Dynamics of the fcc Heisenberg antiferromagnet with nearest-neighbor interactions. Fully occupied lattice

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(Received 26 June 1981; revised manuscript received 23 November 1981)

Numerical techniques are used to investigate the spin dynamics of the nearest-neighbor fcc Heisenberg antiferromagnet on the fully occupied lattice. The dynamic structure factor and the distribution of modes are calculated in the harmonic approximation. Despite the absence of three-dimensional long-range order the dynamics of the disordered antiferromagnet resembles the dynamics of a fcc antiferromagnet with type-III order.

I. INTRODUCTION

The nature of the ground states and the low-lying excited states in face-centered-cubic (fcc) antiferromagnet (AFM) with nearest-neighbor (NN) interactions continues to be an interesting problem in the field of disordered magnetism. The inability to minimize simultaneously all nearest-neighbor bond energies, sometimes referred to as frustration, leads to a highly degenerate ground state with no three-dimensional long-range order. Some years ago Danielian investigated the ground state of the Ising fcc AFM with NN interactions.[1,2] He found a degeneracy of 2^R with $R \simeq N^{1/3}$, N being the number spins. Although in the thermodynamic limit there are an infinite number of ground states, both the specific heat and the susceptibility vanish at $T=0$. The dilute fcc Ising AFM with NN interactions has been investigated by Grest and Gabl.[3] They found a crossover from frustrated AFM to spin-glass behavior as x, the fraction of occupied sites, fell below 0.4. The spin-glass phase extended down to the percolation concentration ($x \simeq 0.18$).

Analogous studies of the fcc Heisenberg AFM began with the work of Anderson who considered possible ground states of the fully occupied lattice.[4] More recently Alexander and Pincus[5] discussed ground-state configurations in the fcc AFM and other fully frustrated models. In an earlier study[6] the present authors reported the results of a numerical investigation of the ground states of the classical site-dilute fcc Heisenberg AFM with NN interactions. The ground-state energy and the rms ground-state magnetization (which vanishes in the thermodynamic limit) were calculated as a function of x for $0.1 \leq x \leq 1.0$. For the fully occupied lattice ($x=1.0$) it was found that the rms magnetization was zero for all ground-state configurations whereas the ground-state energy per spin was always equal to $-2J$, J being the exchange interaction. Both of these results were shown to be in agreement with a theoretical analysis based on the Luttinger-Tisza method[7,8] as formulated by Kaplan *et al.*[9,10]

In this paper we extend the analysis begun in Ref. 6 to the low-lying excited states of the fcc Heisenberg AFM with NN interactions. We use matrix diagonalization[11] and equation-of-motion techniques[12] to calculate the density of harmonic spin-wave modes and the corresponding zero-temperature dynamic structure factor. We limit our analysis to the fully occupied lattice; spin-wave excitations in dilute systems will be considered in a subsequent paper. Additional comments on the ground-state configurations are presented in Sec. II, while the dynamics is treated in Sec. III. Although there have been earlier studies of the ground states of this system, to the best of our knowledge ours is the first investigation of the dynamics.

II. EQUILIBRIUM CONFIGURATIONS OF THE FULLY OCCUPIED LATTICE

In our analysis of the fully occupied lattice we discovered two classes of equilibrium configurations depending on the approach followed in minimizing the energy. Both approaches involved rotating individual spins into the direction of their local field.[6,11] Configurations belonging to the first class (class I) were obtained when the spin being rotated was selected at random from the entire sample with the process being continued until the energy stabilized at $-2NJ$. In addition to class I a second class of configurations was obtained when the spins being rotated were chosen in sequence along a row in a particular (001) plane. After each spin in the row was rotated the process was repeated in a neighboring row in the same plane. When all the spins in the plane had been rotated the corresponding rotations were carried out in an adjacent plane. In all instances periodic boundary conditions were assumed.

The class-I configurations are easily described. Each (001) plane is a square lattice with two-dimensional AFM order in which in-plane nearest-neighbor spins are antiparallel. The disorder arises in the random orientation of the antiferromagnetic axis from plane to plane. Since a spin interacts with equal numbers of "up" and "down" spins in each of the nearest-neighbor planes the interplanar exchange field vanishes leaving a net intraplanar exchange field equal to $-4J$. The degeneracy associated with the orientation of the antiferromagnetic axes gives rise to a ground-state proportional to the number of (001) planes or, equivalently, to $N^{1/3}$.

The class-II configurations, which are the ones discussed in Ref. 6, can be viewed as defective class-I configurations in which the spins in a given (001) plane can be subdivided into at least two interpenetrating antiferromagnetic clusters (Occasionally spins in adjacent planes are found to belong to the same cluster.) All spins in a given cluster are either parallel or antiparallel to the cluster axis. However, the orientations of the cluster axes are correlated so as to ensure that the exchange field for each spin is equal to $-4J$. This condition can be expressed mathematically as

$$\sum_j{}' \cos\theta_{ij} = -4 \quad (i=1,\ldots,N) , \tag{1}$$

where θ_{ij} is the angle between spins i and j and the sum is over the twelve nearest neighbors of site i.

As pointed out in Ref. 6 the clusters differ from configuration to configuration coming in various sizes with unequal numbers of up and down spins. We have not succeeded in determining the degeneracy of the defect states. However, in view of the correlations in the orientations of the axis which are required in order that Eq. (1) be satisfied it would appear that the class-II configurations are no more numerous than those belonging to class I.

Finally we should mention that the ground-state configurations of the fcc Ising AFM have energy equal to $-2J$[1,2] and thus also qualify as ground states for the classical Heisenberg Hamiltonian. However, as far as we can tell our numerical procedures did not generate any Ising-type ground states.

III. DYNAMICS OF THE FULLY OCCUPIED LATTICE

In Ref. 12 it was shown how equation-of-motion techniques can be used to calculate the dynamic structure factor $S(\vec{q},E)$ in the harmonic spin-wave approximation provided the equilibrium spin configurations are available as input. Using the methods outlined in Ref. 12 and the configurations discussed in Sec. II we have calculated $S(\vec{q},E)$ for a 4000 spin array with periodic boundary conditions. To within the accuracy of the method our results are insensitive as to whether we use class-I or class-II configurations. With the exception of the (preferred) [001] direction $S(\vec{q},E)$ displays a sharp peak when plotted as a function of the energy E (see Fig. 1). The width of the peak is determined largely by instrumental effects (the exponential cutoff in the integration of the equations of motion gives rise to a half-width at half maximum of $0.5J$) whiles its position shifts with wave vector $\vec{q}$.

In Figs. 1, 2, and 3 we have plotted the peak po-

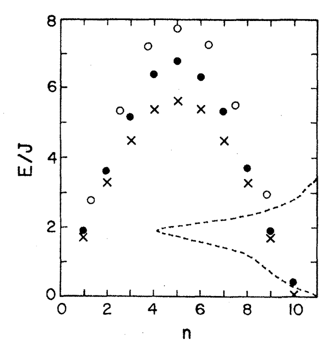

FIG. 1. Position of the spin-wave peak vs $\vec{q}$ for $\vec{q}$ along [100]. The wave vector is related to n through the equation $\vec{q}=(2\pi/10)\,(n,0,0)$. The solid circles are the results for the disordered magnet; the energies of the mode of the type-III AFM [Eq. (2)] are denoted by $\times$. Numerical results are from a single configuration with 4000 spins. Broken curve is $S(\vec{q},E)$ (vertical energy scale) for $n=1$. The results for a single Ising ground-state configuration of an array of 2048 spins are shown as open circles for corresponding q values.

sition for $\vec{q}$ along the [100], [110], and [111] directions. The numerical results, shown as solid circles, are compared with the spin-wave dispersion curve for a fcc Heisenberg AFM with type-III three-dimensional long-range order.[13] In a type-III fcc antiferromagnetic all spins are collinear. The ordering of successive (001) planes is described by the sequence $ab\bar{a}\bar{b}$ where the bar denotes the antiparallel (time reversed) configuration. Thus, equivalent planes are four layers apart. According to Ref. 13 the spin-wave energies in the type-III AFM are given by

$$\omega(\vec{q}) = 4J[\cos(q_x/2)\cos(q_y/2) + \cos(q_x/2)\cos(q_z/2) + \cos(q_y/2)\cos(q_z/2) + 1]^{1/2}[1 - \cos(q_x/2)\cos(q_y/2)]^{1/2} , \quad (2)$$

for unit spin and lattice constant. Here J denotes the exchange interaction in the Heisenberg Hamiltonian

$$\mathscr{H} = \sum_{(i,j)}' J\vec{S}_i \cdot \vec{S}_j , \quad (3)$$

where the prime signifies that the sum is over nearest-neighbor spins. The energies calculated from (1) are denoted by x.

It is apparent that there is a close resemblance between the dispersion curve for the disordered magnet and that associated with type-III order.[14] It is also evident that the disorder associated with having the spin axes of the individual (001) planes point in random directions has a small effect on the dynamics. The dynamical behavior for $\vec{q}$ along [001] is also similar in the two cases. From Eq. (2) we see that the spin-wave energy vanishes when the direction of propagation is along the preferred axis. Similar behavior, i.e., a peak in $S(\vec{q},E)$ centered at $E = 0$, is obtained for the disordered system for both classes of ground-state configurations.

We have also studied the dynamics associated with the Ising ground states. The latter were constructed by the method devised by Mackenzie and Young,[15] in which the integers ± 1 were assigned with equal probability to each of the (001) planes. If the jth plane had $n_j = +1$, spins in corresponding locations in the $j-1$ and $j+1$ planes were parallel; if it had $n_j = -1$, they were antiparallel. When allowance is made for periodic boundary conditions the configuration of an array with L (001) planes ($L+1 \equiv 1$) are specified by sequences of length $L-2$. Calculations carried out on $8\times8\times8$ arrays indicated that the dynamics closely resembled that of the class-I and class-II configurations. This is evident from Figs. 1–3 where the results from a single Ising configuration are plotted as open circles. There is good agreement between the three sets of data for $\vec{q}$ along [110] and [111]. For $\vec{q}$ along [100] the discrepancies are noticeably larger, although the behavior is qualitatively similar in all three cases.

In order to obtain additional insight into the dynamics we have diagonalized the dynamical matrix[11] and obtained the corresponding eigenvalues and eigenvectors. The distribution of modes for five configurations each with 256 spins is shown in the histograms in Fig. 4. As with the dynamic structure factor our results are insensitive as to whether we use class-I or class-II ground states. The data in Fig. 4 are to be compared with the corresponding results for the type-II antiferromagnetic shown in Fig. 5. It is apparent that the dis-

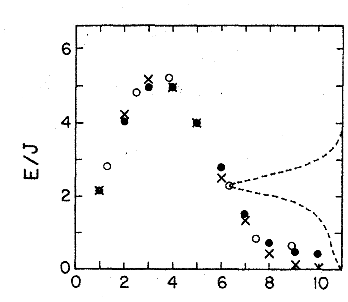

FIG. 2. Same as Fig. 1 except $\vec{q} = (2\pi/10)\,(n,n,0)$. The open circle coincides with the solid circle for $n = 5$.

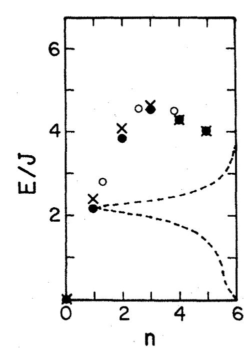

FIG. 3. Same as Fig. 1 except $\vec{q} = (2\pi/10)\,(n,n,n)$. The data are symmetric about $n = 5$. The open circles coincide with the solid circles for $n = 0$ and 5.

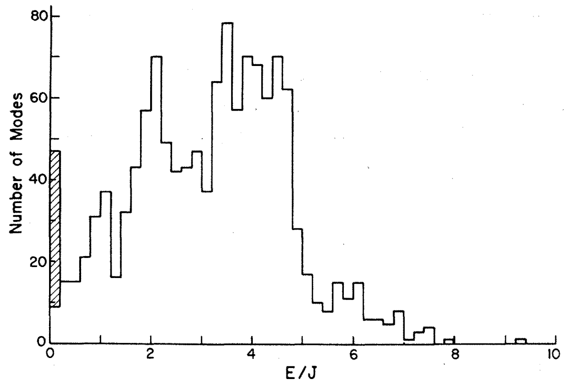

FIG. 4. Histogram for the density of states of a disordered AFM. The data are from five configurations each with 256 spins. The crosshatched area denotes modes with $E \leq 0.001$.

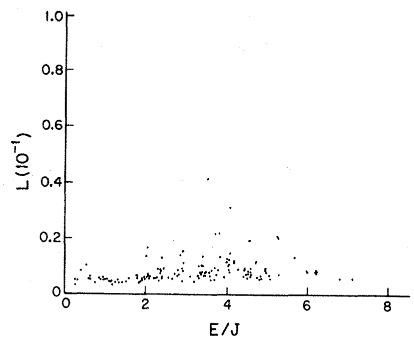

FIG. 6. Localization indices L_ν (Ref. 11) plotted vs mode energy. The data are from a single configuration of a disordered AFM with 256 spins.

tributions of modes in the two figures are qualitatively similar. However, the histogram in Fig. 6 has a cutoff at $E \simeq 6J$ whereas the modes in Fig. 5 extend out to higher energies. Comparison with Fig. 1 shows that the modes with energies above $6J$ are associated with the peak in $S(\vec{q},E)$ for $\vec{q}$ in the neighborhood of $(\pm\pi,0,0)$ or $(0,\pm\pi,0)$. The cross hatched areas in the two figures represent the contributions from the "zero-energy" modes ($E < 0.001$). It is these modes which give rise to the zero-energy response for $\vec{q}$ along [001]. Just as in the type-III AFM (Ref. 16) the presence of these modes indicates that the ground state of the disordered AFM is unstable with respect to spin fluctuations.

We have also calculated the localization indices[11] L_ν associated with the various modes. As discussed in Ref. 11 L_ν^{-1} is a measure of the number

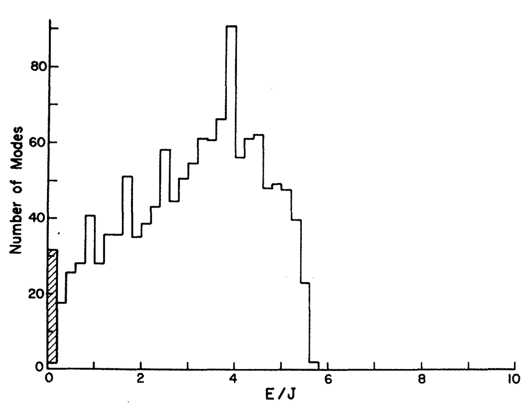

FIG. 5. Histogram for the density of states of a type-III AFM. Results are for an array of 256 spins with periodic boundary conditions. Crosshatched area denotes zero-energy modes.

of spins participating in the νth mode. Our results for L_ν for a single configuration of 256 spins are shown in Fig. 6. It is apparent that nearly all of the modes are delocalized, having indices < 0.02.

IV. DISCUSSION

Despite the absence of three-dimensional long-range order, the ground-state configuration of the fcc Heisenberg AFM with NN interactions on the fully occupied lattice shows a high degree of correlation. This is reflected in the fact that each spin sees the same exchange field and that the magnetization for a finite array is exactly equal to zero.[6] The correlation also carries over to the dynamics. As pointed out in Sec. III the spin-wave modes are delocalized and bear a close resemblance to the excitations of a type-III AFM. Although there is no interplanar exchange field the modes are fully three dimensional, propagating in all directions except along [001]. They are very different from the excitations in various models of Heisenberg spin-glasses.[12] In the spin-glass systems that have been studied so far there is no evidence of weakly damped propagating modes in $S(\vec{q},E)$. Finally we note that removing spins from the lattice weakens the correlations. The rms magnetization becomes finite and there is a distribution in local fields. As will be discussed in a subsequent publication there are corresponding changes in the spin dynamics as well.

ACKNOWLEDGMENT

This research is supported in part by the NSF under Grant No. DMR-7904154.

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[14]Although type-I order which is a collinear spin array with an *abab* sequence of (001) planes is also a possible ground state the dispersion of the disordered magnet bears no resemblance to the type-I spectrum. According to Ref. 13 the energies of the type-I modes are given by

$$\omega(\vec{q})=4J[\cos(q_x/2)\cos(q_y/2)+\cos(q_z/2)\cos(q_x/2)+\cos(q_z/2)\cos(q_z/2)\cos(q_y/2)+1]^{1/2}\times[\cos(q_x/2)\cos(q_y/2)-\cos(q_x/2)\cos(q_z/2)-\cos(q_y/2)\cos(q_z/2)+1]^{1/2},$$

which vanishes for $\vec{q}$ along [100] or [010].
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