Isolatoren entwickelt. Die Ungenauigkeit der Bestimmungen der spezifischen Wärme war unterhalb von 2% bei metallischen und unterhalb von 5% bei isolierenden Materialien.

Резюме — Для экспериментального исследования температурных зависимостей удельной теплоемкости и теплопроводности в области 4—300 К был разработан гелиевый криостат непрерывного потока. Описана его адаптация к низкотемпературному калориметру и использование его для измерения температурной зависимости удельной теплоемкости объёмных образцов металлов и изоляторов. Для NbTi был установлен фазовый переход из нормального состояния в сверхпроводящее и определена его критическая температура. Разработаны два метода определения температурной зависимости удельной теплоемкости металлов и изоляторов. Точность определения удельной теплоемкости в случае металлов не превышает 2%, а в случае изоляторов 5%.