Spin waves in dilute ferromagnets: Cluster–Bethe-lattice approach*

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A calculation of the local density of spin-wave states in a dilute ferromagnet is presented. It is based on a Green's-function formulation solved within the cluster–Bethe-lattice approximation. The method is compared with an exact solution for the one-dimensional dilute ferromagnet. We also study clusters of nine atoms in the bcc structure, and clusters of 7, 19, and 27 atoms in the simple-cubic structure. Our study shows: (i) a stability condition for ferromagnetism in agreement with percolation theory; (ii) a local structure dependence of the spin-wave structure; (iii) the existence of localized spin-wave states due to both isolated magnetic clusters and nonpropagating magnon modes.

I. INTRODUCTION

The study of spin waves in dilute ferromagnets[1] is ideally suited for the application of the cluster–Bethe-lattice method. The Bethe-lattice approximation is a well-known approximation which has been used for the calculation of the partition function of magnetic systems,[2] as well as for the calculation of the density of states in interacting many-electron systems.[3]

This approximation consists essentially in substituting for the infinite periodic lattice an infinite system of connected "atoms," with the same coordination number as the lattice of interest, but without closed rings of bonds. Alternatively, in diagrammatic terms, the Bethe-lattice approximation corresponds to the sum of all paths in the lattice which are self-retracing and contain no closed loops. Within this approximation it is possible to obtain, for instance, an analytic expression for the one-particle Green's function in an interacting electron system.[3]

It is also possible, naturally, to apply the same approximation to obtain the local electronic density of states in noninteracting systems, within the tight-binding scheme. Yndurain *et al.*[4] have extended the Bethe lattice systematically by using it to write down approximate boundary conditions for finite clusters of atoms embedded in infinite systems. They have called this approach the cluster–Bethe-lattice (CBL) method. In this method it is possible to compute, for instance, the local density of electronic states at the central atom of a cluster of atoms, attaching Bethe lattices of the same coordination number as the lattice of interest, to the "dangling bonds" of the atoms at the surface of the cluster. The advantage of the method resides mainly in the fact that the boundary conditions, which allow us to embed the finite cluster in an infinite system, can be expressed in a simple analytic form. In many cases it is then possible to obtain an exact, closed-form solution to the problem. The usefulness of this approach to the study of the local density of electronic states of amorphous systems[5] and alloys[6] has by now been extensively demonstrated.

When we decided to study the spectrum of the one-spin-wave states in alloys and amorphous magnetic systems, we found that the formal similarity of the formulation with the above-mentioned problems naturally led to the application of the CBL method to study these systems. Alloys and amorphous magnets have attracted considerable interest in view of the fundamental problems they pose as to the nature of their collective (magnetic) excitations.[7]

As our preliminary results[1] have shown, the method does yield very reasonable results for the local density of one-spin-wave states in the case of simple clusters. In this work we extend our previous treatment of the dilute ferromagnet to include, in addition to first nearest neighbors in a bcc lattice, clusters of first, second, and third nearest neighbors in a simple-cubic lattice. These clusters have, respectively, 7, 19, and 27 atoms.

It is well known that the Bethe-lattice approximation, when applied to uniform systems, results in a band narrowing.[8] Such a band narrowing can be traced to the restricted number of lattice paths which are summed over. In the CBL approximation, though, states appear outside the Bethe-lattice density of states, as expected intuitively, since we can imagine approaching the uniform system by considering successively larger clusters. In disordered systems, however, the

band narrowing may be a real physical effect, owing to the destruction of phase coherence of the electronic (or magnetic) excitations. Even so, we also expected to find band "tails" extending outside the main body of the density of states, a feature which the CBL method is particularly well suited to reproduce. This point is investigated in the present work, by considering the large clusters mentioned above.

Disordered magnetic systems have also been studied within the coherent-potential approximation, by Harris *et al.*,[9] Theumann,[10] and Tahir-Kheli,[11] among others. Our method provides in a certain sense, a complimentary way of looking at such alloy systems. Approximations like the coherent-potential approximation reintroduce at some point the translational periodicity of the empty lattice, so as to make use of Bloch's theorem. This feature makes them appropriate to the study of extended states but not convenient for the study of local properties. In the CBL approximation, on the other hand, the emphasis is put on the local properties of the system, the extended properties being treated in a very convenient, but nonetheless crude, way.

In the present work, we study the dilute ferromagnet, as an example of the application of the CBL method to disordered magnetic systems. The dilute ferromagnet is a binary substitutional alloy, of the form A_xB_{1-x} $(0 \leq x \leq 1)$. Atoms of type A have a localized magnetic moment, whereas atoms of type B are nonmagnetic. To make use of the power of the method in dealing with local properties we introduce short-range-order effects. We define a parameter p which is the probability that the nearest neighbor of a magnetic atom is also magnetic. Hence xp is the fraction of magnetic-magnetic nearest-neighbor pairs in the alloy.

In Sec. II, we discuss the Hamiltonian, the equation of motion for one-spin-wave states, and the CBL approximation in a form appropriate for the dilute ferromagnet. In Sec. III, we present an exact solution to the problem of a dilute ferromagnet in one dimension and compare it with the Bethe-lattice solution. In Sec. IV, we study the simple clusters of (1 +8) atoms, in a body-centered-cubic lattice. We show the connection between our approach and the usual percolation-theory ideas. In Sec. V, we examine more complicated clusters for a simple-cubic lattice and, finally, in Sec. VI, we present our conclusions.

II. DISORDERED MAGNETIC SYSTEMS

A. Hamiltonian and equations of motion

We want to deal with a disordered system of localized spins. The disorder can originate from the spatial distribution of magnetic atoms, from the different character of magnetic atoms forming the system, or from both. Hence we employ the generalized Heisenberg Hamiltonian:

$$H = -\sum_{j,j'} [J(j,j')S^z(j)S^z(j') + I(j,j')S^+(j)S^-(j')]. \tag{2.1}$$

In (2.1) the summation extends over all magnetic-atom sites (j).

We define two exchange coupling constants, which are position dependent: the longitudinal coupling $J(j,j')$ and the transverse coupling $I(j,j')$. The spin operator $\vec{S}(j)$, corresponding to the spin quantum number $S(j)$, is site dependent in the case of constitutional disorder. It obeys the usual commutation rules

$$[S^+(j), S^-(j')] = 2S^z(j)\delta_{jj'}, \tag{2.2}$$

$$[S^z(j), S^\pm(j')] = \pm S^\pm(j)\delta_{jj'}, \tag{2.3}$$

where

$$S^\pm(j) = S^x(j) \pm iS^y(j). \tag{2.4}$$

To solve the dynamical problem we employ Zubarev's formalism.[12] Since we are interested below in ferromagnetic systems, we define the simple Green's function:

$$G(j,j'|\omega) = \frac{1}{2[S(j)S(j')]^{1/2}} \langle\langle S^+(j); S^-(j')\rangle\rangle_\omega. \tag{2.5}$$

The double angular brackets in (2.5) indicate the time Fourier transform of the thermal average of the retarded commutator of the two operators. For $T = 0$, which is the situation we deal with, the thermal average reduces to the ground-state expectation value.

The equation of motion for the Green's function can readily be derived. Within the one-spin-wave approximation, which corresponds to the decoupling

$$\langle\langle S^z S^+; S^-\rangle\rangle \cong \langle S^z\rangle \langle\langle S^+; S^-\rangle\rangle, \tag{2.6}$$

we obtain

$$[\omega - E(j)]G(j,j'|\omega) = \delta_{j,j'} + \sum_{j''} T(j,j'')G(j'',j'|\omega). \tag{2.7}$$

In (2.7) we have

$$E(j) = 2\sum_{j'} J(j,j')S(j'), \tag{2.8}$$

$$T(j,j') = 2I(j,j')[S(j)S(j')]^{1/2}. \tag{2.9}$$

Several quantities of physical interest can be

computed from $G(j,j'|\omega)$. The inelastic-neutron-scattering cross section by one-spin-wave excitations of the magnetic system described by (2.1), for instance, is related to the dynamic structure factor, defined by

$$S(\vec{k},\omega)=\frac{1}{N}\sum_{j,j'}e^{i\vec{k}\cdot(\vec{R}_j-\vec{R}_{j'})}G(j,j'|\omega), \qquad (2.10)$$

where N is the number of magnetic atoms in the crystal.

The local density of one-spin-wave states at site j is given by

$$\rho_j(\omega)=-(1/\pi)\mathrm{Im}G(j,j|\omega). \qquad (2.11)$$

This is the quantity which we calculate using the CBL method.

It is interesting to observe that in disordered magnetic systems, as can be seen from (2.8) and (2.9), "diagonal" and "off-diagonal" disorder are intimately connected. To solve Eqs. (2.7) we must thus find a way of dealing with the nonperiodic energies $E(j)$ and "overlap integrals" $T(j,j')$.

B. Dilute ferromagnet

As a prototype disordered magnetic system we take a substitutional alloy A_xB_{1-x}. Atoms A have a localized magnetic moment (spin S) and atoms B are nonmagnetic (spin zero). The exchange constants J and I are taken to be of the form

$$J(j,j')=I(j,j')=\begin{cases}J & \text{for nearest-neighbor}\\ & \text{magnetic atoms}\\ 0 & \text{otherwise.}\end{cases} \qquad (2.12)$$

Moreover we assume J to be positive. In the uniform case ($x=1$) our problem reduces then to the case of an isotropic Heisenberg ferromagnet with nearest-neighbor-only coupling.

To describe the short-range-order effects in the alloy we introduce a parameter p, the probability that a given nearest neighbor of a magnetic atom be magnetic. In Fig. 1 we indicate the nature of the magnetic alloys resulting from specific choices of the parameters x and p. In constructing that diagram we restricted ourselves to lattices such that a perfectly ordered binary alloy ($x=0.5$) can be formed. Such lattices can be divided into two interpenetrating equivalent sublattices such that all nearest neighbors of an atom in one sublattice lie in the other one. The following limits are worth pointing out: (i) $p=0, x=0.5$ corresponds to the ordered alloy in which all nearest neighbors of an A atom are of the B type and vice versa; (ii) $p=x$ corresponds to the completely random alloy; and (iii) $p=1$ corresponds to the segregated alloy.

In the spirit of the CBL method we try to solve

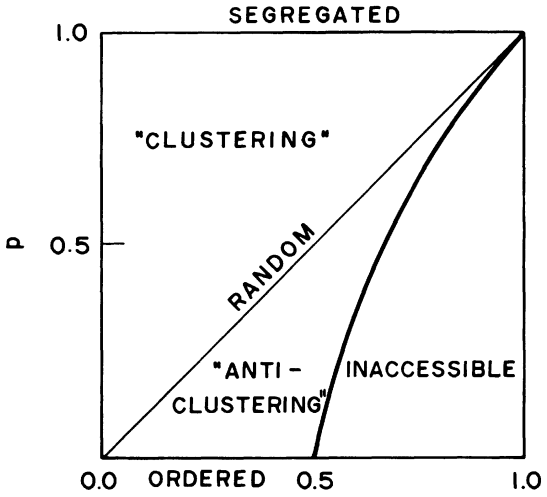

FIG. 1. Clustering properties of alloys in the x-p parameter space. If $p>x$ the magnetic atoms tend to form clusters, if $p=x$ the alloy is random, and if $p<x$ the magnetic atoms tend to stay apart from each other. For a lattice composed of two interpenetrating sublattices, such that all nearest neighbors of an atom in one sublattice lie in the other sublattice, the region $p<2-x^{-1}$, for $x\geq 0.5$, is inaccessible.

Eqs. (2.7) by replacing the real structure with its closed loops of atoms by a Bethe lattice without closed loops outside a finite cluster of atoms. Let us then consider a magnetic atom in the Bethe lattice. There is a single path connecting this atom with the central cluster. Hence, if a magnetic excitation, originating at the central cluster, reaches this atom, the path must be formed by magnetic atoms only. Indeed, the presence of a single nonmagnetic atom along the path is enough to block off the excitation, because of (2.12). Thus we distinguish two types of nearest neighbors for a magnetic atom in the Bethe lattice: the atom immediately preceding it and the $z-1$ atoms immediately following it. Obviously a necessary condition for a given magnetic atom to contribute to the solution of Eqs. (2.7) is that the atom immediately preceding it in the Bethe lattice be magnetic. Keeping the fact above in mind, and using the interpolation scheme of Ref. 6, we write for the magnetic atoms in the Bethe lattice which are connected with the central cluster:

$$E_B=2JS+2p(z-1)JS \qquad (2.13)$$

and

$$T(n,n-1)=-2JS,$$
$$T(n,n+1)=-2pJS, \qquad (2.14)$$

where the index n indicates the corresponding "generation" in the Bethe lattice. There are $z(z-1)^{n-1}$ atoms in that generation.

We observe that this approximation corresponds to saying that a relevant magnetic atom—i.e., one that contributes to the spin-wave spectrum of the central atom—has one magnetic atom preceding it in the tree and $p(z-1)$ magnetic atoms following it. In this respect, we have replaced a fluctuating number of magnetic neighbors in each following generation by an average number. This "mean-field" approximation is therefore a very good one when fluctuations are very small (p very close to 1) or when the number of magnetic neighbors is large, $p(z-1)>1$. The scheme is not very satisfactory when p is small and it gives its worst possible answers in the case of small z, in particular $z=2$, the linear chain. This is so because for small pz fluctuations will occur in the number of magnetic nearest neighbors such that the continuity of the tree will be broken. The finite, "island" character of the resulting structure differs drastically from the average Bethe lattice we use here.

In the following sections we apply the approximation above to the CBL calculation of the one-spin-wave local density of states (SW-LDOS) for several cases: a one-dimensional chain, where our approximation breaks down but where we can test how badly, and two three-dimensional cubic lattices, where our method should give sensible answers.

III. ONE-DIMENSIONAL DILUTE FERROMAGNET

In this section we discuss the one-dimensional dilute ferromagnet with nearest-neighbor interaction only. We also present the corresponding Bethe-lattice solution, based on the scheme proposed in Sec. II B. The Bethe-lattice solution is shown to be a very bad approximation, incapable of reproducing any one of the moments of the exact density of states, except near $p=1$. We argue that this result is directly connected with the absence of magnetic order, a fact which plays an important role in the interpretation of the CBL results for higher dimensionalities. We discuss briefly the connected problem of the critical temperature of the dilute magnetic system.

The one-dimensional dilute ferromagnet is formed by finite chains of magnetic atoms separated from each other by one or more nonmagnetic atoms. If we consider only nearest-neighbor interactions these finite chains do not interact with each other. The conditions for the existence of a ferromagnetically ordered ground state are clearly absent. The finite length and noninteraction of the chains prevents the formation of a long-range ordered state even at $T=0$. The application of an infinitesimal external magnetic field, at $T=0$, causes the spins in the various chains to align along the field direction. Under this circumstance we can then talk about "one-spin-wave" excitations of the system. It must be kept in mind, however, that these excitations can exist only as long as the external field is present.

Since the finite chains of magnetic atoms are isolated from each other, the spectrum of the one-dimensional dilute ferromagnet is the weighted average of the spectra of each finite chain. The probability that a magnetic atom belongs to a chain of n magnetic atoms is given by

$$P_n = x p^{n-1}(1-p)^2 . \tag{3.1}$$

The discrete spectrum of a chain of n atoms can be shown to be

$$\omega_n(\nu) = 4\sin^2(\pi\nu/2n), \quad 0 \leq \nu \leq n-1 \tag{3.2}$$

where we have taken $2JS=1$ and neglected the infinitesimal contribution due to the external field.

The density of one-spin-wave states can then be written down immediately:

$$\rho(\omega) = x(1-p)^2 \sum_{n=1}^{\infty} p^{n-1} \sum_{\nu=0}^{n-1} \delta(\omega - \omega_n(\nu)). \tag{3.3}$$

It is convenient to separate explicitly in (3.3) the contribution of the states for which $\omega_n=0$ ($\nu=0$). We find

$$\rho(\omega) = x(1-p)\delta(\omega) + x(1-p)^2 \sum_{n=2}^{\infty} p^{n-1} \sum_{\nu=1}^{n-1} \delta(\omega - \omega_n(\nu)). \tag{3.4}$$

We observe that the weight of the δ function at the origin vanishes for $p=1$. This δ function is connected with the finite size of the chains of magnetic atoms and hence its contribution disappears only when the chains become infinitely long. We notice that $p_c=1$ is the classical percolation limit in one dimension.

The Bethe-lattice SW-LDOS in one dimension is obtained by solving Eqs. (2.7), (2.13), and (2.14) for $z=2$. We obtain

$$\rho(\omega) = \begin{cases} x\dfrac{1-p}{1+p}\,\delta(\omega), & \omega \sim 0 \\ x\dfrac{1}{\pi}\,\dfrac{[4p-(\omega-1-p)^2]^{1/2}}{\omega[2(1+p)-\omega]}, & \omega_- \leq \omega \leq \omega_+ , \end{cases} \tag{3.5}$$

where

$$\omega_\pm = (1 \pm p^{1/2})^2 .$$

We observe that there is a δ-function contribution to the density of states, which vanishes for $p=1$. It is trivial to show that this density of states tends to the exact result as $p \to 1$, with the proper inverse-square-root singularities at the

TABLE I. Moments of the density of states $\langle(\omega-\bar{\omega})^s\rangle_{av}$ for a one-dimensional dilute ferromagnet.

s	Exact solution of a dilute lattice	Bethe solution of a dilute lattice	Perfect lattice $p=1$
0	$\langle\omega^0\rangle_{av}=xp$	$\langle\omega^0\rangle_{av}=x\frac{2p}{1+p}$	$\langle\omega^0\rangle_{av}=1$
1	$\bar{\omega}=2$	$\bar{\omega}=1+p$	$\bar{\omega}=2$
2	$2p$	$p(1+p)$	2
3	0	0	0
4	$4-2p+4p^2$	$p^2(2+3p+p^2)$	6
5	$-40(1-p)$	0	0
6	$240-238p+12p^2+6p^3$	$p^3(5+9p+5p^2+p^3)$	20
7	$-1120(1-p)$	0	0

band edges.

Comparing (3.4) and (3.5) we notice that the Bethe-lattice results yield the wrong weight for the contribution of the δ function at the origin—and, hence, also for the "band states" ($\omega>0$). In Table I we summarize the results for the weight, center of gravity, and a few lower moments of the density of states for the exact dilute, Bethe-lattice dilute, and perfect one-dimensional "ferromagnets." The following points are worth emphasizing: (i) Both the exact dilute and the Bethe-lattice dilute results tend to the results for the perfect ferromagnet as $x\to 1$, $p\to 1$; (ii) the exact dilute and perfect ferromagnet have the same density-of-states center of gravity, whereas the Bethe-lattice density of states has a center of gravity which shifts towards lower values as $p\to 0$; (iii) the Bethe-lattice and perfect ferromagnets have symmetric densities of states about their respective centers of gravity; (iv) the exact dilute ferromagnet has an asymmetric density of states, the states tending to pile up in the interval $0<\omega<2$ as $p\to 0$; (v) the Bethe lattice is a reasonable approximation to the exact result only near $p=1$.

That the Bethe lattice should be a bad approximation in one dimension may seem particularly surprising in view of the fact that it is a "one-dimensional" type of approximation. For the perfect ferromagnet, indeed, the Bethe lattice yields the same result as the exact calculation (the Bethe lattice *is* the exact calculation). However, for the dilute ferromagnet in one dimension we are always below the percolation limit, i.e., below the value of p such that an infinite chain can exist. This is indicated clearly by the presence of the δ-function contribution to the density of states at the origin. We take, at higher dimensionalities, the appearance of such a contribution—owing to finite isolated clusters of magnetic ions—as an indication of the breakdown of the Bethe-lattice or cluster–Bethe-lattice approximation. Indeed, we restrict ourselves later on only to values of $p>p_c=(z-1)^{-1}$, where z is the lattice coordination number, because for smaller values of p the δ function at the origin appears in the Bethe-lattice density of states. We can say that, for values of $p>p_c$, the critical temperature for the magnetic system is $T_c>0$. The critical temperature approaches zero as $p\to p_c$. In the case of a dilute Ising ferromagnet within the Bethe-lattice approximation, we can compute the transition temperature explicitly.[13] We find

$$k_B T_c = 2J\left[\ln\left(\frac{p(z-1)+1}{p(z-1)-1}\right)\right]^{-1}, \tag{3.6}$$

showing that $T_c(p=p_c)=0$.

IV. $(1+z)$-ATOM CLUSTERS: bcc LATTICE

In this section we apply the CBL formalism to the calculation of the SW-LDOS of a $(1+z)$-atom cluster in a body-centered-cubic configuration ($z=8$). Even in this simple case several interesting features of the method will become apparent.

Our system consists now of a $(1+z)$-site cluster with a central magnetic ion 0 connected to z atoms, ν of which are also magnetic ($\nu=0$ through z). The bonds of each one of these ν magnetic atoms are saturated with Bethe lattices of coordination number z and probability p.

The equations of motion reduce in this case to

$$\begin{aligned}(\omega-E_0^{(\nu)})G_{00}^{(\nu)}(\omega)&=1-\nu T G_{10}^{(\nu)}(\omega),\\ (\omega-E_n)G_{n0}^{(\nu)}(\omega)&=-TG_{(n-1)0}^{(\nu)}\\ &\quad -p(z-1)TG_{(N+1)0}^{(\nu)},\quad n\geq 1\end{aligned} \tag{4.1}$$

where

$$T = 2JS = 1\,, \quad E_0^{(\nu)} = \nu T$$
$$E_n = T + p(z-1)T\,, \quad n \geq 1\,. \tag{4.2}$$

We can solve Eqs. (4.1) for $G_{00}^{(\nu)}(\omega)$ by considering a transfer function defined by

$$\phi(\omega) = G_{n0}^{(\nu)}(\omega)/G_{(n-1)0}^{(\nu)}(\omega)\,, \quad n \geq 1\,. \tag{4.3}$$

Substituting this expression back in Eq. (4.1) we obtain

$$G_{00}^{(\nu)} = 1/[\omega - \nu + \nu\phi(\omega)]\,, \tag{4.4}$$

where

$$\phi(\omega) = \frac{(E_1 - \omega) \pm [(\omega - E_1)^2 - 4p(z-1)]^{1/2}}{2p(z-1)}\,. \tag{4.5}$$

The density of states is obtained by averaging over the clusters with different composition,

$$\rho(\omega) = x \sum_{\nu=0}^{\infty} P_\nu \left(-\frac{1}{\pi}\,\mathrm{Im} G_{00}^{(\nu)}(\omega)\right), \tag{4.6}$$

where the weights P_ν follow a binomial distribution

$$P_\nu = \frac{8!}{\nu!(8-\nu)!}\, p^\nu (1-p)^{8-\nu}\,. \tag{4.7}$$

The ambiguity in the double sign in Eq. (4.5) is solved by requiring the density of states to be positive and the Green's function to have the proper asymptotic behavior for large $|\omega|$.

The results obtained for values of p equal to 0.25, 0.50, 0.75, and 1.0 are presented in Fig. 2. In all cases we obtain a continuous band of states corresponding to those values of ω for which the square root in Eq. (4.5) is negative. The band edges are given by

$$\omega_{BE} = \{1 \pm [p(z-1)]^{1/2}\}^2\,. \tag{4.8}$$

Localized states corresponding to poles of the Green's function appear on both sides of the continuous band. A study of the location of these poles shows that in the low frequency side they appear at frequencies given by

$$\omega_\nu = \nu\left(1 + \frac{1}{\nu - p(z-1)}\right) \tag{4.9}$$

for $0 \leq \nu < \{p(z-1) - [p(z-1)]^{1/2}\}$. The cluster with $\nu = 0$ which corresponds to the isolated central magnetic ion contributes with a mode of zero frequency with residue equal to $(1-p)^8$.

This mode *does not* imply instability of the ferromagnetic system: It only corresponds to the zero energy required to flip a spin of an isolated atom, one that is disconnected from the "bulk" ferromagnet.

In the high-frequency side localized modes appear owing to the contribution of configurations with a large number of magnetic ions. The frequencies at which they occur are given by Eq. (4.9) for $\nu > \{p(z-1) + [p(z-1)]^{1/2}\}$. For values of p lower than the critical value $p_c = (z-1)^{-1}$ there appears an extra pole at the origin which gives a contribution to the LDOS equal to

$$\rho_0(\omega = 0) \sim \frac{1 - p(z-1)}{1+p}\,\delta(\omega)\,. \tag{4.10}$$

This mode with zero energy evidences an instability of the ferromagnetic state, i.e., a "breakdown" of the ferromagnet into isolated, disconnected "islands." This result is in agreement with percolation theory, according to which for values of $p < p_c$ no infinite clusters of magnetic ions exist and consequently the ferromagnetic phase transition cannot occur.

In Sec. V we extend the application to the case of more complicated clusters with more involved topological features.

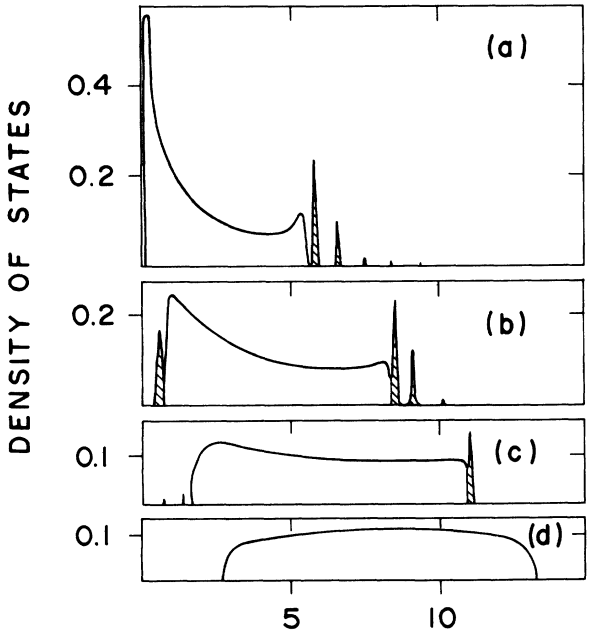

FIG. 2. Local density of one-spin-wave states for a nine-atom cluster in the bcc lattice. The values of p are (a) 0.25, (b) 0.50, (c) 0.75, and (d) 1.0. δ-function peaks in the density of states are indicated by broadened shaded peaks. The density of states is normalized to 1.

V. HIGH-ORDER CLUSTERS: SIMPLE-CUBIC LATTICE

The close relation between ring statistics in the lattice and the structure of the local electronic density of states in perfectly ordered solids has been amply demonstrated in the literature.[8] For alloys, however, even when the overall shape of the LDOS is determined by the local environment,

the correlation with closed-ring topology is apparently not so marked.[6] In order to study this effect it is necessary to go to clusters of higher order than the ones considered so far. We present in the following the results obtained for the SW-LDOS in a dilute ferromagnet with a simple-cubic structure.

To apply the CBL method to the study of these alloys a cluster of 27 atoms was isolated from the material as illustrated in Fig. 3. This cluster contains first, second, and third nearest neighbors of a central magnetic atom. This central atom is labeled with the number zero. Atoms labeled 1–6 have one dangling bond, while atoms 7–18 and 19–27 have two and three dangling bonds, respectively. A Bethe lattice of coordination number 6 is connected to each of these bonds.

Clusters of 19 atoms containing only first and second nearest neighbors and clusters of seven atoms containing only first nearest neighbors were derived from the original 27-atom cluster by removing the appropriate atoms. The different size clusters are used to illustrate the evolution of the SW-LDOS with increasing size of the cluster.

For a cluster with a given distribution of magnetic ions the density of states at the central magnetic ion is obtained using the procedure outlined below. For each frequency the Bethe lattice is solved using a transfer-function method and the problem is reduced to the inversion of a $\nu \times \nu$ matrix, ν being the number of magnetic ions in the cluster. The difficulty that appears, however, is that even in the case of the clusters of only 19 atoms, the number of topologically nonequivalent configurations that needs to be considered and properly weighted is too large to be handled within reasonable computing times. To overcome this problem, a method similar to that employed by Falicov and Yndurain[6] was used.

A set of 200 clusters of 27 atoms is generated using random numbers and weights compatible with the concentration x of the magnetic ions in the material. These clusters are assumed to give an adequate sampling of all clusters in the alloy. Let N_ν be the number of clusters in our sampling composed of a central magnetic ion and exactly ν additional magnetic atoms ($\nu = 0, 1, \ldots, 26$). The number N_ν is defined as

$$N_\nu = \mathrm{Int}[200 W(\nu)], \tag{5.1}$$

where $\mathrm{Int}(x)$ is the integer nearest to x and

$$W(\nu) = \frac{26!}{\nu!(26-\nu)!} x^\nu (1-x)^{26-\nu}. \tag{5.2}$$

In our case we take $x = 0.5$ and clusters are obtained with a number of magnetic ions ranging from 7–21. A value of p is then attributed to each cluster by counting the number of bonds between magnetic nearest neighbors. In Fig. 4 we present a histogram which shows the distribution of clusters with p. This distribution is centered about $p = 0.5$ and shows the theoretical Gaussian trend. In the case of the 19- and seven-atom clusters, the same value of p was attributed to them as the value found for the 27-atom cluster from which they are derived.

To show how the local configuration determines the overall shape of the density of states, three out of the 200 clusters are singled out and their SW-LDOS are presented in Fig. 5. They correspond to values of the probability equal to $p = 0.33$, $p = 0.5$, and $p = 0.63$.

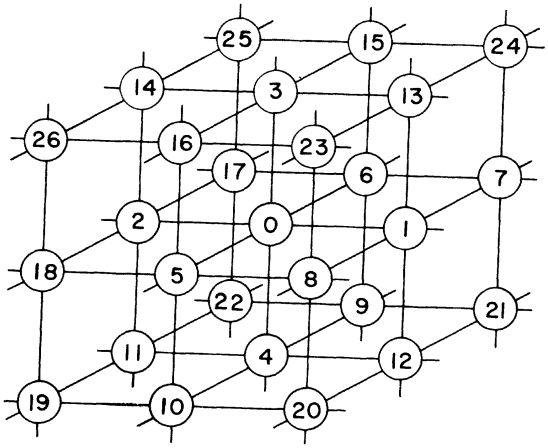

FIG. 3. 27-atom cluster including first (1–6), second (7–18), and third (19–22) nearest neighbors in the simple-cubic structure. Bethe lattices of the type described in the text are attached to the "dangling bonds" of the atoms lying at the surface of the cube.

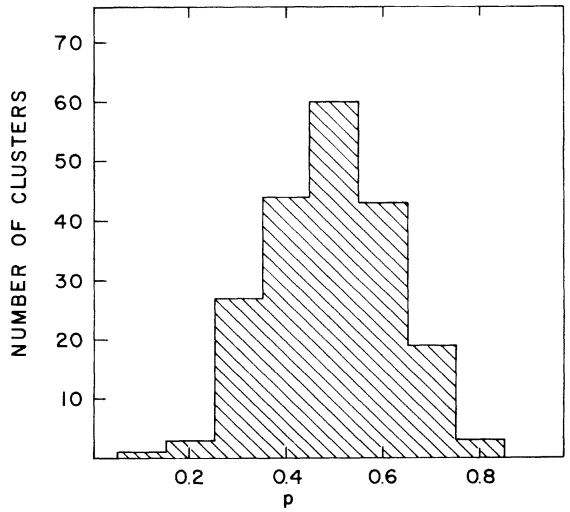

FIG. 4. Histogram showing the distribution of p values for the 200 randomly selected 27-atom clusters in the simple-cubic lattice. The parameter $x = 0.5$.

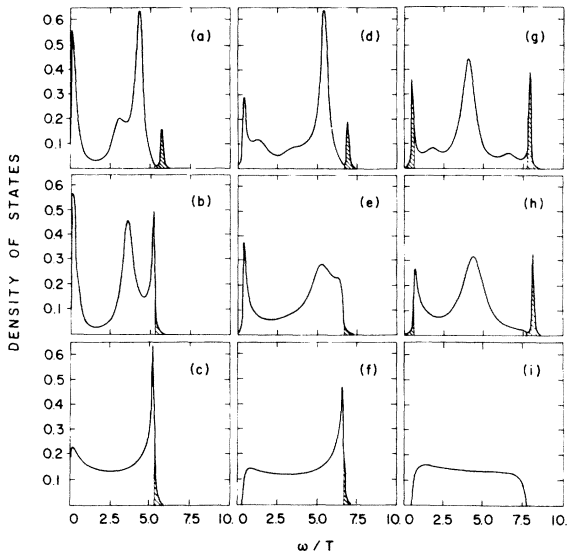

FIG. 5. Local density of one-spin-wave states for three particular clusters. The value of the parameter p increases from left to right: $p=0.33$ (a)–(c), $p=0.50$ (d)–(f), and $p=0.63$ (g)–(i). The size of the cluster increases from bottom to top: seven-atom cluster (c), (f), (i); 19-atom cluster (b), (e), (h); and 27-atom cluster (a), (d), (g). See text for details of the clusters.

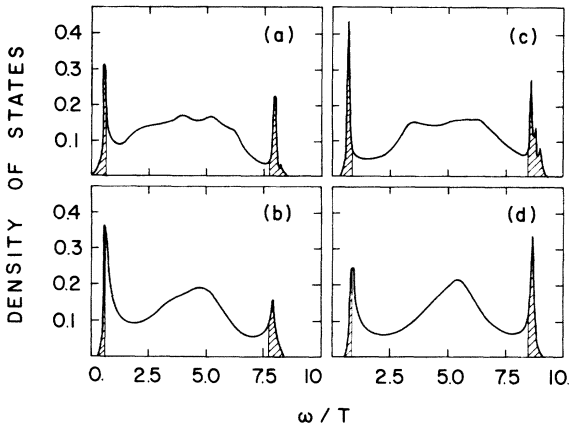

FIG. 6. Restricted-p-average local density of one-spin-wave states. The value of p increases from left to right: $p=0.50$ (a), (b) and $p=0.63$ (c), (d). The size of the cluster increases from bottom to top: 19-atom cluster (b), (d) and 27-atom cluster (a), (c).

The results for cluster I are shown in Figs. 5(a)–5(c). This cluster is composed of three first nearest, three second nearest, and seven third nearest magnetic neighbors. Three out of the latter seven atoms are isolated; i.e., no path of magnetic atoms links them to the central one and consequently they do not contribute to the local density of states at the central atom. No complete rings involving the central atom are present in this configuration. The calculated spectra show that localized states appear in the high-frequency side of the band only for the cluster of 27 atoms, although strong peaks showing in the right edge of the band in Fig. 5(b) and 5(c) seem ready to split off the band.

A strong peak in the edge of the low-energy side of the band is present in the three clusters. A comparison of Figs. 5(a)–5(c) shows a gradual complication of the spectrum, with a wide central peak in the 19-atom cluster that unfolds into two peaks in the 27-atom cluster.

These features are more or less reproduced for cluster II, Figs. 5(d)–5(f). Here, 14 out of the 27 atoms are magnetic although again two of them are isolated. In this configuration one closed four-ring involving the central atom is present.

Cluster III corresponds to the value of $p=0.63$, and the LDOS is shown in Figs. 5(g)–5(i). In this configuration there are four nearest neighbor, seven second nearest, and only two third nearest neighbors of the central atom. This makes the spectra of the 27-atom cluster and the 19-atom cluster very similar, both with a wide central peak, the 27-atom cluster showing additional undulations and a completely split-off mode in the low-energy side. It is interesting to note here that four closed four-rings are present in this configuration. The strong central peak in Figs. 5(g) and 5(h) is compatible with the results of a calculation of the eigenvalues of a closed isolated four-ring. In this case the spectrum consists of two coinciding δ functions located at the center of the permissible energy range plus two additional δ functions, one at each extreme of the range. This trend is apparently maintained in our example.

In Fig. 6 we show restricted averages over all clusters in our 200-cluster sample for which $p=0.5$ and $p=0.63$, and for 29- and 17-atom clusters. They are characteristic SW-LDOS for dilute ferromagnets in which there is a short-range correlation measured by the parameter p. Localized states appear on both high- and low-frequency sides of the continuum band. Some of these, which are very close to each other in frequency, are not resolved in the figure. This is shown by a broadening of the Lorentzian peaks used to represent the δ functions throughout this work.

VI. CONCLUSIONS

The theory presented here as well as the examples of Secs. IV and V clearly point out the suitability of the CBL method for studying disordered magnetic systems. In particular, the method gives a very sensible description of localized magnetic excitations, which appear as δ-function-type anomalies in the local density of states. These anomalies are due to (a) isolated magnetic clust-

ers, which give *only* localized modes including one zero-frequency mode, and to (b) local excitations within the "bulk" ferromagnet which do not propagate beyond a few atoms owing to fluctuations in the local configurations.

Our method shows definite advantages over the coherent-potential approximation, since it naturally treats on the same footing diagonal and off-diagonal disorders, which for disordered magnetic systems cannot be sensibly separated. Since it includes short-range correlations, as parametrized in our case by the quantity p, it permits us to distinguish between different kinds of solids and to determine the attendant changes in localized and extended spectral modes.

Our theory can be extended to study properties other than local density of states. In particular, we have in mind determination of neutron scattering form factors, as given by (2.10), and also thermodynamic properties which include, e.g., temperature dependence of the average magnetization.

These studies can also be extended to other types of magnetic systems. For example, antiferromagnets, amorphous magnets, even mixed valence solids can be tackled with a CBL approach.

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