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The Low Temperature Heat Capacities of Antiferromagnetic MnF_2 and CoF_2*

EDWARD CATALANO AND NORMAN E. PHILLIPS
University of California, Lawrence Radiation Laboratory
Livermore and Berkeley, California, U.S.A.

The heat capacities of MnF_2 and CoF_2 have been measured in the temperature range 1°-4.2°K. The data have been analysed by plotting CT^2 vs T^5 to separate the lattice and hyperfine contributions to the heat capacity from the magnetic contribution. The magnetic heat capacity for MnF_2 is compared with the heat capacity expected from spin waves modified by anisotropy. No apparent magnetic contribution to the heat capacity was found for CoF_2 in this temperature region. This is consistent with a high anisotropy field for this compound.

At low temperatures, the heat capacity of an antiferromagnet can be expected to be the sum of three contributions: a lattice term proportional to T^3; a nuclear hyperfine interaction term proportional to T^{-2}; and, any heat capacity arising from magnetic interactions. In the absence of anisotropy, spin wave theory predicts a T^3 dependence for the latter term. The inclusion of anisotropy modifies this result giving a heat capacity which varies exponentially with temperature. If the anisotropy field is large enough, one can expect a temperature interval in which only the lattice and hyperfine heat capacities are appreciable.

The experimental data for MnF_2 and CoF_2 have been analysed into the three components by plotting CT^2 versus T^5 as shown in Fig. 1. For CoF_2, the experimental points follow a straight line from 4.2° to 1.5°K. Below 1.5°K, the points fall below the line. This behavior is apparently related to a thermal relaxation time observed in this salt. This thermal relaxation time increased rapidly with decreasing temperature reaching a maximum of approximately 10 minutes at 1.5°K and decreasing again at lower temperatures. Both observations are consistent with a nuclear relaxation time which increased with decreasing temperature such that at the lowest temperatures the corresponding hyperfine heat capacity contribution was not observed. The linear relation between CT^2 and T^5 for CoF_2 suggests that there is no significant magnetic contribution to the heat capacity in this temperature region. In the case of MnF_2, the linear region extends only to ~1.8°K. Positive deviations are observed above this temperature. These are assumed to represent the magnetic heat capacity. The hyperfine heat capacity and lattice heat capacity were obtained from the intercept and slope of the straight line for both salts. The T^3 lattice heat capacities obtained in this way are in excellent agreement with an extrapolation of the lattice heat capacity obtained by an analysis of the higher temperature data by Stout and Catalano[1]. This is demonstrated in Fig. 2 as a plot of effective Debye temperature versus temperature.

Kubo[2] has derived a dispersion relation for antiferromagnetic spin waves in which anisotropy is included in a molecular field approximation. Eisele and Keffer[3] have calculated the thermodynamic functions in the same approximation. In the case of the heat capacity, their result is presented as the ratio of the observed magnetic heat capacity to the T^3 heat capacity expected for an isotropic antiferromagnet and is given as a function of T/T_{AE}. T_{AE}, a measure of the anisotropy, is given by $kT_{AE}=2(z|J|K)^{1/2}S$ where $J/2$ is the exchange integral, K is the anisotropy constant, S is the spin, and z is the number of nearest neighbor magnetic ions. The observed magnetic heat capacity for MnF_2, C_M, is compared with the result of Eisele and Keffer[3] in Fig. 3. T_{AE} has been taken as 12.7°K in accord with critical field and magnetic resonance experiments[4-6]. On the whole, the agreement must be considered as reasonable. The experimental and

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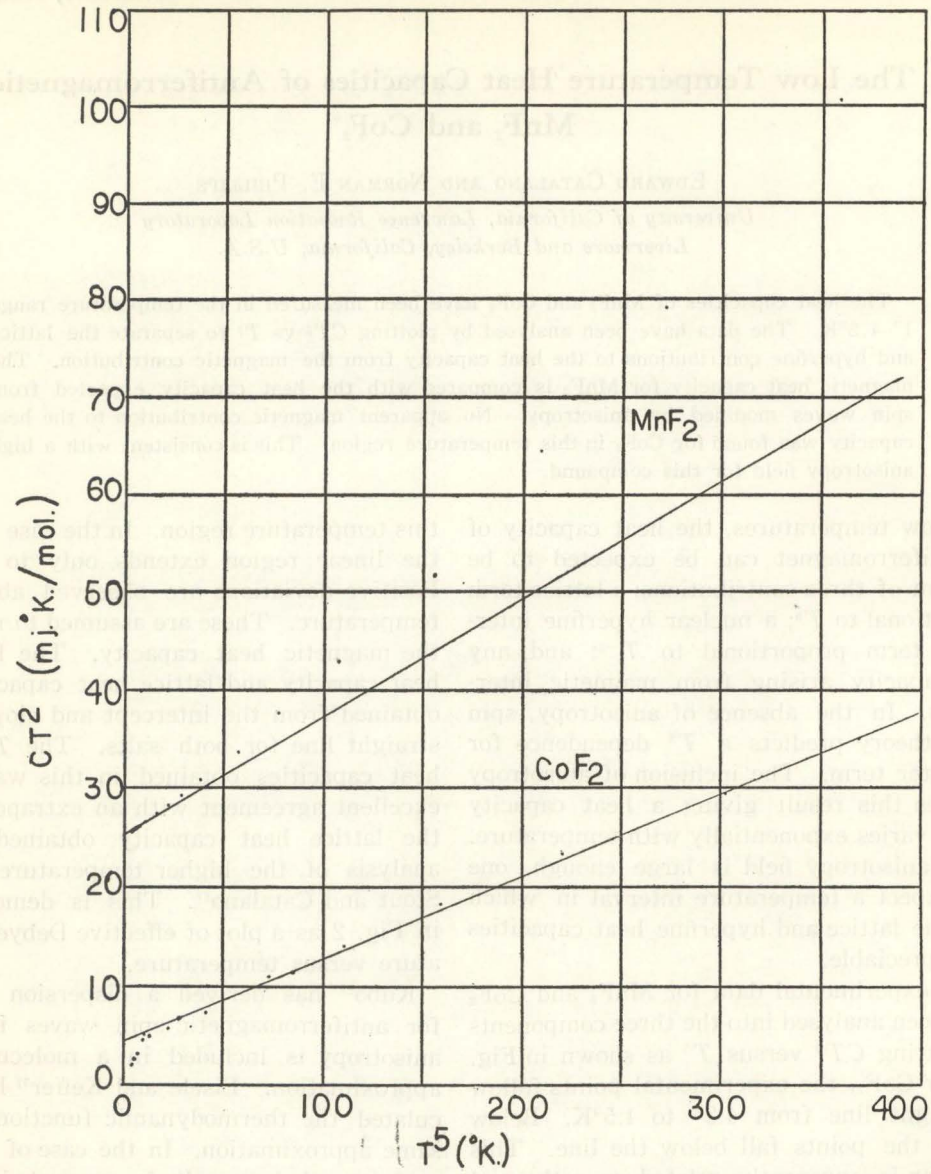

Fig. 1. Total heat capacity, C(mj/mol deg), of MnF_2 and CoF_2 shown as CT^2 versus T^5.

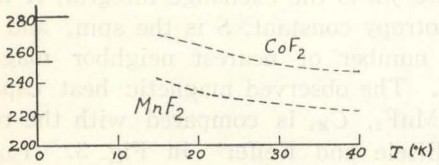

Fig. 2. Apparent Debye temperature. The dashed curve is from the work of Stout and Catalano[1]; the solid lines represent the present measurements.

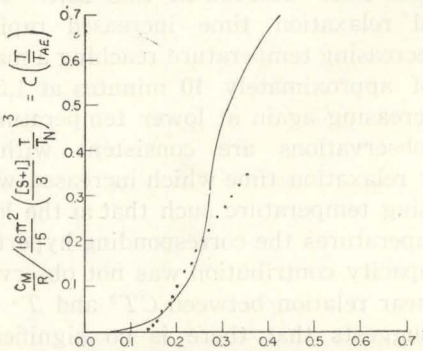

Fig. 3. Magnetic heat capacity, C_M, of MnF_2. The curve is from the calculation of Eisele and Keffer[3]. The points represent the experimental values.

theoretical curves have the same general slope. The magnitude is very sensitive to the value of the exchange integral. This has been estimated from the Néel temperature, T_N, using the usual molecular field formula. At the low temperature end, of course, the error in C_M is quite large and the comparison should be made with this fact in mind.

The absence of magnetic heat capacity and the long nuclear relaxation times observed in CoF_2 are both indicative of a high anisotropy field for this salt.

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