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Fracton excitations in a diluted Heisenberg antiferromagnet near the percolation threshold: $RbMn_{0.39}Mg_{0.61}F_3$

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Abstract. Using neutron inelastic scattering techniques we investigated the magnetic excitations in a diluted Heisenberg antiferromagnet ($RbMn_cMg_{1-c}F_3$ with $c = 0.39$) close to the percolation threshold of $c_p = 0.312$. No indication of propagating spin waves was observed throughout the Brillouin zone, but a broad peak superimposed on Ising-cluster excitations was observed. The origin of this broad peak is attributed to the excitation of fractons in a percolating network.

In recent years, considerable attention has been directed towards the dynamical properties of highly ramified percolating networks that exhibit a fractal geometry [1]. Recent theories and computer simulations of these systems predict the existence of highly localized fracton excitations with huge oscillation amplitudes [2]. A random site-diluted Heisenberg antiferromagnet is an ideal system for probing the existence of these excitations. In such a system and at concentrations close to, but just above, the percolation threshold c_p there should be a crossover from long-wavelength spin-wave excitations to short-wavelength fracton excitations. The origin of this crossover is the fact that the fractal geometry is realized only at length scales shorter than the geometrical correlation length ξ_G (for the percolation problem see [3]). Magnetic excitations in diluted magnetic systems have been extensively studied using neutron inelastic scattering techniques (for a review of experimental results see [4]). However, renewed experimental effort has recently been initiated to characterize the fractal component of the dynamics in these diluted magnetic systems. For this, it is important to distinguish fracton excitations from other localized excitations, such as Ising-like cluster excitations.

High-resolution inelastic neutron scattering studies have been performed on a three-dimensional (3D) diluted near-Heisenberg antiferrogmagnet ($Mn_{0.5}Zn_{0.5}F_2$) by Uemura and Birgeneau [5]. They reported that, as the wavevector q approaches the zone boundary, the observed spectra show a crossover at around $q = 0.3q_{ZB}$ from a spin wave to a fracton. At small wavevectors, although a sharp spin-wave peak is observed, the lineshape is rather asymmetric with a long tail extending towards higher energies. On increasing the wavevector, the intensity of the sharp peak decreases rapidly and the spectra show a characteristic double-peak structure. At the zone boundary, the observed scattering shows a very broad continuous energy spectrum. Uemura and Birgeneau ascribed the asymmetry of the obtained spectra and the double-peak structure at small wavevectors to the coexistence of magnons and fractons, and its wavevector dependence to the magnon–fracton crossover. This interpretation should be viewed with caution, however, because the magnetic

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concentration of the system that they studied (0.5) is far above the percolation threshold ($c_p = 0.245$ or less for a body-centred tetragonal lattice) [6]. 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. An earlier study of the nearly percolating system of $Mn_{0.32}Zn_{0.68}F_2$ was performed by Coombs *et al* [7]. They observed a broad magnetic response over the whole Brillouin zone, although they failed to observed sharp Ising-cluster excitations which should arise in the ordered state of the diluted antiferromagnets. These results were compared with the coherent potential approximation but they may be relevant to the present experiments described below.

The spin dynamics of another system, the 3D diluted pure-Heisenberg antiferromagnet $RbMn_cMg_{1-c}F_3$ (with $c = 0.74$ and 0.63) has recently been studied [6]. The excitation spectra of these samples, which also have concentrations far above c_p ($c_p = 0.312$ for a simple-cubic lattice) are different from the spectrum reported for $Mn_{0.5}Zn_{0.5}F_2$. In the small-wavevector region of this pure-Heisenberg system, the magnetic excitation spectra are dominated by a sharp spin wave with an additional structure. In the vicinity of $q = 0.25q_{ZB}$ the spin-wave peak splits into two main features while additional weak excitations are also evident. In the large-wavevector region the spectra show six well resolved localized excitations (Ising-cluster excitations), reflecting the different local environments in the diluted system. These observations do not reveal any evidence for the existence of fractons, in contrast with the reports on $Mn_{0.5}Mg_{0.5}F_2$ [5], but they only reflect the crossover from propagating spin waves to localized Ising-cluster excitations.

In this paper we report the results of a new experiment that has been performed in order to give the first quantitative measurement of fracton excitations in nearly percolating systems. For this experiment we have chosen a more diluted pure-Heisenberg antiferromagnet $RbMn_{0.39}Mg_{0.61}F_3$, in which the Mn concentration (0.39) is very close to the percolation threshold ($c_p = 0.312$). The geometrical correlation length of this system is defined as $\xi_G = (c - c_p)^{-\nu_G}a_0$ ($\nu_G = 0.88$ for a 3D system, and a_0 is the atomic spacing) and its corresponding wavevector is $q_c = \xi_G^{-1} = 0.024$ reciprocal lattice units (rlu). The excitations with wavevectors smaller than q_c are expected to be spin waves, while the excitations with wavevectors larger than q_c are expected to be fractons. Very recently, a crossover from a resolution-limited Gaussian profile at $q < q_c$ to a q^{-D_f} behaviour at $q > q_c$, has been directly observed in the magnetic Bragg profile measured by high-resolution neutron elastic scattering from nearly percolating 2D and 3D diluted antiferromagnets $Rb_2Co_{0.6}Mg_{0.4}F_4$ ($c_p = 0.593$) and $RbMn_{0.34}Mg_{0.66}F_3$, respectively, where D_f is a fractal dimension [8]. This indicates experimentally that the atomic connectivity in infinite networks takes the form of fractals at wavevectors larger than the respective q_c. 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 paper.

Our experiment was performed on a single crystal (about 1 cm^3 in volume) of $RbMn_{0.39}Mg_{0.61}F_3$ which we grew using the Bridgman method. The pure system $RbMnF_3$ has a cubic perovskite structure and becomes antiferromagnetic at $T_N = 82$ K with the spin directions alternating along the cubic edges. The spin-wave gap of $RbMnF_3$ is less than 0.03 meV [9]; this should be compared with 1.08 meV in MnF_2 [10]. The $RbMn_{0.39}Mg_{0.61}F_3$ 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, with pyrolytic graphite (PG) crystals as both monochromator and analyser, and with collimations of 60′–40′–40′–60′. The crystal was oriented with its $[01\bar{1}]$ axis vertical. The magnetic excitations were measured for wavevectors along the [011] direction from the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ zone centre ($q = 0$), 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. A PG filter of sufficient thickness to reduce the $\lambda/2$ contamination by a factor of 4000 was utilized. The instrumental resolution of this configuration was about 1.0 meV full width at half-maximum (FWHM). The experiments revealed a very broad energy spectrum, and the background and the incoherent scattering centred at $E = 0$ were not negligible compared with the magnetic signal. This was true especially at the Brillouin zone boundary where the background (about 1 count min^{-1} from both neutron and electric noise) at the energy transfer of 5 meV was about one quarter of the total count. We therefore performed both energy-loss ($0 \leqslant E \leqslant 10$ meV) and energy-gain ($0 \geqslant E \geqslant -4$ meV) inelastic measurements. The intensities of these two measurements are proportional to $(\langle n\rangle+1)\chi''(q, E)$+background and $\langle n\rangle\chi''(q, E)$+background, respectively, where $\langle n\rangle$ is the Bose factor and $\chi''(q, E)$ is the magnetic response. The difference between these two intensities gives the energy spectrum of the magnetic response $\chi''(q, E)$. In order to test whether this subtraction process is correct, we measured the background at the (111) nuclear zone. We subtracted this measured background from the energy-loss spectra and then divided the difference by $\langle n\rangle+1$. The values of $\chi''(q, E)$ obtained by this method were indistinguishable from the values obtained from the (energy-loss)−(energy-gain) subtraction. Our measurements were performed at $T = 4.5$ K, well below the Néel temperature. At the energy transfer of 1.5 meV, $\langle n\rangle$ is quite small, i.e. 0.02. Therefore, energy-gain measurements simply give the background counts at energies larger than 1.5 meV. The typical counting time at the zone boundary was 60 min per point.

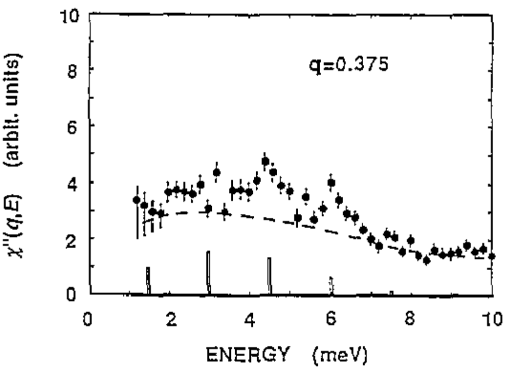

Figure 1. Magnetic response $\chi''(q, E)$ of $RbMn_{0.39}Mg_{0.61}F_3$ observed at the zone boundary at 4.5 K with an energy resolution of 1.0 meV. The vertical bars indicate the probability density of random populations of magnetic neighbours. The broken curve represents the additional magnetic contribution and also the fitted scattering function described in the text. The vertical bars represent error bars.

Figure 1 shows the energy spectrum of the magnetic response at the zone boundary. In antiferromagnets at the zone boundary, the neutron scattering intensity is simply proportional to the density of states available for the excitations. Therefore the intensity must correspond to excitations of individual magnetic Mn^{2+} spin in the molecular fields of the surrounding ions [4]. These are the so-called Ising-cluster excitations. As depicted in the figure, a fine structure due to the Ising-cluster excitations in this system appears and is more pronounced at lower energies because the average number of nearest-neighbour magnetic ions for each spin is rather small in the present system. The vertical bars in the figure give the probability density of random populations of magnetic neighbours for $c = 0.39$. The energy values at the peaks agree with the Ising-cluster energies of Mn^{2+} ions ($2zJS$; $z = 1, 2, 3, 4, 5$ and 6; $S = \frac{5}{2}$) when an exchange constant of $J = 0.30$ meV is used. No measurable intensity is expected at the highest position of 9.0 meV ($z = 6$) because of the small probability for six magnetic neighbours. 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 broken curve in figure 1.

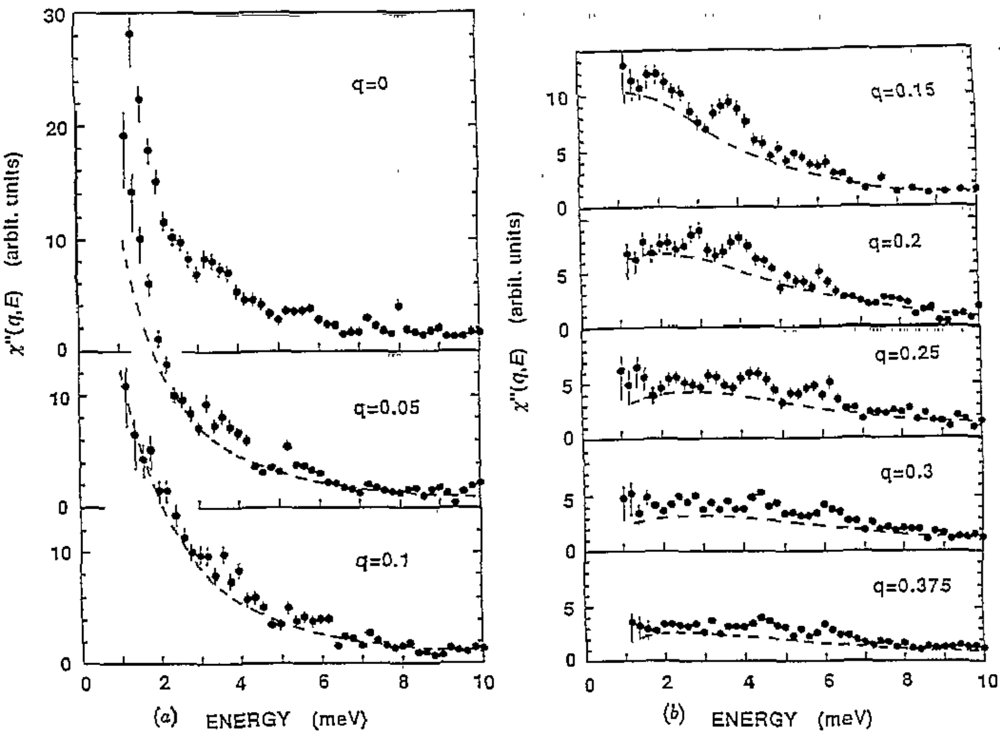

Figure 2. Magnetic response $\chi''(q, E)$ of $RbMn_{0.39}Mg_{0.61}F_3$ observed at (a) $q = 0, 0.05$ and 0.1 rlu and (b) $q = 0.15, 0.2, 0.25, 0.3$ and 0.375 rlu. The symbols are the same as in figure 1. The vertical bars represent error bars.

In figure 2, the energy spectra obtained at several wavevectors from $q = 0$ to $q = 0.375$ rlu (zone boundary) are shown for $RbMn_{0.39}Mg_{0.61}F_3$. The background and incoherent scattering at $E = 0$ meV were subtracted from the data, and the magnetic response $\chi''(q, E)$ is plotted in the figure. As shown in this figure, the observed lineshapes 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. The energy width of this spectrum is much broader than the energy resolution. As the wavevector increases, the intensity of the scattered neutrons decreases rapidly. The energy positions of the Ising peaks shift slightly towards lower energies in the wavevector region below 0.2 rlu.

The overall energy spectra in figures 1 and 2 are quite different from those previously reported for $RbMn_{0.74}Mg_{0.26}F_3$ and $RbMn_{0.63}Mg_{0.37}F_3$ [6], in which the magnetic concentration is far above the percolation threshold. The different feature seen in the energy spectrum for the nearly percolating compound, $RbMn_{0.39}Mg_{0.61}F_3$, is the appearance of a broad lineshape superimposed on Ising-cluster excitations. This contribution to the magnetic response has the following characteristics.

(1) The energy spectra are quite broad compared with the energy resolution.
(2) The linewidth increases with increasing wavevector.
(3) The energy of this broad peak increases with increasing wavevector.
(4) The integrated intensity decreases with increasing wavevector.
(5) The total intensity is much higher than that of the Ising-cluster excitations.

We attribute this magnetic response to fracton excitations, since this appears only in a system close to the percolation threshold, in which the connectivity of the magnetic atoms exhibits fractal geometry. As mentioned earlier, the geometrical correlation length ξ_G of this system leads to the crossover wavevector q_c of 0.024 rlu. The system appears to be fractal at wavevectors larger than 0.024 rlu, and the excitations in this wavevector region are expected to be localized fractons. Our results concerning the broad contribution to the magnetic response in the wavevector region from 0.05 rlu to q_{ZB} are consistent with this argument.

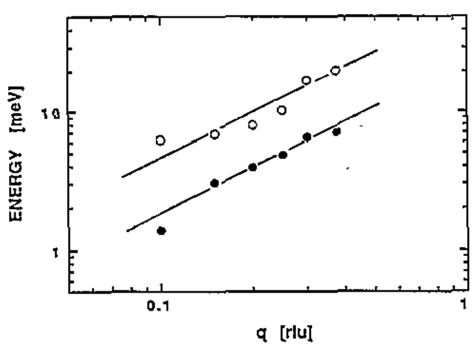

Figure 3. Values of $E_p(q)$ (●) and $\Gamma(q)$ (○) obtained by fitting the energy spectra to the scattering function of the form $\chi''(q, E) = A\Gamma(q)E/[E^2 - E_p(q)^2]^2 + (\Gamma(q)E)^2$ as a function of q. The straight lines show the power-law relationship of $E_p(q) \sim \Gamma(q) \sim q^{1.1}$.

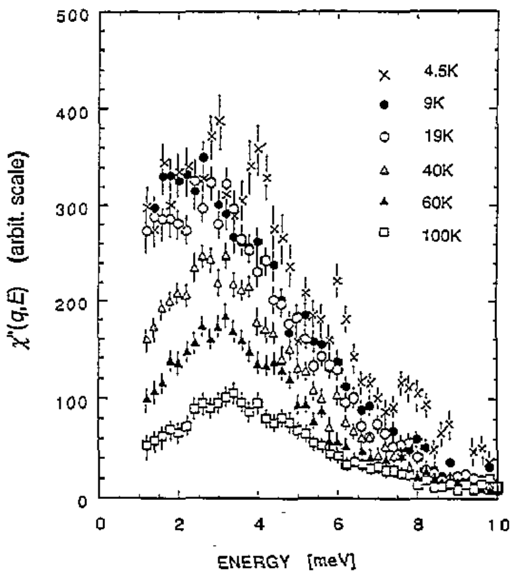

Figure 4. Temperature variation in $\chi''(q, E)$ at $q = 0.2$ as a function of energy observed at $T = 4.5$ K (×), 9 K (●), 19 K (○), 40 K (△), 60 K (▲) and 100 K (□). The vertical bars represent error bars.

We now discuss the main features of the lineshape of the magnetic response of these new excitations obtained at 4.5 K. Unfortunately, at this time there are no analytical theories concerning the magnetic response from fractons; however, since the lineshapes involved are broad and have no singularities, we can model these on 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 $\chi''(q,E) = A\Gamma(q)E/[(E^2 - E_p(q)^2)^2 + (\Gamma(q)E)^2]$, where A is a constant. This form was succesfully fitted to all the data, except for $q = 0.05$ (figures 1 and 2, broken curves), although some ambiguity is inherent in the estimation of a small structure arising from the individual motion of the spins. The fitted values of $E_p(q)$ and $\Gamma(q)$ as functions of q satisfy the power-law relationship with the same exponent, i.e. $E_p(q) \sim \Gamma(q) \sim q^{1.1\pm0.2}$ (figure 3). This relationship holds well in the wavevector region between 0.15 and 0.375 rlu but slightly deviates at $q = 0.10$ rlu. The finding that the data at $q = 0.05$ do not fit the DHO form is consistent with the fact that the energy spectrum should change continuously from a δ-functional form to a DHO form in the small-wavevector region close to the fracton–magnon crossover wavevector q_c (0.024 rlu). It should be noted that, since the value of $\Gamma(q)$ is much larger than $E_p(q)$, the excitations are strongly overdamped over the entire Brillouin zone. The quality of the fits to the data using other functional forms, including a Lorentzian, was worse than that with the DHO form.

A similar analysis using a DHO form has been performed in the nearly percolating antiferromagnet $Mn_{0.32}Zn_{0.68}F_2$ by Coombs *et al* [7]. We presume that their overdamped signal might have contained contributions from both fracton and Ising-cluster excitations, although a separation of these was not pursued.

It has been recently reported that the single length-scale postulate (SLSP) holds for the magnetic response from fractons [2]. This postulate states that the peak energy and the energy width should have the same wavenumber dependence as q^{z_a}, i.e. $E_p(q) \sim \Gamma(q) \sim q^{z_a}$. Very recently, Yakubo *et al* [11] performed numerical simulations for a 3D Heisenberg percolating antiferromagnet assuming that, for antiferromagnetic fractons, $\chi''(q,E)$ has the Lorentzian form. The value of $z_a = 2.5$ was obtained from these numerical simulations. Our results for $E_p(q)$ and $\Gamma(q)$ satisfy the SLSP for the magnetic response from fracton excitations. We therefore believe that our finding concerning the broad energy spectrum over the entire Brillouin zone is new evidence for antiferromagnetic fractons. However, the experimentally obtained exponent z_a ($= 1.1 \pm 0.2$) is much smaller than the current result based on computer simulations ($z_a = 2.5$), although the numerical result was obtained by assuming a Lorentzian magnetic response.

Another piece of evidence for fractons is seen in the temperature variation in $\chi''(q,E)$ at wavevectors larger than q_c. In figure 4, the energy spectra at $q = 0.2$ rlu obtained from $T = 4.5$–100 K are shown. At $T = 9$ K ($T_N/2$), peaks from Ising-cluster excitations almost disappear owing to the much more enhanced thermal fluctuations of molecular fields than at $T = 4.5$ K. On the other hand the overdamped component is predominant at this temperature and the lineshape is the same as that measured at $T = 4.5$ K. In this figure we observe the remarkable fact that the overdamped component can survive even at $T = 100$ K ($> 5T_N$) and the peak energy slightly shifts towards higher energies with increasing T. These features 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. At present, however, in order to give a quantitative explanation for this new phenomenon, we need further developments of the theories in the system with fractal geometry at finite temperatures.

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 analytical theories and/or computer simulations.

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