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## MAGNETIC PHASE BOUNDARIES OF Nicl<sub>2</sub>6H<sub>2</sub>O NEAR THE BICRITICAL POINT\*

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## **ABSTRACT**

Precise determinations of the magnetic phase boundaries near the bicritical point of NiCl<sub>2</sub>6H<sub>2</sub>O are shown to be in good agreement with the predictions of recent scaling theories.

A considerable attention has been lately given to the problem of multicritical points. A recent theory by Fisher and Nelson (1,2) has made predictions for the behavior of the phase boundaries near the bicritical point in Heisenberg antiferromagnets. However, experimental data to compare with the theory are still scarce. The purpose of this letter is to present precise measurements of the phase boundaries near the bicritical point of the monoclinic antiferromagnetic crystal NiCl<sub>2</sub>6H<sub>2</sub>0, made by means of differential magnetization, dM/dH, measurements (modulation field  $\sim$  20 Oe peak to peak, at 155 Hz). The magnetic properties of this salt have been extensively studied.(3-5)

As discussed by Rohrer, (6) the alignment between H and the easy-axis is critical if one wants to compare experimental data with the scaling theories. In the present experiments the external magnetic field (H) was obtained from a NbTi superconducting solenoid

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suspended by a device which permitted external adjustments of the direction of H by ammounts smaller than  $0.05^{\circ}$ . The sample (a single crystal roughly of rod shape elongated in the <u>a</u>-direction) was initially placed inside the pick-up coils with the coil axis approximately parallel to the easy-axis.(5) Then, adjustments of the direction of H were made to obtain optimal alignment. These adjustments were monitored by the height of the dM/dH peak at the spin-flop transition, at a temperature below but close to  $T_b$ . The actual misalignment could afterwords be estimated from curves of peak height versus temperature for various modulation frequencies. For the experimental run presented here, these curves clearly splitted below T=3.85 K thus indicating a misalignment  $\lesssim 0.1^{\circ}$ .(7)

Fig. 1 shows the experimental data. The antiferro-paramagnetic and the antiferro-canted phase boundaries could be followed by observing the peaks in dN/dH, measured by sweeping H at constant T. The boundary between the canted and the paramagnetic phase had to be determined by varying T at constant H. In this case the transition was identified with a kink in dM/dH. The size of the dots in fig.1 represent the resolution of our apparatus:  $\sim 0.003$  K in temperature and  $\sim 30$  Oe in field (the accuracy in the applied field calibration is  $\sim 0.5\%$ ).

To compare our data with the theory of bicritical phase boundaries,(2) a computer fit of the canted-paramagnetic and antiferroparamagnetic boundaries was made to the equations  $\tilde{g} = \omega_1 \tilde{t}^{\varphi}$  and  $\tilde{g} = -\omega_1 \tilde{t}^{\varphi}$  respectively, with

$$\tilde{g} = H^2 - H_b^2 - \left(\frac{dH_{BF}^2}{dT}\right)_b (T-T_b)$$

and

$$\tilde{t} = \frac{T-T_b}{T_b} - \frac{n+2}{3n} \frac{1}{T_b} \left(\frac{dT_c}{dH^2}\right)_{H=0} (H^2-H_b^2)$$

Here, H<sub>SF</sub>(T) is the spin-flop boundary, n is the dimensi onality of the order parameter and  $T_{\mathbf{c}}(\mathbf{H})$  is the antiferro-paramagnetic boundary. From the experimental points between T = 3.85 K and T = 3.93 K,  $(dH_{SF}^2/dT)_b$  could be determined as 174.4  $\pm$  0.1 (kOe)<sup>2</sup>/K. The value of  $(dT_c/dH^2)_{H=0}$  was taken from the calorimetric measurements of Johnson and Reese. (3) We attempted to fit the data with n=3 and n=2. For n=3, the crossover exponent  $\phi$ and the ratio  $\omega_{\perp}/\omega_{11}$  have been theoretically determined as 1.25 and 2.51 respectively, and for n=2,  $\phi$  = 1.18 and  $\omega_{\perp}$ = $\omega_{||}$ .(2) In both cases the adjustable parameters were  $T_{
m b}$  and  $\omega_{
m L}$ . The resulting best curves are seen in fig. 1: - solid line for n=2; - and dashed line for n=3. A quite good fit was obtained with n=2, while the n=3 best curve deviates systematically from the data. For the best curve with n=2 we obtained  $T_b = 3.940 \text{ K}$  and  $\omega_L = \omega_H = 9,962 \text{ (kOe)}^2$ . By playing with parameters we found that almost equally good fits could be obtained with  $\phi$  = 1.25 as weel as  $\phi$  = 1.18, but leaving both  $\omega_{\perp}$ and  $\omega_{H}$  as adjustable parameters. In both cases the best curves had  $\omega_1 \simeq \omega_0$  . In conclusion, although the present data is not accurate enough to precisely determine  $\phi$ , it certainly is in good agreement with the predictions of the theory. The requirement of  $\omega_1 = \omega_{11}$  suggests that the anisotropy energy in NiCl<sub>2</sub>6H<sub>2</sub>0, although predominantly uniaxial (5), has a rhombic component which, in the canted phase, maintains the spins in one plane.

It must be mentioned that the magnetic easy-axis of NiCl $_2^{6\rm H}_2^{0}$  has been reported to change its direction, as temperature changes below  $T_{\rm N}$ , (8) and this could affect our results. To minimize this possibility, the above described fit was made in a temperature interval for which the angular variation estimated from the data shown in ref. 8, was  $\stackrel{<}{\sim} 0.05^{\circ}$ .

## REFERENCES

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## FIGURE CAPTIONS

Fig. 1 - Phase boundaries near T<sub>b</sub>. Solid circles are points obtained by sweeping H at constant T. Open circles were obtained by varying T at constant H. The solid line is the theoretical fit for n=2, and the dashed line, for n=3.

