ing of the antiferromagnet $MnBr_2 \cdot 4H_2O$. They reported $\Delta T \cong -0.125$°K from $T_f = 1.75$°K. K. P. Belov, E. V. Talalaeva, L. A. Chernikova, and V. I. Ivanovskii, Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt. 7, 331 (1968), have recently observed $\Delta T \cong -0.05$°K from $T_f \cong 290$°K in gadolinium iron garnet.

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LOW-TEMPERATURE PHASE TRANSITION IN CERIUM MAGNESIUM NITRATE

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Magnetic-susceptibility measurements on cerium magnesium nitrate covering frequencies from dc to 200 Hz indicate that it undergoes an antiferromagnetic phase transition in the millikelvin range.

We report some recent observations on the low-temperature magnetic phase transition in cerium magnesium nitrate [$Ce_2Mg_3(NO_3)_{12} \cdot 24H_2O$, commonly abbreviated CMN] which lead to a new interpretation of the nature of the transition. Earlier reports on this transition were made by Mess et al.[1,2] and by us,[3] but there remained some doubt as to whether it was a ferromagnetic or an antiferromagnetic transition.[4] Our most recent measurements strongly suggest that it is in fact antiferromagnetic rather than ferromagnetic as suggested by Mess et al. One is especially interested in this point because the CMN transition occurs at a lower temperature than any heretofore observed, with the exception of nuclear magnetic transitions. Furthermore, CMN is widely used as a thermometer at temperatures as low as 1.8 mK,[5,6] which is within the transition region, and consequently the details of the transition bear directly on the thermometry problem.

We have investigated the transition by means of measurements of the susceptibility in the frequency range 0-200 Hz as a function of time (as the sample warmed up following demagnetization) and of the initial entropy. The measurements reported here were performed on single-crystal spheres of 20 mm diam. A most striking feature of these measurements is the observation that the susceptibility goes through a maximum as the sample warms up. This was true over the entire frequency range from dc to 200 Hz provided that the initial entropy $S/R < 0.1$. Similar results were obtained with powder spheres and powder cylinders (of diameter and length both equal to 24 mm).

The same effect was exhibited in another type of measurement. If the salt is adiabatically demagnetized from different initial magnetic fields, and hence at different but constant entropies, the susceptibility attained when the field reaches zero also shows a maximum when plotted against the entropy.

The powder samples were compacted by adding a little water to them at room temperature and then cryopumping off the excess. This resulted in a solid compacted cylinder in which the individual grains were cemented together. The crystal spheres were made from one or two large single crystals. The sample was cooled by single-stage adiabatic demagnetization from helium temperatures (1-1.2 K) and initial fields up to 94 kG. A guard salt of chrome potassium alum was mounted above the sample and thermally isolated from it. A "coil-foil"[7] heat shield, thermally bonded to the guard salt, surrounded the sample but was not in thermal contact with it. A set of coils and a mutual-inductance bridge[8] were used to measure the in-phase (χ') and out-of-phase (χ'') components of the susceptibility in the frequency range between 1.5 and 200 Hz. At lower frequencies, between 0.08 and 1.5 Hz, the amplified and partially balanced signal from the secondary coils was displayed on a two-pen recorder together with the primary signal. The dc measurements were made with an integrating amplifier magnetometer (Magnemetrics Type MF-1) and the susceptibilities obtained from the initial

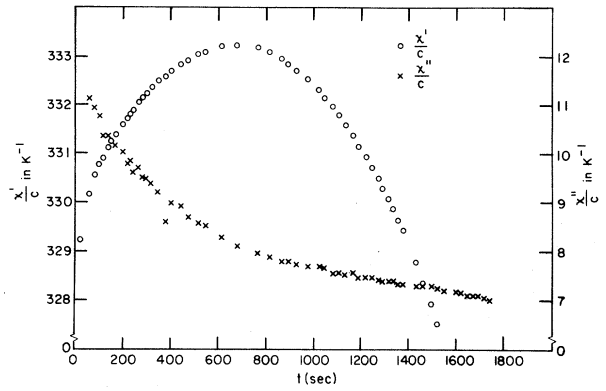

FIG. 1. Magnetic susceptibility as a function of time after demagnetization at a frequency of 15 Hz in an ambient field of 2.6 G. χ' is the in-phase component, χ'', the out-of-phase component, and C, the Curie constant.

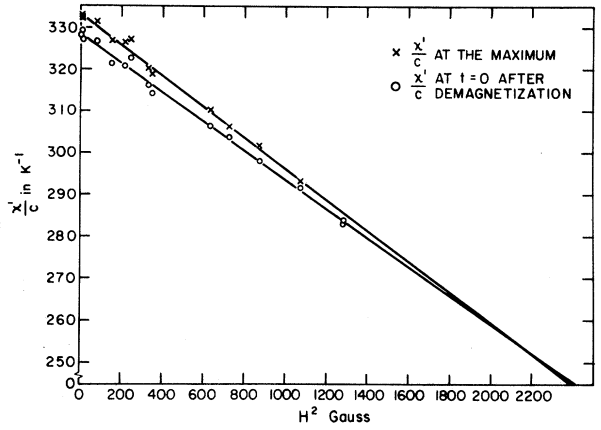

FIG. 2. The location of susceptibility maxima (crosses) and initial points (open circles) as a function of H^2. $S/R = 0.004$ and frequency = 35 Hz.

slopes of the magnetization curves.

In Fig. 1 we show the magnetic susceptibility of a single-crystal sphere normal to the c axis, measured at 15 Hz, as a function of time measured from the instant of complete demagnetization from an initial entropy of $S/R = 0.004$. Similar data were obtained at all the other frequencies from dc to 200 Hz. The size of the maximum defined as $[\chi_{max}'-\chi'(t=0)]/\chi_{max}'$ is about 0.015. At frequencies below 1.5 Hz it increases slightly. Inasmuch as the average sample temperature must increase monotonically with time, there must be a maximum in the in-phase component of the susceptibility as a function of temperature. These observations do not agree with those of Mess et al.[1,2] who reported no maximum in susceptibility when measured by the dc ballistic technique. Below 1.5 K the out-of-phase component is a monotonically decreasing function of temperature, at least in the frequency range between 1.5 and 200 Hz. Our apparatus was not suitable for measuring the two components separately at frequencies lower than 1.5 Hz.

When an external dc field is applied parallel to the ac measuring field, both the observed susceptibility maximum and the initial susceptibility are decreased, as shown in Fig. 2. Both are distinct linear functions of H,[2] where H is the externally applied dc magnetic field. (The leading term in the field dependence should go as H^2 inasmuch as χ' is an even function of H.[9]) The intersection of these two straight lines determines a critical field H_c above which no maximum is observed. The observed value, $H_c = 45 \pm 4$ G at an initial entropy of $S/R = 0.004$, agrees fairly well with the internal field calculated from the specific heat[10] at constant magnetization, i.e., 40 ± 3 G.

In addition we have studied magnetic saturation at temperatures below and above that of the susceptibility maximum. The change in magnetization when an external field is applied is $\Delta M = \int_0^H \chi' dH$. In Fig. 3 we show $\Delta M/C$, where C is the Curie constant, at a temperature below that of the susceptibility maximum, as a function of H. At $T = 0$ the saturation value of $M/C = 2k_B/g\mu$, where k_B, μ, and g are the Boltzmann constant, the Bohr magneton, and the g factor, respectively. Assuming that $g = 1.84$, M/C is equal to 16.19×10^3 G/K. Our experimentally determined $\Delta M/C$, taken below the maximum at an $S/R \cong 0.004$, falls within 4% of the calculated saturation value, which is less than the combined uncertainty in the g value and in our measurements. At this value

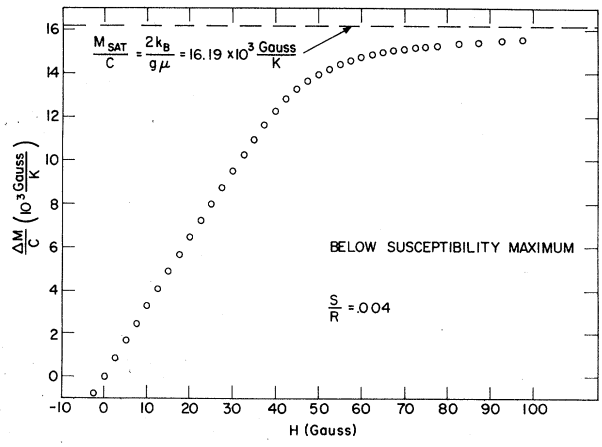

FIG. 3. Change in magnetization as a function of an applied magnetic field. The points were taken at $T^* = C/\chi' = 3.049$ mK at a frequency of 10 Hz. The dashed line denotes saturation magnetization.

of the entropy substantially the saturation magnetization should be attained. We conclude therefore that the initial magnetization (in zero field) was $\cong 0$. This fact implies that no remanent magnetization exists contrary to that expected in the case of ferromagnetism.

As a further check we subjected the sample to cyclic magnetizations and demagnetizations with a peak field of ± 10 G in the temperature region below the susceptibility maximum. No hysteresis or remanence was detected. Our apparatus could have reliably detected a 1% effect. Remanence and hysteresis reported earlier[11] were caused by instrumental errors which were subsequently corrected.

The existence of a susceptibility maximum down to zero frequency, its independence of frequency, and the existence of a critical field strongly suggest that CMN undergoes an antiferromagnetic transition, rather than a ferromagnetic transition with accompanying domain relaxation effects as had been suggested by Mess et al.[1,2] The presence of domain walls would imply hysteresis and remanence phenomena which, however, we did not observe. Furthermore, attributing the final decrease in susceptibility at the lowest temperatures to long relaxation times is implausible. Our first datum point was taken about 10-30 sec after demagnetization. During adiabatic demagnetization one would expect the spins to be aligned parallel to the applied field with a resulting magnetization close to saturation. However, in a time of the order of 10 sec after demagnetization the magnetization becomes zero as shown above in the discussion based on Fig. 3. This suggests a relaxation time shorter than 10 sec whereas we observed the maximum and subsequent drop in susceptibility even for dc.

In summary, the weight of our evidence favors the hypothesis of antiferromagnetism rather than ferromagnetism on four counts: first, the observation of the susceptibility maximum at all frequencies from dc to 200 Hz; second, the existence of a critical field above which the maximum is not observed; third, the pointing of the saturation magnetization obtained from the field dependence of the susceptibility to a zero initial magnetization below the maximum; and fourth, the absence of hysteresis and remanence in cyclic magnetization processes.

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