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Neutron scattering study of spin waves in the antiferromagnet $RbMnCl_3$

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The magnetic structure of the antiferromagnet $RbMnCl_3$ ($T_N = 94K$) consists of ferromagnetic planes stacked in the sequence $A(+)$, $B_1(-)$, $B_1(+)$, $A(-)$, $B_2(+)$, $B_2(-)$ along the C-axis. The two kinds of sites, A and B for the Mn^{2+} ions contain respectively, "lone spins" and (much closer) spin pairs. This study of the spin wave spectrum was undertaken with the objective of determining whether the principal magnetic interactions J_{AB} and J_{BB} are of the same order of magnitude (conventional Heisenberg type) or whether a strong coupling model ($J_{BB} >> J_{AB}$) applies. Two distinct optic branches observed in the [qoo] direction immediately ruled out the strong coupling model. A fit to the data was then performed using the conventional spin wave model with excellent results. We find $J_{BB}/J_{AB} = 1.65$ ($\pm .1$). With spin values $S_A = 2.4$ and $S_B = 2.5$, $J_{BB} = 0.73$ meV and $J_{AB} = 0.44$ meV.

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INTRODUCTION

Compounds of the type RMH_3 (R = Rb, Cs, K, NH_4; M = Mn, Co, Ni, Fe; H = Cl, F, Br) exist with many different crystal and magnetic structures. For example, $RbNiF_3$ is a ferrimagnet with the hexagonal $BaTiO_3$ structure (1), $RbNiCl_3$ is a one-dimensional antiferromagnet with the hexagonal $CsNiCl_3$ structure (2), and $RbMnF_3$ is a cubic antiferromagnet (3). $RbMnCl_3$ is an antiferromagnet (Néel temperature T_N = 94K) with the $BaTiO_3$ structure with six formula units per unit cell (4). The spins on the six maganese atoms are arranged in ferromagnetic layers along the C-axis in the sequence: $A(+)$, $B_1(-)$, $B_1(+)$, $A(-)$, $B_2(+)$ and $B_2(-)$. Figure 1 shows the lower half of the unit cell where the spin direction is shown in a particular direction in the basal plane but all that has been determined so far by neutron diffraction measurements on polycrystalline samples is that the spins are perpendicular to the c-axis (4). The two distinct manganese sites, A and B have very different nearest-neighbor distances d_{AB} = 5.1A and d_{BB} = 3.0A respectively. This has led to a speculation that perhaps $J_{BB} >> J_{AB}$ and hence the magnetic system could be described by a new class of spin structures characterized by strongly coupled B-B spin pairs (considered as a single entity pseudo-spin) alternating with (and weakly coupled to) a lone spin, A(5). Thus there would be only four magnetic entities per unit cell and Samuelsen and Melamud (6) showed that the calculated spin wave spectrum for such a system would have only two distinct branches in contrast with the three (doubly degenerate) branches expected in general for the conventional spin wave treatment. They also showed however that if the ratio J_{BB}/J_{AB} is large ($\sim$20, as indicated by some previous susceptibility measurements (5)) then for the conventional treatment the splitting of the two optic branches is very small within the basal plane but can still be appreciable for the wave vector in the c-direction. The spin wave spectrum as measured by low temperature neutron inelastic scattering on a single crystal sample should therefore provide definitive information to decide the applicability of the strong coupling model as well as to independently determine the magnetic exchange interactions.

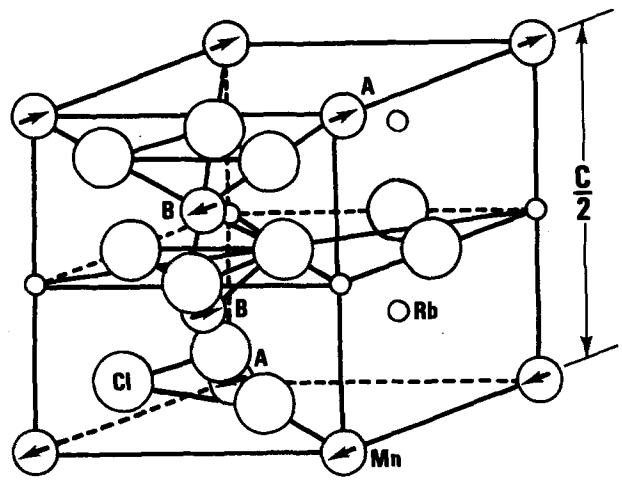

Fig. 1. The lower half of the unit cell for $RbMnCl_3$. Nearest neighbor distances for the two manganese sites A and B are d_{AB} = 5.1 A and d_{BB} = 3.0 A. The spins are collinear and lie in the basal plane but their direction is unknown.

EXPERIMENTAL

The starting materials for $RbMnCl_3$ samples were Merck U.P. RbCl and hydrated $MnCl_2$ (Merck, G.R.). The RbCl was dried in a vacuum dessicator and anhydrous $MnCl_2$ prepared from the hydrated form by heating it in a stream of dried HCl. A tapered quartz ampoule was

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used to grow the crystal by lowering it into a temperature gradient vertical furnace at the rate of 0.1mm/hr. The gradient at the freezing plane was about 30°/cm. The $RbMnCl_3$ single crystals were characterized by chemical and x-ray powder analysis and by density measurements with results in good agreement with expected values. The sample used for this study was cylindrical in shape with a volume $\sim$ 2.5 cm^3.

The experiments were performed on a triple-axis spectrometer with poly-graphite monochromator and analyzer operated in the constant-Q mode (Q = the difference between the incident and scattered wave vectors) and with the final neutron energy scattered into the detector fixed at 14.7 meV. Moderate resolution was employed and a poly-graphite filter before the detector was also used to eliminate higher order wavelength contamination. Data was taken (Fig. 2) in three principal directions of the wave vector q as measured from the Brillouin zone center: [qqo], [qoo], and [ooq]. For the [qqo] data both [001] and [100]-zone crystal orientations were used while for the [qoo] and [ooq] data a [110]-zone orientation of the sample was used. Measurements were performed at liquid helium temperature and repeated for the [ooq] acoustic mode (Fig. 2) at room temperature to verify that the observed peaks (scattered intensity versus energy) were indeed of magnetic origin. The peaks had half-widths varying from 0.5 to 1.0 meV but in general the peak position could be determined more closely than this. The [qoo] and [ooq] data were more accurate than the measurements in the [qqo] direction which shows some scatter which can be taken as a measure of the experimental error.

RESULTS

The energy dispersion measurements taken at liquid helium temperature are shown in Fig. 2 for the momentum transfer vector along the three principal directions of the hexagonal Brillouin zone. No attempt was made to measure the acoustic mode very close to the zone center in order to determine the exchange anisotropy. This would require resolution corrections which were not made. We observe that there are two distinct optic branches and one acoustic branch in all three directions. This immediately rules out $RbMnCl_3$ as a candidate for the strong coupling model described in Reference (6) and suggests that the ratio J_{BB}/J_{AB} is probably much smaller than the value 20 used there, thus giving rise to the hope that the conventional spin wave model will fit the data.

Conventional spin wave theory applies when the two exchange interactions are of the same order of magnitude. Spin wave dispersion relations for low temperature, non-interacting spin waves in $RbMnCl_3$ have been derived in Reference (6) for the case of collinear spins and all interactions neglected except for J_{AB} and J_{BB}. The result is a cubic equation which is written here in the square of a reduced energy $\omega = E/J_{AB}$, with coefficients that depend on the variable q and parameters S_A, S_B and the ratio $r = J_{BB}/J_{AB}$ when the anisotropy field is neglected.

$$\omega^6 + D'\omega^4 + E'\omega^2 + F' = 0$$

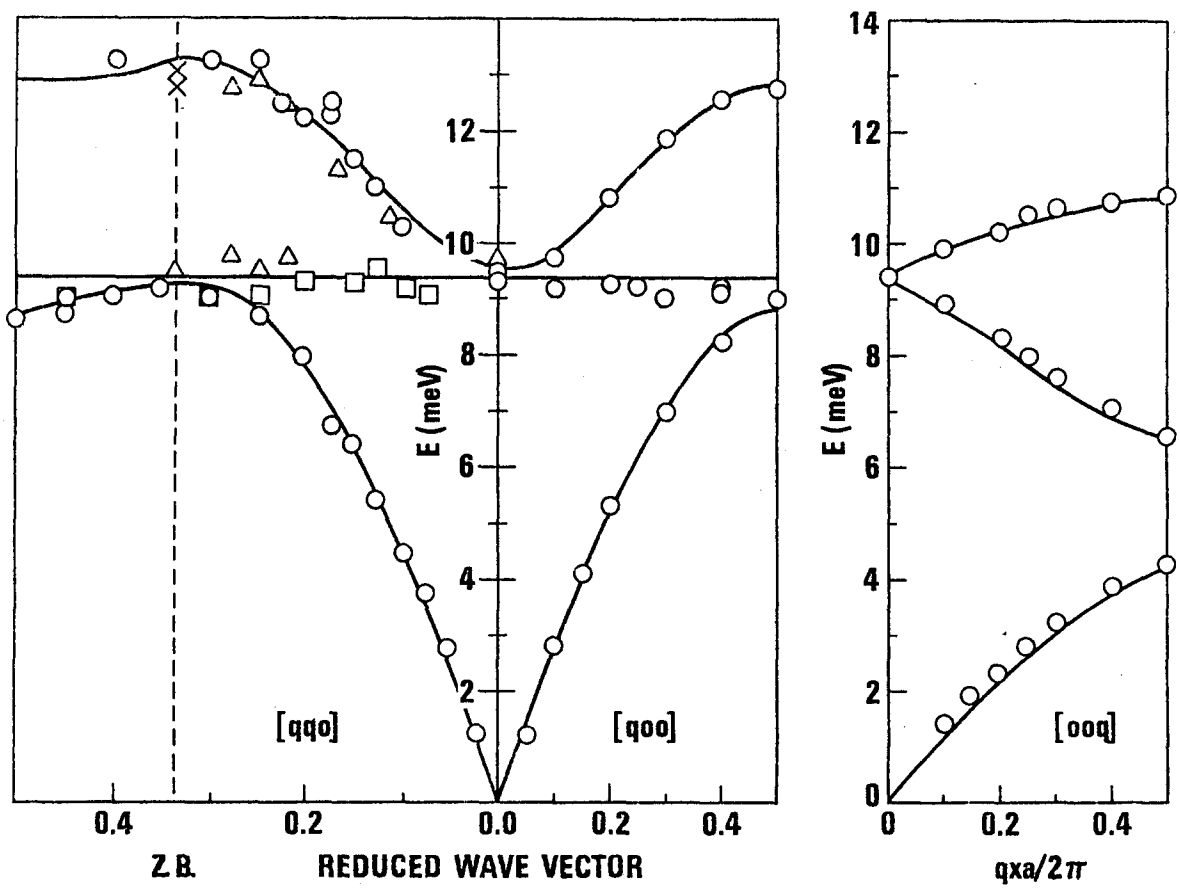

Fig. 2. Spin wave dispersion measurements for $RbMnCl_3$ at 4K. The triangles designate data taken with a [001]-zone crystal orientation. The other [qq0] measurements were made with a [100]-zone orientation. The [00q]-scale is expanded relative to the [qq0] and [q00] scales.

where D', E' and F' are complicated functions of A', B', C' and W where (7)

$$A' = -12S_B,\ B' = -6S_A - rS_B,\ C' = 2rS_B \text{ and}$$

$$W = 3 + 2\,[\cos \underline{a}\cdot\underline{q} + \cos \underline{b}\cdot\underline{q} + \cos (\underline{a} + \underline{b})\cdot\underline{q}].$$

$\underline{a}$, $\underline{b}$, $\underline{c}$ are the hexagonal lattice vectors and S_A and S_B are the spin values for the two sites. The cubic in ω^2 is easily solved and it can be shown that there is only one value of r, namely $r = 1.65 \pm .1$ which will fit the data. However, for the other parameters two sets of values $\{S_A = 2.4,\ S_B = 2.5,\ J_{AB} = .44 \text{ meV}\}$ or $\{S_A = 1.9,\ S_B = 2.0,\ J_{AB} = .55 \text{ meV}\}$ give equally good fits. It is reasonable to choose the first set as we expect the spin value for Mn^{2+} to be close to 5/2. The final dispersion curves calculated then for the parameters $S_A = 2.4$, $S_B = 2.5$, $J_{AB} = .44$ meV and $J_{BB} = .73$ meV are plotted as the solid curves in Fig. 2 and are seen to be in excellent agreement with the measurements.

The interpretation of these results is rather straightforward. $RbMnCl_3$ is a conventional antiferromagnet with the two dominant exchange interactions comparable in magnitude; the A-B interaction takes place via 180° super exchange and the B-B via 90° super exchange, both through the intervening chlorine atoms. (see Fig. 1). The values obtained for J_{AB} and J_{BB} are consistent with the nearest-neighbor values found for other similar Mn compounds, namely $J_{nn} = .29$ meV for $RbMnF_3$ (3), $J_{nn} = .33$ meV for $KMnF_3$ (8), $J_{Mn\text{-}Mn} = .64$ meV for $Rb_2Mn_{.5}Ni_{.5}F_4$ (9).

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