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ノート

General Purpose Cryostat Applicable to Chopped-Light ac Calorimetry with Highly Stable Thermal Condition [†]

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An ac calorimeter was constructed for measurements below room temperature. A cryostat was designed while modifying that for adiabatic calorimetry for highly stable thermal condition. Temperature control comparable to that in adiabatic calorimetry was achieved. Chopped-light was introduced to the cryostat through optical fiber, and lead to the sample through a fused-quartz rod. The cryostat is applicable to general physical measurements keeping its capability for thermal control.

High stability of temperature is essential for any precise measurements of physical properties of materials. In this context, an application of basic design of adiabatic calorimeters is interesting and promising. In this note, an example of such a cryostat designed for chopped-light ac calorimetry is described.

The basic design of the cryostat is essentially the same as that for an adiabatic calorimeter used in this laboratory.[1] The points considered are (1) high thermal stability (controllability and uniformity), (2) interchangeability of the sample stage for many types of measurements, and (3) applicability to ac calorimetry. Besides, for ac calorimetry, (4) use of optical fiber to avoid disturbance on transferring coolant, and (5) ease for sample setting are also considered.

A sectional plan is shown in **Fig.1**. A copper block (D) that works as a heat sink is rigidly hung at the place of a calorimeter vessel for adiabatic calorimetry by three screwed brass rods (G) to avoid fluctuation. The details of the copper block for ac calorimetry is described later. By changing the block to an appropriate sample stage, the cryostat is applicable to many types of measurements. The signal wires are soldered on the terminals (H) to those to the outer side of the sample chamber made of copper (I, 250 g in mass), which is sealed using indium wire vacuum-tight to the lid having a fused quartz window for light entrance. The wires to the outer side are thermally anchored to the lid using varnish (GE7031), and then go through a vacuum seal of epoxy resin. The sample chamber may be evacuated through tubing (c) or filled with some inert gas depending on types of measurements. The chamber is hung by three stainless steel pipes (J) from the bottom of the can for coolant (O). The wiring outside the chamber consists of four miniature coaxial cables (Lake-Shore Cryotronics, SC) and forty doubly silk-insulated copper wires (0.1 mm in diameter) and are thermally anchored to the anchor made of copper (K, 410 g in mass) hung by three stainless steel rods (M) and the bottom of the coolant can, and then go through the tubing (b) outside the cryostat. The coaxial cables are connected to electrically-isolated and

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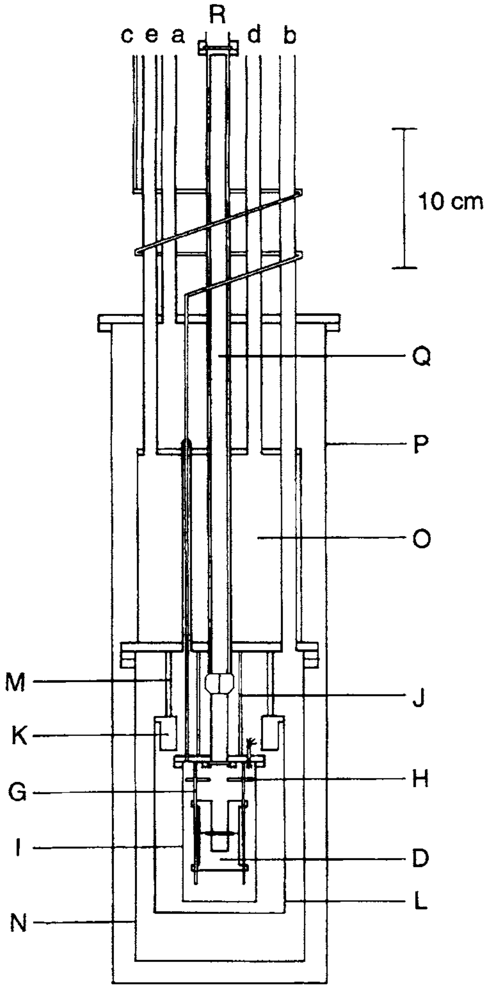

Fig.1 Sectional plan of the cryostat. D, copper block; G, screwed brass rods to support the copper block; H, electrical terminals; I, sample chamber working as an adiabatic shield; J, stainless steel pipe to hung the chamber; K, thermal anchor; L, radiation shield; M, stainless steel supports for the thermal anchor and the radiation shield; N, inner vacuum jacket; O, can for coolant; P, outer vacuum jacket; Q, fused-quartz rod working as light path; R, connector for optical fiber; a, tubing to evacuate the inside of the outer vacuum jacket; b, tubing to evacuate the inside of the inner vacuum jacket; c, tubing to evacuate the sample chamber; d, inlet for coolant; e, outlet for coolant.

vacuum-tight BNC connectors at the top of the cryostat, where normal coaxial cables are connected. The forty copper wires go outside the cryostat through four connectors hermetically sealed at the top of the cryostat. The temperature of the anchor is kept the same as that of the copper radiation shield (L, 530 g in mass). The whole assembly is located inside two vacuum jackets (N and P), which are sealed using indium wires vacuum-tight to flanges. The cryostat is immersed in liquid nitrogen at least up to the upper flange of the cryostat. The insides of the jackets are evacuated separately through the tubing (b) and (a) by a turbo-molecular pump to better than 10^{-3} Pa. The tubing (d) and (e) serve as inlet and outlet for coolant, respectively. The light path consists of a fused-quartz rod (Q) held by a Swagelok. The upper end of the light path is sealed with a window of fused-quartz, which is a part of a connector (R) to optical fiber.

The temperature difference between the copper block (D) and the wall of the sample chamber (I), and between the wall and the lid, and between the wall and the thermal anchor (K) are monitored by thermocouples and used for temperature control. The control is achieved by supplying electrical current to the heaters wound on the wall (70 Ω in resistance), lid (80 Ω), and anchor (and the radiation shield in parallel according to the mass ratio, 70 Ω in total) separately according to PID outputs from laboratory-made controllers. The temperature difference is always set being null between the wall and the lid while the temperatures of the anchor and the radiation shield are set about 2 K lower than that of the wall. To keep the temperature of the copper block, the temperature difference between the block and the wall is also set being null, resulting in the stability within ±1 mK. By changing the setting for the temperature difference between the block and the wall, a very slow temperature scan is possible. A maximum heating rate of scan is limitted to ca. 3 K min^{-1} by the power of the controllers. A step-wise temperature control is also possible in the heating direction by supplying the electrical current to the heater wound on the block while setting null.

A sectional plan of the sample block for ac calorimetry is shown in **Fig.2**. The sample (A) is attached on the junction of the crossed thermocouple of type E (13 μm in diameter), which is thermally anchored

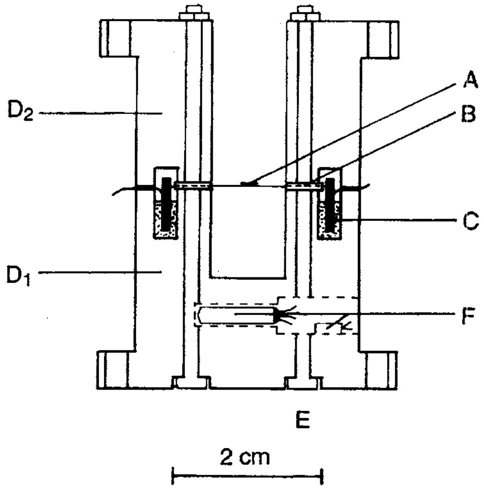

Fig.2 Sectional plan of the sample block for ac calorimetry. A, sample; B, circular plate for anchoring the thermocouples; C, copper terminals; D1, lower part of the heat sink; D2, upper part of the heat sink; E, long brass screws to tighten the block; F, thermometers.

to a circular plate of gold-plated copper (1 mm in thickness and 2 cm in diameter) with a center hole (1 cm in diameter) (B) while isolated electrically using cigarette paper, and connected to four copper terminals (C) using gold paint. The copper rods serving as the terminals are isolated from the block by Teflon. The copper plate (B) is sandwiched by the upper (D_2) and lower (D_1) parts of the gold-plated copper block (310 g in total mass), and tightened gently by two long brass screws (E). Heaters (70 Ω in total) are wound in series according to the mass ratio on the block (D_1 and D_2) for temperature control. The temperature of the block is monitored using platinum (Minco Product, S1055) and germanium (Lake-Shore Cryotronics, GR-200B-500) resistance thermometers, and a diode sensor (Lake-Shore Cryotronics, DT-470-SD) as exemplified as F in **Fig.2**. All the wires are thermally anchored closely to the block. The thermocouple for detecting the temperature difference between the block and the wall of the sample chamber (I) is screwed on the surface of the lower part of the block upon setting-up. A typical dimension of sample necessary for the ac calorimetry is $0.2 \times 0.2 \times 0.02$ mm^3.

Visible light used is from a commercial halogen lamp (12 V, 50 W). The stabilization is achieved by supplying the electricity to the lamp according to the imbalance signal of a resistance bridge circuit where the lamp itself serves as one arm. To avoid possible disturbance on filling coolant, the lamp and chopper are placed 5 m away from the cryostat and the chopped light was transmitted through optical fiber. Some grey filter is necessary for the measurements at low temperatures (below *ca.* 25 K).

Two signals of the thermocouples with the common junction at the sample are transmitted through two miniature coaxial cables to the BNC connectors at the top of the cryostat. The signal of one pair is fed to a step-up transformer (1:100) while being shielded by additional braided copper and then to a lock-in amplifier (Shinku-Riko, LAD-1) to monitor the magnitude of the temperature oscillation. The other signal is used for the measurement of the temperature difference between the copper block and the sample using a high-precision digital multimeter (Keithley, 2001). Since the connection to the digital multimeter exerts high level of noise to the input to the step-up transformer, the connection is allowed only during the measurement using the digital multimeter by the control program. The resistance of the platinum thermometer is measured using an ac thermometer bridge (Automatic System Laboratory, F700), and that of the germanium thermometer using the digital multimeter. The diode sensor is for rough control of the block temperature down/up to a desired temperature by using a commercial temperature controller (Lake-Shore Cryotronics, 330).

The suppression of the noise level is crucial for ac calorimetry because the signal corresponding to the magnitude of the temperature oscillation is less than 1 μV.[2] Besides the primary signal cable, the spatial arrangement of, the way to supply electricity to, and the way to connect the digital bus lines among the measuring instruments and the computer drastically affect the level. These factors were optimized trial and error.

Measurements are carried out automatically by a personal computer operated under Windows95. The connection between the computer and the measuring instruments (thermometer bridge, digital multimeter and temperature controller) is established through an IEEE-488 interface board (Hewlett-Packard, 82341C). The

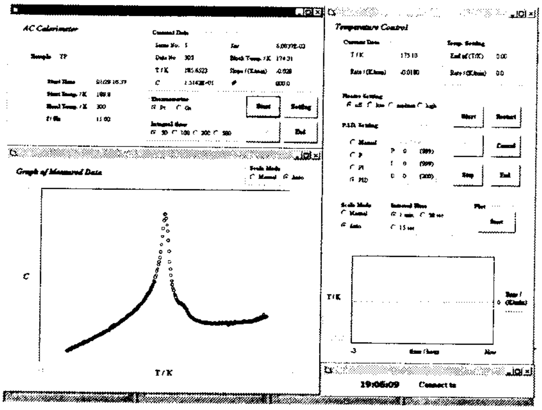

Fig.3 Snapshot of the screen of the computer display during a measurement.

code for the control and the measurement is all written in Visual Basic ver. 4.0 for the ease of future maintenance. The interface and the programming language are rather slow but fast enough for calorimetry. The problems in programming concern, instead, a poor capability for the low-level operation in this set-up. For example, the library supplied with the interface board does not support the interruption from the interface, *i.e.*, no responsibility to the service request from the instruments. This means all schedules should be controlled by the computer. The time required for the measurement of one data point and even for the switching of the internal scanner inside the digital multimeter must be measured by auxiliary experiments before programming. In spite of these strong demands for the timing control, the timer function supported by Visual Basic is capable to trigger an event up to only about 1 min. For a long term control, counting the number of events is necessary. Although such difficulties were encountered on programming, the resultant code works satisfactorily with a good graphical user interface shown in **Fig.3** for example. The results of measurement will be published elsewhere.[3,4)]

As described already, the cryostat is applicable to many types of measurements of physical properties while keeping its thermal stability if an appropriate sample stage is set. The stages for dielectric measurements up to some MHz and for relaxation calorimetry are in preparation.

The final plan of the cryostat was drawn and machined by JECC Torisha Co. Ltd., in accordance with the original design by the present authors. All wiring including winding the heaters and mounting thermometers and thermocouples were made by the present authors. The authors sincerely express their thanks to Mr. T. Hemmi at JECC Torisha for his technical cooperation and Mr. Y. Tozuka at this laboratory for his assistance.

References

1) Y. Yamamura, K. Saito, H. Saitoh, H. Matsuyama, K. Kikuchi and I. Ikemoto, *J. Phys. Chem. Solids* **56**, 107 (1995).

2) I. Hatta and A.J. Ikushima, *Jpn. J. Appl. Phys.* **20**, 1995 (1981).

3) K. Saito, Y. Yamamura, H. Akutsu, M. Takeda, H. Asaoka, H. Nishikawa, I. Ikemoto and M. Sorai, submitted to *J. Phys. Soc. Japan*.

4) H. Akutsu, K. Saito, Y. Yamamura, H. Nishikawa, K. Kikuchi, I. Ikemoto and M. Sorai, in preparation.

要 旨

室温以下で作動する光照射型acカロリメータを製作した。良好な温度制御を実現するために，断熱型熱量計のクライオスタットを改良したクライオスタットをもちいた。光は離れたところでチョップして光ファイバーでクライオスタットへ導入し，クライオスタット内では石英ガラス棒を光路とした．製作したクライオスタットは，良好な熱的安定性を保ったまま多くの物性測定に応用できる。