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Magnetic and Thermometric Properties of $Na_3[Ce(C_7H_3NO_4)_3] \cdot 15H_2O$ in the Millikelvin Range*

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Measurements of the 16-Hz susceptibility of a powdered $Na_3[Ce(C_7H_3NO_4)_3] \cdot 15H_2O$ (CDP) sample, in the shape of a right-circular cylinder with diameter equal to height, have been made as a function of the Johnson noise temperature T_J of a resistor in the range 2 to 20 mK. Pure liquid ^3He was used for thermal contact between the CDP and the noise thermometer in a demagnetization cell. The results are compared with similar data for cerium magnesium nitrate (CMN). CDP appears, like CMN, to have an anomalously low thermal resistance to pure liquid ^3He.

In a recent Letter Doran, Erich, and Wolf[1] introduced the material $Na_3[Ce(C_7H_3NO_4)_3] \cdot 15\ H_2O$, abbreviated CDP, as a possible alternative to cerous magnesium nitrate (CMN) for millikelvin thermometry. CDP has both a smaller interaction specific-heat constant CT^2/R and a smaller Curie constant than CMN and, according to Ref. 1, CDP reached a lower magnetic temperature on demagnetization than CMN. In order to utilize any superior properties of CDP, it is essential to make thermal contact to it. This is usually achieved with CMN by finely powdering it and mixing it with pure ^3He, in which case a magnetic coupling[2,3] is responsible for the low-temperature surface contact. CDP is less stable than CMN, and hence fine powdering might be a problem (no protective oils may be used when good thermal contact at low temperatures is desired). Further, it was not known whether the excellent thermal coupling possible with CMN is possible with CDP as well. In recent measurements we and Giffard[4] studied the thermometric properties of powdered CMN using a noise thermometer as a standard. In the present work we have measured the 16-Hz susceptibility (both χ' and χ'') of CDP against the same noise thermometer in the same experimental arrangement using pure ^3He as the thermal contact agent over the noise temperature range from 2.2 to 20 mK. Hence we can measure the magnetic and thermometric properties of powdered CDP and then compare them with those obtained for CMN.

The demagnetization cell used to obtain the measurements is shown in Fig. 1. It contained 10.7 g powdered CMN as cooling salt and 2.7 cm^3 liquid ^3He at low pressure. The noise thermometer and CDP sample are magnetically decoupled from the cooling salt by a niobium shield but thermally connected to it via a column of pure ^3He.

Although noise thermometry based on a Josephson-effect device is a relatively new technique, it has been used by Kamper *et al.*[5] down to 23 mK and by Giffard and us[4] down to 2.2 mK. We use an improved version of a method described earlier.[6] The noise-temperature sensor was fabricated from copper foil 4 mm wide, 15 mm long, and 0.025 mm thick. Its resistance was observed in a separate experiment to change less than 0.4% from 3 mK to 4.2 K. The measured noise temperature is greater than the actual Johnson noise temperature T_J by an amount called the "device temperature" and determined in this experiment to be 0.05 mK. We believe that the noise temperature, corrected for device temperature, accurately reflects the temperature of the copper

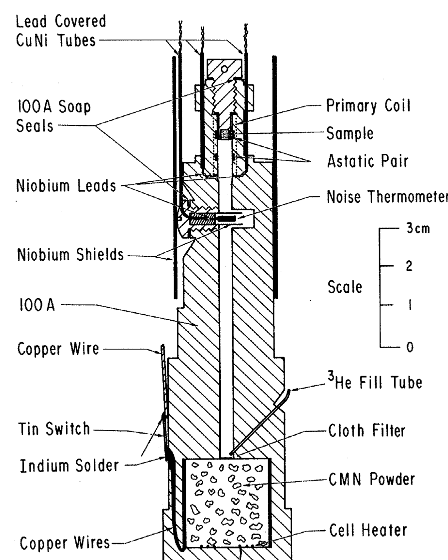

FIG. 1. Schematic diagram of the adiabatic demagnetization cell used for the comparative temperature measurements.

sensor, and we have argued elsewhere[4] that in our apparatus at 2.2 mK the copper noise sensor and the surrounding liquid ^{3}He differ in temperature by no more than 0 to 0.2 mK. The size of this correction, if any, should have no zero-order effect on our intercomparison of CMN and CDP. In equilibrium to obtain values of noise temperature with a precision conservatively estimated at $\pm 1\%$ we had to average the squared noise currents for a 2000-sec interval.

The CDP sample was ground from a single crystal kindly supplied us by Mr. S. Mroczkowski at the request of Professor W. P. Wolf. The crystal was initially stored in liquid paraffin oil, but was cleaned of oil using hexane and then powdered in a helium atmosphere at about 28°C. The powder used passed an NBS-400 sieve (particle size less than 37 μm). The sample had a mass of ca. 10 mg and was in the form of a right-circular cylinder with diameter equal to height. Both χ' and χ'' were measured at 16 Hz using an ac bridge having a superconducting quantum interference device as null detector.[6] The magnetic temperature scale T^* was obtained with an accuracy better than 1% in the range 0.4 to 4.2 K using a germanium resistance thermometer. In auxiliary experiments this thermometer had first been calibrated from 4.2 to 1.2 K against the 1958 ^{4}He vapor-pressure temperature scale. It was then used to calibrate a CMN thermometer in the above range, which in turn was employed to extend the resistance thermometer calibration down to 0.2 K. The absolute susceptibility χ' at high temperatures was taken to be that given by Doran, Erich, and Wolf[1] while χ'' was taken to be zero in that range.

The results of the CDP measurements are shown compositely in Fig. 2, in which $\ln T^*$, $\ln\chi'$, and $\ln 250\chi''$ are displayed as functions of $\ln T_J$ for two demagnetization runs performed over a 3-week interval. We found that the heat leak to our demagnetization cell decreased continuously with a time constant of about 2 weeks so that patience was necessary to achieve the longest warm-up times. There is no systematic deviation between runs even though the initial temperature after demagnetization and the residual heat leak were different for the two. At the lowest temperatures we observed a time constant for the noise thermometer of ca. 800 sec. This is small when compared to the 24.6 ksec the cell stayed below T^* $= 3$ mK. The Johnson noise temperatures T_J were obtained using 2000-sec averaging times below 11 mK and 4000-sec averaging times above 11 mK.

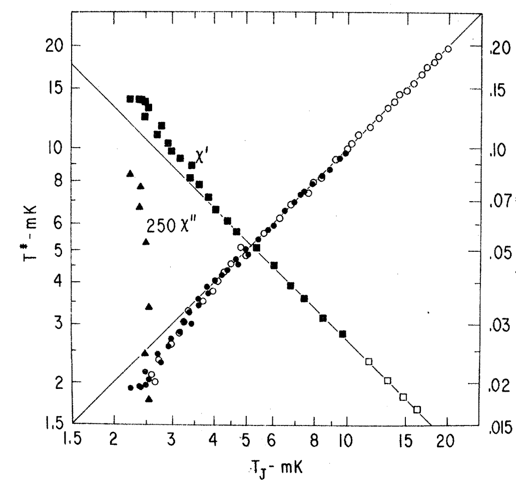

FIG. 2. Magnetic temperature and susceptibility of CDP plotted against T_J, the Johnson noise temperature. Open circles, magnetic temperature data taken after the first demagnetization; solid circles, similar data taken after the second demagnetization. Open and solid squares, 16-Hz in-phase susceptibility χ' taken after the first and second demagnetization, respectively; triangles, 250 times the out-of-phase susceptibility χ'' A 0.05-mK device noise has been subtracted from each T_J. The straight lines are $T_J = T^*$ and $\chi' = C/T_J$, respectively, where C is the Curie constant for powdered CDP given in Ref. 1 at crystalline density.

The thermometric properties of CDP are qualitatively similar to those for CMN.[4] In the range 4.3–19.7 mK we fitted the data with the equation $T_J = T^* + \Delta$ and found $\Delta = 0.10 \pm 0.05$ mK. It is known that for powders[7] variations of 0.1 mK are to be expected for Δ. Measurable deviations of $T^* + \Delta$ from T_J begin at a lower temperature for CDP than CMN, the former occuring at 3.5 mK and the latter at 5.5 mK. Below these temperatures, $T^* + \Delta$ for both powdered samples becomes less than T_J, and at $T_J = 2.4$ mK both have $T^* = 2.0$ mK.

At the lowest temperatures the two salts differ qualitatively, as may be seen on examination of the susceptibility data for CDP in Fig. 2 and for CMN in Fig. 3 taken under similar circumstances. In the case of CDP and for $T_J \lesssim 2.5$ mK the value of χ' becomes insensitive to temperature although χ'' continues to increase. This experimental point is somewhat obscured by plotting χ' (or T^*) versus T_J since the latter is subject to imprecision. In fact the low-temperature points for χ' versus T_J shown were obtained in a single run in which χ' either was constant or decreased monotonically with increasing time. Since χ'' did decrease substantially while χ' was nearly constant,

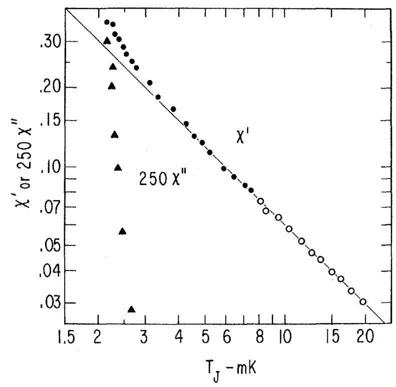

FIG. 3. The 16-Hz in-phase susceptibility χ' for two different demagnetizations (solid and open circles), and 250 times the 16-Hz out-of-phase susceptibility χ'' (triangles) for CMN plotted against T_J. A 0.05-mK device noise temperature has been subtracted from each T_J. The straight line is $\chi' = C/T_J$, where C is the Curie constant for powdered CMN at crystalline density given in Ref. 1.

we feel that the constancy of χ' did not reflect a loss of thermal contact to the ^{3}He. In contrast, both the susceptibilities χ' and χ'' for the CMN powder continue to increase steadily to the lowest temperatures. That the lowest value for T_J is the same within experimental error for both the CDP and CMN measurements increases our confidence in both measurements. Although measurements on powders in the nonellipsoidal shapes of the present samples complicates any interpretation of the magnetic ordering, it does appear that the CDP may be ordering at a higher temperature than the CMN, even though at higher temperatures deviations between T^* and T_J are less for CDP than for CMN.

As thermometers, powdered CDP and powdered CMN are not greatly different from one another, insensitivity of χ' to T at the lowest temperatures and smaller deviations from T at intermediate temperatures being, respectively, the disadvantage and advantage of CDP. It is not known at this time how the two compare as refrigerants nor which has the lower thermal resistance to pure ^{3}He. That we were able to make CDP measurements at 2 mK at all indicates qualitatively a low thermal resistance. We did attempt in crude measurements to detect a thermal time constant associated with the CDP coupling to liquid ^{3}He at 10 mK and found that this time was less than 10 sec. It is known from work of Bishop *et al.*[8] that an internal resistance becomes important in CMN in the low millikelvin range, so experiments planned in this laboratory on thermal coupling to CDP should answer this important question.

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