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Improved sample-holder design for specific-heat measurements in magnetic fields up to 17.5 T

J. C. P. Klaasse,^{a)} I. H. Hagmusa, and E. Brück

Van der Waals-Zeeman Institute, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

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By the use of a gold-plated silver sample holder, and a RuO₂-Cernox combination thermometer, low-temperature specific-heat measurements in the temperature range from 0.35 to 90 K, and in magnetic fields up to 17.5 T, have become possible with remarkably improved accuracy. At 500 mK, the lower limit for the heat capacity that can be measured amounts to 0.02 mJ/K. The sensitivity in that case is about 0.3 μ J/K. The error margin is 1% for large heat capacities (larger than twice the sample-holder contribution) at higher temperatures, increasing to 3% for small heat capacities (half of the sample-holder contribution) around 500 mK. In high fields, for accurate measurements (error <3%) the temperature range is restricted to above 0.5 K. © 1997 American Institute of Physics. [S0034-6748(97)02611-7]

I. INTRODUCTION

Recently, we have reported on newly developed specific-heat equipment in a 17.5 T superconducting magnet, working with the semiadiabatic technique in the temperature range between 0.5 and 60 K, using a mechanical heat switch.¹ This equipment performed well, but, on a few points, improvements were recommended, in particular with respect to (a) the field dependence of the thermometer, and (b) the nuclear contribution to the heat capacity from the copper sample holder.

One of the main problems encountered was the high, negative magnetoresistance at low temperatures of the temperature sensor (Lake Shore Cernox type 1010) which had been used. Below 1 K the strong dependence on the magnetic field B resulted in a remarkable loss of sensitivity: up to about a factor of 2 in 16 T. Therefore, accurate interpolations were not possible (the sensors were calibrated in fields of 0, 4, 8, 12, and 16 T). Furthermore, the low values of the resistance (of the order of 1 k Ω at the lowest temperatures and in high fields) permitted only small excitation voltages on the resistance bridge.

The other important problem was the high nuclear contribution to the heat capacity from the copper of the sample holder, which at 500 mK and in 16 T, was more than one order of magnitude larger than the zero-field contribution, and which increases strongly with decreasing temperature.

In this article, we present the results obtained with a new sample holder made of cold-worked silver, and equipped with a newly developed combination thermometer with a lower magnetoresistance.

II. IMPLEMENTED MODIFICATIONS

A well known alternative for the Cernox temperature sensor is the use of a RuO₂ chip resistor. At low temperatures, these resistors have a low magnetoresistance, in combination with an acceptable sensitivity, S , which is defined by

$$S = d \ln(R) / d \ln(T).$$

At high temperatures, however, the sensitivity of the RuO₂ sensor drops dramatically, resulting in a field dependence of the temperature readings which even increases with increasing temperature.

Therefore, a new sensor was developed, consisting of two resistors in a parallel circuit. One resistor is a RuO₂ chip, and the other is a dedicated Cernox sensor, type 1070. The advantage of using one circuit instead of two separate sensors is that only one set of connecting wires and one thermal anchor are needed. Moreover, switching between different thermometer circuits often causes discontinuities in the resulting specific-heat curves. Nominal resistance values for these sensors at the various temperatures are given in Table I.

At high temperatures, the behavior of the combination is dominated by the Cernox sensor, resulting in a high sensitivity (about -1), whereas at low temperatures the RuO₂ sensor is dominant, resulting in a low magnetoresistance. If we define $T(R, B)$ as the temperature corresponding to a value of R in a field B , then, for 16 T, the absolute value of

$$dT/T = [T(R, 0) - T(R, 16)] / T(R, 0)$$

never exceeds 10%, and rapidly decreases with increasing temperature. With these characteristics it is very possible to interpolate between the calibrated field values for B different from 0, 4, 8, 12, or 16 T, without significant loss of accuracy. The interpolations are performed with a second-order (below 4 T and above 12 T) or third-order polynomial (between 4 and 12 T) through the three or four adjacent calibrated $T(R, B)$ values.

Figures 1 and 2 plot the temperature dependence of the sensitivity S in 0 and 16 T, and dT/T in 16 T, respectively, for this new combination thermometer. As can be observed in Fig. 1, the sensitivity is almost independent of the applied magnetic field.

We attached this combination sensor to a new sample holder made of gold-plated, cold-worked silver. Silver has a low nuclear moment compared to that of copper,² resulting in a remarkable shift of the nuclear Schottky peak to lower

^{a)}Corresponding author; Electronic mail: klaasse@phys.uva.nl

TABLE I. Nominal values of the resistance in $k\Omega$ of the RuO_2 and Cernox resistors used in the combination sensor, and the resulting resistance in a parallel circuit, at various temperatures.

T :	300 K	77 K	10 K	4 K	0.3 K
RuO_2	6.2	6.5	8	9	20
Cernox (1070)	0.065	0.3	5	16	>500
Combination	0.065	0.3	3	6	19

temperatures; the high-temperature tail of the nuclear specific-heat contribution amounts, in the same magnetic field, to about 1/2% of the copper values. At 500 mK and in 16 T, the nuclear contribution to the heat capacity of this holder exceeds that of the holder in 0 T by no more than 30%. This means that it is sufficient to determine the contribution of the empty holder at the same set of fields as for the thermometer calibration, and that, in case of a noncalibrated field value, interpolation procedures for the empty-holder contribution can be performed, without loss of accuracy, just as for the temperature readings.

Moreover, the cryogenic conditions have been improved by:

- (a) reducing electronic magnetic induction noise by improved electronic shielding,
- (b) reducing heat input by improved thermal shielding, and
- (c) heating the cryopump up to 45 K (instead of 40 K) while condensing the 3He , resulting in higher cooling power after switching off the cryopump.

The copper wire connecting the sample holder to the clamp of the heat switch was replaced by a gold-plated silver wire, resulting in improved switch properties. With the heat switch closed, higher cooling rates are observed (numbers between parentheses are from Ref. 1): 40 (25) μW at 2.0 K, 6.5 (2.0) μW at 1.0 K, and 0.6 (0.07) μW at 500 mK. The sample holder can now be cooled to well below 320 mK (this was

FIG. 1. The sensitivity, $S = d \ln(R) / d \ln(T)$ vs temperature, for the combination thermometer consisting of a Cernox sensor, type 1070, in parallel with a ruthenium-oxide resistor.

FIG. 2. Relative deviations (in percent) of the temperature readings for the combination thermometer in a field of 16 T from the readings in 0 T at the same resistance, as a function of temperature.

340 mK, and the equilibrium temperature with the switch open can be brought below 600 mK (this was 850 mK). This means that the resulting heat input with the switch open at 500 mK is reduced to values well below 0.01 μW . The lowest point measured, so far, is at 340 mK; the resulting heat input at this temperature is about 0.03 μW . At the high-temperature side, the range is extended to 90 K.

The modifications listed above resulted in remarkably improved specifications. For instance, we are able to measure smaller heat capacities, leading to more accurate values for the empty-holder contribution, which amounts to less than 20 $\mu J/K$ at 500 mK. Unfortunately, releasing the heat switch still generates an amount of heat of about 3 μJ (somewhat less by step-wise opening). For this reason, the empty-holder measurement is restricted to temperatures above 500 mK. Values below 500 mK have to be determined by extrapolation, which is no problem in low magnetic fields, but generates uncertainties in high fields because of the nuclear contributions. These nuclear upturns are still present, but strongly reduced compared to the heat capacity of the copper sample holder.

III. EVALUATION

We tested the accuracy of the modified equipment with the help of two copper (4N5 purity) samples of 3 and 11 g, an iron (5N purity) sample of 3 g, a platinum (4N purity) sample of 8 g. and a 1.3 g sample of the intermetallic compound UPt. The specific heat of the pure materials is well known from the literature.³⁻⁵

From these tests we could trace, in particular below 3 K, some systematic errors of a few percent (up to 5% below 1 K) in our specific-heat data, obviously due to errors in the thermometer calibration. Because the specific-heat data depend directly on the derivative of the temperature function, only small errors (in our case of the order of a few mK) in these functions account for the observed deviations. Because of the uncertainties in the deviations, it is not possible to

FIG. 3. Relative deviations (in percent) of the raw copper data (11 g sample) from literature values (see Ref. 3) as a function of temperature for two different runs on two different days. The oscillations are ascribed to thermometer-calibration errors.

determine a new, sufficiently accurate, thermometer function by some numerical integration procedure on these deviations. Therefore, we have developed a correction procedure on the data, implemented in the computer routines that control the measurement.

As an example, we present in Fig. 3 the difference between the raw data on the 11 g copper sample in 0 T (two runs on two different days) and the literature values. Above 3 K, this difference is, in general, not more than 1%, whereas at lower temperatures some oscillations (characteristic for errors in thermometer functions) are detectable. The curves for the 3 g copper sample showed roughly the same behavior, although the scatter was somewhat larger, and the obtained specific-heat values between 3 and 25 K were systematically somewhat lower (up to about 1.5%) than those for the 11 g sample.

To test the high-field curves at low temperatures we didn't use copper. We were not sufficiently sure about the accuracy of the nuclear specific-heat contribution of copper because of the rather long relaxation times for thermal equilibrium between the lattice and the nuclear system.⁶ Moreover, the nuclear contribution is strongly dependent on temperature, resulting in two parameters in the correction procedure, the temperature and its derivative, the coupling between which is not straightforward. Therefore, we used pure iron samples. Iron has a very low nuclear contribution,² and the contribution from spin waves is small at low temperatures.

In the temperature range from 0.5 to 3 K, we found in all our test fields oscillations in the observed heat capacity for iron of the same order of magnitude as for copper in 0 T. Below 0.5 K, however, we found, in particular in high fields, upturns for which the origin is not yet fully understood. For this reason, the range of reliable results is limited to above 0.5 K in the highest magnetic fields.

After correcting these errors, small deviations from the

literature values remained due to the temperature drift during the measurements. This drift, however, depends on the heat capacity of the sample, and on the detailed cryogenic conditions, and is, therefore, not reproducible. Correction for these errors, which amount to about 3% for the smallest samples at low temperatures, is for this reason not possible. Moreover, also the empty-holder contributions are affected by the same errors. A complicating point is the evidence we have from a computer simulation of the measuring process, that these errors are not linearly dependent on the drift rate.

It will be clear that the drift rate is proportional to $1/C$, with C the total heat capacity of the sample plus holder. Let C_s be the sample heat capacity, C_h that of the holder, C_{obs} the observed value of the total, and $C_{h,obs}$ that of the holder. If the relative error linearly depends on the drift rate, then, with D_1 some arbitrary constant:

$$C_{obs} = (C_s + C_h) \cdot [1 + D_1 / (C_s + C_h)],$$

$$C_{h,obs} = C_h \cdot (1 + D_1 / C_h).$$

The difference between the two expressions results in exactly C_s , the correct sample heat capacity. This means that, in the case of a linear dependency, subtracting the observed empty-holder contribution from the observed heat capacity should roughly eliminate the drift error. From the simulation, however, we learned that the deviations depend roughly quadratically on the drift rate. This means that we have to write with D_2 again some arbitrary constant:

$$C_{obs} = (C_s + C_h) \cdot [1 + D_2^2 / (C_s + C_h)^2],$$

$$C_{h,obs} = C_h \cdot (1 + D_2^2 / C_h^2).$$

The difference is now $C_s \cdot \{1 - D_2^2 / [C_h \cdot (C_s + C_h)]\}$. From the simulation we learned that, because of the temperature drift, the observed heat capacities are indeed in general too high. Correcting the observed total heat capacities for the as-measured empty-holder values, results, as the formulas show, in too low values for the sample heat capacity, just as observed in Fig. 3. These errors are larger in the case of smaller heat capacities, but they never exceed 3% for the examined test samples.

Our conclusion from the copper and iron test measurements is, that after correcting the data for the oscillatory behavior, we have for large heat capacities, i.e., more than twice the empty-holder contribution, an accuracy of 1% above 3 K, and 2% below 3 K. For small heat capacities, i.e., of the order of half the empty-holder contribution, these numbers are 1% above 25 K, 2% between 3 and 25 K, and 3% between 0.5 and 3 K. For still smaller samples the error margins increase considerably. The smallest difference in heat capacity that significantly can be detected with this new holder amounts to about $0.3 \mu\text{J/K}$ at the lowest temperatures.

In high fields, the same numbers hold for temperatures above 0.5 K. Below this temperature, we have to be very cautious about the results in high fields because of less reliable data for the thermometer and the empty holder, in combination with unknown nuclear contributions and temperature drifts. In particular, the nuclear contributions give rise to problems. Thermal equilibrium between the nuclear system and the phonons is not guaranteed.⁶ For insulators, this is the

FIG. 4. Relative deviations (in percent) of the observed platinum specific heat from the literature values (see Ref. 5) in 0 T as a function of temperature, after implementation of the correction procedures for the thermometer-calibration errors. The upturn at low temperatures is ascribed to impurities, the negative deviations between 2 and 10 K to temperature drift errors.

reason that the nuclear contributions are not seen at all on the time scale of this specific-heat measurement. Although aluminum has a large nuclear contribution (about twice that of copper) no nuclear contribution is reported for sapphire (consisting of alumina) sample holders. For metallic systems, the hyperfine interaction thermally couples the nuclear system to the conduction electrons. The time constant for this coupling, however, is proportional to $1/T$, and can amount to several seconds (sometimes much more) at 0.3 K. Furthermore, strong molecular fields can obscure the picture. For all these reasons, results in high magnetic fields at the lowest temperatures are still the subject of further studies.

In Fig. 4, we present some test results on a sample of 8 g platinum (4N purity) in 0 T, and in Fig. 5 in fields of 5, 10, and 15 T, after implementation of the correction procedures. The chosen fields are different from the calibrated fields. The upturn at lower temperatures observed in the 0 T results is ascribed to impurities. It is well known that the low-temperature heat capacity of platinum is very sensitive to impurities in the sample.^{5,7} A measurement on a 3N pure sample resulted in a much higher upturn. Moreover, the results on the intermetallic compound UPt, which has a high value of the electronic-specific-heat coefficient of about $105 \text{ mJ/K}^2 \text{ mol}$, and therefore, has to show a nearly constant c/T value below 2 K, did not exceed the 2% claim. The observed difference between 3 and 20 K in the results of platinum in 0 T with the literature values can be explained if we realize that at high temperatures the heat capacity, being dominated by phonons, is relatively small compared to the empty-holder contribution. At 5 K, the contribution of the platinum sample amounts to no more than 1.5 times the sample-holder contribution. Moreover, the accuracy of the literature values at low temperatures is not claimed to be better than 1%.⁵

FIG. 5. Relative deviations (in percent) of the observed platinum specific heat from the literature values (see Ref. 5) in magnetic fields of 5, 10, and 15 T as a function of temperature, after implementation of the correction procedures for the thermometer-calibration errors. The nuclear contribution, calculated from nuclear data (Ref. 2), is included in the literature values. The errors show to be smaller than 2%.

In high fields, the impurity contribution in platinum is largely suppressed, and independent of the applied field, as is clearly shown in Fig. 5. In this case, the “literature” values also include the nuclear contribution of platinum, which in our region of interest, never exceeds 5% of the electronic contribution.

Our conclusion is that these test results on platinum underline the above claims with respect to the accuracy of the equipment. From the data it is obviously clear that our statements about the accuracy hold, also in the case of interpolated field values.

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