SPECIFIC HEAT OF THE QUASI 2D-XY HELIMAGNETS BaCo2(AsO4)2 AND BaCo2 (PO4)2

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JOURNAL DE PHYSIQUE Colloque C6, supplément au n° 8, Tome 39, août 1978, page C6-759

SPECIFIC HEAT OF THE QUASI 2D-XY HELIMAGNETS $BaCo_2(AsO_4)_2$ AND $BaCo_2(PO_4)_2$

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Résumé.- Nous avons mesuré la chaleur spécifique C des composés $BaCo_2(AsO_4)_2$ et $BaCo_2(PO_4)_2$ qui présentent un ordre hélimagnétique quasi-bidimensionnel (2-D) du type X-Y. Pour T<2 K, C est proportionnelle à T^2 conformément à la théorie des ondes de spin. A plus haute température C a un comportement exponentiel caractéristique d'une barrière de potentiel. La chaleur spécifique de $BaCo_2(AsO_4)_2$ présente une anomalie lambda étroite à T_N = 5,29 K tandis que pour $BaCo_2(PO_4)_2$ on observe seulement un maximum arrondi vers 5,5 K.

Abstract.- We measured the magnetic specific heat of two quasi-two-dimensional (2-D), XY helimagnets, $BaCo_2(AsO_4)_2$ and $BaCo_2(PO_4)_2$. At temperatures T<2 K, C is proportional to T^2 as expected from spin wave theory. At higher temperatures C has an exponential form characteristic of a potential barrier. The specific heat of $BaCo_2(AsO_4)_2$ exhibits a sharp anomaly at T_N = 5.29 K whereas only a bump is observed around 5.5 K in $BaCo_2(PO_4)_2$.

INTRODUCTION.- The isomorphous compounds $BaCo_2(AsO_4)_2$ and $BaCo_2(PO_4)_2$ crystallize with a rhombohedral structure (space group $R\bar{3}$) /1/. The Co^{2+} ions are located in a honeycomb lattice with a parameter $a/\sqrt{3}$ = 2.89 Å and 2.80 Å respectively; the large distance between neighbouring magnetic planes (c/3 = 7.82 Å and 7.73 Å) is suitable for a quasi 2D-magnetic behaviour. Magnetic measurements on single crystals /2,3,4/ have shown that the magnetic moments lie within the basal plane (X-Y model). Neutron diffraction experiments /2,3,4/ have evidenced a planar helical magnetic ordering, incommensurable for $BaCo_2(AsO_4)_2$ ($\vec{k}$ = [0.261,0,-4/3]) and nearly commensurable for $BaCo_2(PO_4)_2$ ($\vec{k}$ = [0.252,0,1/2]). The interplanar magnetic coupling is non collinear but commensurable with the lattice. In this paper we report low temperature specific heat measurements in order to precise the phase transition and the 2D magnetic interactions.

RESULTS.- The specific heat experiments have been done on sintered polycrystallin samples. The non magnetic compounds $BaMg_2(AsO_4)_2$ and $BaMg_2(PO_4)_2$ have been measured in order to determine the lattice contribution. The results obtained for the four compounds are reported in figure 1a. A sharp lambda-type anomaly is observed at T_N = 5.29 K for $BaCo_2(AsO_4)_2$ whereas for $BaCo_2(PO_4)_2$ only exists a rounded maximum near 5.5 K. Above the transition temperature an important magnetic contribution is present indicating that strong correlations persist.

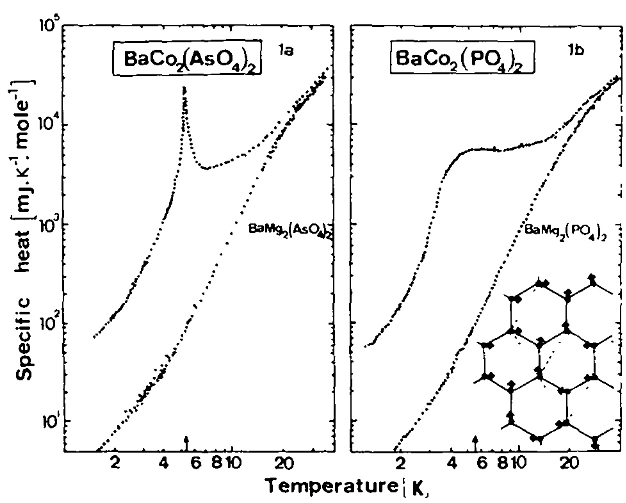

Fig. 1 : Specific heat of $BaCo_2(AsO_4)_2$, $BaMg_2(AsO_4)_2$ (1a) and $BaCo_2(PO_4)_2$, $BaMg_2(PO_4)_2$(1b). In inset is drawn the magnetic structure.

Neutron experiments on $BaCo_2(AsO_4)_2$ single crystal /4/ have shown an abrupt decrease of the staggered magnetization at T_C = 5.33 K and the existence of 2D-correlations up to 40 K. In order to analyse the analyse the magnetic contribution to the specific heat C we have plotted in figure 2a C_m/T^2 versus T. Below T = 2K a $C_m = AT^2$ law is observed, the parameters A are given in table 1. However above T = 2K the specific heat increases much faster than a T^2-law. This additional contribution has in fact an exponential temperature dependence (figure 2b) and C_m can be written : $C_m = AT^2 + Bf(T)e^{-\Delta/T}$

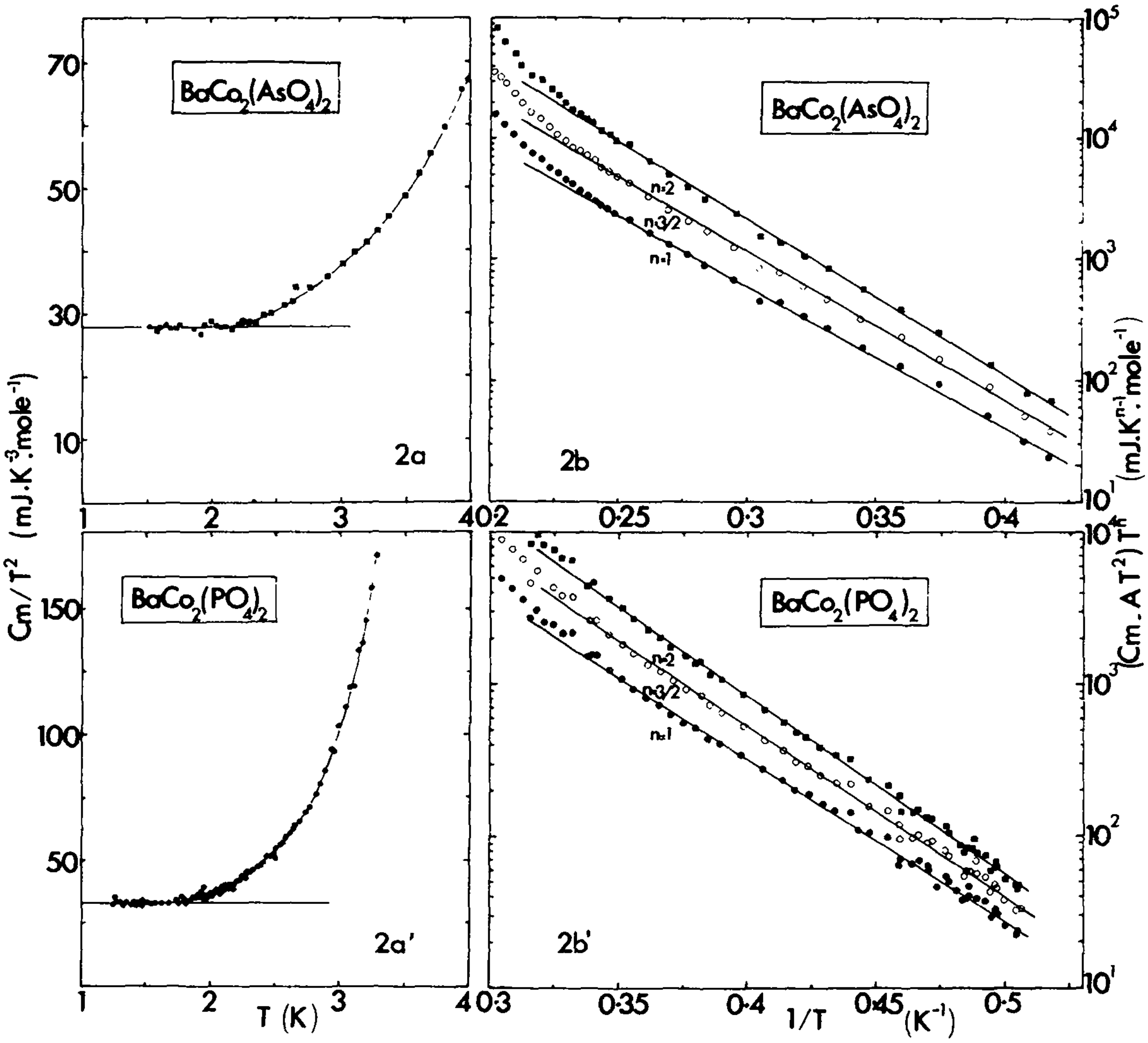

Fig. 2 : Cm/T^2 versus T plot (2a) and semi-log plot to evidence the exponential contribution (2b).

The exponential behaviour does not allow us to determine uniquely the function f(T). Some fits are given in figure 2b for a function $f(T)=T^{-n}$ with n = 1,3/2, 2), the corresponding parameters are reported in Table I.

Table I : Specific heat parameters

Compound	A ($mJ/K^3/mole$)	B ($10^5 mJ.K^{n-1}/mole$) n = 1	n = 3/2	n = 2	Δ (K) n = 1	n = 3/2	n = 2
$BaCo_2(AsO_4)_2$	28	19	67	160	27	29	30
$BaCo_2(PO_4)_2$	33	56	154	250	24.5	25.5	27

The observation of both a power and an exponential laws is unusual for magnetic systems ; however this behaviour has been observed for superfluid helium where the exponential contribution arises from the roton excitations.

DISCUSSION.- The term in T^2 can be accounted for by spin wave excitations. Spin waves in a helimagnet have a linear dispersion law, for small wave vector, which give rise to a low temperature specific heat proportional to T^D, where D is the lattice dimensionality. So the T^2 law means that for low temperatures spin waves propagate only within the magnetic planes, indicating that the interplanar coupling is very weak. Using the exchange integral ratios determined in /2/ for $BaCo_2(AsO_4)_2$ (J_2/J_1=0.12, J_3/J_1=-0.51), the A-coefficient can be accounted for quantitatively with J_1 = 49 K. The exponential contribution observed above 2K is very surprising in the present system (because of the continuous magnetic symmetry) and cannot be explained by otpical magnons, which vould lead to a too large energy gap ($\Delta \simeq 130$ K). Yet other types of excitations can appear in the system if we note that the magnetic structure can be described by quasi-ferromagnetic chains, coupled together nearly at 90°. The coupling between neighbouring chains is very weak and the magnetic excitations consist in turning the moments of segments of chains.

These excitations look like solitons and may be compared with vortex excitations which have been predicted in 2D-XY magnetic systems.

References

/1/ Eymond, S., Durif, A., Mat Res. Bull. 4 (1969) 595.

/2/ Regnault, L.P., Thesis, University of Grenoble (1969).

/3/ Regnault, L.P., Burlet, P., Rossat-Mignod, J., Physica 86-88B (1977) 660.

/4/ Regnault, L.P., Rossat-Mignod, J., to be published.