JOURNAL OF MATERIALS SCIENCE 28 (1993) 1289–1292

# Magnetic susceptibility of the two-dimensional $Cu^{2+}$ complex

A. Z. MOHAMED, S. S. SALEEM, M. A. EI-GHANDOUR
*Physics Department, Faculty of Science, Zagazig University, Zagazig, Egypt*
M. A. AHMED
*Physics Department, Faculty of Science, Cairo University, Giza, Egypt*

The magnetic susceptibility behaviour of $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2$ M $Cl_{4-x}Br_x$ compounds, where M is $Cu^{2+}$ and $x=0$, 1, 2, were studied in the temperature range 300–390 K. The effective magnetic moment, Curie constant and Curie–Weiss constant for $Cu^{2+}$ were calculated. In addition, the effect of replacing $Cl^{-1}$ by $Br^{-1}$ ions in the compounds was studied. Differential scanning calorimeter thermographs were used to check the phase-transition points.

## 1. Introduction

Recent studies of the magnetic interactions for the two-dimensional magnetic systems that have the general formula $(R\text{–}NH_3)_2MX_4$, where R is a linear chain alkyl group, M is a divalent transition metal of the first transition series, and X is $Cl^{-1}$ or $Br^{-1}$, ions have received considerable attention [1–5]. These complexes were formed of infinite sheets (layers) of corner-sharing octahedral anions [6]. These parallel layers were separated by non-magnetic ions. This separation could be increased to the extent that the magnetic interactions between these layers is about $10^{-5}$ times the interaction within the layer [7]. The magnetic behaviour of this layered-type structure could be explained by two models, namely the Ising and Heisenberg: the Ising model is used whenever the magnetic lattice is completely anisotropic, while the Heisenberg model is applicable for the isotropic structures.

The aim of the present work was to study the effect of replacing $Br^{-1}$ ions instead of $Cl^{-1}$ ions (radius of $Br^{-1} = 0.195$ nm, radius of $Cl^{-1}$ ions $= 0.181$ nm [8] on the magnetic susceptibility of the aniline complexes at different magnetic fields.

## 2. Experimental procedure

The aniline complexes in the present study were prepared by dissolving stoichiometric amounts of aniline hydrochloride or hydrobromide and $CuCl_2\text{–}2H_2O$ with ratio of 2:1 in distilled water, respectively, as reported by Remy and Laves [9]. The solution was then allowed to evaporate yielding the compounds. Recrystallization was carried out by using acidified triply distilled water and washing the samples obtained with a mixture of ethanol and ether. Grinding the compounds obtained to fine powders was carried out before filling the susceptibility tube.

The conventional Gouy method [10] has been used to measure the magnetic susceptibility of the powdered samples with an accuracy better than 2%. In addition, the measured values of the susceptibility were corrected for diamagnetism using Pascal's table [11].

The differential scanning calorimetry (DSC) thermographs in the high-temperature region were taken of these compounds before collecting the data.

TABLE I List of Curie–Weiss constants, θ for the compound $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_{4-x}Br_x$, $x = 0, 1, 2$

| Compound | Curie–Weiss constant [6] | | |
|---|---|---|---|
| | 3050 Oe | 3700 Oe | 4200 Oe |
| $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_4$ | 295 | 287 | 285 |
| $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_3Br$ | 327 | 285 | 275 |
| $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_2Br_2$ | 292 | 280 | 257 |

TABLE II List of Curie constants-C, and effective magnetic moments, $M_{eff}$, for the compound $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_{4-x}Br_x$; $x = 0, 1, 2$

| Compound | $C$ (K) | $M_{eff}$ (BM) |
|---|---|---|
| $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_4$ | 0.40 | 1.80 |
| $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_3Br$ | 0.40 | 1.80 |
| $(m\text{-}NO_2C_6H_4NH_3)_2CuCl_2Br_2$ | 0.48 | 1.96 |

## 3. Results and discussion

The results of the corrected molar magnetic susceptibility as a function of absolute temperature for the compound $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_4$ at different magnetic fields are shown in Fig. 1a. The data were collected in the temperature range 295 K upto or near the melting point of the present compound. From Fig. 1a it is clear that the magnetic susceptibility decreased with increasing temperature.

0022–2461 

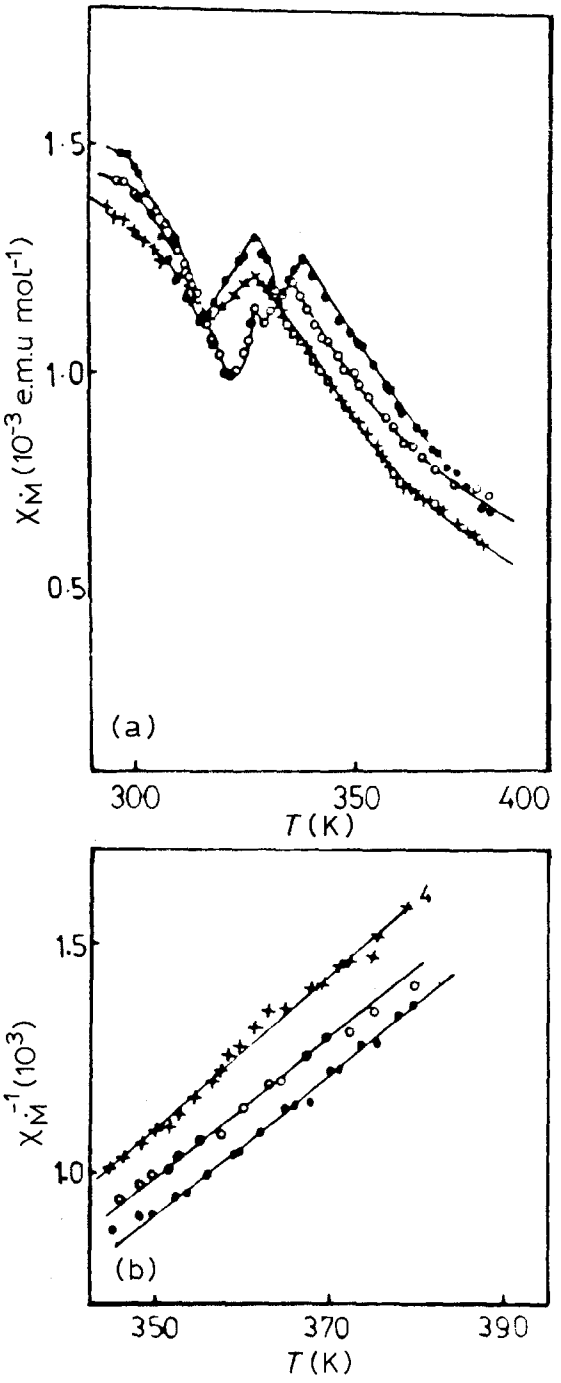


*Figure 1* (a) Temperature dependence of the magnetic susceptibility at different magnetic fields. (b) Relation between temperature versus reciprocal of the magnetic susceptibility at different magnetic fields; for $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2\ CuCl_4$ compound. (●) 3050 Oe, (○) 3700 Oe, (x) 200 Oe.

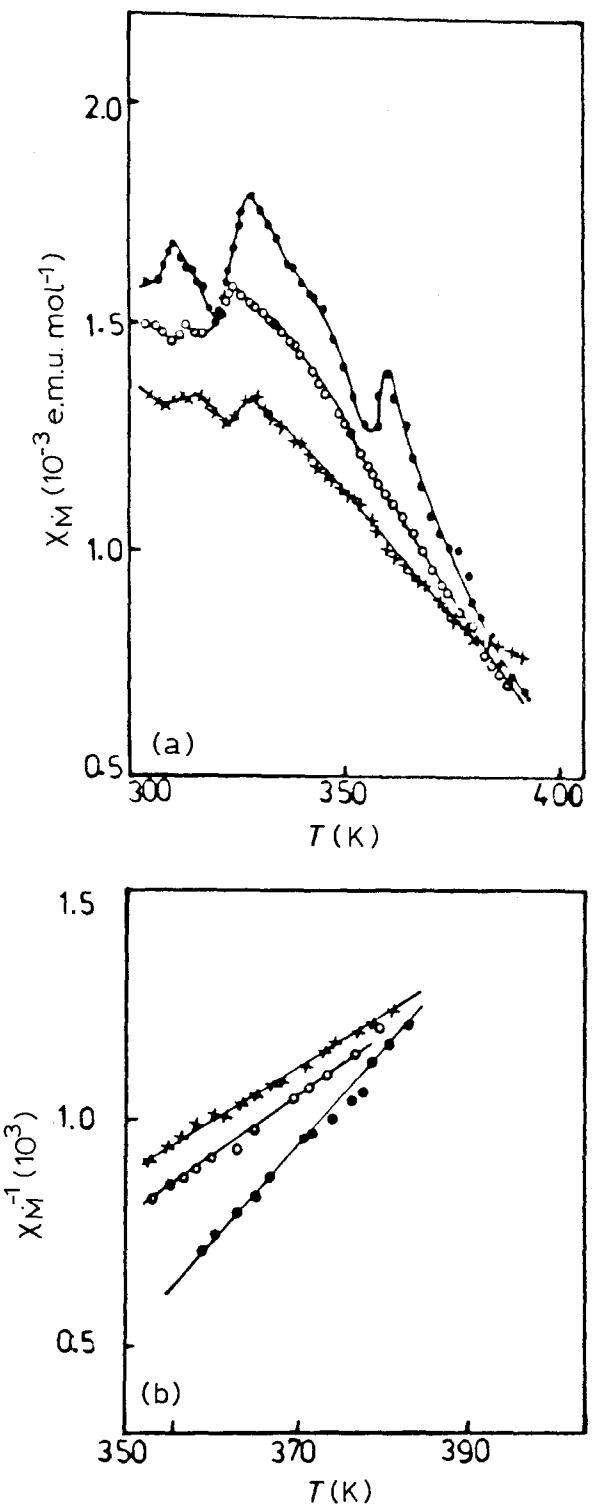


*Figure 2* (a) Temperature dependence of the magnetic susceptibility at different magnetic fields. (b) Relation between temperature versus reciprocal of the magnetic susceptibility at different magnetic fields; for $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_3Br$ compound. (●) 3050 Oe, (○) 3700 Oe, (x) 4200 Oe.

Variation of the magnetic field has a drastic effect on the peak value, to the extent that the peak was decreased by increasing the magnetic field intensity. However, this is a characteristic of ferromagnetic interaction between the magnetic layers, that tend to be saturated with increasing magnetic field [12]. Similar behaviour was observed for the compounds $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_3Br$ and $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_2Br_2$, which is presented in Figs 2a and 3a. From Figs 1a, 2a and 3a it is clear that the susceptibility values for $x = 0$, 1 and 2 are $1.4 \times 10^{-3}$, $1.5 \times 10^{-3}$ and $1.7 \times 10^{-3}$, respectively. The systematic increases in the susceptibility values with $x$ indicates that the magnetic interaction was increased with increasing number of $Br^{-1}$ ions in the compound of the present study. This may be attributed to the larger size of $Br^{-1}$ ions which will take the out-of-plane position. Also, it is clear that the peaks height were increased as the number of $Br^{-1}$ ions increased. The reason behind that was attributed to the change in the crystal structure in addition to the electronic configuration upon replacing $Cl^{-1}$ ions by the $Br^{-1}$ ions.

The phase-transition points which were appeared at 328 and 340 K in the $(m\text{-}NO_2\text{–}C_6H_4NH_3)_2CuCl_4$

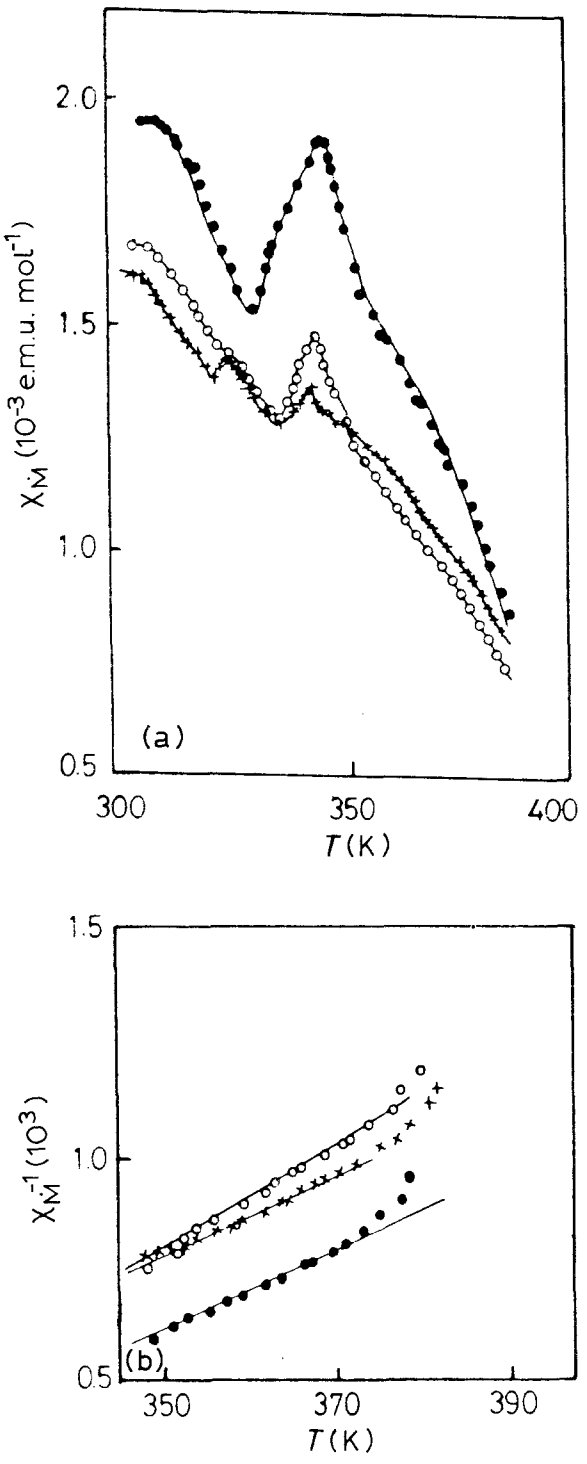


*Figure 3* (a) Temperature dependence of the magnetic susceptibility at different magnetic fields. (b) Relation between temperature versus reciprocal magnetic susceptibility at different magnetic fields; for $m$-$NO_2$–$C_6H_4NH_3)_2CuCl_2Br_2$ compound. (●) 3050 Oe, (○) 3700 Oe, (x) 4200 Oe.

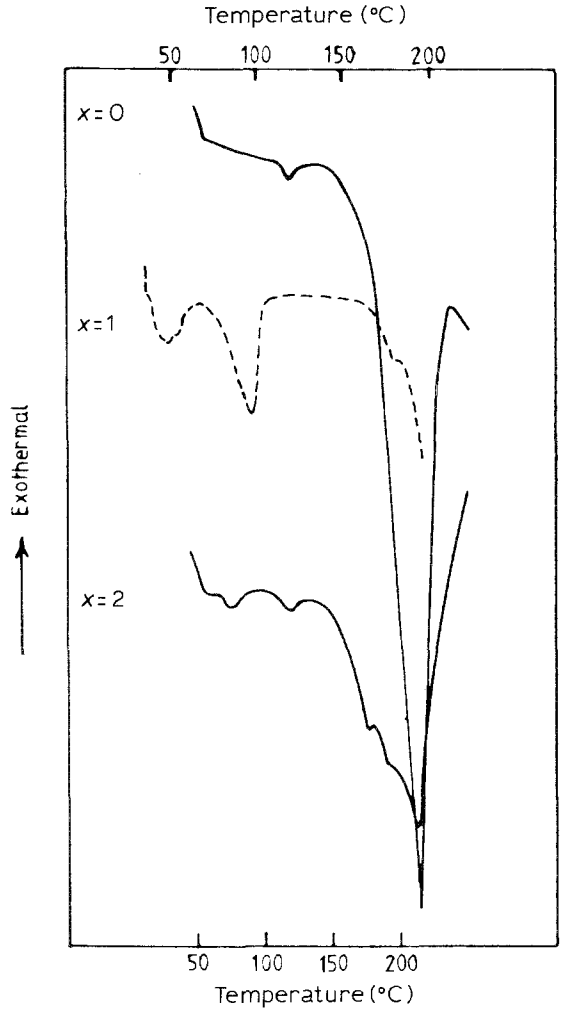


*Figure 4* Differential scanning calorimeter (DSC) thermograph for the compound ($m$-$NO_2$–$C_6H_4NH_3)_2$ $CuCl_{4-x}Br_x$ ($x$ = 0, 1 and 2).

compound, were shifted to 315 and 330 K in the ($m$-$NO_2$–$C_6H_4NH_3)_2CuCl_3Br$. This may be attributed to the existence of $Br^{-1}$ ions in the out-of-plane position which forces the $Cu^{2+}$ ions towards the horizontal plane. The canting angle in this case, as well as the distortion in the system, is changed which affects the magnitude and feature of the magnetic susceptibility. In the case of ($m$-$NO_2$–$C_6H_4$-$NH_3)_2CuCl_2Br_2$ compound, at 3050 Oe, one of the phase-transition points has disappeared. This may be due to varying the interlayer spacing which delays the ordering. The transition temperature which appeared in the compounds ($m$-$NO_2$–$C_6H_4NH_3)_2CuCl_{4-x}Br_x$ ($x$ = 0, 1, 2) are in good agreement with that value which was obtained from the DSC thermograph, Fig. 4.

The reciprocal molar magnetic susceptibility as a function of temperature and magnetic field intensity are presented in Figs 1b, 2b and 3b. The values obtained for the Curie–Weiss constant, θ, are calculated and reported in Table I, which indicates that the nature of the present compound is ferromagnetic. However, from Table I, the values of the Curie–Weiss constant, θ, in these compounds were seen to decrease and to approach the antiferromagnetic character, by increasing the magnetic field strength. The values of the Curie constant $C$, and effective magnetic moment, $M_{eff}$ were calculated from the experimental data and are reported in Table II. The values of the effective magnetic moment for the three compounds ($x$ = 0, 1, 2) are in good agreement with the spin-only value [13].

## References

1. A. Z. MOHAMED, M. A. AHMED, S. S. SALEEM and A. H. BASSYOUNI, in "Proceedings of the Second Cairo

International Symposium on Renewable Energy Sources", Vol. 1, Cairo, Egypt, "Solar energy and applications", 1–4 October 1990.
2. M. A. AHMED , M. M. El-DESOKY and F. A. RADWAN, *Aust. J. Phys.* **41** (1988) 85.
3. M. A. AHMED and M. A. ABDEL-KAFFAR, *Thermochem. Acta* **70** (1983). 217.
4. W. E. HATFIELD and D. J. HODGSON, *Inorg. Chem.* **15** (1976) 3147.
5. R. D. WILLETT and E. F. RIEDEL, *Chem. Phys.* **8** (1975) 112.
6. L. J. De JONGH and W. D. VAN AMSTEL, *J. de Phys.* **32** Cl (1971) 880.
7. J. H. P. COLPA, *Physica* **57** (1972) (397).
8. M. A. SEMARY, M. F. MOSTAFA and M. A. AHMED, *Solid State. Commun.* **25** (1978) 443.
9. H. REMY and G. LAVES, *Ber. Dtsch. Chem. Ges. B* **66** (1933).
10. L. F. BATES, "Modern Magnetism" (Cambridge University Press, Cambridge, 1948) p. 97.
11. F. E. MABBS and D. J. MACHIN, "Magnetism and Transition Metal Complexes", (Chapman and Hall, London, 1973) p. 5.
12. L. J. de JONPH and A. R. MIEDEMA, *Adv. Phys.* **23** (1974) 1.
13. F. A. COTTON, "Progress in Inorganic Chemistry" (Wiley Interscience, New York, 1964) p. 50.

*Received 4 November 1991*
*and accepted 14 August 1992*