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Spin Correlations near the Percolation Concentration in Two Dimensions

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Neutron scattering measurements in the dilute antiferromagnets $Rb_2Mn_cMg_{1-c}F_4$ with $c=0.54$ and 0.57 are presented; for this system the percolation limit is $c_p=0.59$. Two-dimensional critical scattering is observed with inverse width ξ and amplitude χT which diverge as a function of $1/T$; the divergences cut off when ξ exceeds the size of the larger clusters. A simple self-avoiding walk model accounts well for the observed ξ vs T behavior.

Recently, considerable attention has been directed towards the percolation transition as an example of "geometrical" critical behavior.[1] Extensive Monte Carlo computer experiments have been performed[2]; in addition, by exploiting an analogy with the Ashkin-Teller-Potts model, some analytical results have been obtained in two dimensions and in $6-\epsilon$ dimensions using renormalization-group methods.[3] As a model percolative system, one may consider a simple square magnet with only nearest-neighbor bonds. As the concentration c of magnetically active atoms is reduced below some critical concentration $c=c_p$, the system breaks up into finite clusters so that there can be no long-range order. Theoretical work to date has concentrated on the critical behavior of such a magnet at $T=0$ around $c=c_p$. More generally, however, the point $c=c_p$, $T=0$ might be considered a *multicritical point* terminating a line of second-order transitions.[4] Such a system then might be expected to exhibit geometrically driven critical behavior at $T\sim 0$ as a function of $c-c_p$ and thermally driven critical behavior at $c=c_p$ as a function of T. Virtually no experimental information is currently available on the T-dependent fluctuation behavior around $c=c_p$ in any real system.

In this Letter we present the results of neutron scattering experiments in the dilute two-dimensional (2D) antiferromagnets $Rb_2Mn_cMg_{1-c}F_4$. The spin correlations in these crystals are found to exhibit a number of interesting and novel features as we shall discuss in detail below. In particular, the data suggest that the propagation of correlations over long distances is determined mainly by the one-dimensional links in the clusters; we show that the principal features of the correlations can be quantitatively accounted for by a no-adjustable-parameter, self-avoiding walk model. In general, we hope that these new results will provide the impetus for the development of theories of percolation in the concentration-temperature plane, and of course, that they will act as a testing ground for such theories.

Our experiments were performed on two high-quality single crystals of $Rb_2Mn_cMg_{1-c}F_4$. These alloys have the Rb_2MnF_4 structure but with the Mn ($S=\frac{5}{2}$) and Mg (nonmagnetic) atoms arranged randomly on the Mn sites of the pure crystal. In the pure crystal the magnetic interactions are

overwhelmingly between nearest neighbors (nn) only. Measurements of the Rb_2MnF_4 magnon spectra[5] show that the spin system is accurately described by an isotropic exchange Hamiltonian ($\mathcal{H}_{ij}=J_{ij}\vec{S}_i\cdot\vec{S}_j$) with $J_{nn}=0.652$ meV, $J_{nnn}=0.012$ meV, and an anisotropy field $g\mu_B H_A=0.031$ meV; the latter arises mainly from magnetic dipole interactions. In the dilute systems we have carried out a variety of crystallographic measurements; in addition, a detailed study was made of the magnetic excitation spectra at the antiferromagnetic zone boundary. These results will be reported in detail later[5]; for our purposes here, it is sufficient to note that these experiments indicate that the Mn and Mg ions are indeed randomly distributed; they also enable us to estimate $c=0.54\pm0.03$ and $c=0.57\pm0.03$ for the two specimens. From the inelastic spectra we conclude that for the dilute samples $J_{nn}\simeq 0.71$ meV. We cannot extract J_{nnn} directly for the dilute samples. For the square lattice with nn interactions alone, the site percolation limit[1] is $c_p=0.59$. Thus these samples both lie just below this limit. From theory[1] we expect that inclusion of the much weaker nnn coupling would lower c_p to 0.41. However, this nnn coupling has no observable effects down to 1.3 K in these crystals and so we shall not consider it further here.

The critical scattering measurements were carried out on a two-axis spectrometer at the Brookhaven high-flux beam reactor. The crystal orientation, spectrometer configuration, and scattering geometry were identical to those described previously by Als-Nielsen *et al.*[6] In this geometry the measurements yield

$$I(\vec{q})=0.95\mathcal{S}^{\parallel}(\vec{q})+1.05\mathcal{S}^{\perp}(\vec{q}), \qquad (1)$$

where $\mathcal{S}^{\alpha\alpha}(q)=\sum_{\vec{r}}e^{i\vec{q}\cdot\vec{r}}S_0{}^{\alpha}(0)S_{\vec{r}}{}^{\alpha}(0)$. Here $\parallel$ and $\perp$ are with respect to the crystalline c axis. Typical scans on the $c=0.54$ sample are shown in Fig. 1. In none of our measurements is the scattering found to have any 3D character; thus the system can indeed be treated as ideally 2D.

We now consider the analysis of these data. Because of the near-Heisenberg symmetry of the Hamiltonian, we assume as a first approximation that $\mathcal{S}^{\parallel}(\vec{q})=\mathcal{S}^{\perp}(\vec{q})$, and hence $I(\vec{q})=A/(\kappa^2+q^2)$ with $q^2=q_x{}^2+q_y{}^2$; that is, the scattering has the form of a 2D Lorentzian. This expression for the intrinsic scattering cross section is then convoluted with the instrumental resolution function and fitted in a least-squares sense to the experimental data at each temperature. As shown in Fig. 1, satisfactory fits with goodness-of-fit parameters

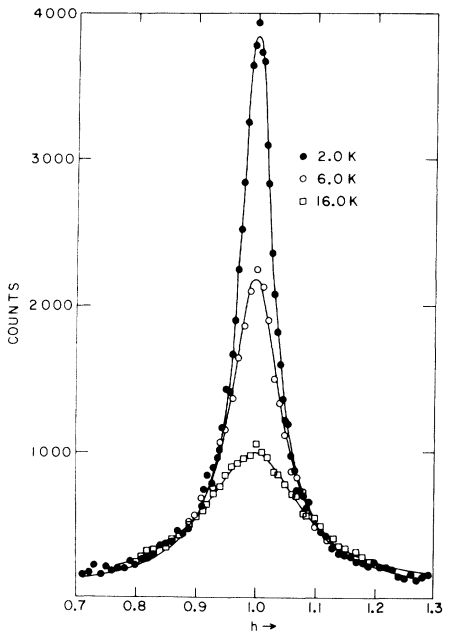

FIG. 1. Critical scattering scans across the ridge at $(h,0,0.4)$ in $Rb_2Mn_{0.54}Mg_{0.46}F_4$ at several temperatures (see Ref. 6 for the geometry). The horizontal instrumental resolution full width at half-maximum is $0.011h$. The solid lines are the fitted Lorentzians as discussed in the text.

$\chi^2\sim 1$ to 1.5 are obtained at all temperatures. This in turn demonstrates that the spin correlations fall off as $e^{-\kappa R}/R^{1/2}$ in real space—an interesting result in itself. We show in Fig. 2 the results so obtained for κ in the two specimens. We

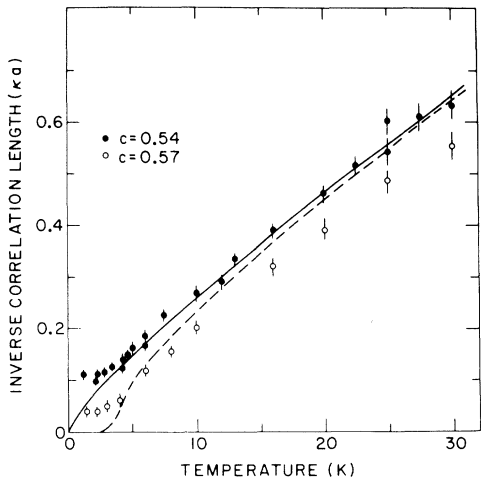

FIG. 2. Inverse correlation length in reduced units ($\kappa a=1/n$, where n is the number of lattice units) versus temperature for $c=0.54$ and 0.57. The dashed and solid lines are the SAW results with and without anisotropy, respectively, as discussed in the text.

shall discuss the significance of these results in detail below.

We consider first, however, the role of the anisotropy. For the pure systems it is known that the anisotropy plays a crucial role in the critical region.[6] However, in those measurements κ is typically an order of magnitude smaller than the values of κ where these data saturate. Ideally, $\mathcal{S}^{\parallel}(q)$ and $\mathcal{S}^{\perp}(q)$ should be measured separately. However, as discussed extensively in Ref. 6, because of restrictions on the quasielastic approximation, only the combination, Eq. (1), can be obtained with confidence. However, by using energy analysis and a small incident neutron energy of 5 meV, the zero-energy (or rather $|\hbar\omega| < 0.08$ meV) part of the full momentum- and energy-dependent van Hove scattering function can be obtained. The results so obtained for $\mathcal{S}^{\parallel}(q^*,E\sim 0)$ and $\mathcal{S}^{\perp}(q^*,E\sim 0)$ are shown in Fig. 3. Here q^* is the superlattice wave vector corresponding to the peak in Fig. 1, and $E\sim 0$ refers to an integral about $E=0$ over the spectrometer resolution function (the full width at half-maximum is 0.16 meV). It is evident that the very small anisotropy does indeed play an important role in the dynamics of the correlations. In particular, $\mathcal{S}^{\parallel}(q^*,E\sim 0)$ becomes Bragg-like at low temperatures, whereas $\mathcal{S}^{\perp}(q^*,E\sim 0)$ decreases to near zero, and a pseudo spin-wave gap seems to open up.[5] The consequences for the instantaneous correlations are less obvious; it is clear, however, that any detailed theory must include the anisotropy explicitly.

We now focus our attention on the inverse correlation lengths shown in Fig. 2. These data possess a variety of interesting qualitative features. Firstly, we note that the effect of dilution below the percolation limit is to suppress drastically the development of the spin correlations. (The mean-field ordering temperature for these samples is ~ 50 K.) Indeed, the results above 4 K are are quite reminiscent of those in the classical linear chain material $(CD_3)_4NMnCl_3$ (TMMC).[7] At high temperatures, the correlation length is dominated by thermal fluctuations; as the temperature is lowered, the correlation length increases until it exceeds the size of the larger clusters; at that point, the correlation length by necessity saturates. The size of the clusters in turn depends critically on c near the percolation concentration c_p. The limiting correlation lengths, which measure directly the mean cluster diameter, are 37 and 102 Å (or 9 and 25 lattice constants) for $c=0.54$ and $c=0.57$, respectively.

It is of interest to see if one can understand the

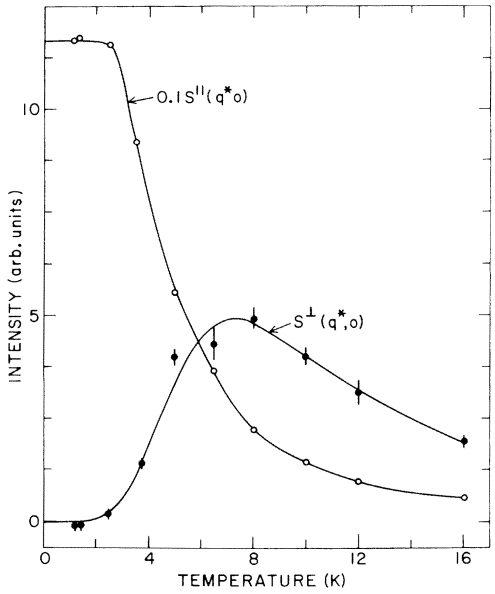

FIG. 3. Transverse and longitudinal dynamical structure factors (integrated over $\hbar\omega \lesssim 0.08$ meV) versus temperature in $Rb_2Mn_{0.54}Mg_{0.46}F_4$. Similar results are obtained in the $c=0.57$ sample.

temperature dependence on a more quantitative basis. In both two and three dimensions for nn interactions, the average coordination number around the percolation concentration is close to 2, that is, the coordination of a linear chain. In any given cluster the local geometry may, of course, be quite complicated. However, over large distances the correlations must propagate through many one-dimensional links, and we expect these one-dimensional "weak links" to dominate. Careful consideration of computer-generated clusters leads one to the heuristic conclusion that the longest paths across the cluster simulate *self-avoiding walks*[8] (SAW) on a square lattice. By exploiting the connection between the diameter of SAW's on a square lattice to the length, one is immediately led to the relation

$$\xi_{\mathrm{SAW}}/a \sim 0.95(\xi_1/a)^{3/4}, \tag{2}$$

where ξ_1 is the one-dimensional correlation length, and a is the nn distance. For the 1D classical Heisenberg model,[7]

$$\xi_1/a = (-u)^{1/2}/(1+u), \tag{3}$$

where $u = \coth[-J_{\mathrm{nn}}S(S+1)/k_BT] + k_BT/J_{\mathrm{nn}}S(S+1)$. The solid line in Fig. 2 is the classical Heisenberg result, Eqs. (2) and (3), for an infinite SAW. It is clear that this simple model with no adjustable parameters accounts surprisingly well for both the absolute magnitude and general tempera-

ture dependence of the correlations in both samples. The nearly exact agreement with the $c = 0.54$ data should be considered accidental since the prefactor, 0.95, in Eq. (2) is very sensitive to the detailed assumptions. The dashed line gives the corresponding result for the longitudinal correlation length $\xi^{\parallel}$ when the anisotropy is included explicitly.[9] Finally, one must incorporate the explicit concentration. It turns out that this can be done most readily for a classical random walk (RW) without the SAW restriction. The random walk gives an exponent of $\frac{1}{2}$ in Eq. (2) which is clearly wrong; however, the qualitative features should still carry over from the RW to the SAW. We shall discuss this RW model in detail in a separate publication.[5] Here it is sufficient to note that the RW calculations indicate that the nearly uniform shift in κ between $c = 0.54$ and 0.57 arises from finite size effects. Further, the approach to saturation at low temperatures is quantitatively accounted for provided that one includes the anisotropy explicitly. From theoretical considerations we believe that $\xi_1^{\parallel}$ should represent an appropriate temperature scaling field. Least-squares fits to the $c = 0.57$ data for 6 K $\leq T \leq 30$ K give $\kappa \sim (\xi_1^{\parallel})^{-0.75\pm0.05}$ and $S(q^*) \sim (\xi_1^{\parallel})^{1.23\pm0.1}$.

In conclusion, we have measured the development of the magnetic correlations close to the percolation transition in a 2D antiferromagnet. The results suggest that the temperature dependence is determined mainly by the 1D weak links in the clusters. Our experiments and calculations show that the falloff of the magnetic correlations is well represented by a simple function of the form $\exp[-(\kappa_T + \kappa_G)/R]/R^{1/2}$, where κ_T and κ_G are the inverse correlation lengths resulting from thermal and geometrical disorder, respectively. A simple SAW model is found to account for κ_T remarkably well. In general, our results are consistent with a multicritical-point description of percolation in magnets. The scaling theories by Stanley *et al.* and by Lubensky,[4] which were developed concurrently with this work, differ mainly in their fundamental assumptions about the cluster geometry; the former uses the SAW *Ansatz*, while the latter presents the consequences of a node-connectivity postulate following de Gennes's work in polymers. For a Heisenberg model in 2D these theories predict $\nu_T = 0.75$ and $\nu_T = 1.2$, respectively. Our experiments favor the former model; however, it is premature to attempt to choose definitively between these theories (and other possible related theories) until the theoretical calculations incorporate explicitly both finite-size and anisotropy effects. It is hoped that these experiments will stimulate further development of the theory of correlations in dilute magnets near the percolation concentration. Experiments on 3D systems are planned in the near future.

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