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Magnetic susceptibility of the compound $(\text{CH}_2)_{10}(\text{NH}_3)_2\text{FeCl}_4$

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Abstract. The magnetic susceptibility of the compound $(\text{CH}_2)_{10}(\text{NH}_3)_2\text{FeCl}_4$ is measured in the temperature range from liquid nitrogen temperature up to room temperature. The effect of thermal and magnetic history on the data obtained is also discussed.

Introduction

During the last few years [1, 2] there was an increasing interest in the examination of two-dimensional magnetic systems. Recently new two-dimensional antiferromagnetic compounds containing Fe^{2+} and Mn^{2+} ions have been found [3–5]. In our laboratory, it was found that, the compounds of the general formula $(\text{C}_n\text{H}_{2n})(\text{NH}_3)_2\text{FeCl}_4$ [6, 7] ($n = 1, 2, 3, \dots$) are known to form good examples of the two-dimensional Heisenberg antiferromagnets with weak ferromagnetic components between the planes. In the compounds of the above general formula, the ferromagnetism appeared below the Neel temperature is attributed to the spin canting in the antiferromagnetically ordered state. Below the ordering temperature, the magnetic properties are found to show unusual dependence on the thermal and magnetic history of the sample.

The crystal structure of all compounds of the series $(\text{C}_n\text{H}_{2n})(\text{NH}_3)_2\text{MCl}_4$, where $n = 3$ and $M = \text{Mn}, \text{Fe}$ and Cu have been determined [1, 8]. The rest of the series where $n = 2, 4, 5, \dots$ are isomorphous to these compounds and crystallizes in the orthorhombic space group Imma or monoclinic. The structure is made up of metal halogen layers each of MCl_6 octahedra shears four corners with neighbouring octahedra in the layer. The layers are separated by $(\text{C}_n\text{H}_{2n})(\text{NH}_3)_2$ groups such that NH_3 groups are hydrogen bonded to the $\text{M}-\text{Cl}$ network. Changing the number of carbon atoms in the series will change the crystal structure of the compounds while in some compounds the symmetry is changed by changing the temperature. It was found that the compounds with even

numbers of carbon atoms have a monoclinic unit cell at room temperature [6, 7] while those with odd numbers of carbon atoms have orthorhombic unit cell except those of $n = 7$ and 8 where both have monoclinic unit cell at room temperature.

In the present work we report on the magnetic susceptibility of $(\text{CH}_2)_{10}(\text{NH}_3)_2\text{FeCl}_4$ compound as related to its magnetic and thermal properties.

Experimental

The sample under investigation is prepared by taking stoichiometric amount of $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ and recrystallized $(\text{CH}_2)_{10}(\text{NH}_3)_2\text{Cl}_2$ with an excess of about one per cent of the alkylammonium chloride. The stoichiometric amounts are dissolved in minimum acidified distilled water under a stream of oxygen free nitrogen gas to prevent oxidation of the sample. Heating the mixture to about one hour at about 80°C is carried out. The solution is slowly cooled down to room temperature during one day. The resulting greenish yellow crystals are redissolved in hot acidified distilled water and the crystallization procedure is repeated again.

Grinding and mounting the sample is carried out in dry glove box under purified nitrogen gas. Complete sealing of sample tube after mounting is also carried out. Chemical analysis of the sample is carried out before collecting the data and the results are inserted in Table I.

The magnetic susceptibility of the powdered sample is measured using the conventional Gouy method which is modified to measure in liquid nitrogen temperature and above. The data obtained are corrected for diamagnetism by measuring the magnetic susceptibility of the isomorphous Cd compound.

Results and discussion

The results of the corrected molar magnetic susceptibility for the compound $(\text{CH}_2)_{10}(\text{NH}_3)_2\text{FeCl}_4$ as a function of temperature is shown in Fig. 1(a). The data are collected in temperature range from liquid nitrogen up to 300 K. The observed peak at ≈ 93 K indicating the antiferromagnetic character of this compound. This peak is partially masked by the superimposed large susceptibility spike of the spin canted interaction. From Fig. 1 it is also clear that the data are

Table 1
Chemical analysis of the compound $(\text{CH}_2)_{10}(\text{NH}_3)_2\text{FeCl}_4$

Compound	C%		H%		N%		Cl%	
	fou.	cal.	fou.	cal.	fou.	cal.	fou.	cal.
$(\text{CH}_2)_{10}(\text{NH}_3)_2\text{FeCl}_4$	32.40	32.27	6.85	6.99	7.56	7.53	38.10	38.19

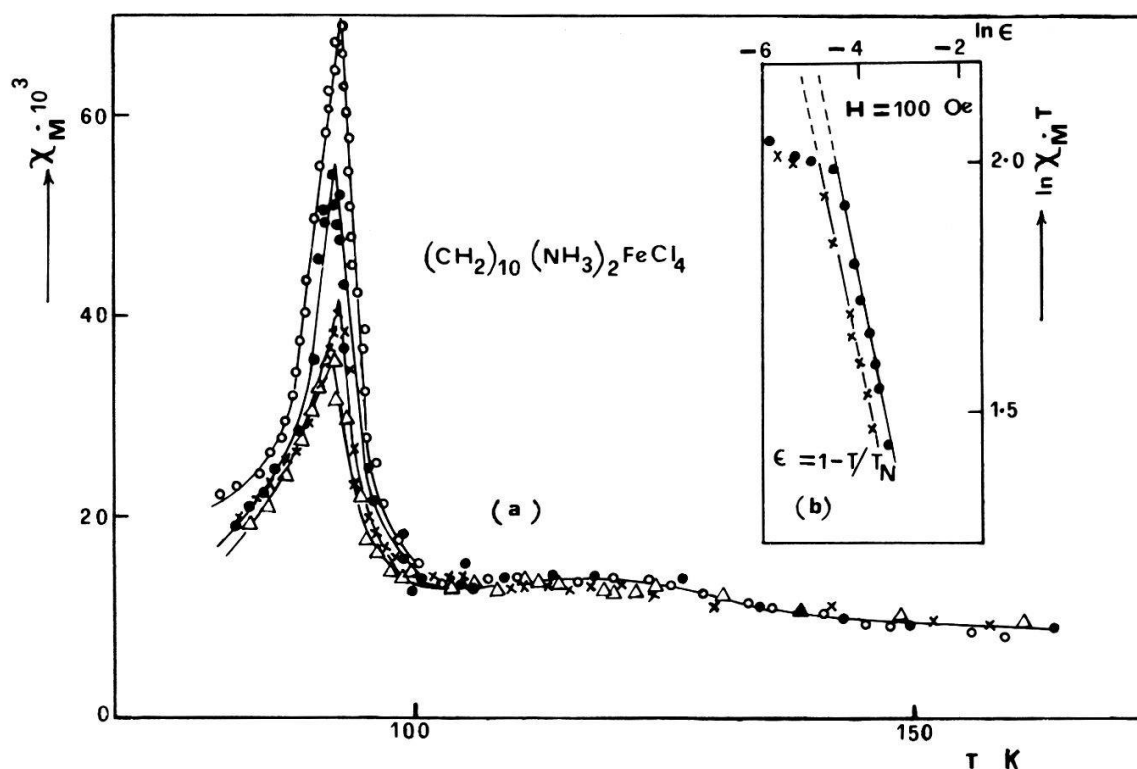


Figure 1

(a) Temperature dependent of the magnetic susceptibility at different values of the magnetic field. $\triangle \triangle \triangle$ 1510 Oe; $\times \times \times$ 1002 Oe; $\bullet \bullet \bullet$ 500 Oe; $\circ \circ \circ$ 250 Oe. (b) Relation between $\ln C$ and $\ln \chi_M \cdot T$ at 100 Oe.

collected at different magnetic field strengths. For clarity only four values of the magnetic field are taken. The effect of variation of the magnetic field on the peak position is small to be neglected but it has a drastic effect on the peak value, such that the peak is decreased by increasing the magnetic field which is a characteristic of ferromagnetic interaction between the magnetic layers. Thus it is to be assumed that the antiferromagnetic intralayer exchange interaction exist at the same time with weak ferromagnetic interlayer interaction. The presence of a broad hump centered around 125 K indicating the presence of short range order interaction in this compound. The values of the critical point exponents γ and γ' as calculated from Fig. 1(b) are found to be 1.85 indicating that the system agrees quite well with the Ising 2-dimensional model. The value of the Curie-Weiss constant $\theta = 150$ K yielding the value of the exchange interaction constant $|J/K| = 28.12$ K as calculated from MF theory [9]. The value of the Curie constant $C = 3.25$ giving the effective magnetic moment of 5.1 BM. The obtained value of the effective magnetic moment agrees nicely with the reported values [10] of Fe^{2+} ions in different symmetry.

Figure 2(a) shows the temperature dependence of the magnetic susceptibility taking into consideration the magnetic history of the sample. In this type of measurement, which is not considered by Blake et al. [11] in case of $(4-ClC_6H_4NH_3)_2FeCl_4$ compound. The sample is cooled from $T > T_N$ down to liquid nitrogen in different magnetic fields and the data are collected at the same

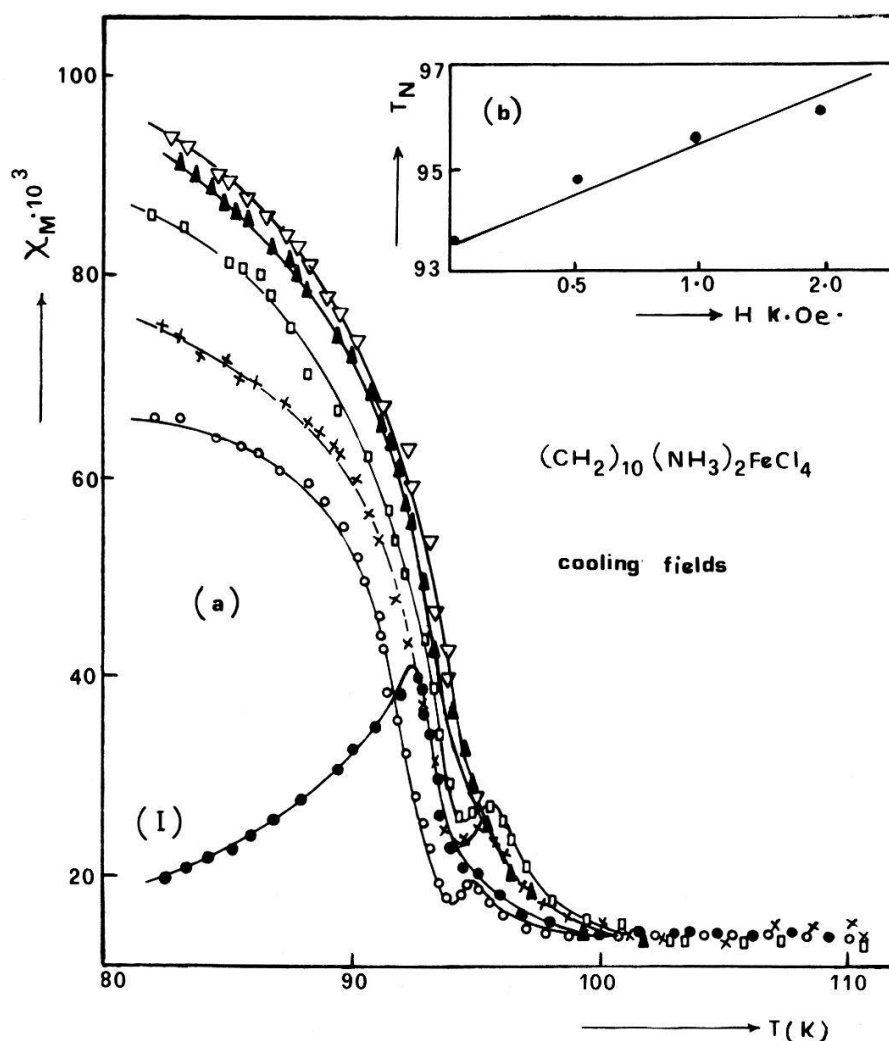


Figure 2
(a) Temperature dependent of the magnetic susceptibility at different cooling fields. ●●● Cooling at zero magnetic field; ○○○ Cooling at 500 Oe; ××× Cooling at 1002 Oe; □□□ Cooling at 1510 Oe; ▲▲▲ Cooling at 1990 Oe; ▽▽▽ Cooling at 2500 Oe; (b) The effect of the cooling fields on the transition temperature (T_N).

magnetic field strength of 1002 Oe. The cooling field will force the spins to be aligned up in its direction. Decreasing the temperature of the sample while the magnetic field is switched on will cause the presence of mixed phases from antiferromagnetic and spin flop up. The relative value of the two phases depends on the cooling field itself. The study of magnetic phase diagram of a group of compounds of the above series is now under study and it will help in the interpretation of this abnormal behaviour. From Fig. 2(a) it is clear that, the cooling field does not affect on susceptibility values after ≈ 100 K, but at the same time affect on the peak position and value as in Fig. 2(b). This is quite different from the case of $(\text{CH}_2)_6(\text{NH}_3)_2\text{FeCl}_4$ [12]. The variation of T_N with the cooling field indicates that the exchange interaction constant J is slightly dependent on such field.

The values of the magnetic susceptibility at liquid nitrogen as a function of the cooling field is shown in Fig. 3. The data indicates that saturation to sublattice magnetization occurs at ≈ 2500 Oe. Over all the data obtained can be discussed

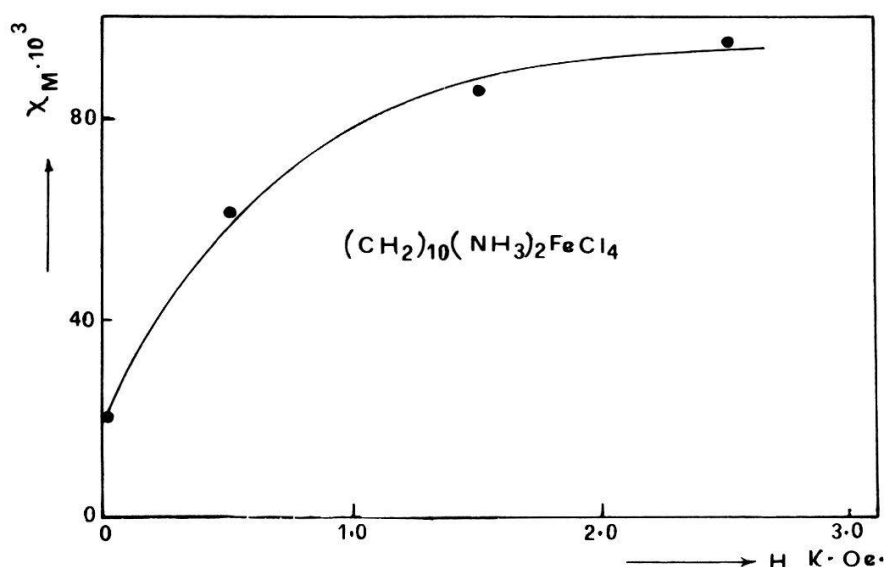


Figure 3

Saturation to the sublattice magnetization at about 2500 Oe, which is cleared from the relation between the magnetic susceptibility at liquid nitrogen and the cooling fields.

on the basis of the domain theory. This theory depends on the increasing of both exchange energy and anisotropy energy at the moment of domain wall formation. At liquid nitrogen temperature (below T_N) the susceptibility value is small as in Fig. 1, due to small dependence of domain wall on the magnetic field at this low temperature. Increasing the temperature of the sample will show temperature dependent susceptibility because the domain wall will be damped at this still low temperature. When cooling the sample in zero external magnetic field [13] as in curve (I) Fig. 2(a), the magnetic moments of the domain walls starts to develop in different directions in different parts of the sample and there will be a net magnetization of the antiferromagnetic domains which is a metastable state. This metastable state will persist unless a large enough magnetic field is applied to allow the domain wall to move from their original positions. But when cooling the sample through T_N in an external magnetic field, one can assume that a weakly ferromagnetic domains accompanied by structural distortion are formed. This structural distortion is appeared in a form of shoulders as in the rest of curves in Fig. 2(a).

In conclusion one can say that, from the magnetic susceptibility behaviour and from the assumed monoclinic structure of this compound, one can say that the compound approximates two-dimensional magnetic system such that the spins aligned antiferromagnetically in the layer with weak ferromagnetic coupling between the layers.

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