Two-Dimensional Antiferromagnetic Fractons in Rb$_2$Mn$_{0.598}$Mg$_{0.402}$F$_4$

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Abstract. The dynamical structure factor $S(q, \omega)$ for diluted two-dimensional Heisenberg antiferromagnet Rb$_2$Mn$_{0.598}$Mg$_{0.402}$F$_4$ was investigated by means of high resolution ($\Delta \varepsilon = 17.5$ meV) inelastic neutron scattering below $T_N$. The dynamical exponents in the peak intensity ($A(q)/q^y$) and the dispersion relation ($E(q)/q^z$) for antiferromagnetic fractons were determined as $y = 2.9 \pm 0.2$ and $z = 1.8 \pm 0.2$, in good agreement with the numerical study. Also, the validity of the scaling based on the single-length-scaling postulate (SLSP) was confirmed.

The concept of fractons was introduced to describe vibrational modes excited in fractal structures, in which the spectral dimension $d$ plays a central role in describing their dynamical properties [1, 2, 3]. The fracton density of states is given by $D(\omega) \propto \omega^{d-1}$ with the frequency $\omega$, the localization length behaves as $\Lambda \sim \omega^{-1/2}$ with the dynamical exponent $z = D_f/d$, where $D_f$ is the fractal dimension, and the dispersion relation of fractons follows the relation $\omega \sim q^z$ with the wavenumber $q$. It has been suggested that the spectral dimension for antiferromagnetic fractons takes a universal value $d_{AF} = 1$ independent of embedding Euclidean dimensions [4]. The dynamics can be characterized on the basis of the single-length-scaling postulate (SLSP), namely, that the localization length, wavelength, and scattering length of fractons collapse onto a single length scale $\Lambda(\omega)$ [5]. The SLSP has concluded the scaling in the dynamical structure factor: $S(q, \omega) = q^{-y}F[q\Lambda(\omega)]$ with an exponent $y$. The validity of the SLSP in the spin dynamics as well as $d_{AF} = 1$ were demonstrated in the three-dimensional (3d) diluted antiferromagnet RbMn$_{0.4}Mg$_{0.6}F$_3$ and the two-dimensional (2d) diluted antiferromagnet Rb$_2$Mn$_{0.598}$Mg$_{0.402}$F$_4$ by high-resolution inelastic neutron scattering (INS) experiments [6, 7]. In this paper, the spin dynamics of antiferromagnetic fractons in the 2d system are described [7].

We have performed high-resolution INS experiments on the 2d diluted antiferromagnet Rb$_2$Mn$_{0.598}$Mg$_{0.402}$F$_4$ [8] with the magnetic concentration close to the percolation concentration of the 2d square lattice ($c_p = 0.593$). The parent system Rb$_2$MnF$_4$ has a layered perovskite structure where Mn spins of $S = 5/2$ are antiferromagnetically ordered at $T_N = 38$ K [9, 10]. Rb$_2$MnF$_4$ is well described by a 2d square-lattice Heisenberg model incorporating the nearest-neighbor exchange interactions ($J$) with a magnetic anisotropy. From INS experiments, $J$ was determined to be 0.34 meV, the second-neighbor exchange constant was smaller than $J$ in two order of magnitude, and the magnetic anisotropy with the gap energy of 0.62 meV was observed at the magnetic zone center [10]. Mn ions can be randomly replaced by nonmagnetic Mg ions,
and in Rb$_2$Mn$_x$Mg$_{1-x}$F$_4$, $T_N$ decreases with $c$ decreasing. At $c = c_p$, $T_N = 0$ K and a percolation network of Mn spins spans in the crystal.

Antiferromagnetic superlattice points in single-crystalline samples separated from a lattice structure were selectively chosen to investigate coherent magnetic properties. The structure factor for fractal structures takes the scaling form $S(q) \propto q^{-D_f}$. In fact, the fractal dimensions $D_f$ of dilute antiferromagnets with the magnetic concentration ($c$) close to the percolation concentration ($c_p$) obtained by elastic neutron scattering experiments [11, 12] agree with the theoretical values for percolation networks [3]. Therefore, a single-crystalline sample of a diluted antiferromagnet with $c$ close to $c_p$ is an ideal material for investigations of fractal nature.

High-resolution INS experiments by using a single-crystalline sample of Rb$_2$Mn$_{0.598}$Mg$_{0.402}$F$_4$ were performed on the IRIS spectrometer at the ISIS spallation neutron source at the Rutherford Appleton Laboratory [7, 13]. On IRIS, a polychromatic neutron beam is incident upon the sample and the scattered neutrons with a final energy $E_f = 1.845$ meV are detected. The energy transfer ($\hbar\omega$) is determined from the time-of-flight of the detected neutrons. The energy resolution ($\Delta E$) was confirmed to be $17.5\mu$eV at full width at half maximum by measuring the elastic incoherent scattering at $T = 100$ K. IRIS is equipped with 51 separate detector elements covering scattering angles ($\phi$) from 25.75° to 158°. The [001] axis of the sample was vertically mounted, and the (010) magnetic reflection was detected at $\phi = 68.25^\circ$. Measurements were performed at $T = 1.5$ K and 100 K. $T_N$ was determined to be 4 K from the temperature dependence of the magnetic Bragg and critical scattering intensities at (010).

Figure 1 shows the observed energy spectra. All spectra at $T = 1.5$ K were well fitted by the sum of one broad dispersive Lorentzian, three Lorentzians centered at $0$ meV, 1.6 meV, a Gaussian component from incoherent elastic scattering with $\Delta E = 17.5\mu$eV, and a constant background. The non-dispersive peaks at 0.66 and 1.6 meV correspond to $2J$ and $5J$, and their origins are excitations in a spin dimer and Ising cluster excitations, respectively [6, 14]. The Lorentzian component centered at 0 meV represents critical scattering. At $T = 100$ K, these signals diminish and only the signal corresponding to incoherent elastic scattering remains. It should be noted that the constant background and the incoherent scattering component centered at 0 meV determined above was in good agreement with that observed at $T = 100$ K.

Figure 2 plots the values of peak positions ($E(q)$) and the peak intensity ($A(q)$) of the broad dispersive Lorentzian component ($I(q, \omega)$), which was assigned to the magnetic fractons. The value of $q$ was determined as the distance between the peak position on the reciprocal plane and the magnetic zone center (010) [7]. As shown in Fig. 2, the dispersion relation and the peak intensity were well fitted to $E(q) = [E_0^2 + E_1^2(q\xi)^2]^{1/2}$ with $\zeta = 1.8 \pm 0.2$, $E_0 = 0.08 \pm 0.01$ meV and $E_1 = 0.38 \pm 0.15$ meV and $A(q) = 1/[A_0^2 + A_1^2(q\xi)^2]^{1/2}$ with $y = 2.9 \pm 0.2$, where $a$ is the lattice constant. Contrary to the 3d system [6], $E_0$ and $A_0$ should be finite, because a 2d system requires an anisotropy energy for the magnetic ordering. The current theory for antiferromagnetic fractons is based on the Hamiltonian incorporating only the nearest-neighbor exchange interactions [4]. To compare the present INS results with the current theory, the data at $q > 0.06$ Å$^{-1}$ should be used, where $E(q)$ and $A(q)$ are approximated to be $q^2$ and $q^{-y}$.

In the vicinity of $c_p$, the inverse correlation length is given by $1/\xi \simeq |c - c_p|^{1/\nu}/a_0$, where $\nu = 1.33$ for the 2d percolating system [3] and $a_0$ is the atomic spacing. The system behaves as a fractal in the length-scale regime $\ell < \xi$ ($q > 1/\xi$) and as a homogeneous system in the regime $\ell > \xi$ ($q < 1/\xi$). The magnetic concentration of $c = 0.598$ was determined by the chemical composition at the single-crystal growth, and the determined $T_N$ is consistent with the value of $c$ [15]. $1/\xi \simeq 0.0003$ Å$^{-1}$ was obtained from $c = 0.598$. Therefore, in the range of $q > 0.06$ Å$^{-1}$, the present system behaves as a fractal. The obtained values of $y$ and $z$ are in good agreement with those obtained by the numerical study, $y = 3.0 \pm 0.3$ and $z = 1.83 \pm 0.08$, and in the range of the theoretical prediction, $z < y < z + d$, where $d = 2$ is the spatial dimension [4]. The value of $z = D_J/d_{AF}$ obtained in the present experiment agrees with $D_J = 91/48 = 1.896$ for
Figure 1. Observed energy spectra of Rb$_2$Mn$_{0.598}$Mg$_{0.402}$F$_4$ at $T = 1.5$ K on IRIS. The solid lines are the fitted curves (see text). The energy spectra at $T = 100$ K were independent of $\phi$.

Figure 2. The dynamical structure factor $S(q, \omega)$ for 2d antiferromagnetic fractons in Rb$_2$Mn$_{0.598}$Mg$_{0.402}$F$_4$ observed at $T = 1.5$ K. The dispersion relation $E(q)$ (a), the peak intensity $A(q)$ (b), and the single-length-scaling analysis for $S(q, \omega)$ (c). The solid lines in (a) and (b) are the fitted curves. The vertical bars represent the statistical errors.

$d = 2$ [3], indicating that $\tilde{d}_{AF} = 1$.

The SLSP for the observed dynamical structure factors in the range of $q > 0.06$ Å$^{-1}$ was examined, assuming the scaling form of $S(q, \omega) = q^{-\nu}F(x)$ with $x = q\Lambda(\omega)$ [4]. The broad dispersive components $I(q, \omega)$ were deduced by subtracting the nondispersive components, the
incoherent elastic scattering and the constant background from observed spectra. The dynamical structure factor is given by \( S(q, \omega) = I(q, \omega)/(n+1)(f(Q))^2 \), where \( n+1 \) is the temperature factor \( [1-\exp(-\hbar\omega/k_BT)]^{-1} \) at \( T = 1.5 \) K with the Boltzmann constant \( k_B \). \( f(Q) \) is the magnetic form factor of Mn \( ^{2+} \) [16] and \( Q \) is the modulus of the scattering vector calculated for each \( \hbar\omega \) on the scan locus. From the value of \( \hbar\omega \) on the scan locus at the corresponding scattering angle \( \phi \), the position on the reciprocal plane was obtained. The value of \( q \) was determined as the distance between the position on the reciprocal plane and the magnetic zone center (010) [7]. Figure 2 (c) shows the scaling function \( F(x) = q^z S(q, \omega) \) as a function of \( x = q\Lambda(\omega) \) with \( \Lambda(\omega) = a(\hbar\omega/E_1)^{-1/2} \) using \( y = 2.9 \) and \( z = 1.8 \) obtained above. Each spectrum collapses onto a single universal curve which unequivocally demonstrates that the SLSP is valid for antiferromagnetic fractons.

In summary, INS experiments were performed at high resolution (\( \Delta E = 17.5 \) meV) for the diluted 2d Heisenberg antiferromagnet Rb\(_2\)Mn\(_{0.9}\)Mg\(_{0.1}\)F\(_4\) at \( T = 1.5 \) K < \( T_N \) (= 4 K). The observed broad dispersive magnetic excitations were attributed to antiferromagnetic fractons. The dispersion relation and \( q \) dependence of the peak intensities of the antiferromagnetic fractons were well described by \( E(q) \propto q^2 \) and \( A(q) \propto q^{-y} \), in the \( q \) range distant from the magnetic zone center (\( q > 0.06 \) Å\(^{-1}\)). In contrast to the 3d system described by only the nearest-neighbor exchange interaction [6], the 2d system requires the anisotropy energy. By using the data points at \( q > 0.06 \) Å\(^{-1}\), the exponents \( y = 2.9 \pm 0.2 \) and \( z = 1.8 \pm 0.2 \) were obtained. These exponents agree well with those determined in a numerical study [4]. The obtained value of \( z \) is identical to \( D_f \), it is concluded that \( \tilde{d}_{AF} = 1 \). This value coincides with that of \( \tilde{d}_{AF} \) previously obtained for 3d diluted Heisenberg antiferromagnet RbMn\(_{0.4}\)Mg\(_{0.6}\)F\(_3\) [6], indicating experimentally that the spectral dimension \( \tilde{d}_{AF} \) is universal and independent of the embedding Euclidean dimensions of the systems. In addition, the observed dynamical structure factors \( S(q, \omega) \) at \( q > 0.06 \) Å\(^{-1}\) were analyzed on the basis of the SLSP, showing that the SLSP for \( S(q, \omega) \) is valid.

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References
[1] Alexander S and Orbach R 1982 J. Phys. (Paris) 43 L625
[2] Rammal R and Toulouse G 1983 J. Phys. (Paris) 44 L13
[3] See a review, for example, Nakayama T, Yakubo K and Orbach R 1994 Rev. Mod. Phys. 66 381
[4] Yakubo K, Terao T and Nakayama T 1994 J. Phys. Soc. Jpn 63 3431
[5] Alexander S 1989 Phys. Rev. B 40 7953
[6] Itoh S, Nakayama T, Kajimoto R and Adams M A 2009 J. Phys. Soc. Jpn. 78 013707
[7] Itoh S, Nakayama T and Adams M A 2011 submitted to J. Phys. Soc. Jpn.
[8] Itoh S, Ikeda H, Yoshizawa H, Harris M J and Steigenberger U 1998 J. Phys. Soc. Jpn. 67 3610
[9] Birgeneau R J, Guggenheim H J and Shirane G 1970 Phys. Rev. B 1 2211
[10] Cowley R A, Shirane G, Birgeneau R J and Guggenheim H J 1977 Phys. Rev. B 15 4292
[11] Ikeda H, Iwasa K and Andersen K 1993 J. Phys. Soc. Jpn. 62 3832
[12] Itoh S, Kajimoto R, Iwasa K, Aso N, Bull M J, Adams M A, Takeuchi T and Yoshizawa H 2006 Physica B 385-386 441
[13] Carlile C J and Adams M A 1992 Physica B 182 431
[14] Takahashi M and Ikeda H 1993 Phys. Rev. B 47 9132; Iwasa K, Andersen K H, Takahashi M and Ikeda H 1994 J. Phys. Soc. Jpn. 63 2862
[15] Birgeneau R J, Cowley R A, Shirane G, Tarvin J A and Guggenheim H J 1980 Phys. Rev. B 21 317
[16] Watson R E and Freeman A J 1961 Acta Cryst. 14 27