

Power-law Form of the Temperature Dependence of the Magnon Specific Heat $C_M(T)$ in Linear, Planar and Spatial Antiferromagnets

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Abstract

The power-law form of the temperature dependence of the magnon specific heat has been calculated for some real, low dimensionality antiferromagnetic crystals. The simple power laws predicted from dimensionality considerations are reproduced in the low temperature, non-interacting spin-wave approximation, but with limited validity.

1. Introduction

Recent neutron scattering investigations on the group of compounds typified by $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$, K_2NiF_4 and the transition metal difluorides have definitely revealed that genuine one-dimensional (linear), two-dimensional (planar) and three-dimensional (spatial) antiferromagnets do exist in nature and as such the study of the behaviour and properties of such compounds is of more than mere academic interest. In this short note we report our findings on the form of the power laws for the temperature dependence of $C_M(T)$ in these materials. The importance of the work lies in the fact that up to now very few workers have really studied:

- (1) the temperature range over which simple power laws of $C_M(T)$ are predicted;
- (2) the validity of the non-interacting spin-wave approximation in such calculations;
- (3) the intrinsic value of such calculations when compared with the results of experiments.

Furthermore it is strongly felt that a knowledge of the behaviour of the specific heat of the real one- and two-dimensional Heisenberg systems must provide a step in the right direction towards the development of a theory for this class of magnetic materials.

The comparison with experimental data is not always quite as easy as it might appear at first sight because of the difficulties which are involved in separating experimentally the various contributions to the total specific heat of a magnetic material.

2. Calculations for $C_M(T)$

It is well known that at low temperatures, i.e. $T \leq \frac{2}{3} T_N$ (Dyson 1956; Bloch 1962, 1963), the magnon contribution to the specific heat comes mainly from magnons

with wavevectors in the vicinity of $k \approx 0$. In the simple theory of antiferromagnetic crystals based on the use of the Heisenberg Hamiltonian and neglecting magnon-magnon interactions etc., the simple power-law expression for $C_M(T)$ for a three-dimensional antiferromagnet is (Kittel 1963)

$$C_M(T) \propto T^3.$$

It follows therefore that $C_M(T)$ should be proportional to $T^{1/\alpha}$ for a linear chain antiferromagnet and proportional to $T^{2/\alpha}$ for a planar antiferromagnet, and that for strictly one- and two-dimensional antiferromagnetic insulators we expect $\alpha = 1$ within the temperature range where the spin-wave theory is valid and for values of T such that $k_B T$ is greater than the energy gap at $k = 0$, i.e. $E_g(0)$.

Table 1. Calculated spin-wave contribution to the specific heat of $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$

Units for T and $C_M(T)$ are K and $\text{mJ mol}^{-1} \text{K}^{-1}$ respectively

T	$C_M(T)$	T	$C_M(T)$	T	$C_M(T)$
0.3	1.16	1.0	151	2.4	909
0.4	4.93	1.2	244	2.6	1020
0.5	13.2	1.4	347	2.8	1140
0.6	27.4	1.6	457	3.0	1250
0.7	48.5	1.8	569	3.2	1360
0.8	76.7	2.0	682	3.4	1480
0.9	111	2.2	796		

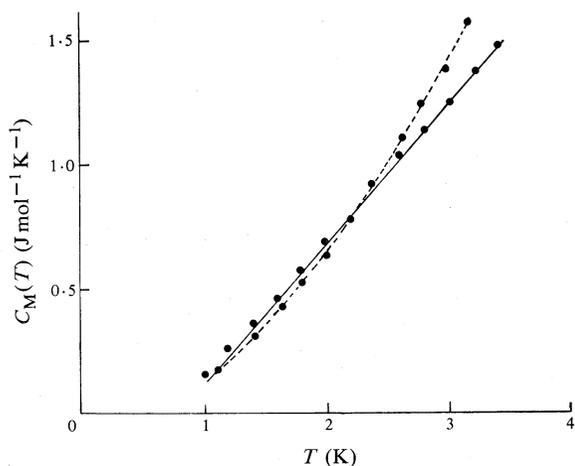


Fig. 1. Variation of the magnon specific heat with temperature in one-dimensional $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$. The solid line is for the present calculation and the dashed line the experimental data of Kopinga (1975).

To verify the existence of such power laws, we calculated the spin-wave contribution to the specific heat in the three materials $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$ (one-dimensional or linear) and K_2NiF_4 and Rb_2MnF_4 which are both planar or two-dimensional. Our earlier studies (Joshua and Cracknell 1969; Begum *et al.* 1969) revealed the existence of such power laws in spatial three-dimensional antiferromagnets and hence we will not repeat the work here.

For $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$ ($T_N \approx 5$ K), we used the following dispersion relation (Skalyo *et al.* 1970):

$$E(\mathbf{k}) = [\{g\mu_B H_A - 4S(J_1 + J_2 + J_3)\}^2 - 16S^2(J_1 \cos \frac{1}{2}ak_x + J_2 \cos bk_y + J_3 \cos \frac{1}{2}ck_z)^2]^{\frac{1}{2}}.$$

Here J_1 , J_2 and J_3 are the nearest neighbour exchange constants for the directions a , b and c respectively and $S = \frac{5}{2}$ for the Mn^{2+} ion. Using the values of the constants quoted by Skalyo *et al.* (1970), we calculated the magnon density of states and then $C_M(T)$. Our results are presented explicitly in Table 1 and shown in Fig. 1 along with the experimental values which were supplied to us by K. Kopinga (personal communication). The experimental data points were obtained (Kopinga 1975) by subtracting the lattice contribution from the total heat capacity. Our calculated results show good agreement with the experimental values within the range $E_g(0)/k_B < T < \frac{2}{5} T_N$ where the spin-wave theory holds good and exhibits an almost perfect T^1 behaviour, justifying the assumption that this material can be regarded as the prototype of a one-dimensional Heisenberg antiferromagnet. It is to be observed however, that our straight line does not pass through the origin. This arises from the fact that the anisotropy energy $g\mu_B H_A$ for $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$ is not quite zero even though it is small in comparison with J_1 . We discuss the question of the power-law form in the vicinity of the origin below.

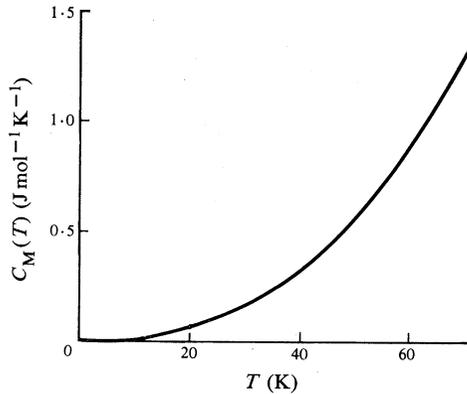


Fig. 2. Variation of the magnon specific heat with temperature in two-dimensional K_2NiF_4 .

For K_2NiF_4 ($T_N \approx 97$ K) we used the dispersion relation given by Parkinson and Loudon (1968) and Suzuki and Kamimura (1970) and the values of the constants given by Skalyo *et al.* (1969). The results are shown in Fig. 2. The experimental value of the total specific heat of K_2NiF_4 has been determined by Salamon and Ikeda (1973), and by comparison with the iso-structural non-magnetic compound K_2MgF_4 the magnetic contribution in K_2NiF_4 has been extracted. However, the Salamon-Ikeda data are not of sufficient sensitivity to make a direct comparison with our calculated values. A plot of $\log C_M$ versus $\log T$ for our values shows that the temperature dependence in the region $E_g(0)/k_B < T < \frac{2}{5} T_N$ gives a perfect T^2 behaviour.

It should perhaps be stressed that the size of the energy gap $E_g(0)$, which in turn is associated with the value of $g\mu_B H_A$, in relation to the dominant exchange constant (J_1 in our case) plays a significant role in determining the power-law forms in the region close to $T = 0$. It is obvious that if a simple power-law behaviour is to be

found, it will not be in the region very close to $T = 0$ but for values of T such that $k_B T$ is greater than this gap energy. The values of $g\mu_B H_A$ and J_1 (in cm^{-1}) for the two materials studied are as follows:

	CsMnCl ₃ ·2H ₂ O	K ₂ NiF ₄
$g\mu_B H_A$	0·03	0·59
J_1	2·5	78

We observe that the anisotropy energy relative to J_1 is much larger for CsMnCl₃·2H₂O than for K₂NiF₄ and as such we expect the power-law behaviour for K₂NiF₄ to be applicable at much lower temperatures than for CsMnCl₃·2H₂O. This is revealed in our calculated results.

Finally it should be noted that in both CsMnCl₃·2H₂O and K₂NiF₄ the values of $g\mu_B H_A$ are relatively small compared with J_1 . However, for the two-dimensional Rb₂MnF₄ ($T_N \approx 38\cdot5$ K), where $g\mu_B H_A$ is of the order of 5% of J_1 (Parkinson 1969), our calculations have revealed no similar T^2 behaviour. In fact the results show that the temperature dependence is of the form T^n where n is not quite constant but varies slowly as a function of T : at $T \approx 4$ K, $n \approx 2\cdot6$ and decreases to give a T^2 behaviour only in the range of T between about 10 and 15 K. This of course arises because of the rather large spin-wave energy gap $E_g(0)/k_B \approx 7\cdot3$ K (deWijn *et al.* 1973), which in turn is associated with the rather large value of $g\mu_B H_A$. However, even for this material the T^2 behaviour is evident in the region $E_g(0)/k_B < T < \frac{2}{3} T_N$. Nevertheless, we conclude that K₂NiF₄ is to be regarded as a better two-dimensional antiferromagnet than Rb₂MnF₄ and should be considered as the prototype of the family of planar antiferromagnets which exhibits the most pronounced two-dimensional features.

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