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Cluster-Bethe-Lattice Study of a Planar Antiferromagnet: Rb_2NiF_4*

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We present a discussion of the Cluster-Bethe-Lattice method for a planar antiferromagnet for which the hamiltonian parameters are known and the one-magnon density of states may be computed exactly. We study all the square clusters of 1 to 121 atoms, both connected to and isolated from the Bethe lattices. We show that, even for the largest cluster treated, the approximation is still far from the exact result. We discuss the liniitations of the method.

Apresentamos uma discussão do método "Cluster-Bethe- Lattice" para um antiferromagneto planar, para o qual os parâmetros do hamiltoniano são conhecidos e a densidade de estados de um-magnon pode ser calculada exatamente. Estudamos todos os núcleos centrais quadrados contendo de 1 até 121 átomos, tanto ligados quanto isolados das redes de Bethe. Mostramos que, mesmo para o núcleo central de 121 átomos, a aproximação ainda se encontra longe do resultado exato. As limitações do método são discutidas.

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1. INTRODUCTION

The Cluster-Bethe-Lattice (CBL) method has been extensively used recently to study amorphous semiconductors[1,2], metallic binary alloys[3,4], impurity centers in ionic crystals[5], phonons[6] and magnons [7,8] in disordered structures, and also charge transfer effects in disordered binary substitutional alloys[9]. The objeçtive of the present work is to study the problem of convergence of the CBL method in a simple, yet quite realistic system. Although we consider an application to magnetic systems, the conclusions which we reach are quite general.

The basic idea of the CBL method is to isolate in a physical system a finite cluster of atoms and attach to the "dangling" bonds of this cluster an infinite Cayley tree, or Bethe lattice, of the same coordination number as the envisaged crystal structure. The Cayley tree is a structure in which, each atom has the same of nearest neighbors, but there are no closed rings of bonds, i.e., between any two atoms in the lattice there is one and only one connecting path. It is thus, as far as the cluster boundary conditions are concerned, a quasi one-dimensional approximation. We can see irnmediately from this description that the method is appropriate for the discussion of local properties, although unlike finite clusters, the boundary conditions give the system some of the properties of an infinite structure. For instance, the one-particle Green's function has always a branch cut indicating the existente of a continuum of excitations. The approach to the real physical system follows from taking clusters of a progressively larger size. However, in the usual three dimensional clusters which have been investigated the number of atoms N_c in the cluster varies very rapidly with cluster radius $R_c (N_c \propto R_c^3)$, so that we are limited, for cornputational reasons, to clusters of rather small dimensions. We have thus decided to look at planar bi-dimensional systems, such as the antiferromagnet Rb_2NiF_4, for which we can take clusters of fairly large radii and yet keep the number N_c within a manageable interval.

In Section 2(a) we review briefly the system we want to investigate and set up the hamiltonian. In Section 2(b) we construct the CBL equations. The results for the local density of one-magnon states are

presented and discussed in Section 3. Finally, in Section 4 we present our conclusions.

2. THE HAIMILTONIAN AND THE CBL EQUATIONS

(a) The Hamiltonian

The transition metal tetrafluoride Rb_2NiF_4 is member of a large family of compounds - A_2TF_4, where A is an alkali and T a transition metal - which have the tetragonal iayered strcuture. In Rb_2NiF_4, the Ni atoms are placed on square lattices, with lattice parameter 4.087 A, stacked up with an interlayer distance of 13.67.A. The exchange constants for the coupling of spins within the same layer are about six orders of magnitude larger than those for the coupling between different layers[10]. Hence we may think of this system as a quasi bi-dimensional one. The spin Hamiltonian may be written:

$$H = \sum_{m,\Delta,\alpha} J(m,m+\Delta)\, \vec{S}(m,\alpha)\cdot S(m+\Delta,\alpha) - 2\mu_0 H_A \sum_{\alpha} \alpha\, S^z(m,\alpha) \qquad (2.1)$$

In (2.1), m labels the primitive cells of the magnetic structure - we are dealing with an antiferromagnet with two ions per primitive magnetic cell - ; a = ±1 labels the magnetic sublattices; $J(m,m+\Delta)$ is the exchange constant for the coupling of nearest neighbors spins only; and H_A is the anisotropy field. The values of the parameters whichenter the hamiltonian (2.1) for Rb_2NiF_4 are quite well-known. It is found that J = 4.15 MeV for nearest neighbors and J=O otherwise, S=I and $2\mu_0 H_A$ = = 0.28 MeV [11]. Since the anisotropy energy is so small we take, in the numerical calculations discussed below, H_A = 0.

(b) The CBL Equations

The CBL method as applied to magnetic systems has been discussed in detail elsewhere[7]. For this reason, we only summarize those results which are important for treating an antiferromagnet.

The one-magnon Green's function is defined by:

$$G^{\alpha\beta}_{mn}(\omega) = (-1)^{\frac{1}{2}(1-\alpha)} (2S)^{-1} \langle\langle S^{+}(m\alpha) | S^{-}(n\beta) \rangle\rangle \qquad (2.2)$$

and its equation of motion at $T=0$, assuming the Néel state as the ground state, is:

$$\{\omega - E(a)\} G^{\alpha\beta}_{mn}(\omega) = \delta_{mn}\,\delta_{\alpha\beta} + \sum_{A} T(m\alpha, m+\Delta\bar{\alpha})\, G^{\bar{\alpha}\beta}_{m+\Delta,n}(\omega) \qquad (2.3)$$

where the summation over h runs over primitive cells of the magnetic lattice and not over atoms. In (2.3), $\bar{\alpha} = -\alpha$ and:

$$E(\alpha) = 8\alpha JS + 2\alpha\mu_0 H_A \equiv \alpha E \ , \qquad (2.4)$$

$$T(m\alpha, m+\Delta\alpha) = \begin{cases} (-1)^{(1+\alpha)} 2JS & \text{for nearest neighbors} \\ 0 & \text{otherwise .} \end{cases} \qquad (2.5)$$

To illustrate the application of the method, we consider the cluster formed by a single spin attached to four Bethe lattices. If the central atom has spin up ($a = +$) we obtain:

$$\begin{aligned} (\omega - E)\, G^{++}_{00} &= 1 - 4\, W\, G^{-+}_{10} \\ (\omega - E) G^{-+}_{2n+1,0} &= W\, G^{++}_{2n,0} + 3W\, G^{++}_{2n+2,0} \\ (\omega - E)\, G^{++}_{2n+2,0} &= -W\, G^{-+}_{2n+1,0} - 3\, W\, G^{-+}_{2n+3,0} \end{aligned} \qquad (2.6)$$

for $n \geq 0$, where $W = 2JS$.

These equations are similar to the ones obtained by Yndurain and Joannopoulos for heteropolar compounds[12], except that the sign of the overlap integral alternates from one equation to the next in the hierarchy above. An equivalent set of equations is obtained for the case of a central spin down atom. The solution of (2.6) requires the introduction of two transfer functions, defined by:

$$\begin{aligned} G^{-+}_{2n+1,0} &= \phi^{-+}\, G^{++}_{2n,0} \\ G^{++}_{2n+2,0} &= \phi^{+-}\, G^{-+}_{2n+1,0} \end{aligned} \qquad (2.7)$$

which are formally related by:

$$(\omega + E)\ \phi^{-+} = -\ (\omega - E)\ \$^{+-} \qquad (2.a)$$

The final result for the density of states is:

$$\rho(\omega) = -\pi^{-1}\ \mathrm{Im}\{G_{00}^{++}(\omega+i0^{+}) + G_{00}^{--}(\omega+i0^{+})\} =$$

$$= \frac{4}{\pi|\omega|}\ \mathrm{i}\ \frac{\omega^2 - 4W^2}{16W^2 - \omega^2}\ \}, \quad \text{for} \quad 2W \leq |\omega| \leq 4W\ . \qquad (2.9)$$

This is plotted in Figure 1.

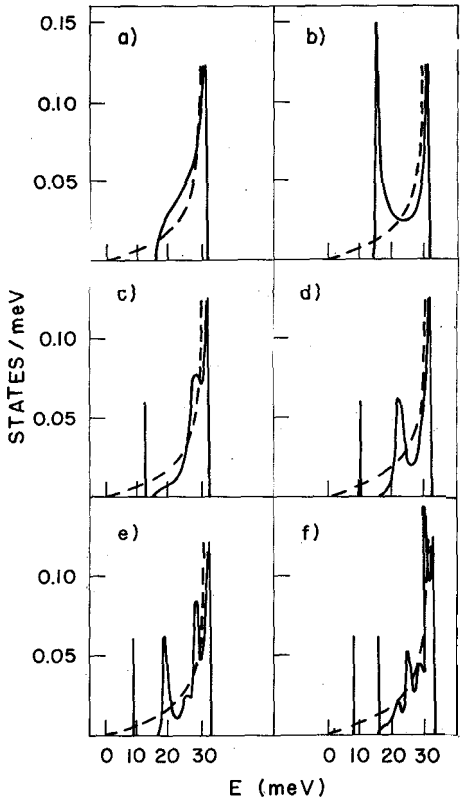

Fig.1 - The exact (dashed line) and CBL (full line) densities of one-magnon states for the antiferromagnet Rb_2NiF_4. The cluster size is (a) N = 1; (b) N_c = 9; (c) N_c = 25; (d) N_c = 49; (e) N_c = 81; and (f) N_c = 121.

3. RESULTS

(a) Exact Density of States

The exact local density of one-magnon states is easily computed for the hamiltonian discussed in Section 2(a). The calculation proceeds along standard lines. It is easily shown that for a square lattice the magnon dispersion relation is given by[13]:

$$\omega(\vec{q}) = W \sqrt{1 - \gamma^2(\vec{q})} \quad , \tag{3.1}$$

where:

$$\gamma(\vec{q}) = \cos\left(\frac{q_x a}{\sqrt{2}}\right) \cos\left(\frac{q_y a}{\sqrt{2}}\right) \tag{3.2}$$

In (3.2), a is the conventional lattice parameter of the square lattice. The density of states is given by:

$$\rho(\omega) = 2N^{-1} \sum_{q} \delta\{\omega - \omega(\vec{q})\} \quad , \tag{3.3}$$

where N is the number of magnetic primitive cells. Equation (3.2) may be rewritten:

$$\rho(\omega) = 4\pi^{-2} \int_0^1 dx \int_0^1 dy \{(1-x^2)(1-y^2)\}^{-1/2} \delta(\omega - W\sqrt{1-x^2y^2}) \tag{3.4}$$

where

$$x = \frac{q_x a}{\sqrt{2}} \quad , \quad y = \frac{q_y a}{\sqrt{2}}$$

This yields an elliptic integral:

$$\rho(\omega) = \frac{8|\omega|}{\pi^2 \sqrt{W^2 - \omega^2}\,(W + \sqrt{W^2 - \omega^2})} K\left\{\frac{W - \sqrt{W^2-\omega^2}}{W + \sqrt{W^2-\omega^2}}\right\} \tag{3.5}$$

where $K(q)$ is the complete elliptic function of second kind.

The sum rule:

$$\int_0^W \rho(\omega)\, d\omega = 1 \,. \qquad (3.6)$$

is easily checked.

The analytic expression for $\rho(\omega)$ is plotted in Figure 1, where it is compared to the approximate CBL results.

(b) CBL Density of States

In this Section we consider six different square clusters containing 1, 9, 25, 49, 81 and 121 atoms. The calculation is performed both for isolated clusters and for clusters connected to Bethe lattices in the usual way. In the former case, the boundary conditions consistin setting the transfer functions connecting the cluster to the Bethe lattice equal to zero. Hence we leave the diagonal matrix elements of the hamiltonian unchanged even for the boundary atoms. These boundary conditions are not physical for a magnetic system, but they allow for an imnediate analysis of the effect of the Bethe lattices upon the finite clusters.

In Table 1 we present the results for the spectrurn of excitations, i.e., position and residues of the poles of $Tr\, G_{00}$, for the isolated clusters. The following points are worth remarking: (i) the residues are simple rational numbers; (ii) as expected, the number of poles increases and they spread out in energy as N_c increases; (iii) the maximum excitation energy is an accumulation point of the spectrum as $N_c \to \infty$, in accordance with the divergence which is found is the exact result at the same energy.

In Figure 1 we show the results for the density of states with attached Bethe lattices. Also shown for comparison is the exact result. There are two important features of the CBL method which show up quite clearly in this Figure. First, the well-known problem of the reduced band-width. For the particular situation studied here the CBL band

TABLE 1

1	9	25	49	81	121
33.200 1/1	33.200 1/2	33.200 1/3	33.200 1/4	33.200 1/5	33.200 1/6
	23.476 1/2	29.926 4/9	31.961 1/4	32.646 4/25	32.921 1/9
		16.600 2/9	30.673 1/8	31.735 4/25	32.355 1/9
			25.136 1/4	29.206 4/25	32.069 1/18
			12.705 1/8	26.859 2/25	31.056 1/9
				21.206 4/25	29.071 1/9
				10.259 2/25	26.247 1/9
					23.476 1/18
					18.192 1/9
					8.593 1/18

Table 1 - Position and residues of the poles of $Tr\, G_{00}$ for the various isolated clusters. At the top of each column is given the number N_c of atoms of the cluster. For each entry, the top number is the position of the pole (in MeV) and the bottom number is the respective residue. For boundary conditions, see text.

-width is only half of the exact one . Second, the appearance of delta function singularities outside the continuum and the oscillations ofthe continuum density as the cluster size increases. The existence of a branch cut in the Green's function is a consequenceofthe thermodynamic limit. The fact that we obtain the wrong branch cut - wrong band-width - is just anwther indication of the problems connected with this limit in the CBL approximation[14]. By artificially increasing the overlap matrix elements in the Bethe lattice it is possible to reproduce the correct band edges for electronic systems. The artificiality of such a procedure is evident, however, when dealing with magnetic systems. Increasing the exchange constant in the Bethe lattice changes both the band edges and the center of the band, so that agreement with the exact result is not improvad. This is a serious limitation of the method that must be kept in mind. The localized states which appear below the edge of the continuum are given in Table 2. As expected, we can establish a correspondence between the poles of $Tr\ G_{00}$ for the isolated cluster and the poles of $Tr\ G_{00}$ for the cluster connected to the Bethe lattices. The overall eff'ect of the continuum is to push down in energy the poles of the isolated cluster. Delta function singularities in the density of states of the cluster plus Bethe lattices show up only when the isolated cluster size is large enough for poles to appear near toor belowthe

TABLE 2

25	26	81	121
13.86 0.14	11.18 0.10	9.28 0.07	16.36 0.06
			7.91 0.05

Table 2 - Position and residues of the poles of $Tr\ G_{00}$ for the clusters connected to the Bethe lattices. At the top of each column is given the number N_c of atoms in the cluster. For each entry, the top number is the position of the pole (in MeV) and the bottom number is the respective residue.

lower continuum band edge. The oscillations in the density of states shown in Figure 1 are also connected to the poles of $Tr\, G_{00}$ forthe isolated cluster. It is noteworthy that, even for the largest cluster studied, they still have appreciably amplitude.

4. CONCLUSIONS

The results presented above indicate that the convergence of the CBL approxirnation to the exact result, inthe case of an ordered solid, as a function of cluster size is quite slow. This is not surprising, since we are, for instance, trying to generate part of the branch cut of the Green's function from a series of isolated singularities (poles). It is interesting to speculate to what extent the approximation id adequate for disordered solids, i.e., alloys and amorphouscornpounds, for which long range order is absent. Since thedetailed structure ofthe density of states depends upon the cluster size strongly, it is clear that not much importance can be attached to it, atleast before the cluster size increases appreciably.

In isotropic magnetic systems, like the one discussedin this paper, the presence of a gap in the spectrum of one-magnon excitations is a clear indication of the shortcornings of the approximation. It means that no excitation can propagate throughout the whole Bethe lattice with energy less than a certain minimum, whereas it is known that the spectrum must extend down to zero. Hence, although the method may be useful as a first approach to many problems for which no better solutions are generally known, conclusions drawn from it must be handled with proper caution.

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