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MAGNETIC PHASE DIAGRAM OF $NiCl_2 4H_2O$ *

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ABSTRACT

The magnetic phase diagram of $NiCl_2 4H_2O$ for $H \| c$ was determined from susceptibility measurements. The exchange and anisotropy fields, obtained from the $T = 0$ spin-flop field $H_{SF} = 23.1 \pm 0.2$ kOe and spin-flop-to-paramagnetic field $H_P = 68.8 \pm 0.2$ kOe, are compared with previous measurements.

$NiCl_2 4H_2O$ is a monoclinic salt isomorphous to the well studied $MnCl_2 4H_2O$ and $MnBr_2 4H_2O$ (1). McElearney, et al. (1) (here after referred to as I) have measured its low temperature specific heat and zero-field magnetic susceptibility from which an antiferromagnetic transition was determined at $T_N = 2.99 \pm 0.01$ K (the easy-axis being the cristalographic c-axis). These data were interpreted in the Molecular Field Approximation (MFA) in terms of: - an exchange parameter $(Jz/k) = -5.25 \pm 0.10$ K ; - and a splitting of the Ni^{++} ground spin triplet of $(D/k) = -11.5 \pm 0.1$ K and $(E/k) = 0.1 \pm 0.1$ K (the minus sign meaning that the almost degenerate doublet lies lower). These numbers reveal an intriguinly high anisotropy energy and an interesting and unexpected axial character of the anisotropy. Moreover, some difficulty was found in reconciling the high value of the exchange

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parameter with the observed Néel temperature.

We have measured the field dependence of the a.c. (155 Hz) magnetic susceptibility, for H parallel to the easy axis (c-axis), at constant temperatures, down to 0.3 K. As can be seen in Fig. 1 the transitions to the paramagnetic and spin-flop phases are quite evident and accurately marked by sharp peaks. Fig. 2 shows the H-T phase diagram thus determined. The sharpness of the spin-flop transition confirms the assignment in I of the c-axis as the easy-axis.

By suitable extrapolation of the phase boundaries we determined: $T_N = 3.00 \pm 0.01$ K in close agreement with I ; the $T = 0$ spin-flop field $H_{SF} = 23.1 \pm 0.2$ kOe ; the $T = 0$ spin-flop-to-paramagnetic field $H_P = 68.8 \pm 0.2$ kOe; and the triple point $T_t = 2.20 \pm 0.02$ K and $H_t = 24.2 \pm 0.2$ kOe. Assuming a two-sublattice model, the phenomenological exchange and anisotropy fields derived from $H_{SF}^2 = 2H_EH_A - H_A^2$ and $H_P = 2H_E - H_A$ [(2)] are: $H_E = 38.3$ kOe and $H_A = 7.8$ kOe . H_E is related to Jz by $g\mu_B H_E = -2Jz$ and taking the spectroscopic factor $g = 2.28$ [(1)] one obtains: $(Jz/k) = -2.9$ K. This value is considerably smaller than that given in I, and has in its favor a much closer prediction of T_N in the MFA, that is $T_N = (2Jz/3k)S(S+1) = 3.9$ K. Really, corrections due to spin-spin correlation effects could very well account for the difference between this and the experimental value [(3)].

On the other hand, the presence of a spin-flop transition at fields of $\sim$23 kOe is surprising in view of the high value of the anisotropy parameter from I. As a comparative value, one should note that $(g\mu_B H_A/k) \simeq 1.2$ K which is one order of magnitude smaller than the reported value of D. Of course, the exact relation between H_A and D depends on the relative orientation between electric and magnetic axes, and two inequivalent Ni^{++} ions are present in the unit cell [1]. However, it is unexpected that this would account for such a big difference. It seems more likely that the interpretation of the data should allow for the inclusion of an anisotropic exchange as has been done for the isomorphic salts $MnCl_2 4H_2O$ and $MnBr_2 4H_2O$ [4]. A comparative study of the field dependence of the susceptibility in these three salts is in progress and will make possible a more detailed analisis of all the data.

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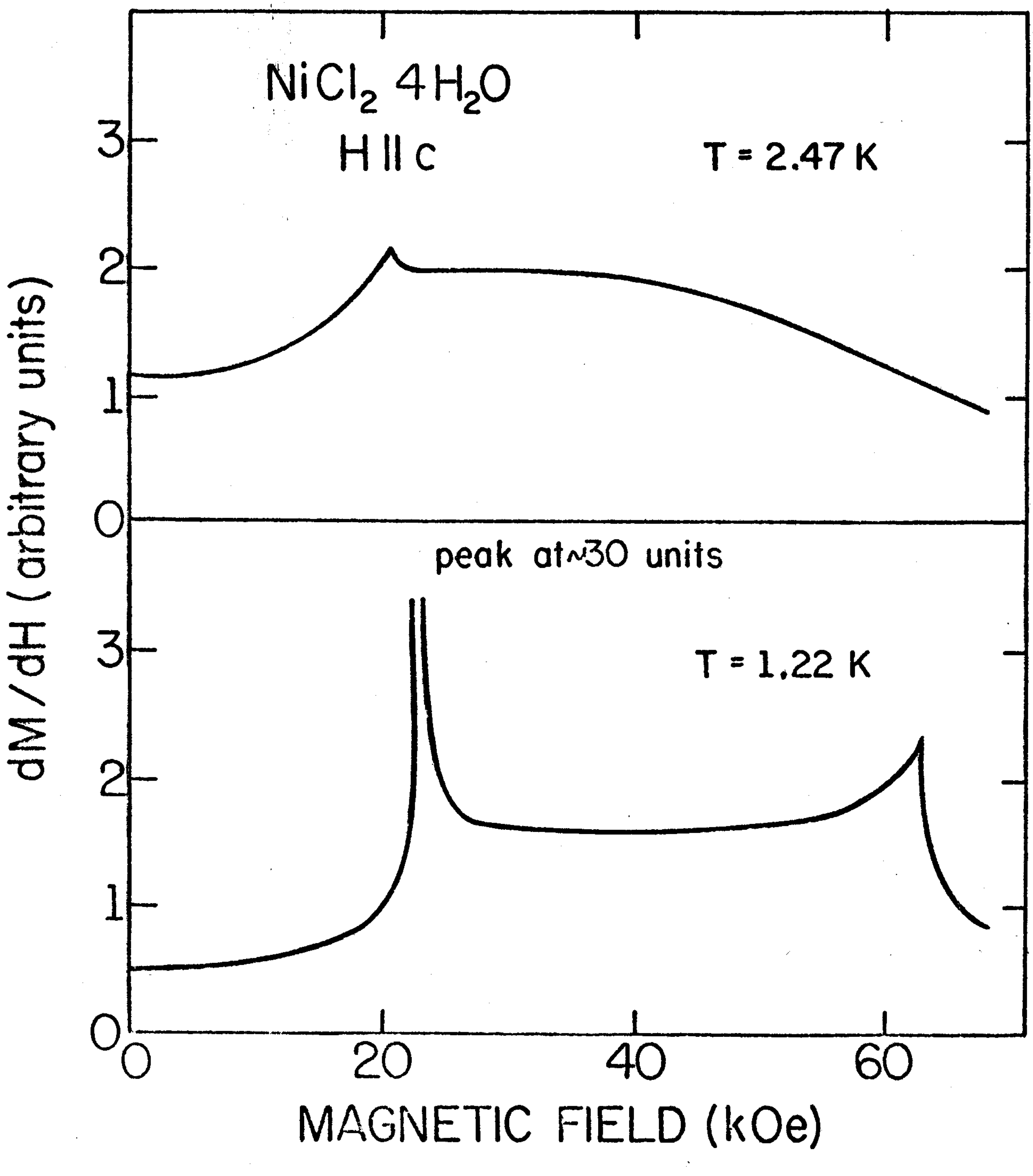
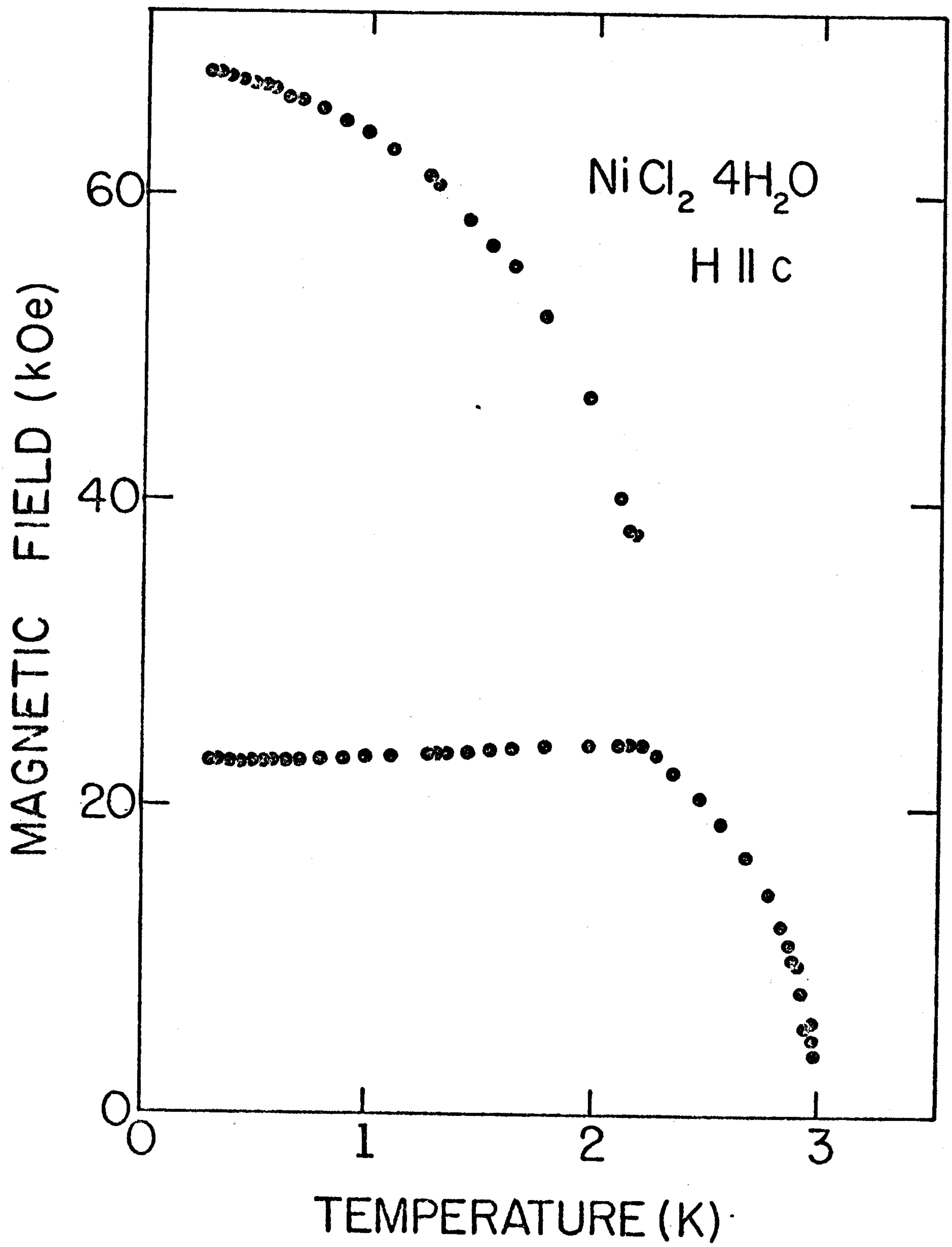
FIGURE CAPTIONS

Fig. 1 - Susceptibility versus field for two constant temperatures below T_N

Fig. 2 - Magnetic phase diagram for $H \parallel c$.



NiCl2 4H2O
H ll c
T = 2.47 K
peak at~30 units
T = 1.22 K
dM/dH (arbitrary units)
MAGNETIC FIELD (kOe)
0
1
2
3
0
1
2
3
0
20
40
60



$NiCl_2\ 4H_2O$
H ll c
MAGNETIC FIELD (kOe)
TEMPERATURE (K)
0
20
40
60
0
1
2
3