

EFFECT OF HYDROSTATIC PRESSURE ON A QUASI-2D

ANTIFERROMAGNET: $\text{Mn}(\text{HCOO})_2 \cdot 2\text{H}_2\text{O}$

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By observing ac-susceptibility and heat capacity of single crystals of $\text{Mn}(\text{HCOO})_2 \cdot 2\text{H}_2\text{O}$ under hydrostatic pressure up to 7 kbar, the phase transition of the second kind is studied at its Néel temperature, T_N , and that of the first kind at its spin-axis reorientation temperature, T_R . Both T_N and T_R increase with pressure giving $dT_N/dP = +0.06$ K/kbar and $dT_R/dP = +0.15$ K/kbar. Surprisingly, the sharpness of both transitions recovers when the pressure is removed, though it is more or less lost when the pressure is applied.

1. Introduction

Study of the low dimensional magnetic systems under hydrostatic pressure is expected to be useful for understanding the mechanism of the phase transition, which depends largely on the strength and arrangement of interaction paths in the crystal. We have studied $\text{Cu}(\text{C}_2\text{H}_5\text{NH}_3)_2\text{Cl}_4$ by the measurements of ac-susceptibility and ESR /1/ and $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ by the measurements of heat capacity and susceptibility. /2/

In this paper we report the simultaneous measurements of susceptibility and heat capacity on $\text{Mn}(\text{HCOO})_2 \cdot 2\text{H}_2\text{O}$. This crystal consists of two kinds of sublattices, A and B, which are alternately piled up along the a-axis. The Mn^{2+} ions of the A-sublattice on (100) plane which couple each other by a strong intralayer exchange interaction through HCOO^- set into a quasi-2D antiferromagnetic order phase at its Néel temperature, $T_N = 3.69$ K, exhibiting a small but sharp peak in susceptibility and heat capacity. A spin-axis reorientation occurs at $T_R = 1.71$ K accompanying a sharp peak both in susceptibility and heat capacity. The Mn^{2+} ions of the B-sublattice on the (200) plane behave almost paramagnetically in the whole temperature region, only giving a Schottky type heat capacity at lower temperatures

due to the small exchange field from the ordered A-sublattice. There are two types of phase transition; The one is that of the second kind at T_N and the other is that of the first kind at T_R . We have observed remarkable pressure effects at both transition points.

2. Experimental Procedures

Hydrostatic pressure up to 7 kbar was applied to the single crystal by clump method. The pressure cell is made of BeCu alloy and heat treated by keeping it at 315°C for 2 hours. The outer diameter of the cell is 15 mm and the length is about 150 mm, whereas the effective working volume is 5 mm in diameter and 10 to 12 mm in length. Silicone oil of high viscosity or Apiezon-J oil is used as the pressure transmitting medium. The pressure actually acting on the specimen at liq. He temperature is determined by the superconducting transition temperature of a small chip of tin or indium placed at the specimen. The temperature is measured by a Ge sensor attached to the outside of the cell.

Single crystal is shaped in a rod of dimension of slightly smaller than the available space of the cell, keeping one of its crystalline axes parallel to the rod axis. The ac-susceptibility is measured by a Hartshorne bridge at 75 Hz and the heat capacity by an adiabatic method.

3. Results

The peak observed in susceptibility along the c-axis in the vicinity of T_N is shown in Fig. 1, without and with applied pressure. Figure 2 shows that observed along the b-axis in the vicinity of T_R . Peaks in Figs. 3 and 4 observed by heat capacity correspond to those given in Figs. 1 and 2. Both transition temperatures become higher with the application of pressure, giving $dT_N/dP = 0.06$ K/kbar and $dT_R/dP = 0.15$ K/kbar, though they have not necessarily a linear dependence on P. The change of T_R is roughly 2.5 times larger than that of T_N .

In addition to the shift of transition temperature, the sharpness at the transition is most apparently lost for the susceptibility peak at T_N and for the specific heat peak at T_R , even under a pressure below 0.5 kbar.

The peak in susceptibility at T_N becomes 10 times broader by applying a pressure of one kbar. Its height becomes smaller at about 3 kbar and turns to larger when an additional pressure

is applied. The heat capacity peak at T_N , however, does not change its sharpness so much by the application of pressure.

The application of only 0.35 kbar causes a sudden rounding of heat capacity peak at T_R to give a broad hump which does not change by additional increase in pressure. In contrast to the heat capacity peak, the susceptibility peak at T_R is not broadened so much by the applied pressure.

The specimen is measured again under atmospheric pressure after the applied pressure was completely removed. In Figs. 5 and 6, it is apparent that the width of the transition completely recovers when it is observed by heat capacity, while that by susceptibility remains to about twice of the initial value.

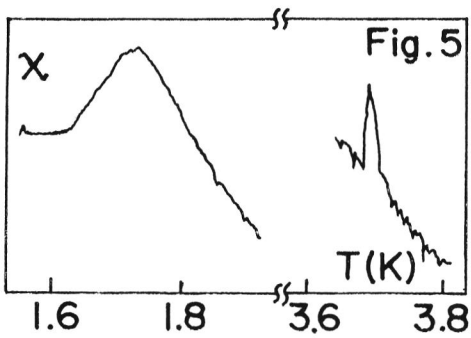
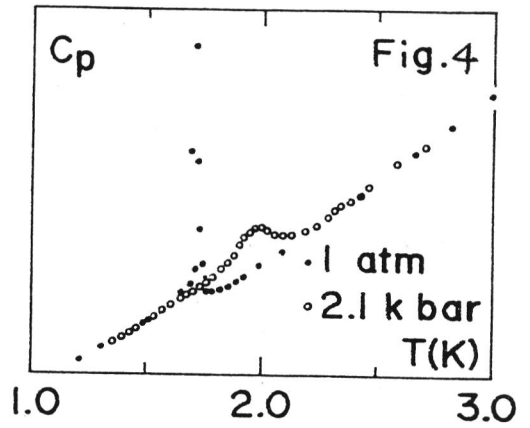
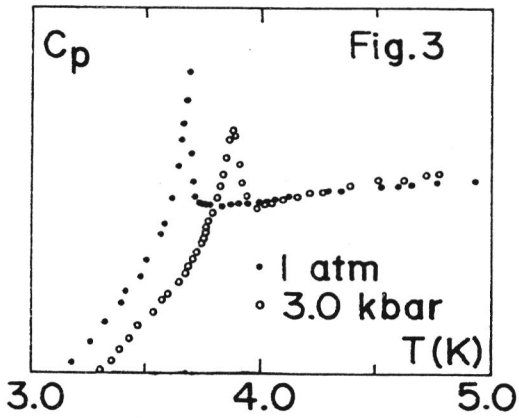
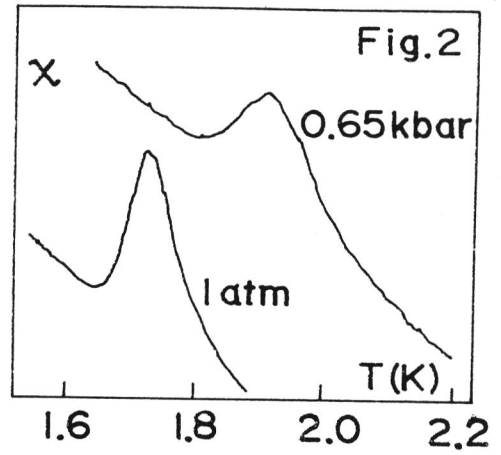
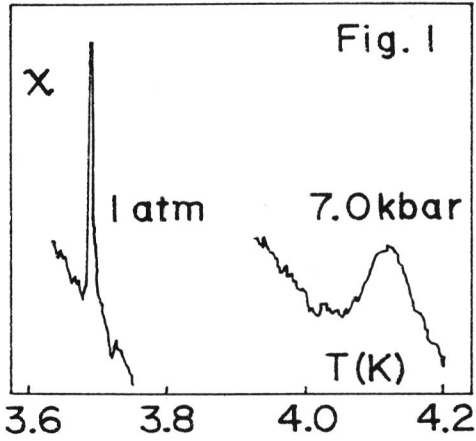
4. Discussions

Experimental facts of the heat capacity suggest that the anisotropic strains and/or continuously changing random fields really exist which might give an effect of weakening the divergence of the singularity of the first kind of phase transition, while the phase transition of the second kind seems to keep the sharp singularity beyond the randomness. In the case of $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, the Néel temperature, T_N , observed under atmospheric pressure does not change after a cycle of compression but the sharpness of heat capacity peak is irreversibly lost even after a pressure cycle of only 3 kbar.

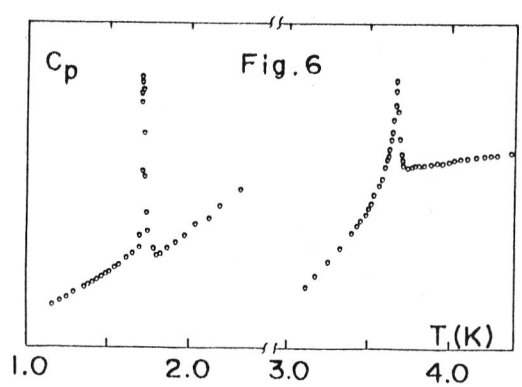
It is surprising that $\text{Mn}(\text{HCOO})_2 \cdot 2\text{H}_2\text{O}$ crystal is very soft and has a complete reversibility of strain.

References

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Compare with Figs. 1 and 2.



Compare with Figs. 3 and 4.