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J. Jamet, J. Ferré, I. Yamada, M. Itoh. MAGNETIC ENERGY IN A 2D-HEISENBERG MIXED CRYSTAL WITH COMPETING EXCHANGE INTERACTIONS:  $K_2\text{Cu}_x\text{Mn}_{1-x}\text{F}_4$ . Journal de Physique Colloques, 1988, 49 (C8), pp.C8-1501-C8-1502. 10.1051/jphyscol:19888691 . jpa-00228925

**HAL Id: jpa-00228925**

**<https://hal.science/jpa-00228925>**

Submitted on 4 Feb 2008

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## MAGNETIC ENERGY IN A 2D-HEISENBERG MIXED CRYSTAL WITH COMPETING EXCHANGE INTERACTIONS: $K_2Cu_xMn_{1-x}F_4$

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**Abstract.** — We measured the  $a$ - $c$  temperature variation of the optical linear birefringence of mixed  $K_2Cu_xMn_{1-x}F_4$  single crystals for several values of  $x$  ( $0 < x < 1$ ). It may be expressed as a combination of static two spin correlation functions for Cu-Cu, Mn-Mn and Cu-Mn pairs of ions. We deduce the Cu-Mn exchange integral  $J(\text{Cu} - \text{Mn}) \simeq -43 \text{ cm}^{-1}$ .

Despite the sustained theoretical interest in low dimensional mixed magnetic systems with competing nearest neighbor exchange interactions there are few experimental data on corresponding compounds [1, 2]. This paper focusses on the study of short range magnetic interactions in  $K_2Cu_xMn_{1-x}F_4$ , a randomly mixed quasi two-dimensional system, by means of the optical linear birefringence.  $K_2MnF_4$  has the same body centered tetragonal structure as  $K_2NiF_4$  while for  $K_2CuF_4$  this structure is slightly modified by a cooperative Jahn-Teller effect providing in  $c$ -planes alternate distortions of the  $F^-$  octahedra surrounding the  $Cu^{2+}$  ions.  $K_2MnF_4$  stands as a good 2d ( $S = 5/2$ ) Heisenberg antiferromagnet which nevertheless orders at  $T_N = 42.1 \text{ K}$ , while  $K_2CuF_4$  may be considered as a good 2d ( $S = 1/2$ ) Heisenberg ferromagnet ( $T_c = 6.25 \text{ K}$ ) since the orbital ordering of  $Cu^{2+}$  ions favors ferromagnetic exchange interactions in  $c$ -plane. The exchange interaction  $J(\text{Mn-Mn})$  in  $K_2Cu_xMn_{1-x}F_4$  remains antiferromagnetic in the whole concentration range  $0 < x < 1$ . Magnetic frustration occurs as long as the Cu-Cu exchange interaction is ferromagnetic, i.e. for  $x \geq 0.4$  [2, 3].

The magnetic ( $x, T$ ) phase diagram of  $K_2Cu_xMn_{1-x}F_4$  has been determined recently [2]. At very low temperature the compound is antiferromagnetic up to  $x = 0.5$  and ferromagnetic for  $x > 0.8$ . A spin glass like phase ( $0.5 < x < 0.8$ ) has been evidenced up to  $T_g = 0.5 \text{ K}$ , as expected in such a frustrated system.

The  $a$ - $c$  magnetic contribution  $\Delta n_m$  to the linear birefringence (LB) of pure or diamagnetically doped uniaxial ( $//c$ ) Heisenberg magnetic materials is proportional to their magnetic energy [4] so the magnetic specific heat can be deduced from the temperature derivation of  $\Delta n_m$  [4]. The magnetic energy of  $K_2Cu_xMn_{1-x}F_4$  is expressed by a linear combination of the individual energies  $E_m^\alpha$  relatively to the different pairs ( $\alpha$ ) of magnetic ions, weighted by their bonding probabilities  $P(\alpha)$ . Limiting ourselves to the strongest

exchange integrals, we can evaluate:

$$P(\text{Mn} - \text{Mn}) = (1 - x)^2 / X, \quad P(\text{Cu} - \text{Cu}) = x^2 / X$$

$$P(\text{Cu} - \text{Mn}) = x(1 - x) / X, \quad X = x^2 - x + 1. \quad (1)$$

It may be shown that  $\Delta n_m(\alpha)$  is proportional to each  $E_m^\alpha$  but the total birefringence:

$$\Delta n_m = \sum_{\alpha} A_{\alpha} \Delta n_m(\alpha) = \sum_{\alpha} A_{\alpha} P(\alpha) E_m^\alpha \quad (2)$$

is never proportional to the magnetic energy since the  $A_{\alpha}$  coefficients are a priori different. The quantity  $d(\Delta n_m)/dT$  is then expressed by a linear combination of the different pair contributions to the specific heat  $C_m$ . The temperature variation of the LB of  $K_2Cu_xMn_{1-x}F_4$  (Fig. 1a) may be decomposed into two contributions: the lattice part  $\Delta n_l$ , the thermal dependence of which being assumed to follow a Debye law, and  $\Delta n_m$ . We have checked the similarity of the  $a$ - $c$  LB for  $a \equiv \langle 100 \rangle$  or  $\langle 110 \rangle$  type of axes. For  $K_2MnF_4$  and  $K_2CuF_4$  pure crystals the variation of  $\Delta n_l$  with temperature is weak compared to that of  $\Delta n_m$  (Fig. 1a). As in the case of  $K_2Cu_xZn_{1-x}F_4$  [5] it is reasonable to suppose that  $\Delta n_l$  is weak for  $K_2Cu_xMn_{1-x}F_4$  and varies monotonously with  $x$  from  $K_2MnF_4$  to  $K_2CuF_4$ . The temperature derivatives of the LB are reported on figure 1b. As already discussed previously [4] the  $d(\Delta n_m)/dT$  curves, obtained for the pure  $K_2MnF_4$  and  $K_2CuF_4$  crystals are similar to the predictions for  $C_m(T)$  in 2d-Heisenberg systems.

The low temperature  $d(\Delta n)/dT$  (or specific heat) anomaly is due to the ferromagnetically coupled Cu-Cu pairs. Its disappearance for  $x \leq 0.38$  is consistent with the absence of cooperative Jahn-Teller effect below  $x = 0.4$  [2, 3]. The manganese 2d anomaly, having a maximum around 50 K for  $K_2MnF_4$ , disappears gradually as  $x$  increases in agreement with the  $P(\text{Mn-Mn})$  variation (Eq. (1)). The sharp peak appearing on  $d(\Delta n)/dT$  curves for  $x = 0, 0.25, 0.38$  reveals the

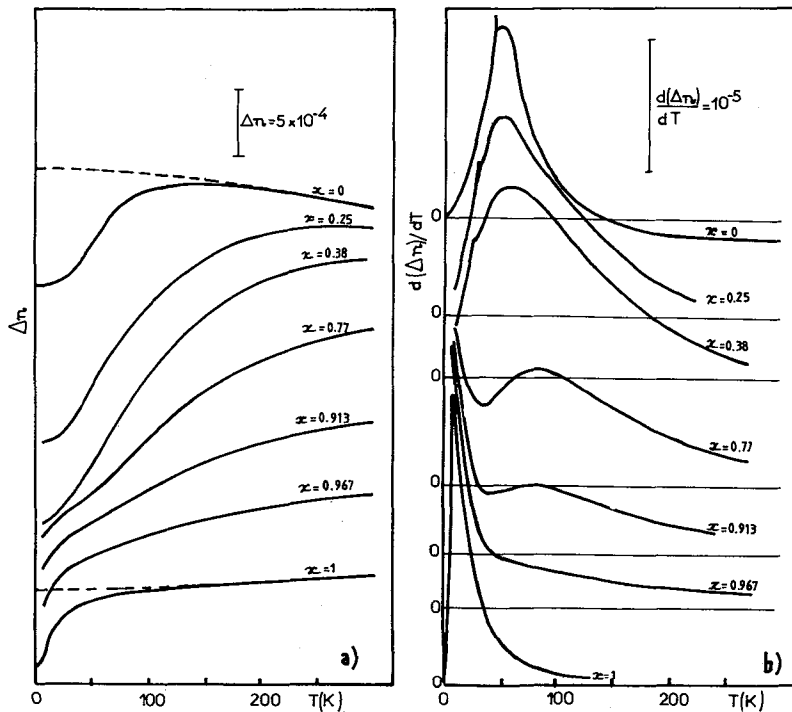


Fig. 1. - Temperature dependence of the  $a$ -c LB (a), and of its derivative (b) for several  $K_2Cu_xMn_{1-x}F_4$  samples. The dashed lines represent the lattice part variation of the LB for pure crystals.

onset of the 3d ordering; the corresponding values of  $T_N(x)$  agree with previous data [2].

A new Schottky anomaly, peaked around 85 K and associated with an increase of the LB, appears in the mixed compound. The dependence of its amplitude with  $x$  is well explained from the  $P(\text{Cu-Mn})$  variation and not by that of  $P(\text{Mn-Mn})$  (Eq. (1)). We interpreted this anomaly within the energy level scheme we introduced to fit the temperature dependence of the Cu-Mn electric dipole optical transitions [6]. From the results of  $x = 0.91$  to  $0.77$ , we have deduced a comparatively large value of the exchange integral:  $J(\text{Cu-Mn}) = -43 \text{ cm}^{-1}$  from  $kT_{\text{max}}(C_m^{\text{Cu-Mn}}) = 1.4 \text{ J}$ . However this value cannot be applicable for  $x < 0.4$ , because the cooperative Jahn-Teller distortion is not effective over this region.  $J(\text{Cu-Mn})$  is also antiferromagnetic so that it explains the rather smooth decrease of  $T_N(x)$  by varying  $x$  from 0 to 0.5 as compared with that of the diamagnetically diluted compound  $K_2Mg_xMn_{1-x}F_4$  (for example  $T_N(x = 0.18) / T_N(x = 0) = 0.5$  and  $0.83$  for the Mg and Cu doped  $K_2MnF_4$  crystal, respectively) [7]. The large Cu-Mn exchange interaction prevents upon the breaking of antiferromagnetic bonds in  $K_2Cu_xMn_{1-x}F_4$  and tends to preserve the

long range order. It is already antiferromagnetic at low temperature for manganese concentrations smaller than that of the percolation threshold.

This work illustrates the use of LB in the study of the different spin correlation functions involved in mixed magnetic materials.

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