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TWO-DIMENSIONAL HEISENBERG FERROMAGNET : SUSCEPTIBILITY

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Résumé. — Des mesures de la susceptibilité magnétique χ de composés de Cu du type $\text{Cu}(\text{C}_n\text{H}_{2n+1}\text{NH}_3)_2\text{Cl}_4$ à ferromagnétisme en couche (n variant de 1 à 10) et de composés isomorphes de Br ont été effectuées. Des anomalies de variation de χ ont été constatées à certaines températures T_c de transition. Pour la plupart des composés à base de Cl, la température T_c est voisine de 7,5 °K, pour ceux à base de Br $T_c \approx 11$ °K, bien que la cellule élémentaire varie d'un facteur 3 dans les séries étudiées. Les résultats de mesure dans le domaine paramagnétique s'ajustent fort bien à un résultat théorique de développement en série de χ . L'ajustement permet de déterminer des valeurs de la constante d'échange J/k . L'existence de transition de phase dans un ferromagnétique à 2 dimensions de Heisenberg est discutée en fonction des valeurs trouvées ainsi pour J/kT_c .

Abstract. — The magnetic susceptibility χ has been measured of the ferromagnetic layer-type Cu-compounds $\text{Cu}(\text{C}_n\text{H}_{2n+1}\text{NH}_3)_2\text{Cl}_4$, n varying from 1-10, and of isomorphous Br-compounds. Transition temperatures T_c have been found at which anomalies in χ occur. For most of the Cl-compounds $T_c \approx 7.5$ °K, for the Br-series $T_c \approx 11$ °K, while the unit-cell varies with a factor 3 through these series. The data in the paramagnetic region may be fitted quite well to the series-expansion result for χ . From these fits values for the exchange-constants J/k have been derived. The consequences of the so obtained J/kT_c — values with respect to the possible existence of a phase-transition for the 2-dimensional Heisenberg ferromagnet are discussed.

The susceptibility of the layer-type Cu-compounds $\text{Cu}(\text{C}_n\text{H}_{2n+1}\text{NH}_3)_2\text{Cl}_4$ and the isomorphous Br-compounds has been studied both in the paramagnetic region using fields of 4 and 10 kOe and near the transition temperature T_c . We define T_c as the temperature at which a peak in χ occurs (at about $kT/J = 0.5$). In the latter region we used an a. c. method (zero field).

In table I we have listed the values for T_c found for the various compounds. It is striking that $T_c \approx 7.5$ °K for the Cl-compounds with $n \geq 3$ and $T_c \approx 11$ °K for the Br-compounds with $n \geq 2$, while the volume of the chemical unit cell varies with a factor 3 through these series. This shows that these salts can be considered to a fair approximation as an assembly of nearly isolated magnetic layers.

TABLE I

Compound	T_c	J/k	J/kT_c	d_1	d_2
$\text{Cu}(\text{CH}_3\text{NH}_3)_2\text{Cl}_4$	8.91	19.2	2.15	5.247	9.97
$\text{Cu}(\text{C}_2\text{H}_5\text{NH}_3)_2\text{Cl}_4$	10.25	18.6	1.81	5.240	11.21
$\text{Cu}(\text{C}_3\text{H}_7\text{NH}_3)_2\text{Cl}_4$	7.65	16.0	2.09	5.297	12.86
$\text{Cu}(\text{C}_4\text{H}_9\text{NH}_3)_2\text{Cl}_4$	7.33	15.4	2.10	5.208	15.83
$\text{Cu}(\text{C}_5\text{H}_{11}\text{NH}_3)_2\text{Cl}_4$	7.30	15.9	2.18	5.265	17.79
$\text{Cu}(\text{C}_6\text{H}_{13}\text{NH}_3)_2\text{Cl}_4$	7.75	17.1	2.20		
$\text{Cu}(\text{C}_{10}\text{H}_{21}\text{NH}_3)_2\text{Cl}_4$	7.91	17.9	2.26	5.239	25.78
$\text{Cu}(\text{C}_1\text{H}_3\text{NH}_3)_2\text{Br}_4$					
$\text{Cu}(\text{C}_2\text{H}_5\text{NH}_3)_2\text{Br}_4$	10.85	19.0	1.75	5.541	11.42
$\text{Cu}(\text{C}_3\text{H}_7\text{NH}_3)_2\text{Br}_4$	10.50	21.32	2.03	5.548	12.76
$\text{Cu}(\text{C}_4\text{H}_9\text{NH}_3)_2\text{Br}_4$	11.33	21.87	1.93	5.530	14.83
$\text{Cu}(\text{C}_5\text{H}_{11}\text{NH}_3)_2\text{Br}_4$	11.4	22.00	1.93		

d_1 and d_2 are respectively the copper-copper distance within a layer and between neighbouring layers.

Also listed in table I are values for the exchange constant J/k , derived from the data in the paramagnetic region. We used the following procedure : by measuring the diamagnetism of isomorphous nonmagnetic Cd- and Pd-compounds, we were able to correct our data for diamagnetism. Our results were then compared with the series-expansion of $\chi T/C$ in powers of J/kT for the quadratic Heisenberg lattice given

by Baker et al. [1]. As only a finite number (10) of terms in this expansion is known it will only be a reasonable representation of the susceptibility down to a certain temperature. We have used the series-result down to $J/kT = 0.6$ because in this region the $\chi T/C$ values calculated with 10 and 7 terms did not differ by more than 1 %, while the uncertainty in the experimental $\chi T/C$ values is a few percent. In the next step we « scaled » the experimental $C/\chi T$ versus T_c/T curves in the region $0.3 < C/\chi T < 0.6$ by a factor J/kT_c , in order to bring them into coincidence with the theoretical $C/\chi T$ versus J/kT curve. The fact that this can be brought about (that J/kT_c does not vary systematically with temperature) again demonstrates the two-dimensional behaviour. This statement is substantiated by the linear temperature-dependence of the specific heat at low temperatures [2]. A typical result is shown in figure 1 where the suscep-

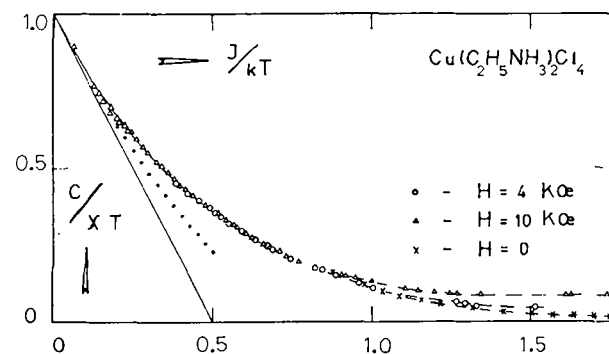


FIG. 1. — Reciprocal susceptibility of $\text{Cu}(\text{C}_2\text{H}_5\text{NH}_3)_2\text{Cl}_4$ versus J/kT (dimensionless units).

tibility of $\text{Cu}(\text{C}_2\text{H}_5\text{NH}_3)_2\text{Cl}_4$ has been fitted to the series result (full curve drawn through experimental points for $J/kT < 0.7$). Also plotted are the molecular field result, which is the straight line

$$C/\chi T = 1 - \theta/T$$

($\theta = 2 J/k$ for the quadratic lattice, $S = \frac{1}{2}$) and the series-expansion result for the b. c. c. lattice which is the dotted curve (abscissa scaled by a factor 2). This has been done to demonstrate the difference in the deviation from the molecular field result in two and three dimensions. It may be seen in figure 1 that the susceptibility is field dependent (for $H = 10$ kOe) for $J/kT > 0.8$.

In figure 2 we have plotted the zero-field suscepti-

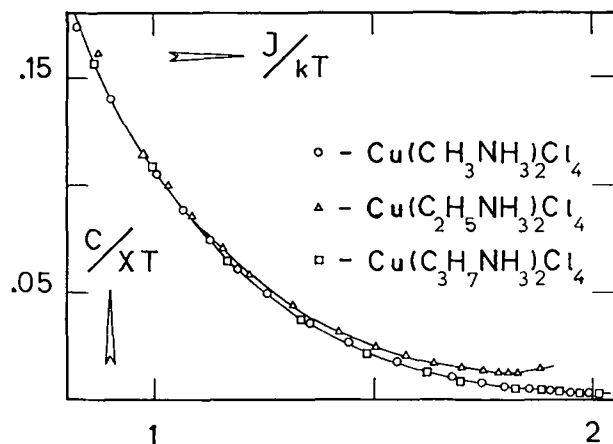


FIG. 2. — Zero-field susceptibility results for three different compounds. The curves coincide for $J/kT < 1.0$ irrespective of the sign of the interlayer interaction J_2 .

bility of powdered samples of three different compounds, in order to show that for $J/kT < 1.0$ all curves coincide. Nearer to T_c the anisotropy energy and the interaction between the Cu-layers J_2 will, however small as compared to the interaction within a layer J_1 , greatly influence the magnitude and the temperature-dependence of the susceptibility by ordering the ferromagnetic planes ferro- or antiferromagnetically with respect to each other (we have also observed weak ferromagnetism). For $\text{Cu}(\text{CH}_3\text{NH}_3)_2\text{Cl}_4$ the sign of J_2 is positive, for $\text{Cu}(\text{C}_2\text{H}_5\text{NH}_3)_2\text{Cl}_4$ it is negative. From figure 2 it follows that for temperatures higher than $J/kT < 1.0$ the influence of the anisotropy and the coupling between the Cu-layers is negligibly small. In this temperature region therefore, our results can be considered to be representative for the ideal square Heisenberg ferromagnet. Since our results extend to a region where the series expansion can no longer be trusted, we have listed in table II

mean values of the $C/\chi T$ versus J/kT curves in the region $0.5 < J/kT < 1$. The $C/\chi T$ -values of all compounds scatter evenly around this curve within a range which is 2 % of $C/\chi T$ at $J/kT = 0.5$, becoming 10 % at $J/kT = 1.05$.

Considering the obtained values for J/kT_c we would like to remark that the mean value $J/kT_c = 1.75 \pm 0.1$ which we have previously [3] reported, has changed to $J/kT_c = 2.0 \pm 0.2$. This difference is due to the fact that the data reported in ref. [2] were taken in the region $0.1 < J/kT < 0.25$ while the present results have been obtained in the region $0.1 < J/kT < 0.6$ (the error in the correction for diamagnetism is negligible for $J/kT > 0.25$). Also, in ref. [2] the isomorphous nonmagnetic salts were not yet available.

The values for J/kT_c we have derived may be of importance because in recent years there has been, mainly theoretical, discussions about the existence or nonexistence of a phase-transition for the two-dimensional Heisenberg model. The specific heat results reported in the preceding paper suggest there is no phase transition in C_M for the ideal model. It is possible however [4] that while long range order is ruled out at any finite temperature, there is a transition point at which the susceptibility, which is proportional to $\sum_R \langle S_0 \cdot S_R \rangle$ diverges. Our results show a variation $1.81 < J/kT_c < 2.26$. This suggests that either the anisotropy or the interlayer exchange J_2 may have an influence. What we need therefore is additional information on the anisotropy in the Cu-plane, which we are studying at this moment, in order to see whether the variation of J/kT_c can be correlated to a variation in the anisotropy. In this respect it is gratifying that the compound $\text{Cu}(\text{C}_{10}\text{H}_{21}\text{NH}_3)_2\text{Cl}_4$, in which thus far we have found the smallest anisotropy in χ (just above and at T_c), has the highest J/kT_c -value, as the effect of anisotropy would be to lower the J/kT_c value. A conclusion which at this stage can be drawn from these susceptibility experiments alone is that if a J/kT_c exists for the quadratic Heisenberg ferromagnet its value will be higher than 2.26, which itself is 4.5 times larger than the molecular field result (See however also ref. [5]).

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TABLE II

J/kT	0.50	0.55	0.60	0.65	0.70	0.75	0.80	0.85	0.90	0.95	1.00
$C/\chi T$	0.36	0.32	0.29	0.26	0.23	0.20	0.18	0.16	0.14	0.12	0.11

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