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Low-Temperature Susceptibility Measurements of Cerium Magnesium Nitrate

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Measurements in Zero Magnetic Field

The susceptibility of a cerium magnesium nitrate (CMN) single-crystal sphere, a powder sphere, and a powder 1×1 cm cylinder were measured in zero magnetic field as a function of specific entropy by adiabatically demagnetizing the CMN from a temperature in the vicinity of 1°K.

The procedure was to demagnetize the CMN from a high temperature, about 1°K, and then observe the ambient warmup of the salt so as to be able to extrapolate the susceptibility to $t = 0$, the time when the magnetic field vanished. The apparatus used was the same as that in Ref. 1.

In order to test the adiabaticity of this process, the samples were cycled several times. In the cycling process the sample was isolated with the magnetic field on, and then the field was reduced linearly to zero in a typical time of 30 min. Susceptibility measurements were then made for about 5 min and the field reapplied over a time span of about 30 min. The process was then repeated. During that time the sample remained isolated. Although we could measure the susceptibility to an accuracy of one part in 10^5, there was no evidence of nonadiabaticity in the process except that which could be attributed to the ambient heat leak. A generous limit can thus be placed on the error due to irreversible processes of about 2%.

At 1°K the magnetic susceptibility of CMN was assumed to be that of an assembly of noninteracting dipoles of spin 1/2,

$$S/R = \ln 2 \cosh (g\mu H/2kT) - (g\mu H/2kT) \tanh (g\mu H/2kT) \quad (1)$$

where the g factor for CMN in the direction of easy magnetization was taken to be 1.84. This assumes that g is a constant and does not change with the applied magnetic

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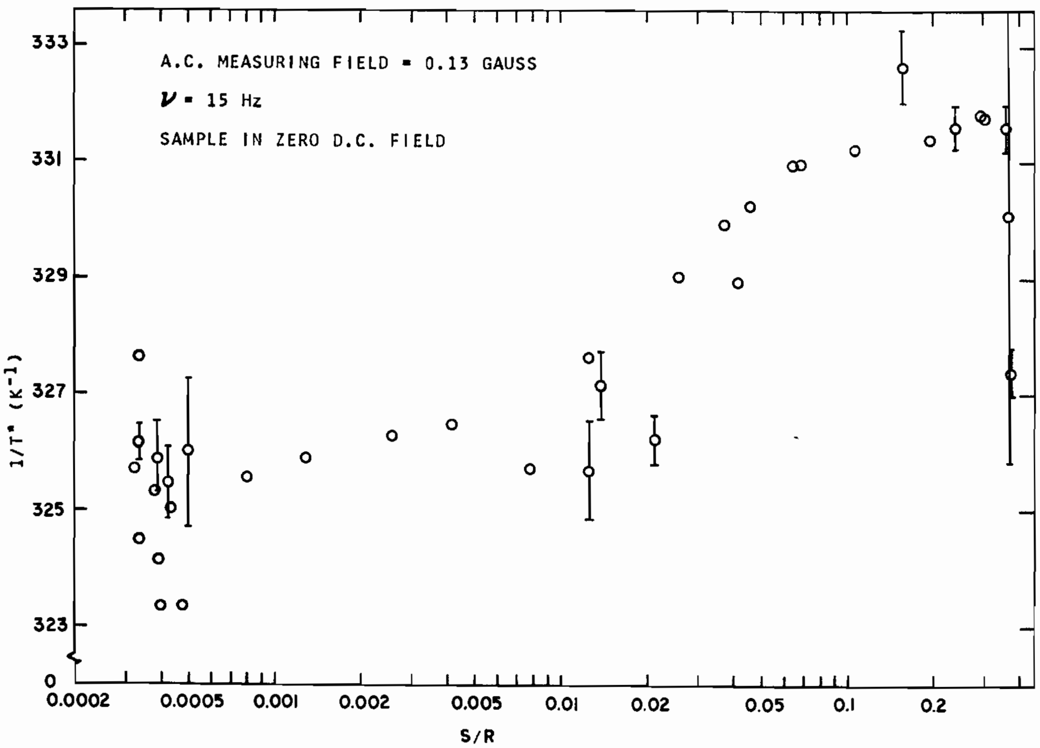

Fig. 1. Low-entropy susceptibility of a single-crystal sphere of CMN in terms of $1/T^* = \chi/C$.

field. This assumption might not be valid in high magnetic fields,[2] where it might be in error by as much as 5 %. The highest magnetic fields used in this investigation were 10^5 Oe. The correction due to the change in the g factor will not be made here, in order to facilitate the comparison of these data with others.

Results of measurements in the low-entropy region on a single-crystal sphere of CMN along the easy magnetization axis are shown in Fig. 1. The data were taken over a period of several months and the reproducibility increased our confidence in this method. The errors are due mainly to the extrapolation of the susceptibility to $t = 0$. One of the failings of our data is that because of apparatus design, our highest entropy was $S/R = 0.55$, which corresponds to a T^* of about 4.5 m°K. The lack of high-entropy data in our experiment presented a problem because the two complete measurements at high entropies, that of Hudson and Kaeser[3] and that of Mess *et al.*,[4] disagree with each other over the entire region of their measurement, and our highest entropy data fall somewhere between the two.

Two least-square fits (to an eight-order polynomial) were then made to our data anchored at high entropy to those of Refs. 3 and 4, respectively. The equation which was fitted is given by

$$\chi/C = \sum_{n=0}^{8} a_n (S/R)^n \tag{2}$$

Table I. Coefficients of Eq. (2) with High Entropy Fitted to Data of Refs. 3 and 4

	Hudson and Kaeser[3]	Mess *et al.*[4]
a_0	3.2521×10^2	3.2536×10^2
a_1	1.5766×10^2	7.4360×10^1
a_2	-1.5138×10^3	1.8108×10^3
a_3	9.5842×10^3	-3.5022×10^4
a_4	-5.9586×10^4	2.1666×10^5
a_5	2.8350×10^5	-6.1015×10^5
a_6	-7.4956×10^5	8.0583×10^5
a_7	9.4740×10^5	-4.2777×10^5
a_8	-4.5370×10^5	2.9397×10^4
RMS residual	0.90	3.858
Maximum deviation	2.60	20.66

with the coefficients and deviations given in Table I. From the standard deviations it can be seen that our data are in somewhat better agreement with those of Ref. 3 than with those of Ref. 4. On the other hand, it should be noted that the least-squares fit is mainly a low-entropy approximation of the data since that is where most of our points were taken, and that at $S = R \ln 2$ the magnetic susceptibility is $\chi/C = 40$, about 13 % of the susceptibility at the maximum.

A similar method to the one above was used in the measurements on the powdered sphere and the powdered cylinder. The powdered sphere, with a diameter of 2 cm, and the powdered cylinder, with diameter and length both equal to 2.54 cm, consisted of ground CMN powder compacted into the respectively shaped nylon containers. In order to increase the thermal bonding between the powder grains, a drop of water was added to each container and subsequently pumped off.

The powder entropy at high temperatures can be computed in two ways, each yielding a different result. In the first instance one can assume an average $g = \frac{2}{3}(1.84)$ and substitute it into (1). On the other hand, if one assumes that the crystallographic axes of the powder grains are distributed uniformly over a sphere, one has

$$S/R = \int_0^{\pi/2} [\ln 2 \cosh (x \sin \theta) - x \sin \theta \tanh (x \sin \theta)] \sin \theta \, d\theta \qquad (3)$$

where $x = g_{\perp} \mu H/kT$. In both cases $g_{\parallel}$ is assumed to be zero. The error of this assumption is assumed to be negligible since $g_{\parallel}$ is of the order of 0.03.

The results of this measurement are given in Fig. 2. The open points are those computed using an average g, while the closed points are those computed by means of Eq. (3). For comparison the single-crystal data are also shown. One can see that at the lowest entropies the single-crystal data and those of the cylinder are in excellent agreement, while those of the powder sphere have a significantly higher $T^* = C/\chi$. At higher entropies the powder cylinder exhibits values which are intermediate between those of the single-crystal sphere and those of the powder sphere.

From Fig. 2 one can compare the T^* of the single-crystal sphere, the powder sphere, and the powder cylinder at constant entropy. This is done in Table II. $T^*_{cs} = 4.5$ m°K is the highest temperature at which we could obtain reliable S/R

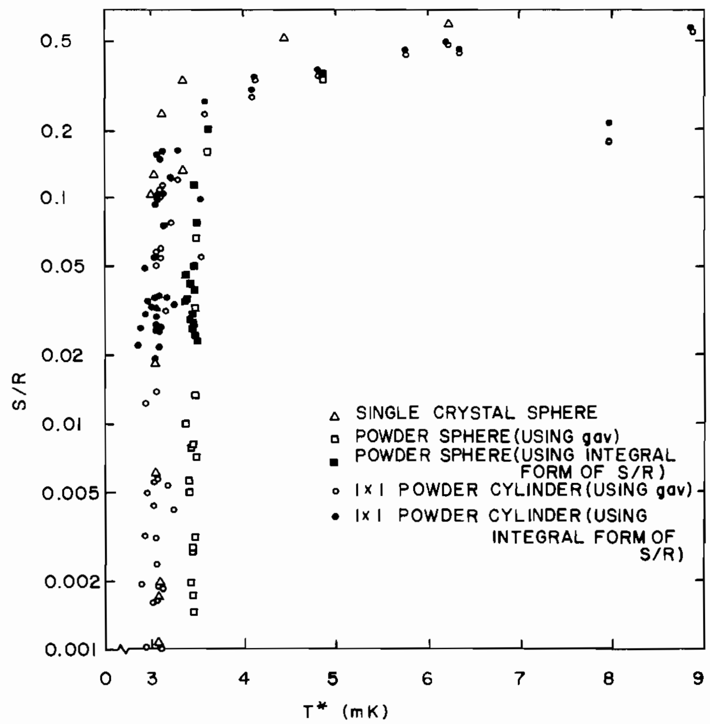

Fig. 2. S/R as a function of T^* for the single-crystal sphere (△), powder sphere (□), and 1 × 1 cm powder cylinder (○). Solid symbols use integral form.

data. One sees that at low temperatures the T^* of the crystal sphere and of the 1 × 1 cm cylinder agree. If we take the single-crystal T^* to be the true temperature, a dubious assumption, then at the lowest T^* the powder cylinder should give a true temperature. At higher temperatures there is a disagreement of as much as 2 m°K, which does contradict the additional 1.7 m°K of Abraham and Eckstein.[5] We thus add another set of data to "the delta campaign."[6]

Measurements in a Magnetic Field

The above measurements were carried out in zero magnetic field with special coils compensating for the ambient field at the apparatus. The reason for this precaution was the observation that the magnetic susceptibility changes markedly in all the temperature regions when a small magnetic field is applied. In order to investigate this phenomenon further, we measured the effect of an externally applied magnetic field on the susceptibility of a single-crystal sphere. Although measurements were made with the field applied both parallel and normal to the measuring field, both of which were normal to the c axis, we shall here report only on the case of the applied field parallel to the measuring field. The results for the other case were similar.

In this measurement the CMN was demagnetized to zero magnetic field and measurements of the susceptibility were made as a function of time as the sample warmed due to ambient heating. Different external magnetic fields were then applied

Table II. Comparison of T^* Values for Crystal Sphere (cs), Powder Sphere (ps), and Powder Cylinder (pc)

T^*_{cs}, m°K	T^*_{ps}, m°K	T^*_{pc}, m°K
3.00	3.50	3.05
3.05	3.55	3.10
3.10	3.60	3.40
3.20	4.10	3.90
3.50	5.00	4.80
4.00	5.75	5.75
4.50	6.50	6.50

adiabatically and the susceptibility in those fields measured. Smooth curves were then drawn through the points at each value of the externally applied field. At each particular time (consequently, constant entropy) the magnetic susceptibility as a function of the applied magnetic field obeyed the equation

$$\chi(H, S)/C = [\chi(0, S)/C] - A(S)\, H^{\eta(S)} \tag{4}$$

where the left-hand side is the magnetic susceptibility in an applied field, the first term on the right is the susceptibility in zero field at that entropy, and A and η are functions dependent on the entropy or susceptibility in zero field. These are shown as a function of T^* for values above the minimum in Fig. 3.

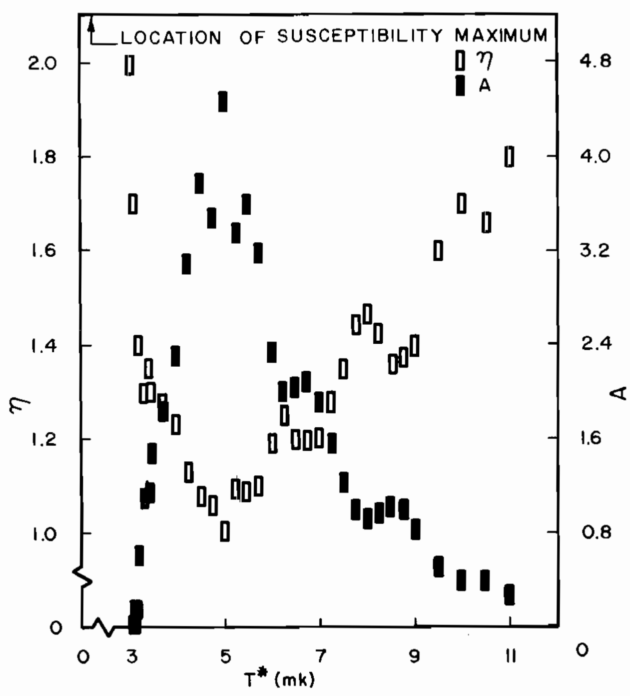

Fig. 3. Exponent η and coefficient A of Eq. (4) as a function of T^*.

As one can see from Fig. 3, A has a maximum at $T^* = 5$ m°K and becomes small, although never zero, at both high and low temperatures. The function η has a minimum of about 1 at that temperature and approaches the classical value of 2 at both high temperature and at the susceptibility maximum. These results are surprising since one would have expected either the value of 2, the classical value, or the value of $-1/5$ or $-1/4$, the scaling law value, for the exponent. Moreover, one would not expect there to be a minimum in the exponent at that high a temperature, a temperature of about 5 m°K as compared with that of about 1 m°K for the susceptibility maximum. We have not been able to come up with a plausible explanation for this behavior since most theories which treat the influence of an applied magnetic field on susceptibility obtain an H^2 dependence.

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