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SPIN WAVE ANALYSIS OF THE TWO-DIMENSIONAL HEISENBERG ANTIFERROMAGNETS Rb$_2$MnCl$_4$ AND (CH$_3$NH$_3$)$_2$MnCl$_4$

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Résumé.- Afin de compléter nos investigations sur la diffraction élastique et inélastique de neutrons des systèmes Rb$_2$MnCl$_4$ et (CH$_3$NH$_3$)$_2$MnCl$_4$, qui présentent un antiferromagnétisme d'Heisenberg à deux dimensions, nous avons étudié les résonances antiferromagnétiques (AFMR) de ces substances en fonction de la température. Pour déterminer ces résonances, nous avons utilisé une diode Impatt, travaillant à 84 GHz, comme source micro-ondes, et fait varier le champ magnétique obtenu avec un solénôide supraconducteur.

Nous avons calculé la relation entre la fréquence de la résonance AFMR, à champ magnétique nul, et la température, en utilisant une théorie d'onde de spin renormalisée sur la base des résultats des expériences AFMR et la relation de dispersion des magnons à 8 K obtenus avec la diffraction inélastique de neutrons.

Dans l'intervalle $0 < T < 0.8 T_N$, l'amplitude de sous-réseau magnétique correspond bien à ce que l'on peut déduire de l'intensité du reflex magnétique (102). Dans l'intervalle $0.5 T_N < T < T_N$, cette amplitude suit une loi exponentielle à exponent critique $\beta = 0.2$.

Abstract.- In addition to recent investigations of coherent elastic and inelastic neutron scattering experiments from E. Neumann of Konstanz University. For both materials, the sublattice magnetization has been investigated as a function of temperature by intensity measurements of the (102) and the magnetic field of a superconducting coil was varied to obtain the resonance condition. The results of these experiments are used to calculate the variation of the AFMR frequency at zero field with temperature. This calculation is performed by means of renormalized spin wave theory on the basis of the AFMR data and the magnet dispersion at 8 K obtained by inelastic scattering. In the range $0 < T < 0.8 T_N$, the calculated values for the sublattice magnetization agree well with the experimental data derived from the intensity of the (102) magnetic Bragg peak. In the range $0.5 T_N < T < T_N$, the sublattice magnetization can well be described by a power law with a critical exponent $\beta = 0.2$.

1. INTRODUCTION.- Perovskite-type layer structures are known to be rather "ideal" model substances for two-dimensional magnetic properties. In materials of this kind, including fluorides like K$_2$NiF$_4$ and K$_2$MnF$_4$ as well as chlorides like Rb$_2$MnCl$_4$ and (CH$_3$NH$_3$)$_2$MnCl$_4$, the dominating exchange interaction is restricted to the perovskite-type layer, a two-dimensional network of corner-sharing octahedra of halogen ions with the paramagnetic ion in the Centre /1,2/.

The manganese compounds usually are Heisenberg antiferromagnets with small anisotropy, and a long range order can exist only in three dimensions. In the compounds considered here, this order is antiferromagnetic. But already above the Neel temperature, a rather high spin correlation in two dimensions has been found by neutron diffraction /2/.

2. NEUTRON SCATTERING EXPERIMENTS.- The results mentioned so far refer mostly to fluoride compounds. We have studied the Heisenberg antiferromagnets of the chloride type Rb$_2$MnCl$_4$ and (CH$_3$NH$_3$)$_2$MnCl$_4$ (= MAMC-D and Rb$_2$MnCl$_4$) for the latter material, we were able to obtain completely deuterated samples for neutron scattering experiments from E. Neumann of Konstanz University. For both materials, the sublattice magnetization has been investigated as a function of temperature by intensity measurements of the (102) or (100) magnetic Bragg peaks. In the range $0.5 T_N < T < T_N$, the temperature dependence of the sublattice magnetization can well be described by

$$M(T) = M(1 - T/T_N)^{\beta}$$

with values $\beta = 0.21$ and $\beta = 0.19$ of the critical exponent for MAMC-D and Rb$_2$MnCl$_4$, respectively.

At 8 K, the magnum spectrum was determined for both materials by inelastic neutron scattering at the high flux reactor of the I.L.L. in Grenoble. As for K$_2$NiF$_4$, K$_2$MnF$_4$ etc. /2,3/, the spin wave spectrum shows also in the case of Rb$_2$MnCl$_4$ and
MAMC no dispersion within the limits of experimental error for \( q \)-vectors perpendicular to the layers. For \( q \)-vectors in the layers, there is a normal magnon dispersion as observed in three-dimensional magnetic materials. In addition, the variation of frequency with temperature was studied for selected magnons.

3. ANTIFERROMAGNETIC RESONANCE.- For Heisenberg antiferromagnets, a theoretical description of the magnetic properties as a function of temperature, e.g. sublattice magnetization, spin wave energies, is mostly performed by means of a renormalized spin wave theory. This treatment usually yields acceptable results in the temperature range \( 0 \leq T \leq 0.8\ T_N \). For such a procedure, it is necessary to know the antiferromagnetic resonance frequency (AFMR) as a function of temperature. We have used an Impatt diode as a microwave source operating at 84 GHz, because the AFMR frequency in Mn-compounds is rather low.

From the transmission of the samples as function of applied magnetic field, the resonance field \( B_{\text{res}} \) was derived. For the AFMR, \( B_{\text{res}} \) shows a variation of frequency and intensity with temperature as it is expected for an easy-axis antiferromagnet with the applied magnetic field parallel to the easy axis. It is interesting to note that we obtain zero field resonance \( (B_{\text{res}} = 0) \) for 84 GHz at about 25 K. For \( T < 25\ K \) or \( T > 25\ K \), resonance is obtained for the lower and the upper branch, respectively, of the two branches for an antiferromagnet with nearly linear Zeeman shift. With the experimental data for the magnetic susceptibilities of MAMC /4/, the zero field AFMR was calculated for various temperatures (Fig. 1). In addition to the AFMR, a temperature independent resonance line has been observed with an intensity rapidly decreasing below \( T_N \). It is assigned to the paramagnetic resonance of Mn\(^{2+}\) spins uncorrelated due to crystalline disorder. \( B_{\text{res}} = 2.98 \) Tesla for this line agrees well with \( g = 2.0023 \) reported Mn\(^{2+}\) /5/.

4. RENORMALIZED SPIN WAVE THEORY.- Along the lines of Oguchi /6/, of Birgeneau et al. /2/ and of de Wijn et al. /7/, we have performed a renormalized spin wave calculation for \( Rb_2MnCl_4 \) and MAMC based on the AFMR-data and the magnon dispersion. In utilizing these experimental data, we assume that there is no difference in the magnetic properties of normal and deuterated MAMC and that the magnetic structure of MAMC shows no deviation from that of \( Rb_2MnCl_4 \) through the crystal structure of MAMC is more complicated at low temperatures.

From these considerations, we obtain information about the strength of the exchange interaction, about the anisotropy, spin deviation etc. For the sublattice magnetization and for some magnon frequencies, experimental data and calculated values agree quite well in the temperature range \( 0 \leq T \leq 0.8\ T_N \).

References

/6/ Oguchi, T., Phys. Rev. 112 (1960) 117

Fig. 1: Zero field AFMR frequency \( \nu_0 \) for MAMC versus temperature.