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Critical dynamics of a spin-$\frac{5}{2}$ two-dimensional isotropic antiferromagnet

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We report a neutron-scattering study of the dynamic spin correlations in Rb_2MnF_4, a two-dimensional spin-5/2 antiferromagnet. By tuning an external magnetic field to the value for the spin-flop line, we reduce the effective spin anisotropy to essentially zero, thereby obtaining a nearly ideal two-dimensional isotropic antiferromagnet. From the shape of the quasielastic peak as a function of temperature, we demonstrate dynamic scaling for this system and find a value for the dynamical exponent z. We compare these results to theoretical predictions for the dynamic behavior of the two-dimensional Heisenberg model, in which deviations from $z=1$ provide a measure of the corrections to scaling.

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Recently, interest in the two-dimensional (2D) square-lattice Heisenberg antiferromagnet has intensified due in large part to the discovery of high-temperature superconductivity in the doped lamellar cuprates and the subsequent realization of the near-ideal 2D Heisenberg nature of their parent compounds.[1] The nearest-neighbor Heisenberg model is defined as:

$$H=J\sum_{<i,j>}\mathbf{S}_i\cdot\mathbf{S}_j\,, \tag{1}$$

where J is the nearest-neighbor coupling which is positive for an antiferromagnet. Classically, $\mathbf{S}_i$ is a three component vector of magnitude $\sqrt{S(S+1)}$ representing the spin at site i, while quantum mechanically $\mathbf{S}_i$ is the quantum spin operator.

As a result of a symbiotic interplay among theory, simulation, and experiment, great progress in understanding the instantaneous spin correlations of the 2D Heisenberg antiferromagnet has been made in recent years. Chakravarty, Halperin, and Nelson (CHN)[2] developed an effective field theory from which an exact low-temperature expression for the instantaneous correlation length, ξ, has been found.[3] While this expression agrees closely with experiments on spin-1/2 Heisenberg systems,[4] measurements on systems with $S>1/2$ display strong deviations from the predicted behavior.[4,5] Subsequent work[6,7] has pointed towards a broad crossover from classical behavior at high temperature to the renormalized classical regime where field theory is valid. Recently, Hasenfratz[8] incorporated cutoff effects into the field theory formalism to describe the behavior in this crossover region.

The dynamics of the 2D Heisenberg antiferromagnet has likewise been the subject of detailed theoretical work. Tyc, Halperin, and Chakravarty (THC)[9] combined renormalization group analysis and the dynamic scaling theory[10] with simulations of the classical lattice rotor model to predict a form for the dynamic structure factor. Classical molecular dynamics[11] and quantum Monte Carlo simulations[12] have lent credence to their predictions; however, due to a lack of suitable systems, comparatively few experimental studies of dynamics in 2D Heisenberg antiferromagnets have been performed. Some of the most ideal 2D Heisenberg systems (La_2CuO_4 and $Sr_2CuO_2Cl_2$) have a very large intersite coupling J ($J\approx 1500$ K), making quantitative results using conventional neutron-scattering techniques difficult to obtain. Consequently, previous experiments have not resolved the quasielastic scattering from the long-wavelength spin-wave excitations.[13–15] In this communication, we present a neutron-scattering study of Rb_2MnF_4, a quasi-two-dimensional spin-5/2 system with an effective spin anisotropy that can be tuned to zero using an external magnetic field. Our results provide a detailed characterization of the dynamic structure factor in the quasielastic region. Previous studies[5,16] indicate that this system behaves like a nearly ideal 2D Heisenberg antiferromagnet. Accordingly, we compare our findings with the current theoretical understanding of 2D Heisenberg critical dynamics.

Following a strategy introduced in our previous work,[5] we exploit the presence of a bicritical point in the field-temperature phase diagram of Rb_2MnF_4 to make possible a study of the dynamic spin correlations of a near-ideal Heisenberg system over a large range of correlation lengths. Rb_2MnF_4 has the tetragonal K_2NiF_4 crystal structure with an in-plane lattice constant of $a=4.215$ Å and an out-of-plane lattice constant of 13.77 Å. The large ratio of the out-of-plane to the in-plane lattice constant combines with the frustration due to the body-centered stacking to make it a nearly two-dimensional magnetic system, with an interplane coupling of less than $10^{-4}J$.

At zero field, Rb_2MnF_4 is a weakly Ising antiferromagnet with $J=0.63$ meV.[17] This interaction energy J is more than two orders of magnitude smaller than that of the lamellar copper oxide Heisenberg systems, thus making the energy scale of the dynamics much more accessible for neutron-scattering studies. The principal spin anisotropy is a magnetic dipole interaction, with $g\mu_B H_A=0.032$ meV (Ref. 17) along the c axis (perpendicular to the magnetic plane). Correspondingly, when a field of approximately 5.5 T (depending on temperature) is applied parallel to the c axis, the spins

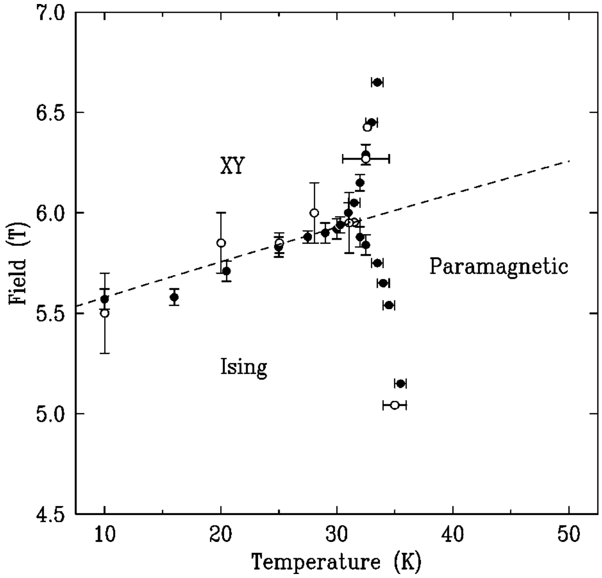

FIG. 1. Phase diagram for Rb_2MnF_4 in an external magnetic field perpendicular to the magnetic planes. Open symbols are our measurements of the phase boundaries; filled symbols are measurements from Cowley *et al.* (Ref. 18) shifted by $+0.15$ T. The dashed line indicates the line of zero effective anisotropy.

flop into the plane. Above this spin-flop transition, the system has XY symmetry. Precisely along the spin-flop line, and on the extension of the line into the paramagnetic phase, the anisotropy is effectively zero, so that the system should be in the 2D Heisenberg universality class (see Fig. 1).

Experiments were conducted at the NIST Center for Neutron Research using NIST's 7 T superconducting magnet. We aligned the c axis within 0.5° of the magnetic field to minimize any induced in-plane anisotropies. We took field scans at several temperatures to confirm the phase diagram and found the line of zero anisotropy (shown in Fig. 1) to be approximately: $H=\sqrt{28.09+0.23T}$ where T is the temperature in K. This is in accordance with the form given by Cowley *et al.*[18]

Studies of the quasielastic scattering were performed with the thermal neutron triple-axis spectrometer BT9 and the cold neutron spectrometer SPINS. At BT9, we used a fixed initial energy of either 13.7 or 14.8 meV with a pyrolytic graphite filter before the sample to remove higher harmonics in the incident beam. Collimations of 40'-27'-Sample-24'-60' were typical, giving an energy resolution of 0.8 meV full width at half maximum (FWHM). For lower temperatures where higher resolution was needed, we used SPINS with a fixed final energy of 4 meV and collimations of guide-20'-S-20'-open, which gave a resolution of 0.12 meV FWHM.

Figure 2 shows the scattered intensity as a function of energy at the antiferromagnetic zone center for several temperatures. The two-dimensional Heisenberg antiferromagnet, in accordance with the Hohenberg-Mermin-Wagner theorem, has no transition to long-range order above zero temperature. At nonzero temperature it has correlated regions whose characteristic length scale diverges exponentially with inverse temperature. These correlated regions have a finite lifetime which translates into a nonzero energy width of the quasi-

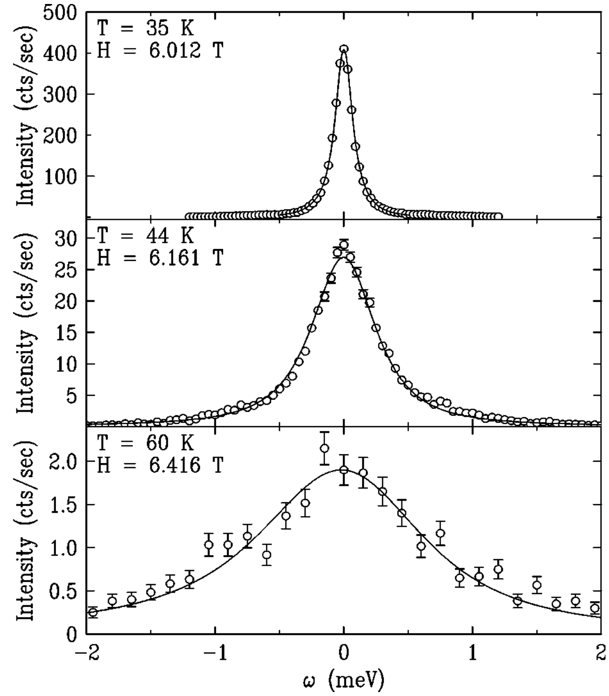

FIG. 2. Energy scans through the quasielastic peak at the antiferromagnetic zone center (0 1 0) at field and temperature values along the zero anisotropy line. The solid lines show fits to Eqs. (2)–(4). Scans shown were taken at BT9.

elastic peak produced in the dynamic structure factor. As the temperature is lowered towards zero, the correlated regions become progressively more stable, and the energy width of the peak decreases. The measurements in Fig. 2 display this critical slowing down.

According to dynamic scaling theory, the functional form of the structure factor is independent of temperature. The temperature dependence enters only through the reduced reciprocal space position k (2D reciprocal space distance from the magnetic zone center) and frequency ω, which are scaled by ξ, the correlation length, and ω_0, the characteristic frequency, respectively:[10] $q\equiv k\xi$ and $\nu\equiv\omega/\omega_0$, so that q and ν are both dimensionless. In addition, the characteristic frequency is predicted to scale with the correlation length to a power $-z$, with $z=d/2$ for a Heisenberg antiferromagnet, where d is the spatial dimension.

In accordance with these predictions, we fit the energy scans through the quasielastic peak at (0 1 0) to the dynamic structure factor:

$$S(k,\omega)=\omega_0^{-1}S(q)\Phi(q,\nu). \tag{2}$$

We took Lorentzian forms for $S(q)$ and $\Phi(q,\nu)$:

$$S(q)=\frac{S_0}{1+q^2} \tag{3}$$

$$\Phi(q,\nu)=\frac{\gamma_q^{-1}}{1+\frac{\nu^2}{\gamma_q^2}}, \tag{4}$$

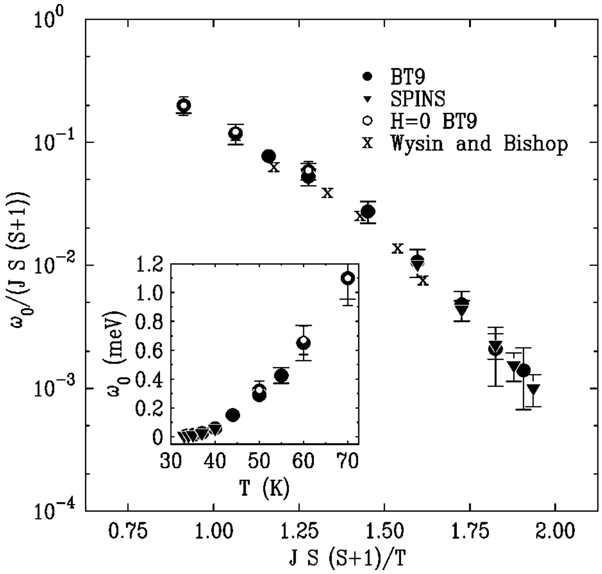

FIG. 3. Measured energy width of the (0 1 0) quasielastic peak as a function of temperature scaled by $JS(S+1)$. Classical simulation data by Wysin and Bishop has been multiplied by an arbitrary constant as per their paper. The inset shows our raw data.

with $\gamma_q=(1+\mu q^2)^{1/2}$ (μ is an arbitrary constant). Due to the finite resolution, q-dependent contributions were needed to reproduce accurately the observed lineshape. $\mu=1.7\pm0.2$ gave the best fit at all temperatures, in agreement with the values (1.4 and 2.0) found by THC in their analyses of the classical lattice rotor model. For the fits, ξ was fixed at the values determined in our previous study,[5] and the temperature dependence of S_0 was found to agree well with the results of those measurements. The correlation length for the temperature range accessible for studying the dynamics varied from $1<\xi/a<60$.

As Fig. 2 demonstrates, fits to this form convolved with the experimental resolution are quite good at all temperatures. Figure 3 shows the results for the energy widths extracted from these fits. Note that data taken at SPINS and BT9 agree closely in the overlapping region. This agreement gives us confidence that we have correctly accounted for the very different experimental resolutions in the two measurements.[19] When the temperature is scaled by $JS(S+1)$, the temperature dependence of $\omega_0(T)$ agrees well with the results of classical molecular dynamics simulations carried out by Wysin and Bishop.[11] They predict a temperature scaling factor of JS^2, but when scaled by this factor, our data show a much stronger temperature dependence than that exhibited by the simulation data. Normalizing temperature by the classical spin stiffness $JS(S+1)$ has been shown[6] to collapse the instantaneous correlation length data for 2D quantum Heisenberg antiferromagnets with $S>1$ onto the classical results at high temperatures, and here again succeeds in reconciling the spin-5/2 data with corresponding results for the classical system. Thus, as with ξ, the dynamic behavior follows classical scaling at high temperature ($1<\xi/a<10$).

Similar measurements of the quasielastic energy width have been performed by Fulton *et al.*[20] on $KFeF_4$, another 2D spin-5/2 antiferromagnet. These results agree with our

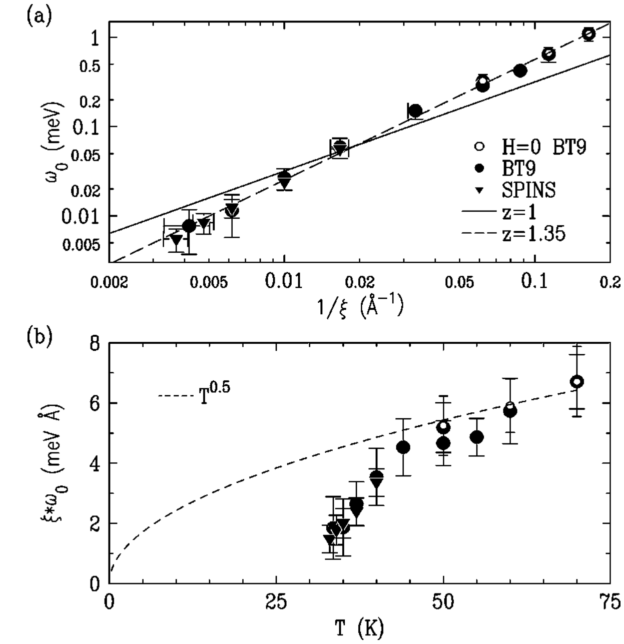

FIG. 4. (a) ω_0 versus ξ showing best fits to $z=1$ and 1.35 to the entire range of data. (b) $\omega_0\xi$ versus T to illustrate remnant temperature dependence if z is assumed to be 1. The dashed line shows the $T^{0.5}$ corrections to scaling predicted by CHN.

data at the highest temperatures, but deviate strongly at lower temperatures. We believe that this discrepancy results from a crossover to Ising critical behavior in $KFeF_4$. Studies of Rb_2MnF_4 in zero field[21] show that the Ising crossover occurs near $1.2T_N$. $KFeF_4$ has nearly the same reduced Ising anisotropy as Rb_2MnF_4, and hence would also be expected to enter a region of Ising critical behavior below $1.2T_N$. All but the highest temperatures from the $KFeF_4$ study therefore lie below the Ising crossover region.

As mentioned above, dynamic scaling theory predicts $\omega_0\propto\xi^{-z}$ with $z=1$ for the 2D Heisenberg antiferromagnet.[10] In addition, CHN predict corrections to scaling which go as $T^{1/2}$: $\omega_0=c\xi^{-1}(T/2\pi\rho_s)^{1/2}$ where c is the spin-wave velocity, and ρ_s is the spin stiffness. Figure 4(a) shows a plot of ω_0 versus $1/\xi$; the best fit to the simple form $\omega_0\propto\xi^{-z}$ gives $z=1.35\pm0.02$. This value for z is intermediate between the values for the 2D ($z=1$) and 3D ($z=1.5$) Heisenberg antiferromagnets. However, as detailed below we believe only a 2D model is relevant here. Figure 4(b), which shows the product $\omega_0\xi$ versus temperature, demonstrates the corrections to scaling if $z=1$ is assumed. Clearly, corrections are stronger than the $T^{1/2}$ predicted by CHN. Simulations of the classical model, as mentioned above, agree with our data for ω_0, yet they claim to see a different temperature dependence for the product $\omega_0\xi$. This is most likely due their use of a form for $\xi(T)$ that has since been shown to be inaccurate in this temperature range. Monte Carlo studies on a spin-1/2 system[12] have also indicated that $z=1$, but with a temperature independent prefactor in agreement with predictions by Arovas and Auerbach.[22]

The data in Figs. 3 and 4, taken at face value, may suggest that $z>1$ for Rb_2MnF_4 along the bicritical line, or that there is a crossover to some other critical behavior with a nonzero phase transition temperature. Explanations involving a crossover to three-dimensional behavior seem unlikely in light of previous studies[16,21] at zero field showing that Rb_2MnF_4 behaves as a nearly ideal two dimensional system to very large correlation lengths. Likewise, 2D Ising or 2D XY behavior are precluded by the high temperature at which we observe deviations from $z=1$ behavior, as compared to the scales at which these crossovers should occur, as well as by our previous results on the statics,[5] which agree very well with theory and simulation for the 2D Heisenberg model.

However, the dynamic scaling near the bicritical point could still conceivably differ from that of the ideal 2D Heisenberg antiferromagnet. While the universality class for static critical behavior is determined solely by the symmetry properties and the spatial dimension, the dynamics can also be affected by conserved quantities and the Poisson-bracket relations they satisfy.[10] The bicritical region differs from a true isotropic system due to the nonzero, conserved uniform magnetization along the applied field direction. Noting this distinction, Dohm and Janssen[23] performed a renormalization group study of bicritical dynamics in $4-\epsilon$ dimensions. They found that dynamic scaling was obeyed, but that the exponent for the 3D bicritical point was larger than that for the 3D Heisenberg model. To explore the possibility that we might be seeing a similar effect in our 2D system, we measured ω_0 in zero field at temperatures above the Ising-Heisenberg crossover. These results, shown in Fig. 4(a), overlap closely with the data taken at the same temperatures on the bicritical line. This indicates that, for this temperature range, the magnetic field itself is not measurably affecting the quasielastic width. Clearly, additional theoretical work on 2D bicritical dynamics and corrections to dynamic scaling for the 2D Heisenberg antiferromagnet would greatly elucidate the findings from these measurements.

With these measurements of the dynamic spin correlations in Rb_2MnF_4 near the bicritical point, we have provided an experimental study of the quasielastic behavior in a 2D isotropic antiferromagnet. These results are largely consistent with the current theoretical understanding of the dynamics of the 2D Heisenberg model, but also raise some questions. The shape of the dynamic structure factor in the quasielastic region obeys a form consistent with dynamic scaling, and the temperature dependence of the characteristic frequency ω_0 is consistent with the anticipated form ξ^{-z}, though with z larger than the predicted value $z=1$. To establish whether the difference in z originates in stronger corrections to scaling than predicted or indicates a distinction between ideal 2D Heisenberg dynamic scaling and the dynamic behavior near a 2D bicritical point will require further theoretical work as well as experimental studies of other Heisenberg antiferromagnets.

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[1] For a review, see M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998).

[2] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. B **39**, 2344 (1989).

[3] P. Hasenfratz and F. Niedermayer, Phys. Lett. B **268**, 231 (1991).

[4] M. Greven *et al.*, Z. Phys. B: Condens. Matter **96**, 465 (1995).

[5] R. L. Leheny, R. J. Christianson, R. J. Birgeneau, and R. W. Erwin, Phys. Rev. B **82**, 418 (1999).

[6] N. Elstner *et al.*, Phys. Rev. Lett. **75**, 938 (1995).

[7] A. Cuccoli, V. Tognetti, R. Vaia, P. Verrucchi, Phys. Rev. Lett. **77**, 3439 (1996).

[8] P. Hasenfratz, Eur. Phys. J. B **13**, 11 (1999).

[9] S. Tyc, B. I. Halperin, and S. Chakravarty, Phys. Rev. Lett. **62**, 835 (1989).

[10] P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).

[11] G. M. Wysin and A. R. Bishop, Phys. Rev. B **42**, 810 (1990).

[12] M. Makivic and M. Jarrell, Phys. Rev. Lett. **68**, 1770 (1992).

[13] S. M. Hayden *et al.*, Phys. Rev. Lett. **67**, 3622 (1991).

[14] K. Yamada *et al.*, Phys. Rev. B **40**, 4557 (1989).

[15] K. R. Thurber *et al.*, Phys. Rev. Lett. **79**, 171 (1997).

[16] R. J. Birgeneau, H. J. Guggenheim, and G. Shirane, Phys. Rev. B **1**, 2211 (1970).

[17] R. A. Cowley, G. Shirane, R. J. Birgeneau, and H. J. Guggenheim, Phys. Rev. B **15**, 4292 (1977).

[18] R. A. Cowley *et al.*, Z. Phys. B: Condens. Matter **93**, 5 (1993). Our results agree quantitatively with this study of the phase diagram of Rb_2MnF_4 provided that we shift the bicritical point by about 0.15 T. This shift likely arises from minor differences in sample quality and small differences in the calibrations of the magnets used for the experiments.

[19] In addition, the sample was remounted in the magnet twice during the experiment. The good reproducibility indicates that any small possible misalignments of the sample in the magnetic field do not adversely affect the data.

[20] S. Fulton, R. A. Cowley, A. Desert, and T. Mason, J. Phys.: Condens. Matter **6**, 6679 (1994).

[21] Y.S. Lee *et al.*, Eur. Phys. J. B **5**, 15 (1998).

[22] A. Auerbach and D. P. Arovas, Phys. Rev. Lett. **61**, 617 (1988).

[23] V. Dohm and H. K. Janssen, Phys. Rev. Lett. **39**, 946 (1977).