Weak ferromagnetism of $RbMnCl_3$

N. V. Fedoseeva and B. V. Beznosikov

Physics Institute, Siberian Division, USSR Academy of Sciences
(Submitted November 16, 1974)
ZhETF Pis. Red. **21**, No. 2, 108–110 (January 20, 1975)

It was observed that the optically transparent rubidium trichlormaganate $RbMnCl_3$ is antiferromagnetic with weak ferromagnetism below $T_N = 94.6$ °K; its magnetic moment at 87 °K is equal to 0.41 G-cm³/g and is directed along a sixfold hexagonal axis.

The compound $RbMnCl_3$ is transparent in the near-ultraviolet and the visible region, and has a space group $D^4_{6h}-P6_3mmc$, with six formula units per cell. Neutron-diffraction investigations by Melamud and Makovsky,[1] in a wide range of temperature from 4.2 to 300 °K, have shown that the magnetic and chemical unit cells coincide, and that the magnetic structure is a set of parallel ferromagnetic layers of Mn^{2+} atoms that are antiferromagnetically bound. According to[1], the magnetic structure of $RbMnCl_3$ can belong to one of two different structures of orthorhombic symmetry, $Cm'c'm$ and $Cmcm$.

The static magnetic properties of single-crystal $RbMnCl_3$ were measured by the Faraday method with accuracy ~4% in the temperature interval from 78 to 300 °K. The accuracy with which the sample was oriented in the holder was 3—4°. The $RbMnCl_3$ single crystals were grown from the melt by the Bridgman method in a vertical tubular oven; the lattice parameters were $a = 7.11 \pm 0.01$ Å and $c = 17.65 \pm 0.01$ Å.

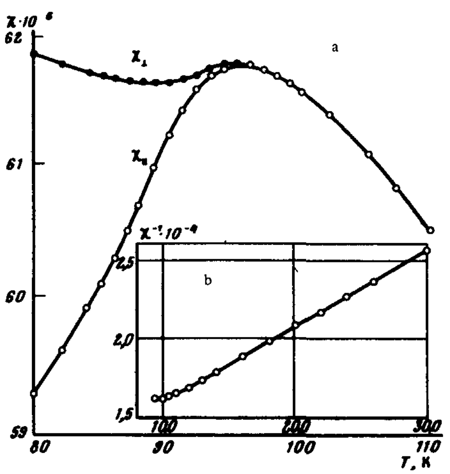

Fig. 1. a) Temperature dependence of the magnetic susceptibility of single-crystal $RbMnCl_3$ in a field 4780 Oe: ●—H directed along [001], ○—H perpendicular to [001]; b) Temperature dependence of χ^{-1}: (solid line—theoretical, points—experimental results).

1. Figure 1a shows the variation of the magnetic susceptibility ($\chi_\parallel$ and $\chi_\perp$) of single-crystal $RbMnCl_3$ with temperature. It is seen that the curves have a broad maximum at 95 °K. The susceptibility is isotropic at temperatures above 95 °K, and anisotropic below this temperature; $\chi_\parallel$ decreases sharply with decreasing temperature, whereas $\chi_\perp$ remains approximately constant. An analysis of the temperature dependence of the magnetic susceptibility χ and of χ^{-1} has shown that above 140 °K the Curie-Weiss law is satisfied, with a molar constant $C_M = 4.84$ and $\theta = -204$ °K, and the maximum on the temperature dependence of the magnetic susceptibility at 95 °K is connected with a transition to the antiferromagnetic state. A reduction of the experimental data was carried out by the high-temperature expansion method of Rushbrooke and Wood.[3] It is seen from Fig. 1b that the experimental data agree well with the theoretical plot of $(Ng^2\mu_B^2/\chi J)(kT/J)$ at $J/k = 9.85$ °K. The value of T_N was determined from the temperature dependence of $\chi_\parallel$ and $\chi_\perp$ below the transition point.[4] When plotted in the coordinates $\ln(\chi T/c)$ and $\ln(\chi T/J)$, the experimental values of $\chi_\parallel$ and $\chi_\perp$ fit well two straight lines whose intersection yields $T_N = 04.6$ °K.

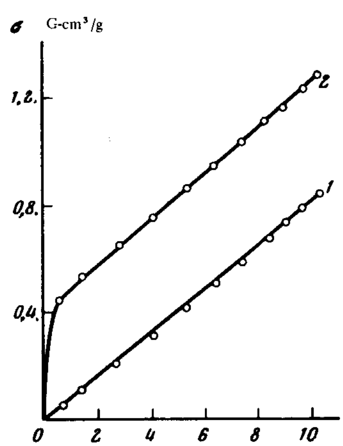

FIG. 2. Dependence of the magnetic moment of $RbMnCl_3$ on the field at 78°K: 1—field perpendicular to [001], 2—field parallel to [001].

2. Figure 2 shows plots of the magnetic moments of $RbMnCl_3$ against the field at $H \perp [001]$ and $H \parallel [001]$, obtained at 78 °K. In the former case this dependence takes the form $M = \chi_{\parallel} H$, where $\chi_{\parallel} = 59.28 \times 10^{-6}$, and in the latter case, in fields stronger than 2.5 kOe, we have $M = \sigma + \chi_{\perp} H$, where $\chi_{\perp} = 61.84 \times 10^{-6}$ and $\sigma = 0.41$ G-cm³/g while the value of σ is obtained by extrapolating the linear section of the strong-field magnetization curve to $H = 0$. When the temperature is increased from 78 °K to T_N, the plots of the moment against H are similar to curve 1 of Fig. 2, but the spontaneous magnetic moment decreases with rising temperature. At temperatures higher than T_N, the $M(H)$ plot is a straight line, i.e., the compound is a typical paramagnet and, as already noted, no anisotropy of the susceptibility is observed in this temperature interval.

Thus, investigations of the field dependence of the magnetic moment of $RbMnCl_3$ at different temperatures at $H \parallel [001]$ and $H \perp [001]$ have shown that rubidium trichlormanganate has below T_N a weak ferromagnetism with a spontaneous magnetic moment equal to 0.41 G-cm³/g at 78 °K and directed along a sixfold hexagonal axis. It should be noted that the weak ferromagnetism connected with second-order invariants in the magnetic Hamiltonian and with the g-factor anisotropy may not exist in antiferromagnets with $6_z^- 2_d^+$ structure, such as $RbMnCl_3$. Nonetheless, the higher-order terms lead to a spontaneous magnetic moment directed along the hexagonal axis of the crystal.[5] Analysis of the symmetry shows that weak ferromagnetism in the direction of a sixfold hexagonal axis can occur in the $Cm'c'm$ magnetic structure but not in $Cmcm$. Therefore our investigations make it possible to eliminate the ambiguity in the determination of the magnetic structure, and allow us to assume that a $Cm'c'm$ magnetic structure is realized in $RbMnCl_3$.

The authors are grateful to K. S. Aleksandrov for suggesting the problem and for constant interest in the work, and to V. V. Uskov for help with the work and for useful discussions.

[1]M. Mclamud, I. Makovsky, and H. Shaked, Phys. Rev. **B3**, 821 (1971).
[2]M. E. Lines, J. Phys. Chem. Sol. 31, 101 (1970).
[3]G. S. Rushbrooke and P. J. Wood, Molec. Phys. **1**, 257 (1958).
[4]D. J. Breed, Physica **37**, 35 (1967).
[5]E. A. Turov, Fizicheskie svoĭstva magnituporyadochennykh kristallov (Physical Properties of Magnetically-Ordered Crystals), AN SSSR, 1963.