Double-peak structure of the dynamical structure factor in diluted Heisenberg antiferromagnets

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We have investigated, in terms of large-scale computer simulations, the spin-wave dynamics of diluted Heisenberg antiferromagnets. The dynamical structure factor has been calculated with high resolution applying the efficient algorithm based on the forced oscillator method. At close to the magnon-fracton crossover energy, our results show no indication of the double-peak structure for the dynamical structure factor $S(\mathbf{q},\omega)$, which has been predicted as the contribution of both magnons and fractons, while $S(\mathbf{q},\omega)$ for highly concentrated systems recover the double-peak structure. The calculated results for the latter case are in good agreement with recent inelastic-neutron-scattering experiments.

I. INTRODUCTION

In the past decade, diluted Heisenberg antiferromagnets have attracted much attention due to their random self-similar (fractal) structures,[1–4] i.e., the spin-wave dynamics reflects the fractal nature of the system.[5–10] At length scale shorter than a correlation length $\xi=\Xi_0|p-p_c|^{-\nu}$ (Ξ_0 is the constant prefactor), where p is the percolation concentration and p_c is the percolation threshold, the self-similarity becomes relevant.[11] While, at larger length scale, the system is considered to be homogeneous. Excitations on such structures exhibit a crossover at a length scale ξ. Conventional magnons with the wavelength larger than ξ cross over to *(antiferromagnetic) fractons* with shorter wavelength λ. Fractons are the characteristic excitations reflecting the self-similarity of the system, and strongly localized. The crossover energy ω_c is related to ξ through the dispersion relation $\omega_c \propto \xi^{-D_f/\tilde{d}_a}$, where D_f is the fractal dimension and $\tilde{d}_a$ is the spectral dimension of *antiferromagnetic* fractons. In previous works,[12–14] we have determined the value of the spectral dimension $\tilde{d}_a$ from the calculation of the density of states of diluted antiferromagnets, namely, the spectral dimension $\tilde{d}_a$ of antiferromagnetic fractons takes a value very close to *unity* for any Euclidean dimension d. This indicates that antiferromagnetic fractons belong to a different universality class from that of vibrational or ferromagnetic fractons ($\tilde{d}\approx\frac{4}{3}$).[15] Yet, there still remain unresolved problems in regard to the dynamics of antiferromagnetic fractons, especially for the analysis of the dynamic structure factor observed by neutron-scattering experiments.

Scattering experiments on diluted Heisenberg antiferromagnets have been reported by several authors.[2,3,16–20] Uemura and Birgeneau[2,3] have performed inelastic-neutron-scattering experiments for $Mn_xZn_{1-x}F_2$ for $x=0.50$ and 0.75. $Mn_xZn_{1-x}F_2$ is $d=3$ diluted antiferromagnet with spins $S=\frac{5}{2}$, but has weak anisotropy. They have obtained asymmetric line shape at small wave vectors with a long tail extending towards higher energies. With increasing wave vectors, the sharp peaks shift higher-energy region with diminishing the intensity, and the peak width increases rapidly. Another broad and damped component grows at higher energy. The effective-medium approximation (EMA) has predicted for $S(\mathbf{q},\omega)$ the appearance of the double-peak structure at the crossover region.[21–23] Chen and Landau have performed numerical simulations for site-diluted bcc antiferromagnets at $p=0.50$, and their calculated results have also supported the above arguments.[4] However, there is no clear explanation on the origin of this double-peak structure.

In the previous study,[14,24] we have demonstrated the asymptotic form of the dynamical structure factor at p_c for diluted Heisenberg antiferromagnets and verified that antiferromagnetic fractons satisfy the single-length-scaling postulate. This implies that all length scales such as the wavelength, the localization length and the scattering length, collapse to an unique characteristic length scale $\lambda(\omega)\sim\omega^{-\tilde{d}_a/D_f}$.[24,25] It is important to clarify the profiles of $S(\mathbf{q},\omega)$ for diluted Heisenberg antiferromagnets at the concentration $p>p_c$, because most of experiments have been performed for the systems at $p>p_c$.

In this paper, we present the numerical results of the dynamical structure factor $S(\mathbf{q},\omega)$ of $d=2$ square (sq) and $d=3$ simple cubic (sc) diluted Heisenberg antiferromagnets varying the concentration p in order to clarify the behavior of $S(\mathbf{q},\omega)$ around the magnon-fracton crossover. Our results show, for the system with low concentration p, but $p>p_c$, that there is only one peak in the ω dependence of the dynamical structure factor at the magnon-fracton crossover. The dynamical structure factor for the system with higher concentration ($p=0.75$) shows the double-peak structure, which is in accord with recent inelastic-neutron-scattering experiments on $RbMn_{0.74}Mg_{0.26}F_3$ performed by Takahashi and Ikeda.[20] This system is an ideal isotropic Heisenberg antiferromagnet because the magnitude of anisotropy is very small and the exchange interaction is dominant only for the nearest neighbors.[20,26]

This paper is organized as follows. In Sec. II, we present the numerical results of $S(\mathbf{q},\omega)$ on the diluted antiferromagnets near p_c, which were obtained by applying the efficient algorithm based on the forced-oscillator

method. This numerical method enables us to work with much larger systems than the exact diagonalization approach, and the details of this method are given in Ref. 13. In Sec. III, the results of $S(\mathbf{q},\omega)$ on the system far from p_c are presented, and confirm the existence of double-peak structure. We also analyze the calculated results of $S(\mathbf{q},\omega)$ at the zone boundary for several percolation concentrations. Conclusions are given in Sec. IV.

II. DYNAMICAL STRUCTURE FACTOR AT THE MAGNON-FRACTON CROSSOVER

The Hamiltonian for diluted Heisenberg antiferromagnets is given by[23,24]

$$\mathcal{H}=\sum_{\langle mn\rangle} J_{mn}\mathbf{S}_m\cdot\mathbf{S}_n \ , \tag{1}$$

where $\mathbf{S}_m$ denotes the spin at the site m, and J_{mn} the exchange coupling between nearest-neighbor sites m and n. We choose J_{mn} as $J_{mn}=1$ if sites m and n are connected on the percolating network, and $J_{mn}=0$ otherwise. Finite cluster effects are neglected. This is because the scattering intensity arising from finite clusters are negligibly small. The dynamical structure factor $S(\mathbf{q},\omega)$ for antiferromagnetic spin waves is given by[13,24]

$$S(\mathbf{q},\omega)\propto\langle n+1\rangle\chi''(\mathbf{q},\omega)$$

$$\propto\langle n+1\rangle\pi\sum_{\lambda}\delta(\omega-\omega_\lambda)\left\{\sum_m e^{-i\mathbf{q}\cdot\mathbf{R}_m}\sigma_m v_m(\lambda)\right\}$$

$$\times\left\{\sum_n e^{i\mathbf{q}\cdot\mathbf{R}_n}u_n(\lambda)\right\}, \tag{2}$$

where $\langle n+1\rangle$ is the Bose factor expressed by $1/(1-e^{-\beta\omega})$, and $\chi''(\mathbf{q},\omega)$ is the imaginary part of generalized susceptibility, $\mathbf{R}_n$ is the positional vector of the site n, and σ_n is a variable taking $+1$ at the site n belonging to the up-spin sublattice or -1 for the down sublattice. $u_n(\lambda)$ [and $v_n(\lambda)$] are the right (and left) eigenvectors of the dynamical matrix describing the transverse-spin deviations under the linearized spin-wave approximations (see Ref. 13). In the following, the Bose factor is omitted without loss of generality. Theoretical studies based on the EMA suggest the characteristic features of $S(\mathbf{q},\omega)$ around the magnon-fracton crossover.[21–23] Namely, for $\xi/\lambda<1$, $S(\mathbf{q},\omega)$ has a narrow peak due to the magnon dispersion law,

$$\omega=c(p)k \ , \tag{3}$$

where $c(p)$ is the concentration-dependent stiffness coefficient. At high energies ($\omega\approx\omega_c$), a small and broad hump appears with a long tail extending to higher ω, reflecting the fracton contribution. For $\xi/\lambda>1$, namely, in the self-similar regime, magnons cannot survive, and the contribution from fractons with broad peak become relevant. The EMA theory claims that the broad line shape results in the Ioffe-Regel condition for fractons.[27]

In this section, we present numerical results for the dynamical structure factor $S(\mathbf{q},\omega)$ on $d=2$ and 3 diluted Heisenberg antiferromagnets around the magnon-fracton crossover at $p>p_c$. In particular, we pay attention to the system at the concentration p close to p_c but $p>p_c$, and investigate the profile of $S(\mathbf{q},\omega)$ around $\omega\approx\omega_c$ in detail in order to clarify the above prediction. This system has the correlation length ξ larger than the previous numerical studies for $S(\mathbf{q},\omega)$.[4,28] The algorithm based on the forced oscillator method is used to calculate the dynamical structure factor on diluted Heisenberg antiferromagnets with high resolution.[13] Calculations are performed in units of $S/\hbar=1$ and under periodic boundary conditions. The calculated results are presented in Figs. 1(a) and 1(b). Figure 1(a) shows the ω dependence of $S(\mathbf{q},\omega)$ for $d=2$ bond-diluted antiferromagnets at $p=0.58$ ($p_c=0.50$) for five different wave vectors $\mathbf{q}$. We have calculated $S(\mathbf{q},\omega)$ with $\mathbf{q}$ along the [10] direction from the magnetic zone center. The solid lines are only guides to the eye. The ensemble average has been taken over six

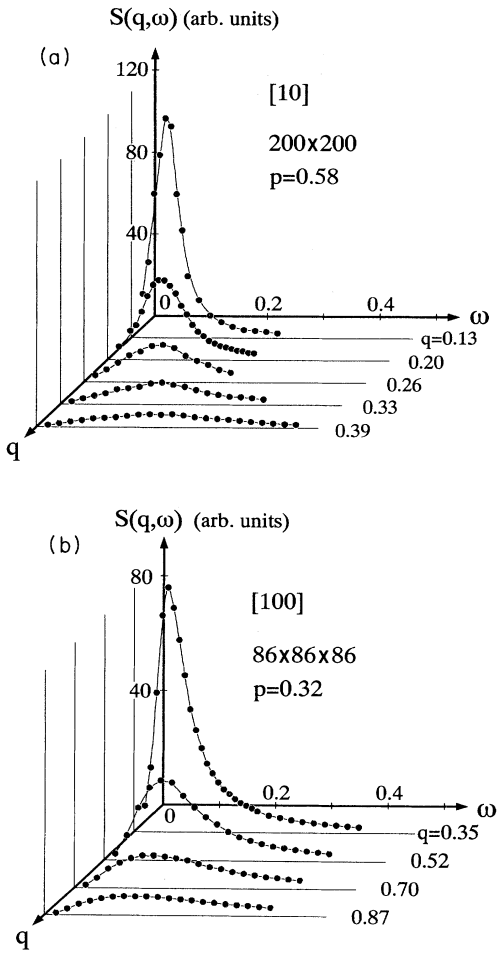

FIG. 1. (a) The ω dependence of $S(\mathbf{q},\omega)$ for $d=2$ bond-percolating networks at $p=0.58$ formed on 200×200 square lattices. The results were obtained by averaging over 6 realizations of bond-percolating networks. The solid lines are only guides to the eye. (b) The ω dependence of $S(\mathbf{q},\omega)$ for $d=3$ bond-percolating networks at $p=0.32$ formed on $86\times86\times86$ cubic lattices. The results were obtained by averaging over realizations of bond-percolating networks.

realizations of bond-diluted (BP) networks formed on 200×200 square lattices. The resolution for ω is $\delta\omega\approx0.02$, which is small enough compared with the linewidth of each peak in Fig. 1(a). The largest network has 37 449 spins under the periodic boundary conditions. The correlation length of this system is $\xi\approx29a$ (a is a lattice constant). ω_c is estimated by $\omega_c\approx0.12$ from the data corresponding to $q(\equiv|\mathbf{q}|)=2\pi/\xi\approx0.22$ (in units of $a=1$) in Fig. 1(a), so the correlation length ξ becomes large enough compared with the lattice constant a, and make the system size much larger than ξ.

The calculated results of $S(\mathbf{q},\omega)$ for $d=3$ bond-diluted antiferromagnets at $p=0.32(p_c=0.25)$ is shown in Fig. 1(b). The ensemble average is taken over two realizations of BP networks formed on $86\times86\times86$ cubic lattices under periodic boundary conditions. The largest network has 501 400 spins. The correlation length of this system is $\xi\approx10a$ and the crossover energy ω_c is estimated to be $\omega_c\approx0.12$. We have calculated $S(\mathbf{q},\omega)$ for four different $\mathbf{q}$ along the [100] direction. The resolution of ω is taken as $\delta\omega\approx0.02$. Figures 1(a) and 1(b) indicate that, for small wave vectors ($q<q_c$), the sharp asymmetric peak exists at small energy with a tail extending towards higher energies and there is no additional peak (nor shoulderlike) structure at higher ω. As $\mathbf{q}$ increases, peak widths increase rapidly and peak positions shift to higher energies beyond $\omega\approx\omega_c$. This corresponds to the magnon-fracton crossover at $\omega\approx\omega_c$. That the q dependence become irrelevant at $\omega>\omega_c$ comes from the strongly localized nature of antiferromagnetic fractons. Figures 1(a) and 1(b) confirm that there is only a single peak of $S(\mathbf{q},\omega)$ *at the magnon-fracton crossover* for the above systems.

III. DYNAMICAL STRUCTURE FACTOR FOR THE SYSTEMS WITH HIGHER CONCENTRATIONS

The calculation was extended to diluted Heisenberg antiferromagnets with higher concentration, on which most of experiments have been performed so far. Note that p_c of $d=3$ bcc site-percolating network takes a value of 0.25, while p_c of sc site-percolating network takes 0.312. The ω dependence of $S(\mathbf{q},\omega)$ for $d=3$ site-diluted sc antiferromagnets at $p=0.75$ are shown in Figs. 2(a) and 2(b). The network is formed on $40\times40\times40$ cubic lattice. The percolation concentration is far from $p_c=0.312$ and the range of length scale treated as self-similar (fractal) is quite narrow. Figure 2(a) shows the ω dependence of $S(\mathbf{q},\omega)$ with wave vectors along the [100] direction from the zone center. In this figure, the maximum energy of spin-wave $\omega_{\max}$ is $\omega_{\max}=6.0$, and the resolution of ω is taken as $\delta\omega\approx0.1$. At the small wave vectors, the line shape shows a sharp peak with a shoulder. At $q=0.79(q=0.25q_{\mathrm{ZB}})$, we can realize the well-resolved double-peak.

Next, the dynamical structure factors with wave vectors along the [110] direction are calculated, and the similar results are obtained [Fig. 2(b)]. From these, the double-peak structure of $S(\mathbf{q},\omega)$ reported by the inelastic-neutron-scattering experiments can be interpreted as the coexistence of a magnon peak at lower ω and discrete peaks from Ising-cluster excitations at higher ω, which reflect different coordination numbers of each site in percolating networks.[2,28–30] This result is in accord with the results of the recent experiment by Takahashi and Ikeda,[20] which have been done for $RbMn_xMg_{1-x}F_3$ ($x=0.74,0.63$).

The line shape of $S(\mathbf{q},\omega)$ at the *zone boundary* have been studied by several authors. Buyers, Pepper, and Elliott[31] have predicted a multiple-peaked line shape by the extended coherent potential approximations. Holcomb and Harris[28] have numerically studied a $d=3$ bcc lattice with 8192 sites and Kirkpatrick[29] calculated $S(\mathbf{q},\omega)$ for $d=2$ sq lattice by the matrix-inversion method. Thorpe and Alben[30] have studied a $d=2$ diluted antiferromagnets by the equation-of-motion method. Their results show multiple-peaked line shape of $S(\mathbf{q},\omega)$ at the zone boundary.[28–30] They have explained the origin of these peaks using Ising-cluster model. Recent neutron-scattering experiments, in which the behaviors of the zone boundary were studied, claim that the intensity dis-

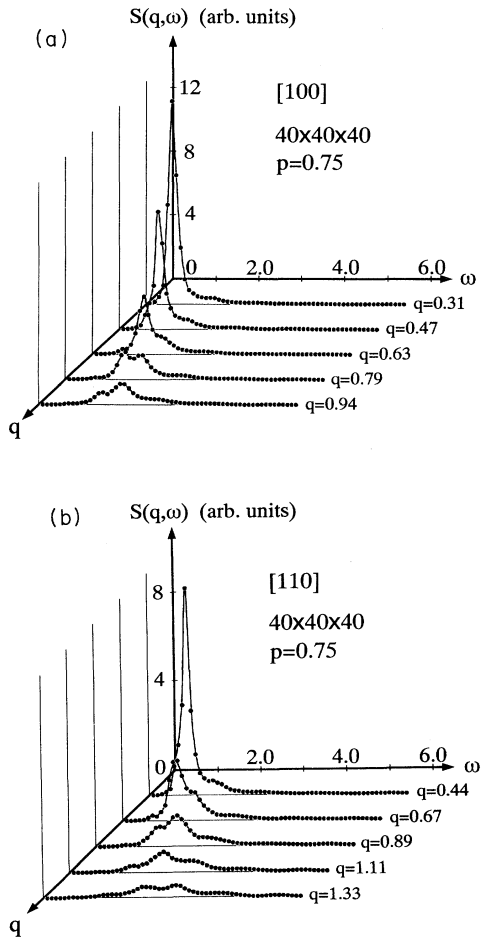

FIG. 2. (a) $S(\mathbf{q},\omega)$ for $d=3$ site-percolating networks at $p=0.75$ formed on $40\times40\times40$ cubic lattices. The wave vectors $\mathbf{q}$ are taken in the [100] direction from the zone center. (b) $S(\mathbf{q},\omega)$ for $d=3$ site-percolating networks at $p=0.75$ formed on $40\times40\times40$ cubic lattices. The wave vectors $\mathbf{q}$ are taken in the [110] direction from the zone center.

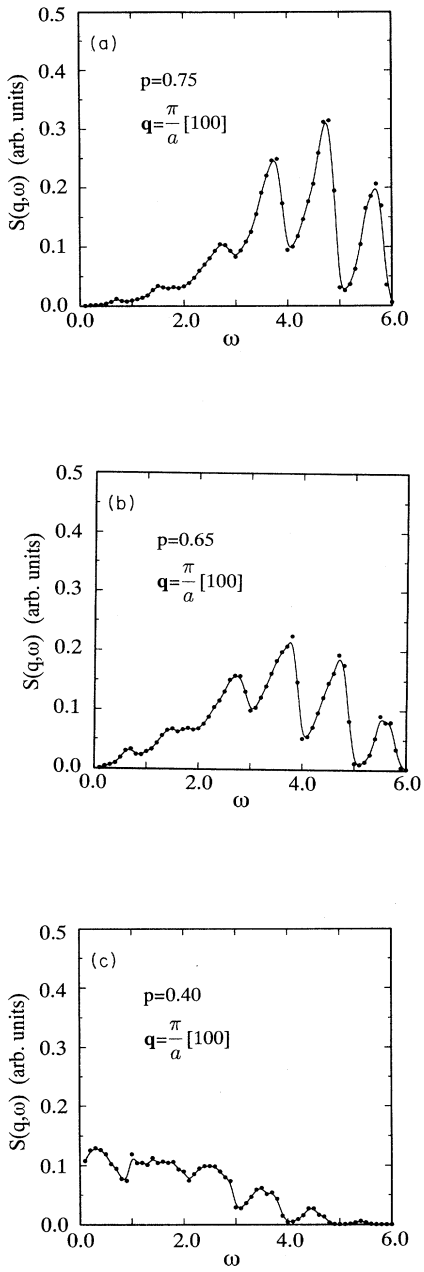

FIG. 3. (a) The ω dependence of $S(\mathbf{q},\omega)$ for $d=3$ at $p=0.75$ calculated at the zone boundary, for $d=3$ site-percolating networks on $60\times60\times60$ cubic lattices. The solid lines are only guides to the eye. The ensemble average is taken over three samples. (b) The ω dependence of $S(\mathbf{q},\omega)$ for $d=3$ at $p=0.65$ calculated at the zone boundary, for $d=3$ site-percolating networks on $60\times60\times60$ cubic lattices. The solid lines are only guides to the eye. The ensemble average is taken over three samples. (c) The ω dependence of $S(\mathbf{q},\omega)$ for $d=3$ at $p=0.40$ calculated at the zone boundary, for $d=3$ site-percolating networks on $60\times60\times60$ cubic lattices. The solid lines are only guides to the eye. The ensemble average is taken over three samples. The peak observed at low energy ($\omega\approx0.3$) is due to the contribution from localized fractons.

tribution of each peak is not in accord with the probability of local distribution of magnetic neighbors in diluted Heisenberg antiferromagnets, and the scattering intensity at low ω is much larger.[3,20]

To understand the features of the discrete peaks at higher energy observed in Figs. 2(a) and 2(b), we have also calculated $S(\mathbf{q},\omega)$ of diluted Heisenberg antiferromagnets at the *zone boundary* $\mathbf{q}_{ZB}=(\pi/a)[100]$ for several percolation concentration ($p=0.75,0.65,0.40$) on $60\times60\times60$ cubic lattice. The system sizes treated are much larger than previous numerical studies. Figure 3(a) shows the ω dependence of $S(\mathbf{q}_{ZB},\omega)$ at $p=0.75$. The multiple peaks appear at energies smaller than "Ising energy" $\omega=n$ ($n=1,2,\ldots,6$). This is because the single-spin excitation is not an eigenstate of the Heisenberg systems.[28,29] Figure 3(b) shows $S(\mathbf{q}_{ZB},\omega)$ at $p=0.65$ and exhibits similar behaviors. The weight of the peaks shifts to lower energy region. Figure 3(c) is the calculated results at $p=0.40$, where the concentration p is close to p_c and the correlation length of this system is $\xi\approx9a$. Much larger intensity is obtained at $\omega\approx0.3$ in Fig. 3(c), compared with the value expected from the probability distribution of magnetic neighbors. This excess intensity results in the contribution of fractons, because fractons are strongly localized modes and they make a prominent contribution also at $\mathbf{q}=\mathbf{q}_{ZB}$. The additional peak at $\omega=1$ is due to the Kirkpatrick-Eggarter state which is the specific eigenstate for percolating systems.[32]

IV. CONCLUSIONS

We have investigated the dynamical structure factor $S(\mathbf{q},\omega)$ of diluted Heisenberg antiferromagnets using the efficient algorithm based on the forced-oscillator method, which is the powerful computational technique for the systems described by non-Hermitian matrices.[13] Our results demonstrate the behaviors of $S(\mathbf{q},\omega)$ at the magnon-fracton crossover, for which the correlation length ξ is large enough, namely, the order of $\xi\geq10a$. It has been confirmed that there appears only a single peak at the magnon-fracton crossover. In addition, we have calculated $S(\mathbf{q},\omega)$ for the system with higher concentration, and reproduced the double-peak structure of the line shape obtained quite recently by Takahashi and Ikeda.[20] To summarize, there is no double-peak structure for the dynamical structure factor $S(\mathbf{q},\omega)$ in diluted Heisenberg antiferromagnets at close to the magnon-fracton crossover. The double-peak structures observed by inelastic neutron-scattering experiments are not due to the magnon-fracton crossover, but the contribution from both magnons and Ising-cluster excitations.

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