Brief Reports

Brief Reports are short papers which report on completed research which, while meeting the usual **Physical Review** *standards of scientific quality, does not warrant a regular article. (Addenda to papers previously published in the* **Physical Review** *by the same authors are included in Brief Reports.) A Brief Report may be no longer than 3½ printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Spin correlations in the two-dimensional $S=1$ Heisenberg antiferromagnet

R. J. Birgeneau
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 1 September 1989)

K_2NiF_4 is a well-characterized planar antiferromagnet that represents a good approximation to the idealized two-dimensional (2D) $S=1$ Heisenberg antiferromagnet. In 1970, Birgeneau, Skalyo, and Shirane measured the 2D antiferromagnetic structure factor in K_2NiF_4 over a wide range of temperatures. In this Report we compare their experimental results with the predictions of a recent theory for the 2D quantum Heisenberg antiferromagnet by Chakravarty, Halperin, and Nelson. The theory describes the temperature dependence of both the absolute value of the correlation length and the relative structure factor peak intensity quantitatively. The crossover temperature to 2D Ising critical behavior is also properly predicted.

Before the advent of high-temperature superconductivity, K_2NiF_4 was considered to be the model example of a two-dimensional (2D) Heisenberg antiferromagnet.[1] Accordingly, in the late 1960s and early 1970s this material was studied quite thoroughly. In particular, the author, together with Skalyo and Shirane (BSS) carried out an extensive set of neutron scattering measurements of the static structure factor $S(\mathbf{Q})$ over a wide range of temperatures.[2] However, only a qualitative interpretation of the data was possible at that time since there was no adequate theory for the 2D quantum Heisenberg antiferromagnet. Furthermore, the data were complicated by a crossover from 2D Heisenberg to 2D Ising behavior which occurs somewhere above the 3D Néel transition at 97.23 K.

Stimulated by the experiments in La_2CuO_4,[3] Chakravarty, Halperin, and Nelson[4] (CHN) among others[5] have developed a quantitative theory for the instantaneous correlations in the 2D quantum Heisenberg antiferromagnet. Yamada *et al.*[6] have shown that the CHN theory describes the structure factor in the $S=\frac{1}{2}$ system La_2CuO_4 quite well. The theory contains as a parameter the spin quantum number S explicitly. It is therefore important to compare theory and experiment in 2D Heisenberg antiferromagnets with S taking on varied values. In this Brief Report we reanalyze the measurements of BSS in K_2NiF_4 utilizing the CHN theory for $S=1$.

The crystal and magnetic structure of K_2NiF_4 are discussed in the BSS paper.[2] For the purposes of this addendum it is sufficient to note that there are isolated sheets of NiF_2 which exhibit pure 2D magnetic fluctuation behavior above the Néel temperature $T_N=97.23$ K. The Ni^{2+} ($S=1$) ions form a simple square lattice. The spin Hamiltonian for a single NiF_2 sheet may be written[7]

$$H=\sum_{\substack{\langle i,j\rangle \\ i,j \text{ nearest} \\ \text{neighbors}}} J_{\rm NN}\mathbf{S}_i\cdot\mathbf{S}_j+\sum_i g_i\mu_B H_i^A S_i^z\,, \tag{1}$$

with $S=1$, $J_{\rm NN}=104\pm1$ K, and the reduced anisotropy $h_i^A=g\mu_B H_i^A/\sum_{j=\rm NN}J_{\rm NN}S_j=0.002$.

The CHN theory[4] predicts for the spin-spin correlation length and structure factor, respectively,

$$\xi/a=C_\xi\frac{e^{2\pi\rho_s/k_BT}}{1+k_BT/2\pi\rho_s} \tag{2}$$

and

$$S(0)=C_s\frac{(k_BT/2\pi\rho_s)^2e^{4\pi\rho_s/k_BT}}{[1+(k_BT/2\pi\rho_s)]^4}\,, \tag{3}$$

where ρ_s is the spin stiffness constant and C_ξ and C_s are constants. For general S the spin stiffness constant may be written as[4]

$$2\pi\rho_s=2\pi J_{\rm NN}S^2(1+0.158/2S)^2(1-0.552/2S)\,. \tag{4}$$

Hence, for K_2NiF_4, $2\pi\rho_s=552\pm5$ K. For $S=\frac{1}{2}$, CHN estimate $C_\xi\simeq0.5$ to an accuracy of about 30%. C_ξ depends on S as $1/[S(1-0.552/2S)(1+0.158/2S)]$ so that for $S=1$ one expects $C_\xi\simeq0.17$, again to an accuracy of 30%.

We now compare the predictions of Eqs. (2) and (3) with the data of BSS. The experiments were carried out using neutrons with energy 77 meV and with the scattering geometry chosen such that a proper energy integration was performed.[2] To a very good approximation the mea-

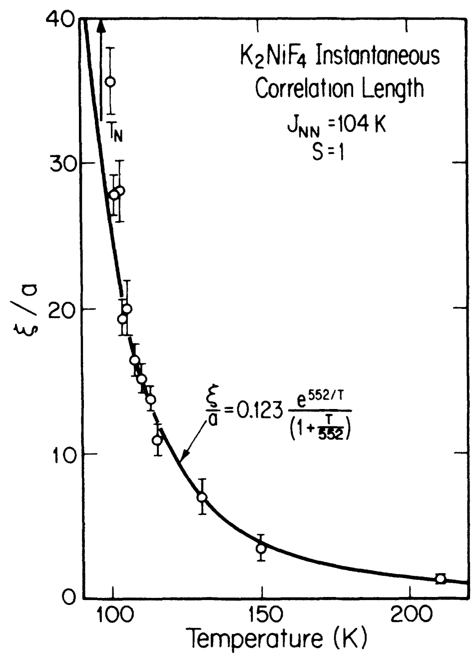

FIG. 1. Instantaneous spin-spin correlation length vs temperature in K_2NiF_4. The data are from Ref. 2. The solid line is Eq. (2) with $2\pi\rho_s$ fixed at 552 K; the amplitude, 0.123, was fitted from the data between 110 and 150 K.

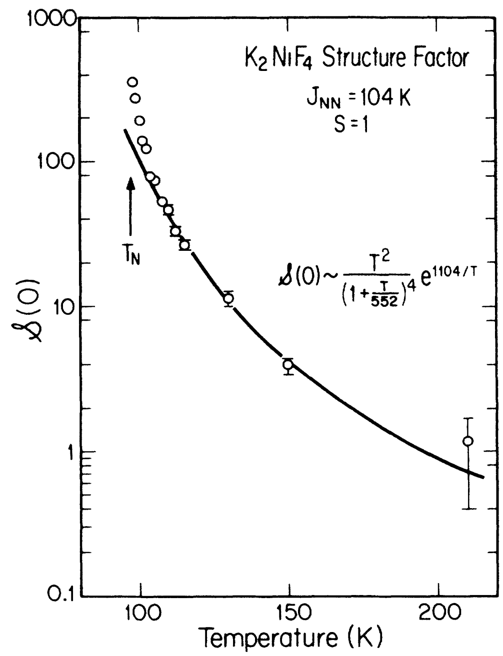

FIG. 2. Structure factor peak intensity, $S(0)$ vs temperature in K_2NiF_4. The data are from Ref. 2. The solid line is Eq. (3) with $2\pi\rho_s$ fixed at 552 K; the amplitude was fitted from the data between 110 and 150 K.

surement yielded

$$I(\mathbf{Q}) \sim S^{\parallel}(\mathbf{Q}) + S^{\perp}(\mathbf{Q}) , \qquad (5)$$

where $\parallel$ and $\perp$ are, respectively, parallel and perpendicular to the Ising anisotropy axis. We expect that $I(\mathbf{Q})$ will provide a reasonable measure of the Heisenberg structure factor $(1/N)\langle \mathbf{S}(-\mathbf{Q})\cdot\mathbf{S}(\mathbf{Q})\rangle$ provided that one is on the Heisenberg side of the 2D Heisenberg to 2D Ising crossover; this crossover in turn should occur for $h^A(\xi/a)^2 \sim 1$.

BSS fit their profiles to the simple Lorentzian form

$$I(\mathbf{Q}) = \frac{A}{\xi^{-2} + |\mathbf{Q} - \mathbf{Q}_{AF}|^2} . \qquad (6)$$

The BSS results for ξ/a and $I(Q_{AF})$ $[\equiv S(0)]$ are shown in Figs. 1 and 2, respectively, together with the CHN predictions, Eqs. (2) and (3) with $2\pi\rho_s$ given by Eq. (4). For both ξ/a and $I(Q_{AF})$ the amplitudes were fixed by fits to the data between 110 and 150 K. There are no additional adjustable parameters. It is evident that the CHN theory describes the BSS data for K_2NiF_4 extremely well. Indeed, for the correlation length, ξ/a, even the prefactor 0.123 ± 0.02 agrees with the CHN value 0.17 ± 0.05 to within the combined experimental and theoretical uncertainties. Manifestly, this is a notable success for the CHN theory especially given that the experiments were done twenty years ago.

According to our heuristic argument, the crossover from 2D Heisenberg to 2D Ising behavior should occur when $h^A(\xi/a)^2 \sim 1$; for $h^A = 0.002$ this implies $(\xi/a)_{cr} \sim 23$. From Fig. 1 it may be seen that $(\xi/a) = 23$ for $T \sim 102$ K and indeed the data depart from the predicted Heisenberg behavior at that temperature. It is shown in Ref. 8 that for $|T/T_N - 1| < 0.05$, K_2NiF_4 exhibits 2D Ising critical behavior as one expects on general symmetry grounds. The crossover to 3D behavior occurs immeasurably close to T_N.

In summary, the CHN theory successfully predicts the temperature dependences of both the absolute value of the spin-spin correlation length and the relative static structure factor for the 2D $S = 1$ Heisenberg antiferromagnet K_2NiF_4. Indeed, these 1970 K_2NiF_4 experiments represent the most stringent test of the theory to date.

This analysis was stimulated by a conversation with A. Aharony and T. R. Thurston; we are grateful to both of these for the stimulus and for helpful conversations. We also thank S. Chakravarty for his enthusiastic response and for helpful conversations. This work was supported by the National Science Foundation–Low Temperature Physics Program under Contract No. DMR85-01856.

[1]See, for example, L. J. de Jongh and A. R. Meidema, Adv. Phys. **23**, 1 (1974).
[2]R. J. Birgeneau, J. Skalyo, Jr., and G. Shirane, Phys. Rev. B **3**, 1736 (1971).
[3]G. Shirane, Y. Endoh, R. J. Birgeneau, M. A. Kastner, Y. Hidaka, M. Oda, M. Suzuki, and T. Murakami, Phys. Rev. Lett. **59**, 1613 (1987); Y. Endoh, K. Yamada, R. J. Birgeneau, D. R. Gabbe, H. P. Jenssen, M. A. Kastner, C. J. Peters, P. J. Picone, T. R. Thurston, J. M. Tranquada, G. Shirane, Y. Hidaka, M. Oda, Y. Enomoto, M. Suzuki, and T. Murakami, Phys. Rev. B **37**, 7443 (1988).
[4]S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. Lett. **60**, 1057 (1988); Phys. Rev. B **39**, 2344 (1989).
[5]D. P. Arovas and A. Auerbach, Phys. Rev. B **38**, 316 (1988).
[6]K. Yamada, K. Kakurai, Y. Endoh, T. R. Thurston, M. A. Kastner, R. J. Birgeneau, G. Shirane, Y. Hidaka, and T. Murakami, Phys. Rev. B **40**, 4557 (1989).
[7]J. Skalyo, Jr., G. Shirane, R. J. Birgeneau, and H. J. Guggenheim, Phys. Rev. Lett. **23**, 1394 (1969).
[8]R. J. Birgeneau, J. Als-Nielsen, and G. Shirane, Phys. Rev. B **16**, 280 (1977).