Spin Correlations in an Isotropic Spin-5/2 Two-Dimensional Antiferromagnet

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We report a neutron scattering study of the spin correlations for the spin-5/2 two-dimensional antiferromagnet Rb_2MnF_4 in an external magnetic field. Choosing fields near the system's bicritical point, we tune the effective anisotropy in the spin interaction to zero, constructing an ideal $S = 5/2$ Heisenberg system. The correlation length and structure factor amplitude are closely described by the semiclassical theory of Cuccoli *et al.* over a broad temperature range, but show no indication of approaching the low-temperature renormalized classical regime of the quantum nonlinear sigma model. [S0031-9007(98)08151-4]

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Magnetic systems with reduced dimensionality have provided a basis for numerous insights into the varying roles quantum and thermal fluctuations play in driving phase transitions. The two-dimensional quantum Heisenberg antiferromagnet (2DQHA) represents a particularly important example of such a system, impacting problems in disciplines ranging from particle field theory to material science. For instance, recent work has illustrated the possible connections between the 2DQHA and the origins of superconductivity in the layered copper oxides [1]. Specifically, the parent compounds to the high T_c superconductors, such as La_2CuO_4, are good examples of 2DQHA's on a square lattice with spin, $S = 1/2$, and the magnetism persists into the superconducting state [1]. As a consequence of this connection, focus on the 2DQHA has intensified in recent years, and the combined efforts of experiment, theory, and simulation have yielded an increasingly cohesive picture of its behavior.

In mapping the 2DQHA onto the quantum nonlinear sigma model (QNLσM), Chakravarty, Halperin, and Nelson (CHN) have produced an effective field theory for the system which predicts renormalized classical behavior at low temperatures. Calculations from this theory (labeled CHN-HN [2]), describe a quasiexponential growth in the correlation length ξ with inverse temperature that matches measurements on $S = 1/2$ systems [3] with good absolute agreement over a remarkably large temperature range. However, high temperature results from systems with $S > 1/2$ vary markedly from the CHN-HN predictions [3–6]. These discrepancies have led to questions regarding the applicability of the effective field theory in the temperature range probed in these experiments and have motivated new approaches to describing the 2DQHA at high temperatures. For example, Elstner *et al.* [7] have applied series expansion techniques to characterize the onset of correlations with good accuracy. In addition, Cuccoli *et al.* [8] have recently proposed a semiclassical

theory, the pure quantum self-consistent harmonic approximation (PQSCHA). This theory calculates thermodynamic quantities, such as ξ, with respect to their classical values, with the effects of quantum fluctuations entering as a renormalization of temperature. While this semiclassical picture should be valid only at high temperature for $S = 1/2$, the authors predict that the theory should work over a broad range of temperatures for $S > 1/2$ [8].

A major obstacle to forming a unified picture of the 2DQHA, in which the relative merits of these theoretical approaches can be distinguished, has been the absence of experimental systems with $S > 1/2$ that model the 2DQHA over a broad range of length scales and, concomitantly, temperatures. In particular, the materials that have been studied, which include K_2NiF_4 [3] and La_2NiO_4 [4] with $S = 1$ and Rb_2MnF_4 [5] and $KFeF_4$ [6] with $S = 5/2$, possess appreciable Ising anisotropies that generate crossovers from 2D Heisenberg to 2D Ising behavior at quite short length scales. For example, in both Rb_2MnF_4 and $KFeF_4$ the crossover to Ising behavior is first apparent at $\xi/a \simeq 6$, where a is the lattice spacing. In contrast, the 2D $S = 1/2$ material $Sr_2CuO_2Cl_2$ exhibits model 2DQHA behavior in its spin correlations for length scales up to $\xi/a \simeq 200$ [3]. As spin number is the central parameter controlling the strength of quantum effects, measurements over a broad range of ξ away from the extreme quantum limit, $S = 1/2$, are crucial to enhance our understanding of the 2DQHA.

In this paper we present a study of Rb_2MnF_4 under conditions in which its spin space anisotropy is effectively reduced to zero, thus allowing us to characterize the Heisenberg behavior of this $S = 5/2$ system over a range of length scales which rivals those probed in the experiments on the $S = 1/2$ systems. Rb_2MnF_4 has the K_2NiF_4 crystal structure, in which magnetic planes of MnF_2 form a square lattice with nearest-neighbor spacing, $a = 4.215$ Å at 5 K. The Mn^{2+} ions ($S = 5/2$) interact

antiferromagnetically with nearest neighbors on the lattice with an isotropic exchange, $J = 7.36$ K, plus a uniaxial, dipolar field along the $\hat{c}$ axis (perpendicular to the planes) of strength, $g\mu_B H_A \approx 0.005J$ [9].

To reduce the effective anisotropy we place Rb_2MnF_4 in an external magnetic field, H, parallel to $\hat{c}$. As studied in detail by Cowley *et al.* [10], the phase diagram for Rb_2MnF_4 in a uniform magnetic field contains a spin-flop transition between the low-field Ising phase and a high-field XY phase. Results from Ref. [10] along with new measurements, shown in Fig. 1, reveal the $T = 0$, 2D bicritical point, where the boundaries between these phases and the paramagnetic phase meet, at $H_B = 5.30 \pm 0.12$ T [11]. The effective anisotropy, g, in the spin interaction varies with field as $g \sim H^2 - H_B^2 - CT$ [10], with $g < 0$ denoting easy axis anisotropy and $g > 0$ denoting easy plane. Thus, tuning the field at a given temperature provides a powerful mechanism for controlling the symmetry and magnitude of the effective anisotropy. In particular, for the fields at which $g = 0$ (the dashed line in Fig. 1), the effective anisotropy vanishes, and all spin components are critical. In our experiment we have carefully chosen fields to match this condition as closely as possible, thus avoiding a crossover to a lower symmetry transition. Specifically, we obtain $H_B = 5.30 \pm 0.12$ T and $C = 0.23 \pm 0.03$ T^2/K from the spin-flop line at low temperature ($T < 25$ K) and extrapolate this line into the paramagnetic region to determine the appropriate fields. This strategy has allowed us to track the Heisenberg behavior in Rb_2MnF_4
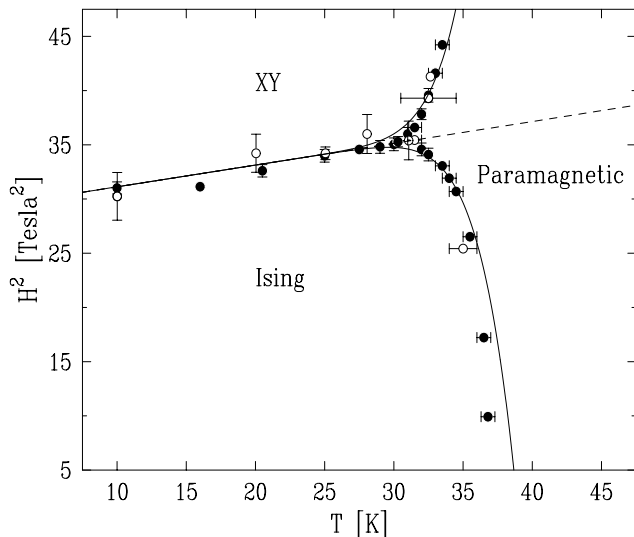
to significantly lower temperatures and, concomitantly, larger length scales than otherwise possible.

We have characterized the instantaneous magnetic correlations of this Heisenberg system by performing two-axis, energy-integrating neutron scattering measurements with the BT9 spectrometer at the NIST Center for Neutron Research. To accommodate the relative orientations of the spectrometer axes and the external field, we oriented the crystal such that $\hat{c}$ was perpendicular to the scattering plane. To guarantee that the quasielastic approximation was satisfied, we employed a large incident neutron energy, $E_{in} = 100$ meV. We created the 100 meV incident beam using the $(0,0,4)$ reflection from a pyrolytic graphite monochromator. The collimator sequence for the spectrometer was $40'$-$13'$-sample-$10'$, and a sapphire filter after the monochromator removed higher order contamination. We model the static structure factor, $S(\mathbf{Q}_{2D})$, where $\mathbf{Q}_{2D}$ is the momentum transfer in the magnetic planes, with a simple Lorentzian line shape

$$I(\mathbf{Q}_{2D}) \propto S(\mathbf{Q}_{2D}) = \frac{S_0}{(1 + \mathbf{q}_{2D}^2 \xi^2)}, \qquad (1)$$

where $\mathbf{q}_{2D}$ is the wave vector measured from the antiferromagnetic zone center, $\mathbf{q}_{2D} = \mathbf{Q}_{2D} - (1/2, 1/2, 0)$. Figure 2 displays scattering profiles from the two-axis scans along the direction $(K/2, -K/2, 0)$ at three representative temperatures. We fit the scattering intensity to Eq. (1) convolved with the spectrometer resolution function together with a sloping background. The fit results,

FIG. 1. Phase diagram for Rb_2MnF_4 in a magnetic field H parallel to the $\hat{c}$ axis. The solid circles (shifted by 0.15 T) are from Cowley *et al.* [10], and the open circles are from the current experiment. The solid lines are a fit to the phase boundaries, $g \sim T^{-2}\xi^{-2}$, using the results for ξ from Fig. 3a. Measurements of the spin correlations at zero effective anisotropy are taken along the dashed line, which extends from the $T = 0$, 2D bicritical point at $H_B = 5.30 \pm 0.12$ T.
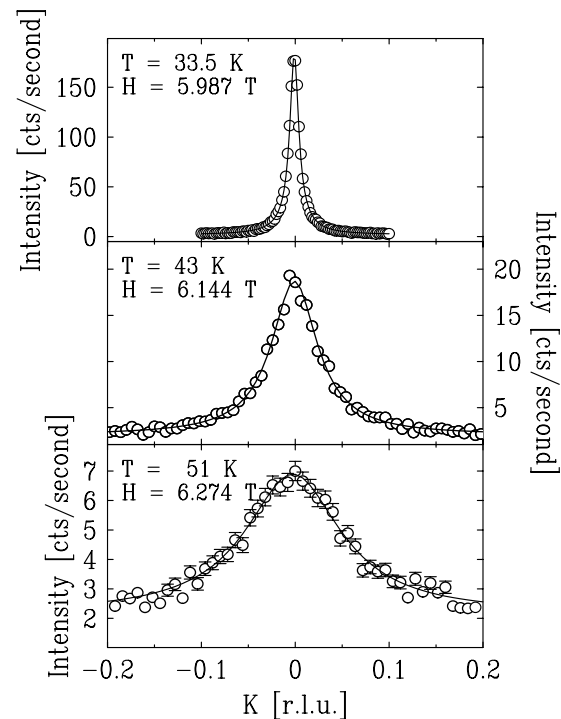

FIG. 2. Two-axis scans along the direction $(K/2, -K/2, 0)$ at field and temperature values for which the effective anisotropy in the spin interaction is near zero. The solid lines are fits to Eq. (1) convolved with the instrumental resolution function.

shown with solid lines in Fig. 2, describe the data very well at all temperatures. In addition, to test the validity of the quasielastic approximation, we have characterized the shape of the dynamic structure factor, $S(\mathbf{Q}_{2D}, \omega)$, in energy transfer, ω, by performing additional three-axis measurements scanning ω at $\mathbf{q}_{2D} = 0$. A Lorentzian line shape in ω fits these three-axis scans well. An analysis of the two-axis scans which accounts explicitly for the shape of the dynamic structure factor gives results for ξ and S_0 that agree with those obtained assuming the quasielastic approximation to within 5% at all temperatures, thus confirming the validity of the approximation for our measurements.

Figure 3a shows ξ/a extracted from the fits on a log scale versus $JS(S + 1)/T$. We choose $JS(S + 1)$ rather than JS^2 for the energy scale following the observation by Elstner et al. [7] that ξ depends on S primarily through $JS(S + 1)$ at high temperature for $S > 1$. Also plotted
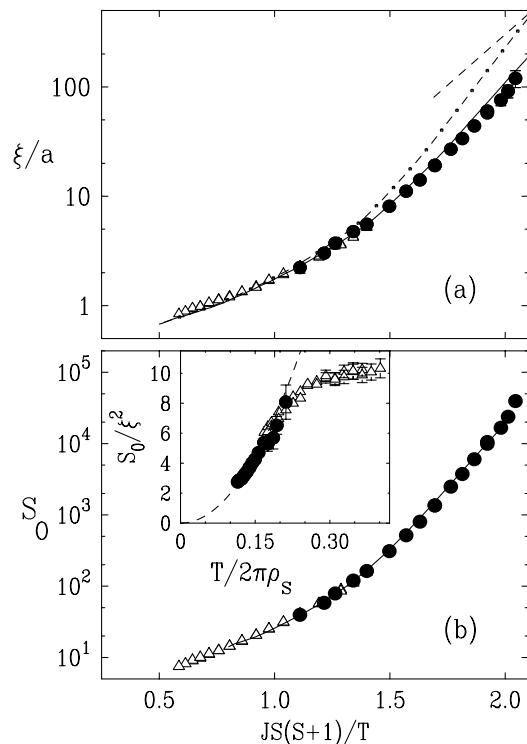

FIG. 3. (a) The magnetic correlation length, ξ, in units of the lattice spacing, a, from the present experiment (solid circles) and measured at zero field in the traditional scattering geometry (open triangles) [5]. The solid line is the prediction from the semiclassical theory of Cuccoli et al. [8]. The dash-dotted line is the correlation length for the classical Heisenberg model obtained from simulation [7,12]. The dashed line is the prediction from low-temperature QNLσM effective field theory of CHN-HN [2]. (b) S_0, the amplitude of the static structure factor at the ordering wave vector, from the present experiment (solid circles) and measured at zero field in the traditional scattering geometry (open triangles) [5]. The solid line is the prediction from the semiclassical theory of Cuccoli et al. [8]. The inset shows S_0/ξ^2 versus temperature. The dashed line is a fit to the solid circles assuming $S_0/\xi^2 \sim T^2$.

are the results from Lee et al. [5] for Rb_2MnF_4 in zero magnetic field. The good agreement between the two measurements in the temperature region in which they overlap confirms that our method to determine the instantaneous spin correlations is accurate. In addition, as the plot illustrates, by reducing the effective anisotropy to zero we have been able to extend significantly the temperature range of the Heisenberg behavior. At our lowest temperature ξ/a exceeds 100, rivaling the experimental range for the best $S = 1/2$ Heisenberg systems.

The solid line in Fig. 3a is the prediction for ξ/a from the PQSCHA [8]. As the figure demonstrates, the calculated behavior from the semiclassical theory agrees closely with the measured correlation lengths over the full experimental range. As mentioned above, the PQSCHA calculates ξ with respect to its value for the classical Heisenberg model. For reference, the dash-dotted line shows the correlation length for the classical model, ξ_{CL}, as obtained from simulation [7,12]. At high temperature ($\xi/a < 5$), ξ closely approximates ξ_{CL} when the energy scale is taken as $JS(S + 1)$. However, as the figure demonstrates, with decreasing temperature quantum effects become increasingly important, and this simple $S(S + 1)$ scaling breaks down. The renormalization of temperature that the PQSCHA provides accounts for the quantum effects nicely.

Sokol et al. [13] have argued that the $S = 5/2$ 2DQHA should cross over from classical scaling to renormalized classical behavior near $T \sim \Lambda$, where Λ is the upper bound of the spin wave energy spectrum (approximately 5.5 meV at 35 K). Thus, we estimate this temperature for our system to be near 60 K [$JS(S + 1)/T \approx 1.1$], roughly where the deviations between ξ_{CL} and the experimental results in Fig. 3a become noticeable. However, the low-temperature, renormalized classical prediction for the QNLσM from CHN-HN [2], shown with the dashed line in Fig. 3a, deviates significantly from the measured correlation lengths even for ξ/a exceeding 10^2. This result contrasts with those for the $S = 1/2$ systems which show good agreement with the theory over length scales down to $\xi/a \sim 1$. However, recent quantum Monte Carlo simulations combined with finite size scaling have indicated that this experimental agreement is in part accidental due to a cancellation among the higher order corrections to the QNLσM [14]. In addition, Elstner et al. have argued that the onset of the renormalized classical behavior described by the QNLσM should occur at progressively higher ξ for larger S [7]. Our measurements on this $S = 5/2$ 2DQHA, showing deviations from the projected asymptotic behavior even as the measurements extend well below the classical scaling regime, support the view of an extended crossover region between the classical and renormalized classical regimes.

In Fig. 3b we display S_0, the amplitude of the static structure factor at the ordering wave vector, extracted from the measurements. The zero field results [5], shown

with open triangles, have been placed on an absolute scale by setting the high temperature limit to $S(S + 1)/3$. Our results, placed on the same scale, extend the data smoothly to lower temperature. The solid line in Fig. 3b is the prediction from the PQSCHA with no adjustable parameters. As with ξ, the PQSCHA predicts S_0 accurately over a broad temperature range.

For both the classical [15] and quantum [2] 2DQHA's, low-temperature theories predict $S_0/\xi^2 \propto T^2$. However, experiments for $S = 1/2$ [3] and $S = 1$ [4] over the range $0.2 < T/2\pi\rho_s < 0.4$, where ρ_s is the zero-temperature spin stiffness, have found S_0/ξ^2 to depend weakly on T. The inset to Fig. 3b shows the results for this ratio for Rb_2MnF_4 ($\rho_s = 43.6$ K). As the figure illustrates, at high temperature S_0/ξ^2 is roughly constant, but at lower temperature the ratio has a strong temperature dependence, qualitatively consistent with the low-temperature theories. The break from temperature independent S_0/ξ^2 occurs near $T/2\pi\rho_s = 0.2$, the lower limit of results on other experimental systems. Thus, the measurements to low temperature ($T/2\pi\rho_s \simeq 0.11$) in Rb_2MnF_4 appear to have captured a crossover from this high temperature regime.

Finally, we note that the bicritical phase boundaries themselves should reflect the 2D correlations. Specifically, the Ising and XY critical lines should approach each other tangentially with $g \sim T^{-2}\xi^{-2}$ [10]. The solid lines in Fig. 1 correspond to this form with ξ from Fig. 3a. In the fit the $g = 0$ line is fixed by the low temperature spin-flop data, and only the Ising and XY amplitudes are adjusted. Clearly, the agreement is good for temperatures below ~35 K.

In conclusion, the presence of a bicritical point in the phase diagram of Rb_2MnF_4 has provided a direct and convenient mechanism for controlling the effective anisotropy in the spin interaction of this two-dimensional $S = 5/2$ antiferromagnet. In the present study we have focused on conditions in which the effective anisotropy is essentially zero, allowing us to track the growth of correlations in this large-spin, two-dimensional Heisenberg system to correlation lengths exceeding 100 lattice units. The success of this approach suggests that similar studies near the bicritical point in tetragonal systems with other spin quantum numbers would be fruitful in further extending our under-

standing of the 2DQHA. Our measurements for $S = 5/2$ demonstrate the success of a semiclassical theory, the PQSCHA, over a broad temperature range. The disagreement between the measured behavior and the predictions of the effective field theory based on the QNLσM, even as ξ becomes large, confirms speculations that the temperature range applicable to this theory lies well below that accessible in experiment. However, the growing departure from the simple scaling to classical behavior in Fig. 3a along with the marked temperature dependence of S_0/ξ^2 suggests behavior distinct from that seen at high temperature. Thus, the experiment reveals an extensive temperature region for the crossover between the classical and renormalized classical regimes. A quantitative theory for this crossover would be most valuable.

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