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E. Lammers, J. Verstelle, A. van Duynveldt, C. Lowe, R. Carlin. MAGNETIC ORDERING IN [CH<sub>3</sub>CH<sub>2</sub>NH<sub>3</sub>] FeCl<sub>4</sub>. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-1465-C8-1466. 10.1051/jphyscol:19888673 . jpa-00228905

**HAL Id: jpa-00228905**

**<https://hal.science/jpa-00228905>**

Submitted on 4 Feb 2008

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## MAGNETIC ORDERING IN $[\text{CH}_3\text{CH}_2\text{NH}_3]\text{FeCl}_4$

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**Abstract.** – The magnetic ordering of  $[\text{CH}_3\text{CH}_2\text{NH}_3]\text{FeCl}_4$  is studied through ac susceptibility and magnetization measurements. The results can be interpreted as ferromagnetic chains that order antiferromagnetically. Non-linear susceptibilities are reported to be an elegant technique to examine this kind of systems.

The compound  $[\text{CH}_3\text{CH}_2\text{NH}_3]\text{FeCl}_4$  (MEIC) contains magnetic Fe(III),  $S = 5/2$ ,  $g = 2$  ions which are expected to show magnetic interactions of the Heisenberg-type due to the isotropic character of the Fe ion. The crystal structure of MEIC is not known, but the almost similar compound  $[(\text{CH}_3\text{CH}_2)_4\text{N}]\text{FeCl}_4$  belongs to space group  $P6_3m$  and is chainlike as the  $\text{FeCl}_4$ -tetrahedra are placed on top of each other with one Cl-atom pointing along the chain. It seems likely to assume that the MEIC-compound also consists of discrete  $\text{FeCl}_4$ -tetrahedra [1] and low temperature studies of the magnetic properties should elucidate the interactions between the Fe ions [2].

The present research deals with measurements of the ac-susceptibility at low frequencies by means of the mutual inductance technique [3]. The experiments were concentrated on the temperature regime below 4 K and external magnetic fields up to 1 kOe.

The zero-field susceptibility  $\chi_0$ , shown in figure 1, is characteristic for 3-dimensional ordering of the antiferromagnetic-type. The curve in figure 1 extrapolates

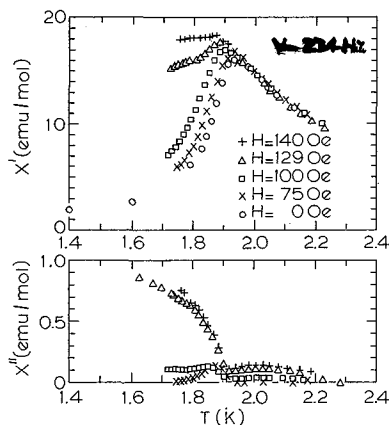


Fig. 1. – Measured in phase susceptibility  $\chi'$  and out-of-phase susceptibility  $\chi''$  as a function of temperature for various external magnetic fields. The frequency of the ac-field is 234 Hz.

down to below 1 emu/mol at 0 K, indicating an alignment parallel to the easy axis to within 4 degrees. Considering the difficulty of mounting the fragile crystal, it seems justified to conclude the crystal  $c$  axis to be the easy axis of the antiferromagnetic ordering. The ordering temperature was obtained from the maximum  $d\chi_0/dT$  as  $T_c = 1.89(3)$  K. The maximum susceptibility in figure 1 is quite large, which shows the necessity for correcting for demagnetizing effects. Our estimate for the demagnetizing factor  $1/N$  on basis of the approximate crystal dimensions agrees well with the average experimental value of 20 emu/mol. The large susceptibility can be explained if, despite the antiferromagnetic order, a large ferromagnetic interaction occurs also. Such interactions are indeed present as the high temperature zero-field susceptibility shows a positive  $\theta$ -value of 2.52 K.

The correction of the zero-field data with  $1/N = 20$  emu/mol leads to a maximum susceptibility of 83 emu/mol. Applying the mean field relation for a two sublattice antiferromagnet with antiferromagnetic interactions between ferromagnetic sublattices [4]  $\chi_{\max} = Ng^2\mu_B^2 / 4z_{AF}|J_{AF}|$  one obtains  $z_{AF}|J_{AF}|/k = 5 \times 10^{-3}$  K. The above results allow a magnetic linear chain to be present. A fit of the data to the so-called Fisher curve [2] is indeed possible, and leads to  $z_F J_F / k = 0.5$  K, but the temperature region is not particularly wide. In fact deviations at the low  $T$  side suggest another significant ferromagnetic interaction to be present.

The data for  $\chi$  vs.  $T$  in a non-zero external magnetic field showed a behaviour that resembled the zero-field curve up to  $H = 100$  Oe. Above that field the susceptibility does not decrease at temperatures below  $T_c$ , indicating the character of the ordering to be different. This effect was further analysed by measuring  $\chi$  vs.  $H$  at temperatures below  $T_c$ . The characteristics of the data can be seen from the example in figure 2. At about 105 Oe the in-phase susceptibility rises to about 20 emu/mol, decreasing again at fields of several hundred oersteds. At the same field range out-of-phase signals are present.  $M$  vs.  $H$  data were also performed

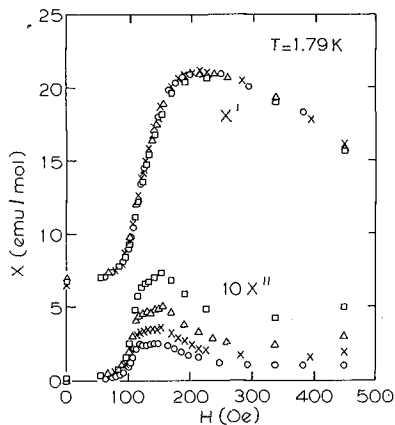


Fig. 2. -  $\chi'$  and  $\chi''$  as a function of external magnetic field at  $T = 1.79$  K. (o):  $\nu = 21$  Hz, (x):  $\nu = 41$  Hz, ( $\Delta$ ):  $\nu = 83$  Hz and ( $\square$ ):  $\nu = 234$  Hz.

and it is clear that up to about 450 Oe a mixed phase exists. Presumably at a field of 105 Oe a first order transition to a ferromagnetic state occurs, the mixed phase arises due to demagnetizing effects. Applying mean field theory, the field of the first order phase transition leads to  $z_{AF} |J_{AF}| / k = 3 \times 10^{-3}$  K. The relevant results for the transition temperatures and fields were collected in a phase diagram. The smooth curve starting at 1.79 K ends at  $H = 105$  Oe at 0 K, and allows the determination of the temperature  $T_t$  where the transition changes from second order to first order. Using the relation  $T_t / T_c - 1 = z_{AF} |J_{AF}| / 3z_F J_F$  [2] leads to  $z_F J_F / k = 3 \times 10^{-2}$  K in agreement with the analysis of the Fisher curve, where it became clear

that, apart from the one of 0.5 K, another significant ferromagnetic contribution should exist.

The study of non-linear susceptibilities is not a standard experiment in low temperature magnetism. In general, non-linear effects are expected whenever the magnetization shows a discontinuity. We performed some measurements in which the primary coil was activated at a frequency  $\nu_1$ , while the output of the secondary ones was detected at  $2\nu_1$  in order to determine  $\chi_2$ . At 105 Oe a sharp spike in  $\chi_2$  was found, indicative for the first order phase transition. In fact our preliminary results demonstrated that an experiment involving non-linear susceptibilities is a simple but elegant way to determine a magnetic phase diagram. For instance, the study of  $\chi_2$  should give a direct way to decide upon the order of the phase transition where usual methods through  $\chi$  or  $M$  are complicated by relaxation effects [5]. We propose a further analysis of the use of non-linear susceptibilities in the study of ordered magnetic compounds as an additional tool for the determination of the phase diagram from which the microscopic exchange interactions can be estimated.

- [1] Zora, J. A. and Seddon, K. R., private communication.
- [2] Carlin, R. L. and van Duyneveldt, A. J., *Magnetic Properties of Transition Metal Compounds* (Springer Verlag, New York) 1977.
- [3] Van Duyneveldt, A. J., *J. Appl. Phys.* **53** (1982) 8006.
- [4] Algra, H. A., de Jongh, L. J., Huiskamp, W. J. and Carlin, R. L., *Physica B* **92** (1977) 187.
- [5] Groenendijk, H. A. and van Duyneveldt, A. J., *Physica B* **115** (1982) 41.