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Magnons and factons in diluted antiferromagnets (invited)

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Site-diluted antiferromagnets with short-range interactions can serve as a model magnetic percolation system. A length scale ξ exists such that for length scales $(1/q) > \xi$ the structure is continuous (hydrodynamic limit), and the magnetic excitations are antiferromagnetic magnons, with a stiffness constant which depends critically upon the magnetic concentration. For length scales $(1/q) < \xi$, the structure is fractal, and the magnetic excitations are fractons, with a characteristic dispersion law. The fracton excitations are (strongly) spatially localized. The magnons are expected to be extended in the very long length scale limit, with the possibility of Anderson (weak) localization for length scales longer than ξ . The magnetic excitations cross over from magnon to fracton at an energy $\omega_c \propto \xi^{-[1+(\theta/2)]}$, where θ is the anomalous diffusion exponent. We have calculated the scattering form factor $I(q, \omega)$ for magnetic excitations of a $d = 3$ simple cubic antiferromagnet within the effective medium approximation (EMA), as a function energy transfer ω and momentum transfer q . We find that at small fixed q (within the magnon regime), $p > p_c$, $I(q, \omega)$ is sharply peaked at the magnon frequency. Near to p_c , a second (but small) peak at ω_c is visible on the asymmetric high-energy tail of $I(q, \omega)$. For larger fixed q (within the fracton regime), $I(q, \omega)$ is centered about the fracton frequency, although it is quite broad, reflecting the strong spatial scattering of fractons.

I. INTRODUCTION

The percolation network is the prime example of a random fractal geometry.¹ Site percolation is constructed from a fully occupied lattice by withdrawing sites at random until a fraction p remain occupied. Sites are said to be "connected" for short-range interactions if two occupied sites are near neighbors. A critical concentration p_c exists such that, for $p > p_c$, a connected "infinite cluster" spans the entire structure.²

A model example of a percolation network is the site-diluted antiferromagnet. Nonmagnetic ions are substituted at random for the magnetic ions. For $p > p_c$, the existence of an infinite cluster allows for long-range order in the antiferromagnetic state.³ The density of states for magnetic excitations in what we now refer to as the fracton regime (see Sec. II) was first obtained by Shender for both dilute ferromagnets and dilute antiferromagnets.⁴ The dynamical properties of the band-diluted antiferromagnet were first formulated within the effective medium approximation (EMA) by Tahir-Kheli,⁵ and later by the present authors with explicit reference to the fracton density of states and dispersion relation.⁶ Very recent neutron scattering experiments by Uemura and Birgeneau⁷ on the site-diluted antiferromagnet $(\text{Mn}_{0.5}\text{Zn}_{0.5})\text{F}_2$ have investigated the magnetic excitations throughout the Brillouin zone for concentrations p (here, $p = 0.5$) somewhat near to p_c (~ 0.25).

The purpose of this paper is to calculate the scattering form factor $I(q, \omega)$ for magnetic excitations of a diluted antiferromagnet. We shall have to resort to the EMA for a full calculation, although a more general formulation⁸ is now available which appears capable of predicting nearly all the

important experimentally observable features. The experimental results of Uemura and Birgeneau⁷ appear quite similar to the EMA result for $I(q, \omega)$ contained in this paper, even with regard to some initially unexpected structure. The similarity between the experimental and theoretical determination of $I(q, \omega)$ lends credence to a "fracton" interpretation for short length scale excitations on a percolating network.⁹

Section II presents a review of what is expected for excitations on a fractal network. Section III briefly outlines the EMA calculation of $I(q, \omega)$, and Sec. IV presents the results and a comparison with experiment.

II. EXCITATIONS ON A FRACTAL NETWORK

There are two independent parameters which are necessary to describe fully excitation dynamics on a fractal network. The first is the fractal dimension D , which allows for the distribution of occupied sites in space. In particular, the number of occupied sites within a sphere of radius r is proportional to¹

$$N(r) \propto r^D. \quad (1)$$

The choice of the second independent parameter is somewhat a matter of taste. Here, we select θ , the exponent which determines the range dependence of the diffusion constant on a fractal network¹⁰:

$$D(r) \propto r^{-\theta}. \quad (2)$$

With these definitions, one is led⁹ to the definition of the fracton dimension,

$$\bar{d} = 2D / (2 + \theta). \quad (3)$$

An alternative would be to define \bar{d} as an independent parameter, and thence to obtain θ using Eq. (3). The fracton dimension controls the energy density of states and the fracton "dispersion" law.⁹

The percolating network serves as a very rich model for materials which can exhibit fractal geometry. A characteristic length scale ξ exists which distinguishes between the conventional ("Euclidean") mass distribution and excitation spectrum, and the fractal equivalents. For the percolating network, $\xi \propto (p - p_c)^{-\nu}$. We refer to such behavior as "critical."

The Euclidean dimension d describes the embedding dimension in which the fractal network is constructed ($d = 3$ for three-dimensional solids). Then, the mass distribution for the infinite cluster on a percolating network behaves as

$$M(r) \propto \begin{cases} r^d, & r > \xi \\ r^D, & a < r < \xi, \end{cases} \quad (4)$$

where a is the size of the atomic cell.

In general, $D \leq d$, with $D \approx 2.5$ for a percolating network in $d = 3$. The short length scales are governed by fractal geometry, while the long length scales are governed by Euclidean geometry. Similarly, the fracton dimension $\bar{d} = 2D / (2 + \theta)$ for $a < r < \xi$, while $\bar{d} = d$ for $r > \xi$ ($D = d$ and $\theta = 0$ for Euclidean space).

The fracton dimensionality was originally introduced for vibrational excitations on a fractal network,⁹ but it can be immediately generalized to magnetic excitations as well.^{6,11} For long length scales (Euclidean space), the excitation spectrum behaves as usual, but with a stiffness coefficient which is critical (i.e., depend on the difference $p - p_c$). This has been worked out in detail for phonons,¹² and was developed for antiferromagnetic magnons first by Harris and Kirkpatrick¹³ and later by Kumar and Harris.¹⁴ Thus one expects a magnon dispersion law $\omega = J(p - p_c)qa$, where $J \rightarrow 0$ as $(p - p_c)^\tau$ (Ref. 14). Apart from weak localization effects brought about by Anderson localization,¹⁵ the magnons can be thought of as extended states, with a scattering lifetime primarily due to Rayleigh scattering off the fluctuations in site occupancy.

The new insights provided by fractal geometry apply to the short length scale regime. Here, the fundamental nature of the excitation spectrum is altered. The spin-wave states are strongly localized, in the Ioffe-Regel sense.^{16,17} In addition, there exists a known relationship between the localization length l and the spin-wave excitation energy⁹:

$$\omega \propto l^{-D/\bar{d}}. \quad (5)$$

This enables us to define a crossover frequency ω_c by the relationship

$$\omega_c \propto \xi^{-D/\bar{d}} \equiv \xi^{-[1 + (\theta/2)]}. \quad (6)$$

Hence, as a function of excitation energy, one crosses over from magnon to fracton excitations at the crossover frequency ω_c . This will play an essential role in the interpretation of the neutron scattering data of Uemura and Birgeneau.⁷

III. EFFECTIVE MEDIUM APPROXIMATION CALCULATION OF $I(q, \omega)$

Most scattering experiments measure the structure factor $I(q, \omega)$. For neutron scattering from antiferromagnetic

spin waves,

$$I(q, \omega) = \langle n + 1 \rangle \chi''(q, \omega), \quad (7)$$

where $\langle n \rangle$ is the thermal average of the Bose function at frequency ω , and χ'' is the imaginary part of the spin-deviation operator Green's function,¹⁸

$$\begin{aligned} \chi''(q, \omega) &= \text{Im } G^{\alpha\beta}(q, \omega) \\ &= \text{Im} \langle T a_l^{\alpha\dagger}(t) a_{l'}^\beta(0) \rangle_{q, \omega} \\ &= \text{Im} [(-1)^{l+l'} + \alpha/2 (S_l S_{l'})^{1/2}] \\ &\quad \times \langle T S_l^{\alpha\dagger}(t) S_{l'}^\beta(0) \rangle_{q, \omega}, \end{aligned} \quad (8)$$

where the superscripts α, β label the two interpenetrating antiferromagnetic sublattices ($+1$ for up spins, -1 for down spins), l, l' the position of the l, l' spins, T the time-ordering operator, and the subscripts q, ω denote the spatial and time Fourier transform, respectively.

The equations of motion for $G^{\alpha\beta}(q, \omega)$ are given in Refs. 5 and 6 for a bond-percolation model. The EMA procedures are detailed, and the complex algebraic equations for both the spin-wave density of states and dispersion law are exhibited therein. A magnon regime is found for $\omega < \omega_c$, and a fracton regime for $\omega > \omega_c$. The density of states for the two regions joins on continuously, as opposed to the "steplike" structure found for the percolating ferromagnet.¹¹ This is undoubtedly due to the fact that antiferromagnetic excitations belong to a different universality class than do ferromagnetic excitations (and lattice vibrations).^{13,14} Within EMA, one finds $D = 2$ and $\bar{d} = 1$ for $d = 3$.

Neutron scattering experiments measure the structure factor $I(q, \omega)$ directly. Using the Green's functions obtained from Refs. 5 and 6, and carrying out the operations of Eqs. (8) and (9), we are able to compute $I(q, \omega)$ for fixed q at various values of ω , and vice versa. For simplicity, the former is exhibited in Figs. 1 and 2, for different bond concentrations p . Within EMA, $p_c = 1/d$, so that Fig. 1 is for $p - p_c = 0.17$, while Fig. 2 is for $p - p_c = 0.057$. The former is not too far from the relative concentration used in the experiments of Uemura and Birgeneau⁷ ($p - p_c \sim 0.25$).

IV. DISCUSSION AND COMPARISON WITH EXPERIMENT

The shapes of the $I(q, \omega)$ curves are very instructive. For each p , there exists a crossover wave vector q_c , corresponding to the crossover frequency ω_c . At fixed $q < q_c$, a sharp peak is observed at the corresponding magnon frequency. The peak is broadened on the high energy side, with structure at ω_c . This structure takes the form of a peak for p close to p_c (see Fig. 1) and a shoulder for p further away (see Fig. 2). The relative amplitude of this feature to the magnon peak height increases with increasing q .

This somewhat surprising result has actually been anticipated.¹⁹ Entin-Wohlman *et al.* calculated the asymptotic form for the structure factor for lattice vibrations on a fractal network and found a decreasing amplitude for $\omega > \omega_c$. In combination with the delta function at the phonon frequency (no damping was contained within that calculation), it suggests structure at ω_c . Physically, this is caused by the "softening" of the vibrational spectrum as one crosses over

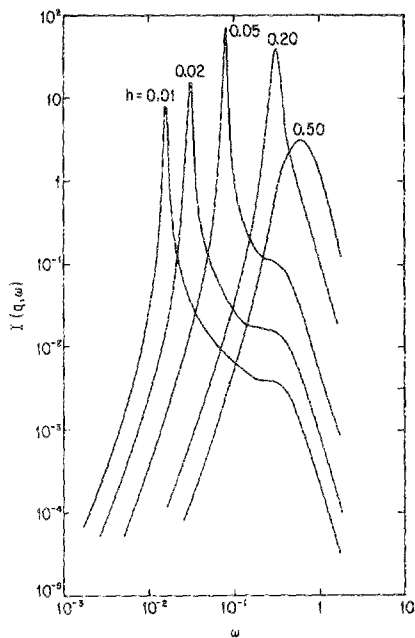


FIG. 1. A plot of the structure factor $I(q, \omega)$, for fixed q as a function of ω , for a simple cubic bond-percolation antiferromagnet calculated within the effective medium approximation (EMA). The concentration $p = 0.50$ ($p_c = 1/3$), leading to $\omega_c = 0.306$ and $q_c = 0.39\pi$. The reduced wave vector labeled on the figure, $h = q/\pi$, so that $h_c = 0.39$. The sharp peak for $h < h_c$ corresponds to the magnon dispersion law. Within EMA, the spin-wave stiffness coefficient C , defined from $\omega = Cq$, is proportional to $p - p_c$. This is consistent with the EMA values for the critical exponents: $t = 1$ and $\beta = 0$. The smaller peak at higher energies lies at ω_c for all $q < q_c$.

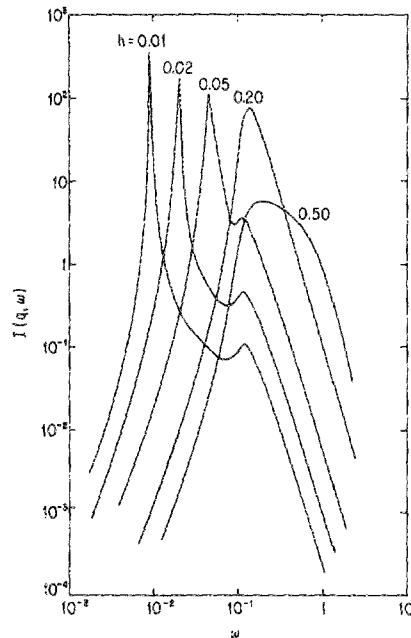


FIG. 2. Same as for Fig. 1, but with $p = 0.39$ ($p_c = 1/3$), leading to $\omega_c = 0.104$ and $q_c = 0.227\pi$ (so that $h_c = 0.227$).

into the fracton regime. A more general treatment, which obtains analytic forms for most of the features of $I(q, \omega)$, has recently been developed.⁸

As q crosses over into the fracton regime, $q > q_c$, the shape of the curve changes completely. $I(q, \omega)$ becomes a broad maximum with no structure at ω_c . This is as it should be, since in the fracton regime scaling arguments^{9,12} require that the states are not critical. They do not "know" that a crossover length ξ exists, and hence cannot depend upon ω_c .

These calculations for $I(q, \omega)$ have been performed for a simple cubic lattice, and for bond percolation. Nevertheless, the general shapes appear quite consistent with those of Uemura and Birgeneau,⁷ even to the extent of the feature at ω_c . We believe this is the strongest evidence yet for the supposition that fracton dynamics, both in the magnon and fracton regimes, are essential to the description of dilute antiferromagnets. It also suggests an immediate test for the validity of the conjecture²⁰ that vibrational excitations of amorphous solids can be described above a crossover frequency by fracton excitations: does $I(q, \omega)$ (q in the phonon regime) contain structure at ω_c , the latter being determined by the onset of the Ioffe-Regel¹⁶ limit as determined from an analysis²¹ of the thermal conductivity?

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