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Spin correlations in percolating networks with fractal geometry

H.Ikeda, K.Iwasa,^a J.A.Fernandez-Baca^b and R.M.Nicklow^b

Booster Synchrotron Utilization Facility, National Laboratory for High Energy Physics, Oho 1-1, Tsukuba 305, Japan

^a*Department of Physics, Keio University, Hiyoshi 3-14-1, Yokohama 223, Japan*

^b*Solid State Division, Oak Ridge National Laboratory, Tennessee 37831, USA*

Using neutron scattering techniques, we investigated the magnetic correlations in diluted antiferromagnets close to the percolation threshold in which the magnetic connectivity takes a fractal form. Recent experimental results concerning the self-similarity of the magnetic order, and magnetic excitations in two-dimensional Ising and three-dimensional Heisenberg antiferromagnets are presented.

Keywords: Percolation, Fractal, Spin Dynamics

Postal Address: H.Ikeda; Booster Synchrotron Utilization Facility (BSF), National Laboratory for High Energy Physics (KEK), Oho 1-1, Tsukuba 305, Japan (FAX: +81-298-64-3202)

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1. Introduction

It is generally accepted that the atomic connectivity of a percolating cluster takes the form of a fractal.¹⁾ The theory of percolation was formulated by many authors and can now be used to interpret an exceptionally wide variety of physical and chemical phenomena. The concept of fractals has contributed significantly to the understanding of percolation.²⁾ The simplest ideal percolating networks are realized by substitutionally diluting magnetic systems by non-magnetic atoms. At a critical magnetic concentration (c_p), a single infinite cluster (a percolating network) spans the full space. With a further decreasing concentration of magnetic atoms, the system splits into an assembly of only finite clusters. The percolating networks exhibit a self-similarity, and can be characterized by a non-integer mass dimension, i.e., a fractal dimension D_f . In a square and a simple cubic lattice, it has been numerically estimated that the D_f of the percolating network is 1.896 and 2.48 respectively, i.e. D_f is smaller than the Euclidean dimension, D .³⁾

As is well established, neutron scattering is a powerful tool to investigate the static and dynamical aspects of magnetic systems. By observing the

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scattering function which is the space-time Fourier transform of the spin pair-correlation function, we can obtain detailed information concerning the structural and dynamical properties of these systems.

In this report, we present recent experimental results on neutron scattering experiments in two- and three-dimensional (2D and 3D) percolating antiferromagnets; a direct observation of the self-similarity of the magnetic order in a percolating cluster,^{4,5)} and investigations of the magnetic excitations in percolating antiferromagnets.^{6,7)}

2. Magnetic structure of a percolating cluster

The scattering law for a fractal structure which is observed by neutrons or photons is quite simple. The small-angle neutron scattering intensity ($I(q)$) is simply proportional to q^{-D_f} , where q is the wave vector. This relationship has been actually observed by small-angle neutron scattering from silica aerogels.⁸⁾ However, no experimental observation of the self-similarity of the percolating network in diluted magnets has been successful until now.

Very recently, we achieved the first experimental observation of the fractal structure in diluted antiferromagnets. In the vicinity of the

percolation threshold, the geometrical correlation length (ξ_G) is defined as $\xi_G = |c - c_p|^{-\nu_G} a_0$, where ν_G is a numerical constant which depends on the lattice geometry and a_0 is the nearest-neighbor lattice spacing. It is now believed that the system is a fractal at length scales smaller than ξ_G ; conversely, the system appears to be homogeneous at length scales larger than ξ_G . The geometrical correlation length (ξ_G) is related to the crossover wave vector by $q_c = \xi_G^{-1}$. The systems in which the magnetic concentration (c) is very close to, but above, the percolation threshold, order antiferromagnetically at low temperatures. The magnetic elastic scattering of neutrons from the infinite cluster in these systems, at the antiferromagnetic superlattice position, can be written as follows: $I(q) \sim \delta(Q - \tau)$ for $q < q_c$, and $I(q) \sim q^{-D_f}$ for $q > q_c$; here, $q = Q - \tau$, and Q and τ are the scattering vector and the antiferromagnetic reciprocal vector, respectively. Below the transition temperature, the spins belonging to the infinite network order magnetically, while those belonging to only finite clusters do not order, even at 0 K. It is important to note that magnetic correlations between an infinite cluster and finite clusters are absent due to the lack of magnetic interactions between them. This makes possible the observation of the fractal structure of an infinite cluster by a

high-resolution measurement of the elastic scattering profile near the magnetic reciprocal point.

Our experiments were performed on the 2D diluted Ising antiferromagnet $\text{Rb}_2\text{Co}_{0.6}\text{Mg}_{0.4}\text{F}_4$ ⁴⁾ and the 3D diluted Heisenberg antiferromagnet $\text{RbMn}_{0.34}\text{Mg}_{0.66}\text{F}_3$,⁵⁾ whose magnetic concentrations are very close to the percolation threshold of a square lattice (0.593) and a simple cubic lattice (0.312), respectively.

The pure Rb_2CoF_4 system becomes antiferromagnetic at $T_N=103.0$ K with the spin direction along the c -direction and alternating in the basal c -plane.⁹⁾ The magnetic-exchange interaction dominates within the plane and is limited to the nearest neighbors. The measured Neel temperature of $c=0.60$ sample was 20.0 K. Using the value of v_G (1.33) for a square lattice system we can estimate the geometrical correlation length (ξ_G): $730 a_0$ for $c=0.60$, which implies crossover wave vectors (q_c) of $3.1 \times 10^{-4} 2\pi/a$, where a ($\sqrt{2}a_0$) is taken as a unit length of the $[100]$ direction. This sample is then expected to reveal a crossover at q_c , from a homogeneous structure at low q to a self-similar fractal structure at large q . Neutron elastic scattering experiments were performed on triple-axis spectrometers (C1-1 and T1-1) installed at the

cold- and thermal-neutron beam guides of the JRR-3M reactor at JAERI, Tokai.

The magnetic intensity distribution was measured at $T=4.5$, 7.5 and 10 K from $\text{Rb}_2\text{Co}_{0.60}\text{Mg}_{0.40}\text{F}_4$. The absence of a temperature dependence in the scattering well below T_N indicates that the measured intensity arises only from the ordered magnetic structure. The critical scattering decreases very rapidly with decreasing temperature, and is clearly negligible at these temperatures. A remarkable feature of the line shape observed in the near-percolating system is the very large width of the peak.

The data taken at 4.5 K were plotted in a double-logarithmic scale. The slope of the observed intensity versus the wave vector at $q > 0.01 \ 2\pi/a$ is 1.95 ± 0.07 . This value is in good agreement with the fractal dimension ($D_f=1.896$) of an infinite cluster in a $2D$ percolating network. At q smaller than $0.005 \ 2\pi/a$, a Gaussian peak shape due to long-range order is observed. The crossover from a homogeneous structure at small q to a fractal structure at large q is thus clearly seen in this experiment.

Neutron elastic scattering experiments on a $3D$ diluted antiferromagnet $\text{RbMn}_{0.34}\text{Mg}_{0.66}\text{F}_3$ were performed on a high resolution diffractometer (HRP) installed at the pulsed neutron source (KENS)

in KEK, Tsukuba. The crystal was oriented with its $[0\ 1\ -1]$ axis vertical so as to measure the magnetic elastic scattering centered at the $(1.5\ .5\ .5)$ position. The measured Neel temperature of $c=0.34$ sample was 9.0 K.

Figure 1 shows the magnetic intensity distribution (plotted in a double-logarithmic scale) measured at $T=3.0$ K (closed circles) together with the instrumental resolution function (open circles) estimated from the intensity distribution near the $(3\ 1\ 1)$ nuclear position. The same magnetic-intensity distribution was obtained at $T=7.0$ K. From this, we conclude that the measured intensity at $T=3.0$ K is only from the ordered magnetic structure. In the $c=0.34$ sample, since v_G is 0.88 for a simple cubic lattice, the geometrical correlation length (ξ_G) is $21.9\ a_0$, which implies a crossover wave vector (q_c) of $7.3 \times 10^{-3}\ 2\pi/a_0$. In Fig.1 the resolution-limited line shape at small q and the $I=q^{-D_f}$ dependence of the intensity at larger q (with $D_f=2.48$, which is the $3D$ fractal dimension) were clearly seen. This corresponds to the crossover from a homogeneous structure at small q to a fractal structure at large q .

3. Magnetic excitations

3-1. Ising system

The magnetic excitations in diluted Ising antiferromagnets have so far been elucidated in systems whose magnetic concentration is close to, but above the percolation concentration. A typical result was obtained in the 2D antiferromagnets $\text{Rb}_2\text{Co}_c\text{Mg}_{1-c}\text{F}_4$ ($c=0.89$ and 0.70), using a neutron inelastic scattering technique.¹⁰⁾ In Ising systems with nonmagnetic impurities, the excitation energies split into z dispersion-less branches, z being the coordination number. In diluted magnets there are z different local configurations in which the number of magnetic neighbors is different, thus producing z different molecular field energies. These are known as Ising-cluster excitations. In magnetic systems including diluted ones, it is now believed that magnetic excitations such as spin waves disappear at higher temperature than T_N in systems with two or three dimensions. In fact, no spin-wave excitations in a pure antiferromagnet with Ising symmetry, Rb_2CoF_4 , were observed above T_N .¹¹⁾

Independent of this, in a highly diluted system whose magnetic concentration is very close to the percolation threshold, the percolating network exhibits a self-similarity. Due to the fact that the percolating systems comprise highly ramified clusters and therefore the time scale of spin fluctuations becomes extremely long compared to

the corresponding pure system, the temperature variation of the spin correlations would be most interesting to study.

In order to measure the magnetic excitations over the entire temperature region in a percolating Ising magnet, we have performed extensive inelastic neutron scattering experiments on a 2D diluted Ising magnet, $\text{Rb}_2\text{Co}_{0.60}\text{Mg}_{0.40}\text{F}_4$.⁶⁾ Neutron scattering experiments were performed at the HET spectrometer installed at the pulsed spallation neutron source at ISIS, Rutherford Appleton Laboratory. The incident energy was selected to be 40 meV in order to cover energy transfers up to 35 meV.

The energy spectra were taken at 1.5, 10, 20, 30, 40, 55, 70, 100 and 150 K, and four Ising peaks at 7.0, 13.6, 19.9, and 27.0 meV are detected. Intensities from Ising-cluster excitations decrease with increasing temperature. Surprisingly, these can be seen even at 100 K ($T/T_N=5.0$) which is far above T_N . As mentioned before, the spin wave peak in the pure compound, Rb_2CoF_4 , corresponds to the highest-energy peak at 27 meV, and it does not persist above T_N .

In order to elucidate this anomalously long-lived spin-wave excitation in the system close to the percolation threshold, we performed similar

measurements on a sample with higher concentration, $\text{Rb}_2\text{Co}_{0.70}\text{Mg}_{0.30}\text{F}_4$ ($T_N=45$ K). Again, the magnetic excitations can be seen in the paramagnetic phase, but the intensities almost disappear at $T=90$ K ($T/T_N=2.0$). We show in Fig.2 the normalized magnetic intensity which is the sum of the four peak intensities against normalized temperature (T/T_N). In the figure, we also show the spin-wave intensity of Rb_2CoF_4 which has been recently measured at the LAM-D spectrometer installed at KENS.

Early neutron scattering measurements of $\text{Rb}_2\text{Co}_{0.58}\text{Mg}_{0.42}\text{F}_4$, whose magnetic concentration is just below c_p (0.593) have been performed only at the lowest temperature of 5 K and revealed four Ising peaks although this temperature is above T_N .¹²⁾ This observation is in accord with the present experiments.

From these observations it is clear that the spin waves in the near-percolating systems with Ising symmetry can be excited even far above T_N . This phenomenon could be explained by the extremely slow dynamics of spins in the percolating networks as has been suggested by experiments on real-time kinetics of ordering in the $c=0.60$ sample.¹³⁾ If the time scale of the fluctuations of local spins is very long compared with the neutron's travelling time,

the system appears ordered. In this case, each spin feels the molecular field from the spins surrounding it as a static field.

3-2 Heisenberg system

We have recently performed an experiment aimed to give the first quantitative measurement of fracton excitations in near-percolating Heisenberg systems.⁷⁾ For this experiment we have chosen a diluted pure-Heisenberg antiferromagnet ($\text{RbMn}_{0.39}\text{Mg}_{0.61}\text{F}_3$), in which the Mn concentration (0.39) is very close to the percolation threshold ($c_p = 0.312$). The corresponding cross-over wave vector is $q_c = \xi_G^{-1} = 0.024$ reciprocal lattice units (rlu). The excitations with wave vectors smaller than q_c are expected to be spin waves, while the excitations with wave vectors larger than q_c are expected to be fractons. Since q_c in the present system is very small, we do not expect to observe the spin-wave region in a standard neutron inelastic scattering experiment. The fracton region, however, is observable in such an experiment as we report in this article.

Our experiment was performed on a single crystal (about 1 cc in volume) of $\text{RbMn}_{0.39}\text{Mg}_{0.61}\text{F}_3$. The pure system RbMnF_3 has a cubic perovskite structure and becomes antiferromagnetic at $T_N = 82\text{K}$.

with the spin directions alternating along the cubic edges. The $\text{RbMn}_{0.39}\text{Mg}_{0.61}\text{F}_3$ sample orders at 18 ± 1 K with the same magnetic configuration as in the pure system. The inelastic scattering measurements were performed on a triple-axis spectrometer installed at the HFIR reactor at the Oak Ridge National Laboratory. The crystal was oriented with its $[0\ 1\ -1]$ axis vertical. The magnetic excitations were measured for wave vectors along the $[0\ 1\ 1]$ direction from the $(1/2\ 1/2\ 1/2)$ zone center ($q=0$), to the zone boundary ($q=0.375$ rlu). The spectrometer was operated in the constant- Q mode with the final-energy fixed at 13.7 meV.

Figure 3 shows the energy spectrum of the magnetic response, taken at 4.5 K, at the zone boundary corrected for instrumental background. As depicted in the figure, a fine structure due to the Ising-cluster excitations in this system appears. The vertical bars in the figure give the probability density of random populations of magnetic neighbors for $c=0.39$. The energy values at the peaks agree with the Ising-cluster energies of Mn^{2+} ions when an exchange constant of $J=0.30$ meV is used. No measurable intensity is expected at the highest position of 9.0 meV ($z=6$) due to the small probability for 6 magnetic neighbors. The value of J is slightly larger than that of a pure crystal ($J=0.29$

meV). It should be noted, however, that the observed magnetic response extends beyond 8 meV where no magnetic intensities from the cluster excitations are expected. This observation indicates that the energy spectrum at the zone boundary, in this system, cannot be described solely by a simple Ising-cluster model. Since the energy resolution (1.0 meV FWHM) is finer than the peak interval (1.5 meV), the contribution from the cluster excitations can be resolved from the additional magnetic scattering. This additional magnetic contribution is indicated by the dotted line in Fig.3.

The energy spectra obtained at several wave vectors from $q=0$ to $0.375(\text{ZB})$ rlu reveal that the observed line shapes are not smooth but have some small structure, originating from the cluster excitations, throughout the Brillouin zone. Also, over the entire Brillouin zone the energy spectrum of the additional contribution shows a very broad, smooth shape, and its magnetic response extends to the highest energies measured in our experiment (10 meV). The energy width of this spectrum is much broader than the energy resolution. As the wave vector increases, the intensity of the scattered neutrons decreases rapidly.

We have analysed the line shape of the magnetic response of additional magnetic contribution obtained

at 4.5 K. We modeled these to some simple analytical functional forms. From a physical point of view, the damped harmonic oscillator (DHO) would be the most reasonable model to fit these excitations. We therefore fitted the broad excitation data to the functional form $\chi''(q,E) = A \Gamma(q)E / ((E^2 - E_p(q))^2 + (\Gamma(q)E)^2)$, where A is a constant. This form was successfully fitted to all of the data. The fitted values of $E_p(q)$ and $\Gamma(q)$ as a function of q satisfy the power-law relationship with the same exponent, i.e. $E_p(q) - \Gamma(q) \sim q^{1.1 \pm 0.2}$. It should be noted that since the value of $\Gamma(q)$ is much larger than $E_p(q)$, the excitations are strongly over-damped over the entire Brillouin zone. It has been recently reported that the single length-scale postulate (SLSP) holds for the magnetic response from fractons.³⁾ This postulate states that the peak energy and the energy width should have the same wave-number dependence as q^{z_a} , i.e. $E_p(q) - \Gamma(q) \sim q^{z_a}$. The present result is in accord with this theory.

A similar analysis using a DHO form has been performed in the nearly percolating antiferromagnet $\text{Mn}_{0.32}\text{Zn}_{0.68}\text{F}_2$ by Coombs et al.¹⁴⁾ We presume that their over-damped signal might have contained contributions from both fracton and Ising-cluster excitations, although a separation of these was not pursued.

Another evidence for fractons is seen in the temperature variation of $\chi''(q,E)$ at wave vector larger than q_c . In Fig.4, the energy spectra at $q=0.2$ rlu obtained from $T=9$ K to 100 K are shown. At $T=9$ K ($T_N/2$), peaks from Ising-cluster excitations almost disappear due to the much more enhanced thermal fluctuations of molecular fields than at $T=4.5$ K. On the other hand the over-damped component is predominant at this temperature and the line shape is the same as that measured at $T=4.5$ K. In this figure we observe the remarkable fact that the over-damped component can survive even at $T=100$ K ($>5 T_N$) and the peak energy slightly shifts towards higher energies with increasing T . These are completely different from the traditional spin-wave excitations in the Heisenberg system. The increase in the peak energy could be related to the localized nature of fractons, and thus the excitation energy (oscillating frequency) could be increased by obtaining the thermal energy. The existence of the magnetic excitations in near-percolating Heisenberg antiferromagnet at temperatures far above T_N is similar to those in percolating Ising antiferromagnet mentioned before.

On the other hand, high-resolution inelastic neutron scattering studies have recently been performed on a three-dimensional (3D) diluted

near-Heisenberg antiferromagnet ($\text{Mn}_{0.5}\text{Zn}_{0.5}\text{F}_2$) by Uemura and Birgeneau.¹⁵⁾ They ascribed the asymmetry of the obtained spectra and the double-peak structure at small wave vectors to the coexistence of magnons and fractons, and its wave-vector dependence to the magnon-fracton crossover. This interpretation should be taken with caution, however, because the magnetic concentration of the system that they studied (0.5) is far above the percolation threshold ($c_p=0.245$ or less for a body-centered tetragonal lattice).¹⁶⁾ At this relatively high magnetic concentration it is difficult to obtain conclusive evidence of fractons since the system is not self-similar even at small length-scales.

Our arguments, based on the DHO function, are qualitatively in good agreement with the current theory for fractons, although a quantitative description for the physical properties of the antiferromagnetic fractons in a Heisenberg system has not yet been completely resolved. We hope that an accurate description, even for the magnetic response function itself, will be available in the near future from analytic theories and/or computer simulations.

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Figure Captions

- Fig.1. Elastic neutron scattering intensities for $\text{RbMn}_{0.34}\text{Mn}_{0.66}\text{F}_3$ near to the (1.5 0.5 0.5) antiferromagnetic reciprocal position measured at 3.0 K (solid circles) together with instrumental resolution (open circles) in a double-logarithmic plot.
- Fig.2 Normalized magnetic intensity, defined as the sum of the four peak intensities, against normalized temperature (T/T_N) in three systems: $\text{Rb}_2\text{Co}_{0.6}\text{Mg}_{0.4}\text{F}_4$ (closed circles), $\text{Rb}_2\text{Co}_{0.7}\text{Mg}_{0.3}\text{F}_4$ (closed triangles) and Rb_2CoF_4 (open circles).
- Fig.3 Magnetic response ($\chi''(q,E)$) of $\text{RbMn}_{0.39}\text{Mg}_{0.61}\text{F}_3$ observed at the zone boundary at 4.5K with an energy resolution of 1.0 meV. The vertical bars indicate the probability density of random populations of magnetic neighbors. The dotted line represents the additional magnetic contribution and also the fitted scattering function described in the text. Vertical bars represent error bars.
- Fig.4 Temperature variation of $\chi''(q,E)$ at $q=0.2$ as a function of energy observed at $T=9$ K (closed circles), 40 K (open circles), 60K (closed triangles) and 100 K (open triangles). Vertical bars represent error bars.

Fig. 2

Fig 3

Fig 4