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Percolation in antiferromagnetic insulators

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The use of neutron scattering techniques to study problems in statistical mechanics is illustrated by describing recent experiments on magnetic–non-magnetic transition metal fluorides. The concentration of the magnetic ions is chosen to be close to the percolation point for the onset of long-range magnetic order. The results have confirmed that the percolation point is a multicritical point at which the long-range order may be destroyed by either geometrical or thermal disorder. The temperature scale of the thermal disorder is determined by the one-dimensional weak links in the large clusters. Results are obtained for the exponents in systems of dimensionality two and three, and with Ising and Heisenberg symmetry. In some systems the results agree with theoretical predictions, but there remain some discrepancies between theory and experiment and some aspects of the results which are not yet understood.

1. Introduction

One of the most rewarding applications of neutron scattering techniques during the last few years has been to study problems in statistical mechanics. Neutron scattering provides a uniquely detailed way of studying the phase transitions of magnetic systems, and by choice from the wide variety of magnetic systems it is possible to perform experiments on systems that closely approximate many of the simple models studied in statistical mechanics. Neutron scattering enables measurements to be made of the development of the long-range order below the phase transition, and the changes in the amplitude and correlation length of the magnetic fluctuations both above and below the transition temperature.

The study of the antiferromagnetic transition metal fluorides has proved to be particularly useful. The magnetic interactions are well known from low temperature spin wave measurements and are largely between only nearest neighbour magnetic ions; they can be of either Heisenberg character ($J\boldsymbol{S}_1 \cdot \boldsymbol{S}_2$ in Mn^{2+} salts) or Ising character ($JS_1^z S_2^z$ in Co^{2+} salts). Furthermore, systems can be found in which the magnetic ions are arranged in effectively one-dimensional chains, two-dimensional sheets or three-dimensional networks. In brief, classic experiments have now been performed on many of these systems. We note only the work of Birgeneau *et al.* (1971) on tetramethyl ammonium manganese chloride, which gave excellent agreement with Fisher's (1964) theory of linear chains of classical spins, the work of Ikeda & Hirakawa (1974) on K_2CoF_4, which agrees with Onsager's (1944) theory of the two-dimensional Ising model, and the work of Tucciarone *et al.* (1971) on $RbMnF_3$, which gave agreement with theories of the three-dimensional Heisenberg model. More recently this work has been extended to study more complex situations such as crossover phenomena and tricritical points.

The transition metal antiferromagnets have also proved to be excellent systems on which to study the effect of disorder on the excitations. Mixed single crystals can be grown in which the magnetic interactions are well known and of short range, while the effects of the disorder can be studied in detail by using neutron scattering techniques. The first experiments were

studies of the low concentration limit, and demonstrated the existence of local spin modes near some impurities and resonant distortions of the spin wave spectrum near other impurities as reviewed by Cowley & Buyers (1972). More recently, work has been on more concentrated systems and has elucidated the nature of the excitations in highly disordered systems and the extent to which they can be described by theories based on the coherent potential approximation and by computer simulation techniques. The results of this have been reviewed (Cowley 1976; Cowley *et al.* 1979).

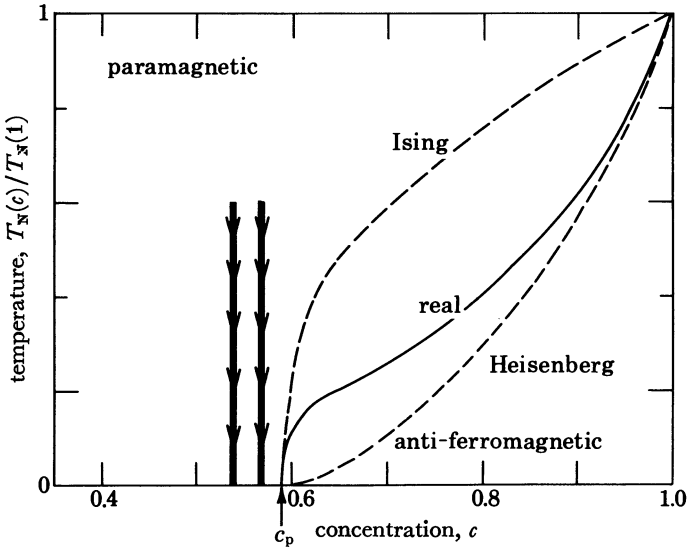

FIGURE 1. Schematic phase diagram of a two-dimensional square lattice close to percolation. The phase boundaries are sketched for Ising interactions, Heisenberg interactions and for the mixed interactions characteristic of $Rb_2Mn_cMg_{1-c}F_4$. The heavy vertical lines represent the locus of points at which the scattering was measured.

In this paper I wish to review some experiments where these two aspects of the usefulness of transition metal fluorides have been combined; namely, the study of the phase transitions of disordered systems, and in particular of systems close to the percolation limit in which sufficient non-magnetic ions have replaced the magnetic ions that the long-range magnetic order is destroyed. The phase diagram of this type of system is illustrated in figure 1.

The results described in this paper were obtained over the past few years at the Brookhaven National Laboratory, in collaboration with Dr R. J. Birgeneau and Dr G. Shirane. Full details of the experimental method and of the results are given in the full papers, which have now been prepared (Birgeneau *et al.* 1980; Cowley *et al.* 1980*a*, *b*) and of which this report is a brief review.

The relevant aspects of percolation theory are reviewed in §2, together with a brief account of the neutron scattering from these systems. The experimental results are described in §3 and the conclusions of the study given in a final section.

2. Percolation theory and neutron scattering

When a magnetic system with nearest neighbour interactions is diluted with sufficient non-magnetic ions, the system breaks up into separate clusters at a well defined concentration, c_p; this is known as the percolation point, and a schematic phase diagram of a magnetic system

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as a function of concentration and temperature is shown in figure 1. The percolation point is believed to be an example of a multicritical point (Stauffer 1976; Stanley *et al.* 1976) in the sense that the long-range magnetic order for $c = c_p$ and $T = 0$ may be destroyed geometrically by changing c to less than c_p or thermally by raising the temperature. Much of the work on percolation has concentrated on the geometric aspects at $T = 0$, as reviewed by Essam (1972). Initially, the problem was studied by Monte Carlo computer methods and by series expansion, while more recently renormalization group theory has been applied to the Hamiltonian found by Kastelyn & Fortuin (1969), who mapped the percolation problem onto a lattice gas model.

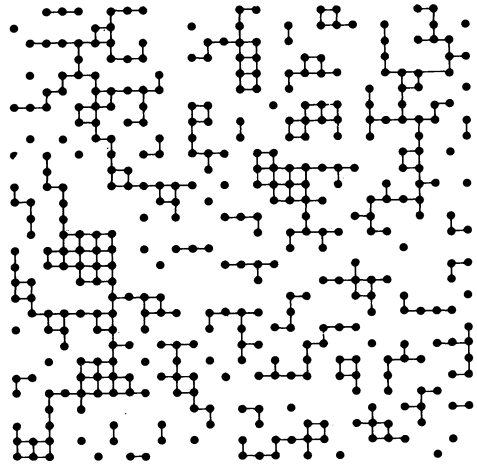

FIGURE 2. Clusters on a two-dimensional square lattice for $c = 0.50 < c_p = 0.59$.

There is therefore a correspondence between the properties close to the percolation point and the thermodynamic functions of a normal magnetic transition. In particular, the correlation length, ζ, is defined in terms of the pair connectedness, $S(\boldsymbol{r})$, which gives the probability that a site distance $\boldsymbol{r}$ away is in the same cluster as the site at the origin. Our experiments suggest that as with thermodynamic functions the Fourier transform of $S(\boldsymbol{r})$ is a Lorentzian.

$$S(\boldsymbol{q}) = A/(\kappa^2 + \boldsymbol{q}^2), \tag{1}$$

where $\kappa = 1/\zeta$ is the inverse correlation length.

The mean size of the finite clusters plays the same role as the susceptibility at thermodynamic phase transitions and is given by $S_0 = S(0)$. Finally, the fraction of sites in the infinite cluster, $P(c)$, plays the same role as the magnetization at a thermodynamic phase transition.

Not surprisingly, the behaviour of these quantities as $c \to c_p$ is described by exponents. In particular, percolation theory suggests that for geometrically driven behaviour, $T = 0$ of figure 2, then

$$\kappa(c, T = 0) \approx (c_p - c)^{\nu_G}, \tag{2a}$$

$$S_0(c, T = 0) \approx (c_p - c)^{-\gamma_G}, \tag{2b}$$

$$P(c, T = 0) \approx (c - c_p)^{\beta_G}, \tag{2c}$$

where the best values (Reynolds *et al.* 1977; Klein *et al.* 1978; Stanley 1977) of the exponents are listed in table 1.

To discuss the thermal critical behaviour it is first necessary to choose the appropriate temperature scale. In figure 2 I show a computer-generated two-dimensional square lattice with $c = 0.5$, which is less than $c_p = 0.59$. It is apparent that the large clusters consist of blobs joined by almost one-dimensional parts. These one-dimensional parts will dominate the thermal behaviour at low temperatures. Thus it is plausbile that the appropriate temperature scale (Stauffer 1976; Stanley *et al.* 1976; Birgeneau *et al.* 1976) is the inverse correlation length of the one-dimensional parts of the clusters, $\kappa_1(T)$. In the Ising and Heisenberg limits,

$$\kappa_1(T) \approx 2 \exp(-2J/kT) \quad \text{and} \quad \kappa_1(T) \approx kT/J, \quad \text{respectively.}$$

TABLE 1. EXPONENTS AND AMPLITUDES FOR PERCOLATION (Cowley *et al.* 1980*b*)

	ν_T	ν_G	γ_T	γ_G	D
2d Ising	1.32 ± 0.04	1.356 ± 0.015	2.40 ± 0.1	2.435 ± 0.045	0.96 ± 0.06
2d Heisenberg	0.90 ± 0.05	1.356 ± 0.015	1.50 ± 0.15	2.435 ± 0.045	1.00 ± 0.05
3d Ising	0.85 ± 0.10	0.845 ± 0.021	—	1.66 ± 0.07	—
3d Heisenberg	0.95 ± 0.04	0.845 ± 0.021	1.73 ± 0.15	1.66 ± 0.07	1.09 ± 0.03

Once the temperature scale has been established, we can introduce thermal exponents in analogy to (2) for $c = c_p$ and varying temperature by

$$\kappa(c_p, T) \approx (\kappa_1(T))^{\nu_T} \tag{3a}$$

and

$$S_0(c_p, T) \approx (\kappa_1(T))^{-\gamma_T}. \tag{3b}$$

Within the multicritical point picture, the exponents of the geometrically driven behaviour and the thermally driven behaviour are related by a crossover exponent, ϕ, so that $\gamma_T = \gamma_G/\phi$, and $\nu_T = \nu_G/\phi$. The exponent ϕ has been obtained as $\phi = 1$ for Ising systems by Wallace & Young (1978); more recently, Stinchcombe (1979) has suggested that this might be the case for Heisenberg systems as well.

Neutron scattering measurements give the magnetic spin–spin correlation function. The intensity for a wavevector transfer, $\boldsymbol{Q}$, and integrated over all frequency transfers, ω, is determined by the static correlation function

$$I(\boldsymbol{Q}) = \sum_{mn} \exp[i\boldsymbol{Q}\cdot(\boldsymbol{R}_m - \boldsymbol{R}_n)]\langle \boldsymbol{S}_m \cdot \boldsymbol{S}_n \rangle, \tag{4}$$

where m and n denote the sites of the magnetic ions, and $\boldsymbol{R}_m$ and $\boldsymbol{R}_n$ and their positions. It is then apparent that $I(\boldsymbol{Q})$ is given in terms of the pair correlation function $S(\boldsymbol{q})$ of (1) when $I(\boldsymbol{Q}) = S(\boldsymbol{q})\,\delta(\boldsymbol{Q} - \boldsymbol{q} - \boldsymbol{\tau}) + N^2 P(c)^2\,\delta(\boldsymbol{Q} - \boldsymbol{\tau})$, where $\boldsymbol{\tau}$ is the nearest magnetic reciprocal lattice point to $\boldsymbol{Q}$. These results show that neutron scattering may be used to determine A, κ and $P(c)$ and hence, in principle, the geometric and thermal exponents describing the percolation multicritical point.

3. EXPERIMENTAL RESULTS

The single crystals of mixed magnetic and non-magnetic transition metal fluorides were grown by Dr H. J. Guggenheim of Bell Laboratories and by the late Dr D. A. Jones of Aberdeen University. The samples were grown with concentrations close to c_p. Unfortunately, one of the difficult aspects of this work was determining the concentration and uniformity of the samples. Use was made of lattice parameter measurements, chemical analysis and the

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excitation spectra, but frequently the most accurate method was from the magnetic critical scattering at low temperatures and the use of percolation theory as described in the full accounts of this work. Unfortunately, the uncertainty in the concentration prohibited the experimental determination of the geometrical exponents. The experiments therefore consisted of measuring the temperature dependence of the correlations as a function of temperature for certain crystals or fixed concentrations as illustrated in figure 1.

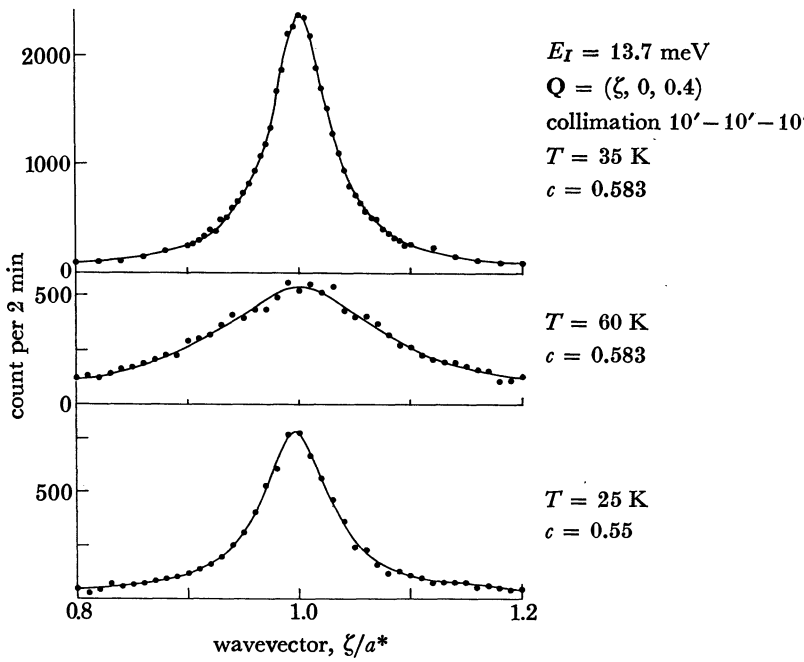

FIGURE 3. Neutron scattering distributions in $Rb_2Co_cMg_{1-c}F_4$ and fits to a two-dimensional Lorentzian, $S(q) = A/(q^2+K^2)+B$ (——, fitted curve). (From Cowley *et al.* (1980*a*).)

The experiments were performed with a two-axis neutron spectrometer with the crystals mounted in a variable temperature cryostat. No evidence was found in any of the examples for chemical ordering of the different transition metal ions. The experiments were conducted with a sufficiently high incident neutron energy that the intensity gave a direct measure of the frequency integrated intensity, $I(\boldsymbol{Q})$.

Some of the results for the $Rb_2Co_cMg_{1-c}F_4$ system are illustrated in figure 3. The results show diffuse scattering, which decreases in width and increases in intensity as the system is cooled. The results for each temperature were fitted by assuming that the scattering is given by (1) and (4), convoluted by the experimental resolution function. A and κ were then determined to give the best fit, in a least-squares sense, to the experimental results. In each case a good fit was obtained, showing that the scattering is determined by a Lorentzian in reciprocal space so that the correlations decay exponentially in real space.

Initially, we discuss the results for $c < c_p$ when there is no long-range magnetic order. The temperature dependence of κ is shown in figure 4 for different concentrations in the two-dimensional Ising system, $Rb_2Co_cMg_{1-c}F_4$. In this system the exchange interactions are known

from spin wave measurements (Ikeda & Hutchings 1978; Cowley *et al.* 1980*a*). The results suggest that the inverse correlation length is given by the simple form

$$\kappa = \kappa_G + \kappa_T, \tag{5}$$

where κ_G depends solely on concentration and κ_T solely on the temperature. The thermal part is determined by the known one-dimensional correlation length, $\kappa_1(T)$, as

$$\kappa_T a = D(\kappa_1(T)\, a)^{\nu_T}, \tag{6}$$

from which we can determine D and ν_T as given in table 1. Similarly, the exponent γ_T can be found from the intensity of the scattering. The results clearly confirm the multicritical point picture of percolation and (5) and (6), showing that the temperature scale is indeed determined by the one-dimensional weak links in the clusters. Furthermore, the measured exponents ν_T and γ_T are within error in agreement with the calculated geometrical exponents ν_G and γ_G (table 1), confirming that the crossover exponent, ϕ, is unity as obtained theoretically by Wallace & Young (1978).

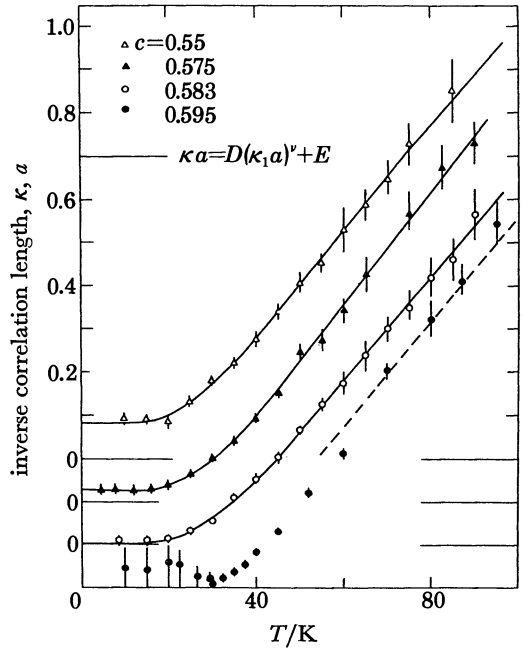

FIGURE 4. The temperature dependence of κ for four concentrations of $Rb_2Co_cMg_{1-c}F_4$. Note that for $c = 0.595$, long-range order occurs below 31 K. The solid lines show fits to (5) and (6). (Cowley *et al.* (1980*a*).)

Similar measurements were made on the analogous two-dimensional system, $Rb_2Mn_cMg_{1-c}F_4$, in which the magnetic interactions are predominantly of Heisenberg character (Cowley *et al.* 1977*a*). The results are illustrated in figure 5. The results show that the inverse correlation length decreases with decreasing temperature, until below about 4 K when the scattering becomes almost independent of temperature. This behaviour arises because at low temperatures the magnetic dipolar energy becomes comparable with kT. Consequently, although at high temperatures (20 K) the system is behaving like an isotropic Heisenberg system, on cooling the magnetic dipolar forces tend to align the spins along the unique c-axis and the system takes on Ising character. The inverse correlation length $\kappa_1(T)$ of an anisotropic linear chain was

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calculated by using a computer program written by Blume *et al.* (1975), and as shown in figure 5, equations (5) and (6) give a good account of the results. The value of ν_T is, however, quite different for this Heisenberg-like system from that for the Ising system discussed above. The value of the constant D is, in contrast, very similar.

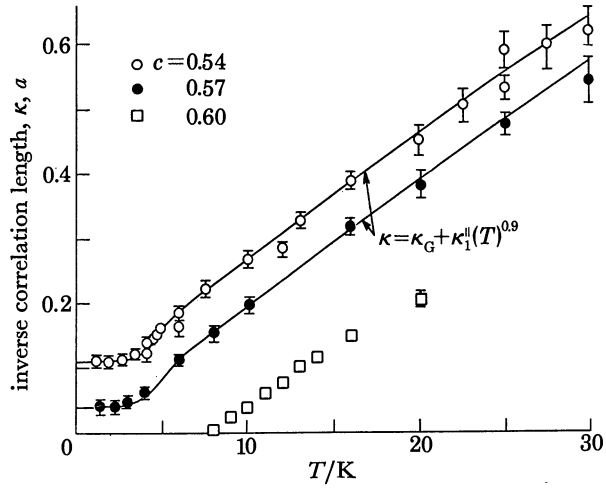

FIGURE 5. The inverse correlation length, κ, as a function of temperature for $Rb_2Mn_cMg_{1-c}F_4$. The crystal with $c = 0.60$ showed long-range order below 8.0 K. The solid lines show fits to (5) and (6). (Birgeneau *et al.* (1980).)

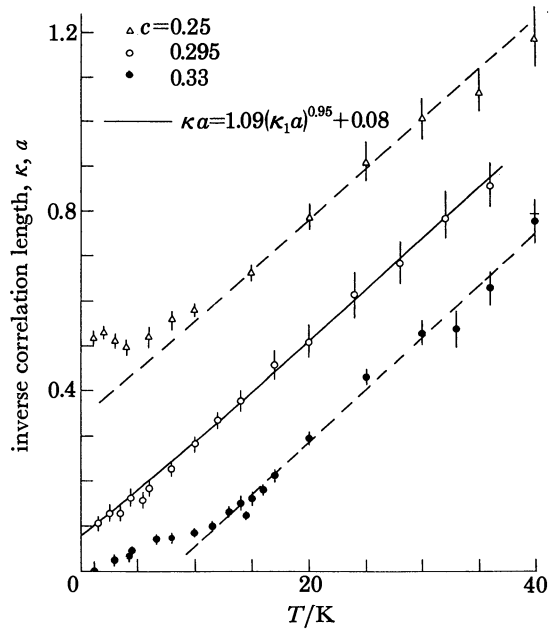

FIGURE 6. The inverse correlation length, κ, for three concentrations of $KMn_cZn_{1-c}F_3$. The solid line shows a fit to (5) and (6) the dotted lines are this fit with a different constant chosen to give agreement with other concentrations at high temperature. (Cowley *et al.* (1980*b*).)

Two three-dimensional systems have been studied. The $Mn_cZn_{1-c}F_2$ structure is tetragonal. In the pure system the magnetic interactions are of Heisenberg character but the magnetic dipolar forces produce a significant energy gap owing to the tetragonal structure (Nikotin *et al.* 1969). In the mixed system these forces give rise to a crossover to Ising behaviour at low

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temperatures, but the tetragonal structure makes it possible to determine the longitudinal and transverse scattering independently Consequently, it was shown (Cowley *et al.* 1977*b*) that both inverse correlation lengths were given by (5) and (6), if $\kappa_1(T)$ was replaced by the appropriate longitudinal and transverse inverse correlation lengths of the anisotropic linear chain. The parameters D and ν_T were the same for both longitudinal and transverse scattering.

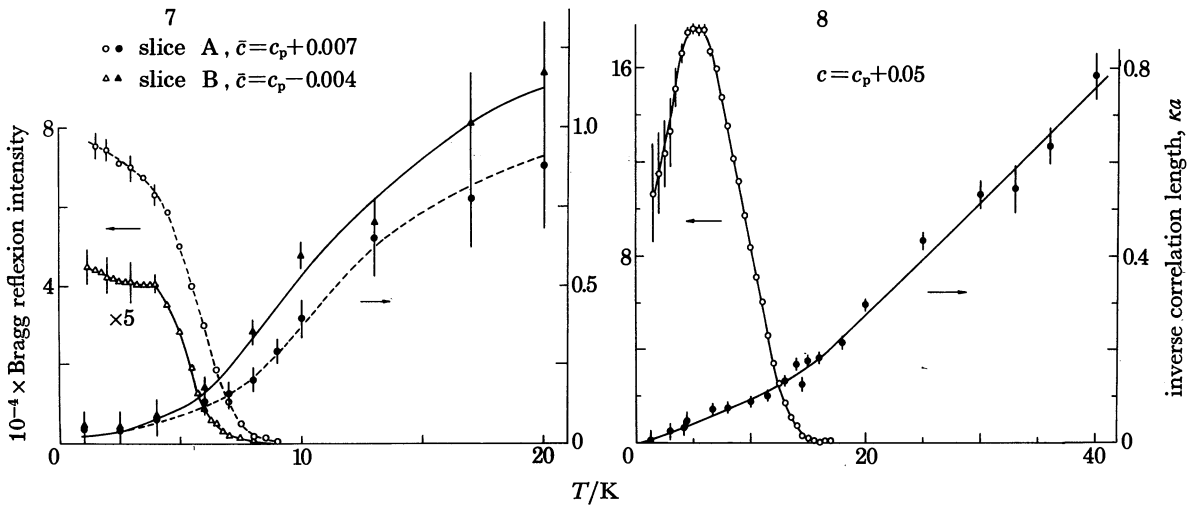

FIGURE 7. The Bragg scattering and the inverse longitudinal correlation length for two slices of the $Mn_cZn_{1-c}F_2$ system with concentrations as shown. Note that for neither slice is K zero at the onset of long-range order. (Cowley *et al.* (1977*b*).)

FIGURE 8. The Bragg scattering and inverse correlation length of $KMn_cZn_{1-c}F_3$ for $c > c_p$. (Cowley *et al.* (1980*b*).)

The other three-dimensional system is $KMn_cZn_{1-c}F_3$, which has the cubic perovskite structure. The exchange interactions in the pure material are of Heisenberg character between nearest neighbours and in the cubic symmetry there is no anisotropy gap in the spin wave spectrum owing to the magnetic dipolar forces. The results for the inverse correlation length in the mixed systems are shown in figure 6.

Once again, they are consistent with (5) and (6), and give results for the exponent ν_T and constant D as listed in table 1.

We now turn to discuss the results for $c > c_p$ when long-range order occurs at low temperatures. In the two-dimensional systems the behaviour for $c > c_p$ is at least qualitatively similar to expectations: the inverse correlation length decreases towards zero at the transition temperature, $T_N(c)$ (figures 4 and 5), and the long-range order as measured by the intensity of the magnetic Bragg reflexion increases monotonically with decreasing temperature. It is difficult to determine the exponents close to $T_N(c)$ because $T_N(c)$ changes rapidly with concentration c. Nevertheless, the indications are that the exponents are consistent with those occurring in pure K_2MnF_4 (Birgeneau *et al.* 1977) and pure K_2CoF_4 (Ikeda & Hirakawa 1974).

The results for $c > c_p$ for the two three-dimensional systems are considerably more difficult to understand. In neither $Mn_cZn_{1-c}F_2$ nor $KMn_cZn_{1-c}F_3$ is the measured critical scattering described by an inverse correlation length which decreases to zero at $T_N(c)$. Indeed, figures 7 and 8 show that κ is non-zero at $T_N(c)$ and continues to decrease as the temperature is lowered.

The scattering in these systems arises from three parts: the finite clusters, the backbone or multiply connected part of the infinite cluster, and the dangling ends of the infinite cluster. Presumably only the backbone orders at $T_N(c)$, and if this is a small enough part of the whole crystal then the critical scattering from the backbone might be obscured by the scattering from the finite clusters and the dangling ends. Although computer calculations (Kirkpatrick 1978) show that for a given $c-c_p$ the proportion of spins in the backbone is less in three dimensions than in two, the numbers suggest that it is surprising that no critical scattering was observed associated with $T_N(c)$.

The other surprising feature is the temperature dependence of the long-range order in $KMn_cZn_{1-c}F_3$ as shown in figure 8. The intensity of the Bragg reflexion increases on cooling from $T_N(c)$ *ca.* 13.5 to 6 K, but then decreases on further cooling. This result is difficult to understand. Our only suggestion is that 6 K is about the temperature at which the magnetic dipolar forces begin to play a role in these systems. Although these cancel in the cubic environment of pure $KMnF_3$, the random environments of the mixed crystal will lead to randomly directed dipolar forces. Possibly these destroy the long-range order as suggested by Aharony (1978), leading to the development of a spin glass phase at low temperatures.

4. Conclusions

Neutron scattering measurements have been made on a number of transition metal antiferromagnets close to the percolation concentration. The results have substantiated the multicritical point picture of percolation as being a competition between geometrically and thermally driven disorder. The temperature scale of the thermal disorder has been identified as being determined by the one-dimensional weak links in the large clusters. Measurements have been made of the thermal exponents γ_T and ν_T for various different systems. In Ising systems these are within error the same as the best estimates of the geometrical exponents as suggested by Wallace & Young (1978). In the Heisenberg systems the results differ from the geometrical exponents in contradiction to the suggestion of Stinchcombe (1979). Measurements have also been made of the amplitudes, but there are as yet no theoretical results with which to compare these results.

In the two-dimensional systems the onset of long-range order occurs in a manner qualitatively similar to that of the pure systems, but in the three-dimensional systems this is not the case. The critical scattering does not show the usual divergences as $T \to T_N(c)$, but is relatively smoothly varying near $T_N(c)$. This result and the reason for the behaviour of the long-range order at low temperatures in $KMn_cZn_{1-c}F_3$ (figure 8) must await further work. We hope that these results will stimulate further work on the multicritical point picture of percolation, so that it will be possible to compare the results with theory in more detail. Finally, it is clear that neutron scattering results have provided very significant and timely results for our understanding of the statistical mechanics of percolation phenomena, in addition to the earlier successes at other types of phase transition.

I am grateful to my collaborators on this project, Dr R. J. Birgeneau and Dr G. Shirane, and to the many others who have assisted with the experiments and with the understanding of their significance. Financial support at Brookhaven was provided by the Division of Basic

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