without damping. However, for the SB model the required phonon amplitude ($\sim sb$) is unrealistically high. For the more realistic PWL model discussed above, we find analogous solutions with a much smaller phonon amplitude (again, $\sim sb$), especially for conditions corresponding to the minima of s (see Fig. 1). Such solutions are currently under investigation.

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[6]The above calculations for the strain can also be checked by using the boundary conditions $D(z) = \beta$ and $D(-z) = 1-\beta$, which give s as in Eq. (4) except that $|H(x)|^2$ is changed to $H(x)\cos zx$ (Ref. 4). For the two expressions to be equal, it is not necessary that $H(x)$ be equal to $\cos zx$. The numerical results agree quite well. We can also check the displacement field far from the core by a simple generalization of the analytic techniques in Ref. 3.

[7]See, for instance, L. S. Jacobsen and R. S. Ayre, *Engineering Vibrations* (McGraw-Hill, New York, 1958), Chap. 4.

Critical Scattering near the Percolation Threshold in $Mn_cZn_{1-c}F_2$

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We report critical-neutron-scattering measurements as a function of concentration and temperature in a crystal of $Mn_cZn_{1-c}F_2$ which has a concentration gradient such that c spans the percolation threshold c_p. The results are consistent with a multicritical-point description of percolation in magnets and illustrate explicitly the importance of spin-space crossover effects on percolation. We observe no critical scattering associated with the three-dimensional phase transition for $c \geq c_p$.

Recently, considerable attention has been directed towards the problem of percolation in magnets. On the experimental side, Birgeneau *et al.*[1] have reported a study of the spin-spin correlations in the two-dimensional (2D) antiferromagnetic system, $Rb_2Mn_cMg_{1-c}F_4$, with the concentration c just below c_p, the percolation threshold. Several theoretical models and calculations have also been reported.[2-5] It is now believed that the point, $c = c_p$ temperature $T = 0$, may be regarded as a multicritical point terminating the line of continuous phase transitions of the infinitely connected cluster.[2-5] There are, however, very few experimental results on real magnets especially in three dimensions against which to test the theoretical results. In particular, the role of the dipolar anisotropy and the nature of the ordering process in the infinitely connected cluster are still not understood.

In this Letter we report a neutron-scattering study of the spin-spin correlations in the three-dimensional (3D) anisotropic antiferromagnet $Mn_cZn_{1-c}F_2$. The sample studied has a concentration gradient along its length and, by a very fortunate coincidence, c equals c_p approximately half way down the crystal so that only the bottom half of the sample exhibits a 3D phase transition. Hence by masking off thin slices we have been able to study the spin correlations as a function of concentration and temperature in the neighbor-

hood of c_p. From these studies we find (a) the general behavior of the longitudinal correlation length is consistent with a multicritical-point picture; (b) the spin anisotropy plays a central role—in particular, it causes a crossover from power-law to exponential behavior at low temperatures; and (c) the 3D ordering appears not to be accompanied by any separately observable critical fluctuations.

The single crystal of $Mn_cZn_{1-c}F_2$ has previously[6] been described and characterized. The growth axis of the crystal along which there was a concentration gradient was about 20° away from the (010) axis which was placed vertical for the measurements. By masking off narrow horizontal slices it was thus possible to study the neutron scattering as a function of concentration. The relative concentrations were determined accurately from the lattice constants of the individual slices.

The measurements were performed with a two-axis spectrometer at the Brookhaven National Laboratory high-flux reactor. A monochromatic beam of neutrons with an energy of 40 meV was obtained by reflection from the (002) planes of pyrolytic graphite and higher-order neutrons were largely eliminated by a pyrolytic-graphite filter. The horizontal collimations throughout the spectrometer were 10 min. The experiments were carried out using essentially the same method as Schulhof *et al.*[7] in their study of the critical scattering in pure MnF_2. MnF_2 is a body-centered tetragonal antiferromagnet with the spins oriented along the c axis. Hence around the (001) position pure transverse fluctuations are measured whereas around (100) equal contributions from the transverse and longitudinal susceptibilities are obtained. At the phase transition only the longitudinal component behaves critically. A similar anisotropy might reasonably be anticipated in the diluted system.

The experimental results at all temperatures and at all concentrations are qualitatively similar to those reported previously[1] in the 2D system $Rb_2Mn_cMg_{1-c}F_4$. For the slices with $c < c_p$, somewhat broad Lorentzian scattering is observed at (001) while at (100) at low temperatures the scattering is the sum of a narrow and a broad Lorentzian as expected from the pure MnF_2 results. For two slices, one with $c \simeq c_p$ and the second with $c = c_p + 0.007$ a 3D magnetic phase transition occurs at ~6 and ~7 K, respectively. The temperature dependence of the order parameter for these slices is shown in Fig. 1. Neutron photographs show that only about one-third of the $c \simeq c_p$ slice actually orders.

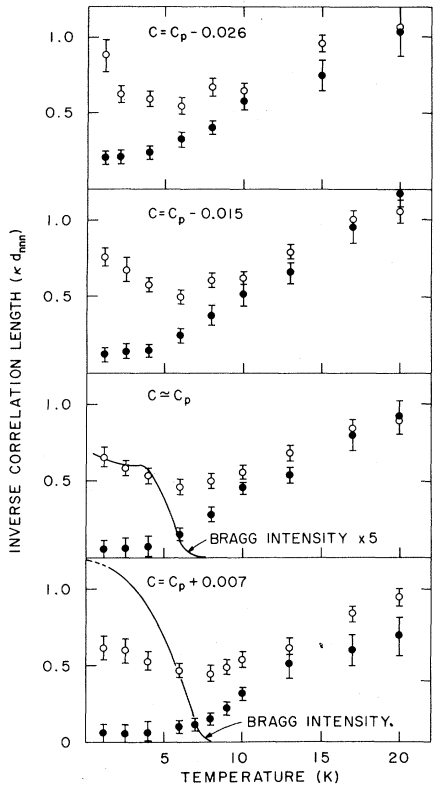

Fig. 1. Transverse (○) and longitudinal (●) inverse correlation lengths in reduced units (d_{nnn} = corner to body center distance) in $Mn_cZn_{1-c}F_2$. In addition at $c = c_p - 0.037$ and $T = 1.2$ K we find $\kappa_{\parallel} d_{nnn} = 0.3 \pm 0.1$, $\kappa_{\perp} d_{nnn} = 1.2 \pm 0.2$. The solid lines give the magnetic Bragg intensity and hence the order parameter squared in the $c \simeq c_p$ and $c = c_p + 0.007$ slices.

Following Schulhof *et al.*[7] we have fitted the data at (001) with a Lorentzian $2\mathcal{S}_{\perp}(\vec{q}) = 2A_{\perp}/(\kappa_{\perp}^2 + q^{*2})$ and the data at (100) with the sum of two Lorentzians, $\mathcal{S}_{\parallel}(\vec{q}) + \mathcal{S}_{\perp}(\vec{q})$, where $\mathcal{S}_{\parallel}(\vec{q}) = A_{\parallel}/(\kappa_{\parallel}^2 + q^{*2})$. In general the goodness of fit parameters χ^2 are close to 1 thus showing that the Lorentzian cross sections adequately describe the data. The best-fit values of the transverse and longitudinal inverse correlation lengths are shown in Fig. 1. The amplitude factors $A_{\parallel}$ and $A_{\perp}$, expressed as $\mathcal{S}^{\alpha}(\vec{q}) \cdot \kappa_{\alpha}^2$ for $c \sim c_p$ are shown in Fig. 2. Similar results for $A_{\parallel}$ and $A_{\perp}$ are obtained at the other concentrations. A number of important features are immediately apparent from Figs. 1 and 2. Firstly it is evident that below 10 K the anisotropy plays a central role and that only the longitudinal correlation length diverges at c_p as $T \to 0$. Indeed, the transverse correlation length is rel-

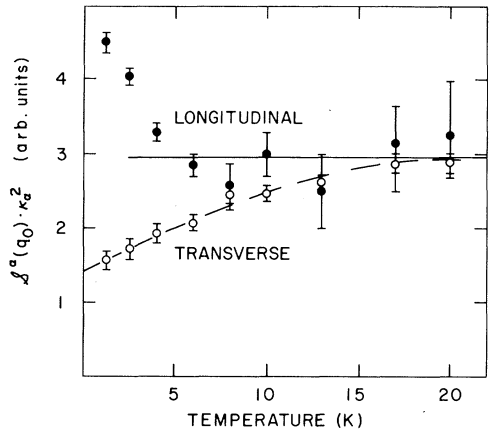

FIG. 2. Amplitude factors $A_{\parallel}$ and $A_{\perp}$ vs temperature in $Mn_{c_p}Zn_{1-c_p}F_2$. The solid and dashed lines are guides to the eye.

atively insensitive to concentration through c_p. We shall discuss the unusual temperature dependence of $\kappa_{\perp}$ below. Secondly, as expected, $\kappa_{\parallel}(1.2\ \mathrm{K})$ decreases continuously as $c \to c_p$ and apparently goes to zero for $c = c_p$. The best fit value of $\kappa_{\parallel}(1.2\ \mathrm{K})$ is finite although the error bars include zero. We believe that the actual value at c_p is zero and that the apparent finite value is a result of the variation of c within the slice. Thirdly, in the $c \approx c_p$ and $c \approx c_p + 0.007$ slices, $\kappa_{\parallel}$ is not zero at T_N but continues to decrease on further cooling. No evidence of critical scattering associated with this phase transition was obtained.

We now consider these results in the context of current theories of percolation in magnets. Pure MnF_2 has the tetragonal rutile structure and the magnetic interactions are adequately described by Heisenberg exchange interactions,

$$H = \sum_{i>j} J_{ij} \vec{S}_i \cdot \vec{S}_j,$$

between nearest- and next-nearest-neighbors with $J_{nn} = -0.055$ meV, $J_{nnn} = 0.304$ meV, and more-distant-neighbor interactions negligible.[8] There is, in addition, an anisotropy field $g\mu_B H_A = 0.092$ meV which arises largely from dipolar interactions. From computer results[9] we estimate that c_p should be close to, but somewhat smaller than 0.245 for $Mn_cZn_{1-c}F_2$ which is consistent with the concentration of the specimen estimated from chemical analysis and lattice constants.

For $c < c_p$ the system is made up of finite clusters and $\kappa_{\parallel}(T=0)$ provides a measure of the mean cluster size.[1] The data shown in Fig. 1 may be interpreted in terms of a power-law behavior $\kappa_{\parallel}(T=0) \sim (c_p - c)^{\nu_p}$ with ν_p slightly less than 1. This is consistent with the value of $\nu_p = 0.86$, deduced[9] from Monte Carlo calculations in 3D.

We now consider the temperature dependence of $\kappa_{\parallel}$ and $\kappa_{\perp}$ for $c = c_p$. In our earlier work we suggested[1] that to a good approximation the correlations in the clusters propagate along paths which simulate self-avoiding walks (SAW), and showed that this gave a very good account of our 2D results. Furthermore we suggested that the spin-space crossover effects could be incorporated by using the underlying one-dimensional correlation lengths κ^1 as the temperature variables, as independently suggested by Lubensky.[4] Here we predict that

$$\kappa_{\parallel} = \kappa_0(\kappa_{\parallel}^1)^{\nu_T}, \quad \kappa_{\perp} = \kappa_0(\kappa_{\perp}^1)^{\nu_T},$$

where κ_0 should be of order unity when the κ^1 are expressed in reduced units. The exponent ν_T is predicted to be 0.60 for the SAW model[1,3] and 0.86 by Stephen and Grest[5] using renormalization-group calculations near one and six dimensions.

We have calculated the one-dimensional correlation lengths $\kappa_{\parallel}^1$ and $\kappa_{\perp}^1$ using a program developed by Blume, Heller, and Lurie[10] for *classical* spins and using spin-Hamiltonian parameters deduced from pure MnF_2. These calculations predict that the minimum in $\kappa_{\perp}$ should occur at 2.5 K rather than the 6 K observed. Simple arguments suggest, however, that quantum effects (specifically, detailed balance) will shift this minimum to higher temperatures and so we have increased the anisotropy in the model by a factor of 6 so as to give the minimum in $\kappa_{\perp}^1$ at 6 K. We then choose κ_0 to fit the data at 20 K. With these two parameters fixed the calculations give the results shown in Fig. 3. It is evident that both theories account very well for the experimental results but that because of uncertainties connected with the spin-space crossover effects, the experiments do not select between them. In any case, given the simplicity of our model, it clearly accounts for the essential physics of the problem.

For $c > c_p$ the system is made up of finite clusters together with a single infinitely connected cluster. Specifically, for the $c = c_p + 0.007$ slice we estimate from the Monte Carlo simulations that from 25% to possibly as high as 50% of the spins belong to the infinite network. One then might expect diffuse scattering similar to that observed for $c < c_p$ arising from the finite clus-

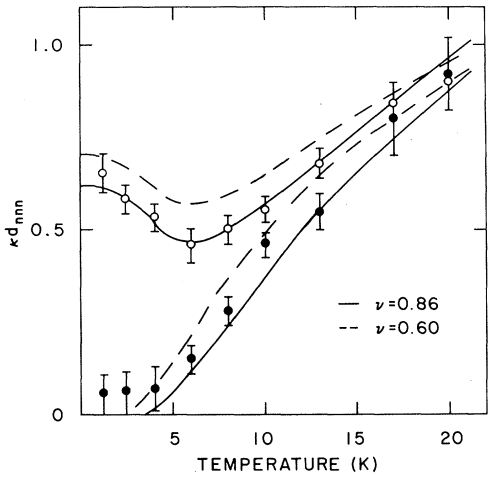

FIG. 3. Transverse (○) and longitudinal (●) inverse correlation lengths vs temperature in $Mn_{c_p}Zn_{1-c_p}F_2$. The solid and dashed lines are the Stephen and Grest $\varphi = 1$ ($\nu = 0.86$) and SAW ($\nu = 0.60$) theories, respectively, as described in the text.

ters together with critical scattering from the infinite network. By analogy with concentrated systems, the latter should diverge at the transition temperature and should then disappear at low temperatures. Somewhat surprisingly, no divergence is observed in the diffuse scattering near T_N in either the $c \simeq c_p$ or the $c = c_p + 0.007$ slices. Rather $A_\parallel$ is approximately constant through T_N while $\kappa_\parallel$ decreases continuously in a manner very similar to that in the $c < c_p$ slices. We do not understand this result and are not aware of any theory for the critical fluctuations for $c \gtrsim c_p$ which might help with the interpretation. Finally, the close similarity in T_N for the $c \simeq c_p$ and $c = c_p + 0.007$ slices presumably arises from the spin-space crossover effect which should also initiate a changeover from a power-law to a logarithmic dependence of T_N on $c - c_p$.

In conclusion, our experiments have verified that most aspects of our picture of the percolation problem in magnetic materials are correct, and have illustrated the essential role played by the anisotropy in real systems. We hope that our results will stimulate theoretical studies on the correlations for $c > c_p$ which we do not understand at present.

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