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LETTER TO THE EDITOR

A phase transition in $CsMnBr_3$ at 2.7 K

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Abstract. Specific heat and magnetic susceptibility of the triangular-lattice XY antiferromagnetic $CsMnBr_3$ have been measured. Results of our experiments show that a phase transition occurs at around 2.7 K in addition to one at 8.31 K. Findings of neutron diffraction experiments suggest that Mn spins still lie on the basal plane with 120° angles between the neighbouring spins.

A number of theoretical and experimental studies have recently been devoted to examining the magnetic properties of triangular-lattice antiferromagnetic $CsMnBr_3$ (space group $P6_3/mmc$, with lattice parameters $a = 7.61$ Å and $c = 5.62$ Å [1–4]). These are mainly motivated by the particular features of the material: a combination of the quasi-one-dimensionality and a non-collinear magnetic structure [2].

It is well known [3] that, below 8.31 K, $CsMnBr_3$ has a stacked triangular structure with spins lying on the basal plane, aligned antiferromagnetically along the c axis with 120° angles between the neighbouring spins. A suitable Hamiltonian to describe this system can be written as:

$$= 2J \sum_{ij} S_i S_j + 2J' \sum_{ij} S_i S_J + D \sum_i \left(S_{iz}\right)^2 - g\mu_B H \sum_i S_i \qquad (1)$$

where J and J' are the antiferromagnetic super-exchange interactions between the neighbouring Mn spins along the c axis and in the c plane, respectively, and $J = 0.88$ meV, $J' = 0.0019$ meV, and D is the in-plane anisotropy, $D = 0.014$ meV [4].

Using (1), Chubukov [5] calculated the magnetization $\boldsymbol{M}$ and the antiferromagnetic resonance frequency ω_i of $CsMnBr_3$ in an applied magnetic field. Chubukov's results pointed out that, in contrast to a simple two-sublattice antiferromagnet, the spin rotation in a transverse magnetic field is accompanied by a magnetic phase transition at a relatively small field. It is supposed that spin flip of two magnetic sublattices in the c plane causes the phase transition. Later, Kotyuzhauskii *et al* [6] also found that the spin deviates from the c plane through experiments. According to their conclusion, an intermediate phase with convergent sublattices between triangular and paramagnetic phases exists at $H_c = 64$ kOe below 2 K.

We have found, through NMR experiments, that there is a possibility that a phase transition occurs below 4.2 K without external field. In order to see whether the transition does exist, we have examined the specimen by specific heat and magnetic susceptibility experiments.

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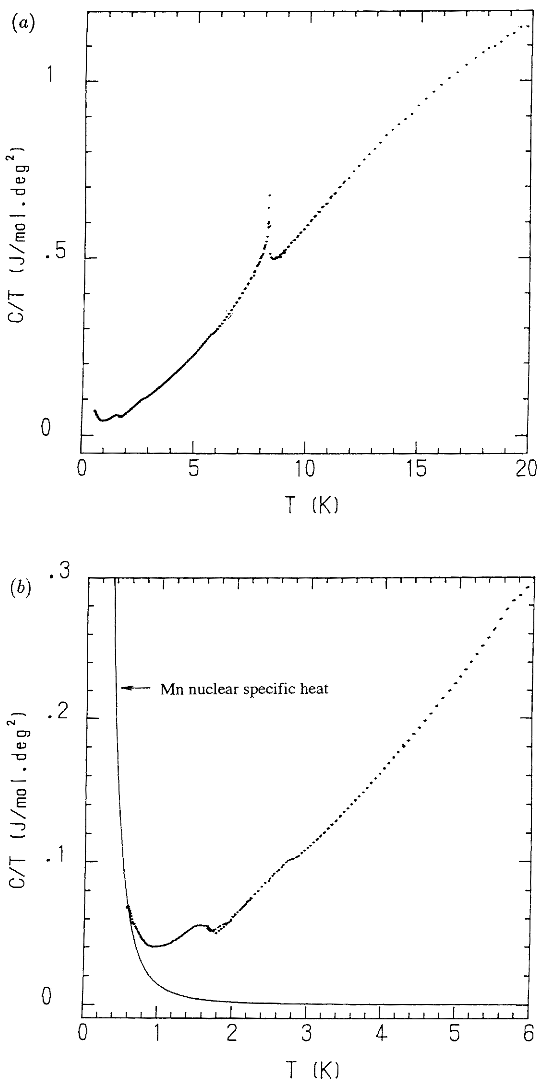

Figure 1. (a) The temperature dependence of C/T, from 0 to 20 K. (b) The temperature dependence of C/T, from 0 to 6 K. The solid curve shows the calculation of Mn nuclear specific heat, and the dots represent the experimental data (see text).

Single crystals of $CsMnBr_3$ were grown by the Bridgman method [7]. The specific heat was measured by the pulse method with ^3He vaporization refrigeration in the temperature range from 22 to 0.5 K. The magnetic susceptibility was measured with a SQUID magnetometer in the temperature range from 16 to 1.7 K and with the external field ranging from 100 to 50 kOe, in both parallel and perpendicular polarization with respect to the c axis.

The results of the specific heat measurements are shown in figure 1(a). Small peaks are observed at $T = 5.7$, 2.7, 1.6 and 0.5 K in addition to the Néel temperature $T_N = 8.31$ K. Assuming the NMR Mn nucleus frequency is 600 MHz, we have obtained the Mn nuclear

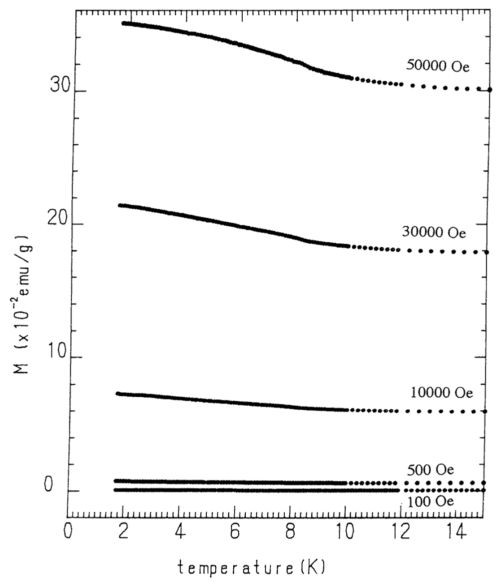

Figure 2. The temperature dependence of the magnetization $M(T)$ in fields of 100, 500, 1000, 3000 and 5000 Oe.

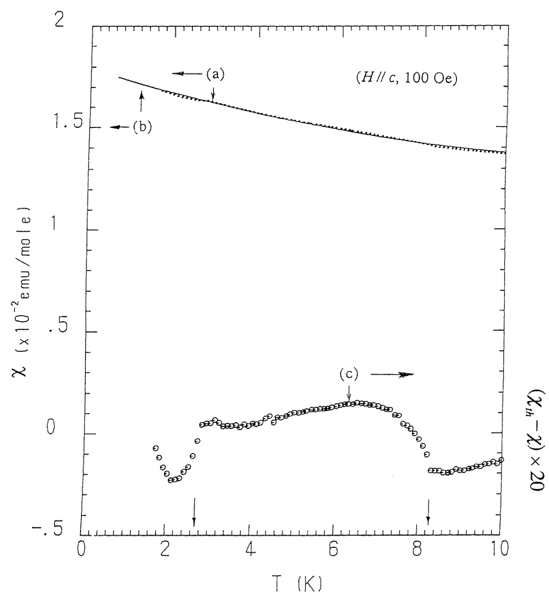

Figure 3. (a) The temperature dependence of $\chi(T)$ in the case of $H \parallel c$, at $H = 100$ Oe. The dots represent the experimental data. (b) The solid curve is the calculation of $\chi(T)$ as a quadratic function. (c) Difference between the experimental and calculated $\chi(T)$ as described in the text. Note that the ordinate has been enlarged by a factor of 20.

specific heat:

$$C_N = R\theta^2\left[-\left(I+\frac{1}{2}\right)^2\frac{1}{\sinh^2\theta(I+\frac{1}{2})}+\frac{1}{4}\frac{1}{\sinh^2\frac{\theta}{2}}\right] \tag{2}$$

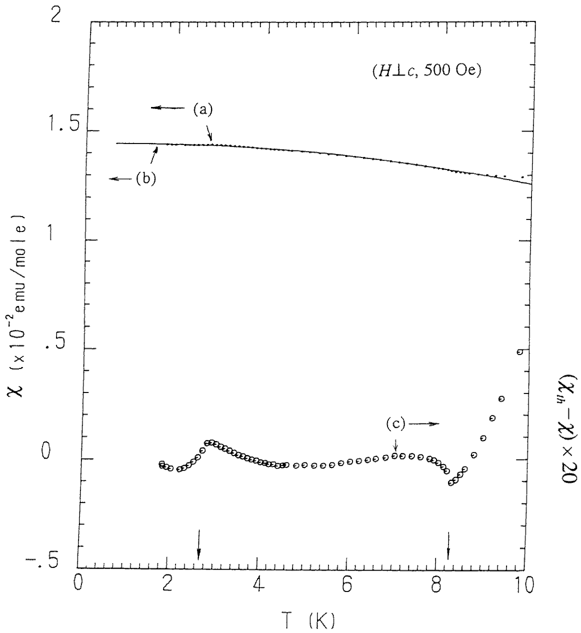

Figure 4. (a) The temperature dependence of $\chi(T)$ in the case of $H \perp c$, at $H = 500$ Oe. The dots represent the experimental data. (b) The solid curve is the calculation of $\chi(T)$ as a quadratic function. (c) Difference between the experimental and calculated $\chi(T)$ as described in the text. Note that the ordinate has been enlarged by a factor of 20.

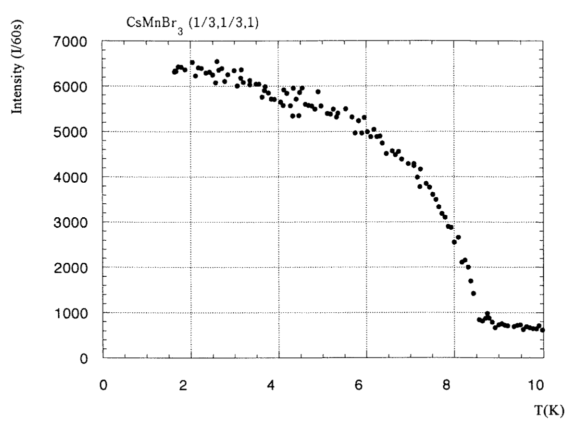

Figure 5. The temperature dependence of the $(\frac{1}{3}, \frac{1}{3}, \frac{1}{3})$ Bragg peak.

where I is Mn nuclear spin, $\theta = \gamma_N h H/\kappa_B T$, and R is the molar constant, and the calculated result is shown in figure 1(b). The increment of the specific heat below 0.5 K may be understood as the Mn nuclear specific heat. By our calculation, the total entropy (S) of $CsMnBr_3$, $S = Nk_B \ln(2s+1)$, where s is the Mn electric spin, is about 15 (J mol^{-1} K^{-1}); and the entropy at 8.31 K is only about 1.6 (J mol^{-1} K^{-1}). That is, about 10% entropy remained below 8.31 K. Thus, between 8.31 and 0 K, there is a possibility that some phase transitions occur.

The results of magnetization measurements for various external fields are shown in figure 2 (in the case of $\boldsymbol{H}$ parallel to the c axis). The result of the magnetic susceptibility measurement, in the same polarization, is shown by the dots (a) in figure 3 which also show an anomaly at about 2.7 K in addition to the one at $T_N = 8.31$ K. We observe that below the Néel temperature, the susceptibility χ depends on temperature slightly. Neglecting higher-order contributions, we assume that χ can be represented as a quadratic function of the temperature χ_{th}. Determining the three coefficients in χ_{th} by the least-squares method, we have a solid curve (b) shown in figure 3. The difference between the experimental result (a) and a quadratic approximation (b) is shown by the dots (c) in the same figure. Note that in plotting the difference, we have magnified the ordinate 20 times. We observe two distinct dips at 2.7 and 8.31 K; the dips suggest phase transitions. The same phenomena were also obseved in the case of $\boldsymbol{H}$ perpendicular to the c axis, as shown in figure 4. A detailed examination of this new phase will be given elsewhere.

It is claimed theoretically [5] that at $H_c = 64$ kOe and below 2 K, Mn spins deviate from the basal plane. Later, a phase transition was recognized under the conditions above [6]. In order to confirm whether or not the structure of the phase transition in our experiments is similar to that of their structure [5, 6], we have performed neutron diffraction experiments in zero field at temperatures ranging from 1.7 to 25 K.

The results of the neutron diffraction experiments are shown in figure 5. No changes were observed below 8.31 K and hence, the neutron diffraction data do not give evidence for a change of magnetic structure below T_N. This means that in zero field, the Mn spins are still lying in the basal plane with 120° angle between neighbouring spins. Therefore, in order to find the type of this intermediate phase transition, we need to use the complete Hamiltonian to calculate the exchange interaction between next neighbouring Mn spins in the c plane or the biquadratic exchange interaction in $CsMnBr_3$.

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