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J. Ferré, J. Jamet, C. Landee, K. Reza, J. Renard. TEST OF THE HALDANE CONJECTURE IN A 1D-HEISENBERG ($S = 1$) ANTIFERROMAGNET BY OPTICAL LINEAR BIREFRINGENCE. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-1441-C8-1442. 10.1051/jphyscol:19888661 . jpa-00228892

HAL Id: jpa-00228892

<https://hal.science/jpa-00228892>

Submitted on 4 Feb 2008

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TEST OF THE HALDANE CONJECTURE IN A 1D-HEISENBERG ($S=1$) ANTIFERROMAGNET BY OPTICAL LINEAR BIREFRINGENCE

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Abstract. – We report on optical linear birefringence measurements in the nearly ideal $S = 1$ Heisenberg antiferromagnetic chain compounds: $\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{NO}_2(\text{ClO}_4)$ and $\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{NO}_2\text{PF}_6$. The temperature dependence of the magnetic contribution, proportional to that of the magnetic energy, indicates $J/k = -53$ K and supports the Haldane conjecture.

Considerable interest has been recently stimulated by Haldane's conjecture [1] that a gap appears in the excitation spectrum of quantum Heisenberg antiferromagnetic (HAF) spin chains with an integer spin. In counterpart a gapless spin wave spectrum is expected for a half integer spin. These predictions have spurred many theoretical investigations and despite some minor points of controversy they are now widely accepted. The pioneering work of Botet *et al.* [2] has confirmed numerically these conclusions for $S = 1$ HAF chains by using a finite size scaling method. Such a system is described by the hamiltonian:

$$H = -J \sum_i [(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + \lambda S_i^z S_{i+1}^z] + \sum_i D (S_i^z)^2 \quad (1)$$

where J is the exchange constant and D the single ion anisotropy coefficient. The calculated energy gap $\Delta = 0.41 |J|$ for the $S = 1$ HAF ($\lambda = 1$, $D = 0$) chains is large enough to be experimentally evidenced [3] and exists over a wide anisotropy range $-0.25 \leq D/|J| \leq 0.8$.

Experimental check of the Haldane predictions is in its infancy. The choice of adequate materials is very limited because they must show very good one dimensional HAF properties. In particular the interchain interaction J' has to be small enough to avoid the occurrence of a 3d-long range order at finite temperature. Haldane showed that the gap decays exponentially with increasing S and therefore it is easier to detect it for Ni^{2+} ($S = 1$) compounds. The verification of the Haldane's conjecture has been tentatively experienced on the quasi 1d-HAF CsNiCl_3 [4, 5] and on the most promising compound $\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{NO}_2\text{ClO}_4$ [3, 6], hereafter abbreviated NENP. CsNiCl_3 is an imperfect 1d-HAF system because it orders antiferromagnetically below 4.8 K. NENP stands as a nearly ideal $S = 1$, 1d-HAF with a large intrachain interaction $|J|/k \simeq 55$ K. It crystallises in the orthorhombic structure

(space groupe Pnma), the spin chains being parallel to the b -axis. The weak interchain interaction ($J'/J = 4 \times 10^{-4}$) is unable to establish 3d-long range order in NENP, as demonstrated experimentally down to 1.2 K. The existence of an energy gap gives rise to drastic effects on the magnetic susceptibility which drops to nearly zero when lowering the temperature down to 0 K, independently of the magnetic field orientation. Neutron scattering data also strongly support this conjecture and give evidence to two energy gaps corresponding to spin fluctuations parallel ($\Delta_{\parallel}/|J| = 0.58$) or perpendicular ($\Delta_{\perp}/|J| = 0.26$) to the chain axis, resulting from a planar anisotropy $D \simeq 0.25 |J|$ [3].

In this paper we report on optical linear birefringence (LB), measured between the chain (b) and a axes at $\lambda = 0.6328 \mu\text{m}$, in NENP (Fig. 1a) and in the $\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{NO}_2\text{PF}_6$ analog compound (NENF) in the 5-300 K temperature range. The LB can be decomposed into two contributions (7): $\Delta n = \Delta n_l + \Delta n_m$. The lattice part Δn_l is related to crystallographic deformations and to the electron-phonon interactions. The magnetic part Δn_m is known to be proportional to the magnetic energy for uniaxial Heisenberg magnets. The temperature variation of Δn_l is usually small as compared to that of Δn_m , giving the opportunity to deduce the thermal dependence of the magnetic energy, and consequently that of the magnetic specific heat C_m , without making ambiguous assumptions on the lattice contribution as in the case of direct heat capacity measurements. The thermal dependence of the temperature derivative of the LB, $d(\Delta n)/dT$, in NENP is reported in figure 1b. We suggest a sudden ordering of the perchlorate groups below 278 K in NENP, as evidenced by the significant anomaly on the $d(\Delta n)/dT$ curve shown in the insert of figure 1b. On another hand a very large and sharp peaked anomaly occurs on $d(\Delta n)/dT$ for NENF close to 235 K; it corresponds to a phase transition, recently evidenced by structural measurements done in the Willett's group [8], which lowers the space group symmetry to monoclinic and twins the crystal.

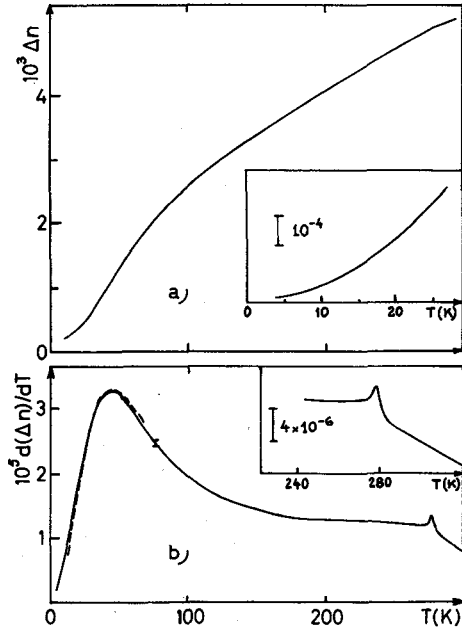


Fig. 1. - (a) Temperature dependence of the linear birefringence in NENP and (b) of its temperature derivative. Fit (---) of the broad anomaly following theoretical calculations (9) with $J/k = -53$ K. In insert: (a) detail of the low temperature variation of Δn , (b) detail showing the crystallographic transition at 278 K.

As for CsNiCl_3 [9], the temperature variation of $d(\Delta n)/dT$ for the two compounds, is mainly due to the magnetic contribution which clearly reveals their low dimensional character. As expected for 1d-HAF the $d(\Delta n)/dT$ (or specific heat) broad anomaly shows a maximum located near 43 K for NENP and NENF (Fig. 1b), and short range order manifests itself up to at least 180 K. The magnetic specific heat numerical calculations based upon extrapolated estimations for finite size systems [10, 11] give $KT(C^{\text{max}}) = 0.82|J|$ in the case $D = 0$. The relatively small anisotropy $D = 0.25|J|$ in NENP [3] does not change significantly this result [10]. Neglecting the lattice contribution to the LB we deduce $|J|/k = 53$ K for the two compounds confirming the previously reported value of the exchange integral $|J|/k = 55$ K [6]. The fit of the thermal variation of $d(\Delta n)/dT$ curve by the law predicted in [11] is very satisfactory (Fig. 1b).

Our data confirms the absence of a transition to a 3d-ordered phase down to 3.8 K since no sudden break in slope appears on the temperature variation of Δn , as found in CsNiCl_3 [12]. The occurrence of an Haldane gap may be found from the deviation of the thermal variation of the specific heat (or of the magnetic energy) from the $S = 1$ HAF chain spin wave theory. In fact, the predicted $C_m(T)$ variation calculated in

[10, 11] takes the gap into account implicitly. In particular, fixed Padé approximants in the finite size scaling calculation of thermodynamic functions give better results than usual Padé approximants at low temperature [11] and predict $C_m(T) = C_0(KT/|J|)^\alpha$ for the $S = 1$ HAF chain model with $\alpha = 1.6$, i.e. strongly different from spin wave calculations giving $\alpha = 1$. Then the low temperature variation of Δn_m may be approximated by:

$$\Delta n_m(T) - \Delta n_m(0) = A(kT/|J|)^{\alpha+1}. \quad (2)$$

Our precise data shown in the insert of figure 1a can be fit by a power law with $\alpha = 1.4 \pm 0.1$. This significant deviation of α from spin wave predictions is consistent with the Haldane conjecture. More work has to be done to relate theoretically this deviation to the energy gap.

Acknowledgments

We wish to thank R. Jullien for enlightning discussions. We also thank M. Bond and R. D. Willett for their analysis of the crystallographic phase changes in NENP and NENF.

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