Excitations in Dilute Magnets Using the Coherent-Potential Approximation

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A study of the excitations in randomly dilute Heisenberg ferromagnets and antiferromagnets using the coherent-potential approximation (CPA) is summarized. It is pointed out that Tahir-Kheli's recent work corresponds to the use of the CPA in a model where isolated bonds are cut at random. This model gives unsatisfactory features if the neutron-scattering spectra are calculated. An attempt to include the full scattering by a nonmagnetic impurity in the CPA is described. It also leads to an unsatisfactory description at low frequencies.

In two recent papers[1,2] Tahir-Kheli (T-K) has discussed an approximate treatment of spin waves in dilute ferromagnets and antiferromagnets. In the course of some recent work on the interpretation of experimental results on excitations in disordered antiferromagnets,[3,4] we have also considered a similar approximation to that used by T-K. The purpose of this note is to briefly report a somewhat different derivation of his main results using the coherent-potential approximation (CPA) and to give further details of certain properties. In particular, we calculate the general response function $G(\vec{k}, \omega)$ associated with neutron scattering and show that the results are not those one would expect for a dilute magnet on physical grounds. We also briefly report our attempts to use the CPA, including the full scattering from nonmagnetic sites, which have also proved unsatisfactory in some ways.

The results of T-K may be obtained by considering a ferromagnetic crystal in which a concentration $c = 1 - m$ of the nearest-neighbor exchange interactions (bonds) is removed at random. The Hamiltonian for the ferromagnet is

$$\mathcal{H} = -\sum_{mn}{}' J\eta_{mn}\vec{S}_m \cdot \vec{S}_n , \tag{1}$$

where the sum extends over all nearest-neighbor pairs (mn), and η_{mn} takes the value 1 if the bond (mn) is intact, and 0 if it is cut. The spin-wave Green's function for this Hamiltonian

$$G_{ll'} = \langle\langle S_l^+, S_{l'}^- \rangle\rangle (S_l S_{l'})^{-1/2} \tag{2a}$$

satisfies a Dyson equation

$$\underline{G} = \underline{g} + \underline{g}\,\underline{V}\,\underline{G} , \tag{2b}$$

where $\underline{g}$ is the Green's function in the lattice with all bonds intact, and $\underline{V}$ is the matrix describing the scattering of spin waves by the cut bonds. Each missing bond produces a 2×2 block in $\underline{V}$ of the form

$$\underline{v} = 2JS\begin{bmatrix} -1 & 1 \\ 1 & -1 \end{bmatrix} . \tag{3}$$

We neglect the overlap of the matrices of two or more adjacent cut bonds.

In order to find the configurationally averaged Green's function $\langle G \rangle$, we employ the coherent-potential approximation[5] (CPA). Takeno[6] has shown that this may be used in cases where one defect produces an extended block of elements in $\underline{V}$; the self-energy $\underline{\Sigma}$ is then written as a sum over all sites

$$\underline{\Sigma} = \sum_n \underline{w}_n , \tag{4}$$

where $\underline{w}_n$ has a small block of nonzero elements given by

$$\underline{w} = c\underline{v}[\underline{1} - \underline{G}_0(\underline{v} - \underline{w})]^{-1} , \tag{5}$$

and $\underline{G}_0$ is the appropriate block of $\langle G \rangle$. The CPA has also been used for bond problems by Blackman *et al.*[7] In our case $\underline{v}$ is given by (3), and

$$\underline{G}_0 = \begin{bmatrix} G_{00} & G_{01} \\ G_{01} & G_{00} \end{bmatrix} , \tag{6}$$

where G_{01} links nearest-neighbor sites. Since $\underline{v}$ is a singular matrix with equal off-diagonal elements, the only possible form of solution for (5) is

$$\underline{w} = \epsilon\underline{v} , \tag{7}$$

where ϵ is a scalar function of energy. Putting this into (5), we obtain a scalar equation

$$\epsilon = c[1 - 4JS(1 - \epsilon)(G_{01} - G_{00})]^{-1} . \tag{8}$$

As the denominator of $G(\vec{k}, \omega)$ is now

$$G(\vec{k}, \omega)^{-1} = \omega - 2JSz(1 - \epsilon)(1 - \gamma_{\vec{k}}) , \tag{9}$$

we may identify $J(1 - \epsilon)$ with T-K's $\tilde{J}$, and (8) is identical, after some manipulation, to Eq. (7.15) of Ref. 1.

It is clear that T-K's result corresponds to neglecting the correlations between scattering processes from the z bonds of one nonmagnetic defect. This coherent scattering must be treated properly if the spin waves are to be adequately described. In fact, he has treated an approximation to the "bond" problem in percolation theory rather

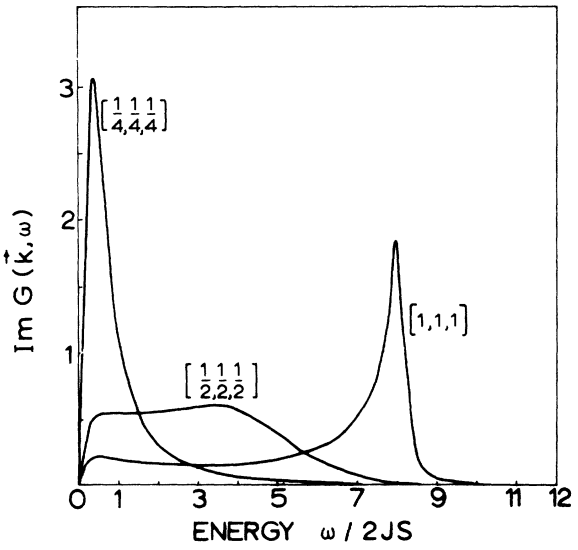

FIG. 1. Response function $\mathrm{Im}G(\vec{k}, \omega)$ for the simple cubic ferromagnet with $m = 0.5$ at three points in $\vec{k}$ space.

than the "site" problem which is appropriate to a dilute magnet. The critical concentration is quite different in these two problems.[8] Moreover, the low-c limit of this theory differs from the correct result obtained by Iziumov.[9]

T-K concentrated his attention on the density of states, but clearer evidence of the unsatisfactory nature of the model is found if it is used to predict the neutron scattering spectra and spin-wave dispersion relations. Figure 1 shows the response function $\mathrm{Im}G(\vec{k}, \omega)$, given by Eq. (9), for the simple cubic ferromagnet with $m = 0.5$ at three points in the first Brillouin zone. For small $\vec{k}$, the peak is as we should expect, but for $\vec{k}$ well away from the zone center an unphysical tail appears, with a subsidiary peak which remains at almost constant energy. This tail is responsible for the low-energy peak in the density of states. Figure 2 shows dispersion curves deduced from the maxima in $\mathrm{Im}G(\vec{k}, \omega)$. The spin-wave curve below $\zeta = 0.5$ is progressively distorted until the second peak appears, shown by the broken lines. The fall in the position of the top of the dispersion curve is proportional to $1 - m$ but is much smaller than we should expect: for $m = 0.5$ the maximum $\omega(\vec{k})$ is 66% of the pure-crystal value.

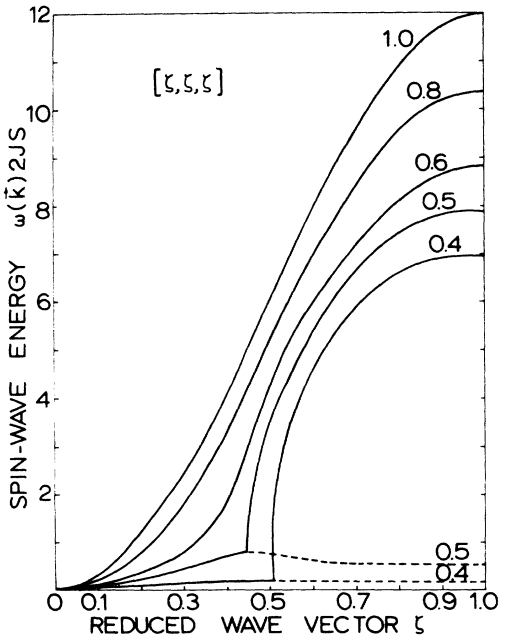

FIG. 2. Spin-wave dispersion curves for the simple cubic ferromagnet with the values of m indicated. The broken lines show the position of the low-energy hump in $\mathrm{Im}G(\vec{k}, \omega)$.

In the case of a two-sublattice antiferromagnet, the Green's functions of both the perfect and imperfect crystal depend on the sublattice. $\langle G \rangle$ and $\underline{g}$ can be written as 2×2 matrices. If the Néel ground state is assumed, (3) is replaced by

$$\underline{v} = 2JS\begin{bmatrix} -1 & -1 \\ 1 & 1 \end{bmatrix} \tag{10}$$

and (6) by

$$\underline{G}_0 = \begin{bmatrix} G_{00}^{++} & G_{01}^{+-} \\ G_{01}^{-+} & G_{11}^{--} \end{bmatrix} . \tag{11}$$

Substituting these into (5) and (7) gives

$$\epsilon = c[1 - 2JS(1-\epsilon)(G_{01}^{+-} - G_{01}^{-+} - G_{00}^{++} + G_{00}^{--})]^{-1} \tag{12}$$

and

$$G_{00} = \begin{bmatrix} G_{00}^{++} & G_{00}^{+-} \\ G_{00}^{-+} & G_{00}^{--} \end{bmatrix}$$

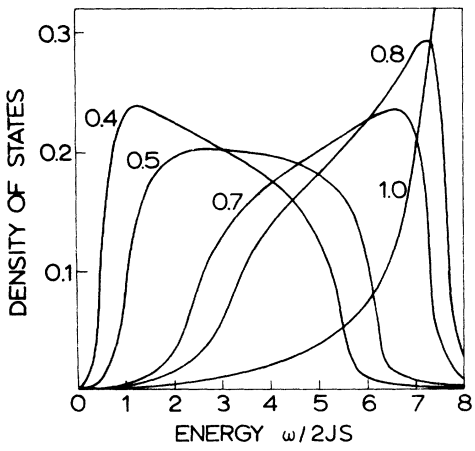

FIG. 3. Densities of states for the body-centered-cubic antiferromagnet with values of m as shown.

$$= \begin{bmatrix} \omega + 2\tilde{J}Sz & -2\tilde{J}S\gamma_{\vec{k}} \\ 2\tilde{J}S\gamma_{\vec{k}} & \omega - 2\tilde{J}Sz \end{bmatrix}$$

$$\times [\omega^2 - (2\tilde{J}Sz)^2 (1 - \gamma_{\vec{k}}^2)]^{-1}; \quad (13)$$

g_{00} has the same form as this with J replacing $\tilde{J}$ $= J(1-\epsilon)$. Equation (12) is the same as T-K's equation (6.5) in Ref. 2.

Using these results, we have calculated the density of states for a body-centered-cubic lattice as shown in Fig. 3. Even for $m = 0.8$ there has been a large transfer of weight to the center of the band and only a small shift in the peak, whereas we should expect the density to be approximately the same shape as in the pure crystal but scaled by a factor m. The appearance of the peak at low energy as m is reduced is similar to the peak found by T-K in the ferromagnet (cf. Fig. 3 or Ref. 1), although here the curve continues to fall to zero as ω^2. The low-energy peak in $\mathrm{Im}G(\vec{k}, \omega)$, shown in Fig. 4, is more prominent than in the ferromagnetic case; indeed for $m \leq 0.6$ the normal spin-wave response is seen only as a shoulder on the low-energy peak, which is almost independent of $\vec{k}$. For m close to unity, the spin-wave energy again falls slower than m.

We have compared these line shapes with corresponding ones calculated by means of a theory developed to describe antiferromagnets containing either magnetic[3] or nonmagnetic[4] impurities. This uses the CPA on the diagonal parts of $\underline{v}$, not only on impurity sites, but also on neighboring sites. Thus $\underline{v}$ depends on the arrangement of impurities. However the theory treats the off-diagonal parts of $\underline{v}$ in a more approximate way. The results of this theory are shown in Fig. 5, and are physically more reasonable than those described above. The low-energy peak is absent, and the spin-wave energy is approximately m times its value in the pure crystal. The total weight of the response is proportional to m. This is expected in the site problem but is not true in the bond theory, because all the atoms there remain magnetic.

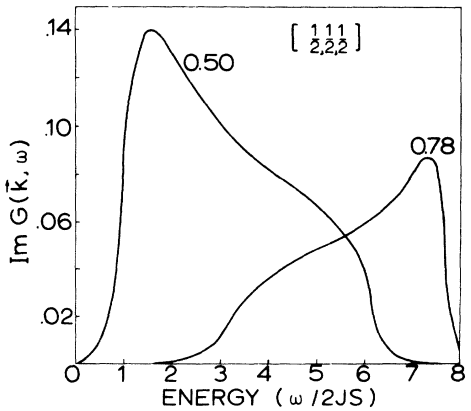

FIG. 4. Response function $\mathrm{Im}G(\vec{k}, \omega)$ for the body-centered-cubic antiferromagnet for the values of m shown, at the boundary of the first magnetic Brillouin zone.

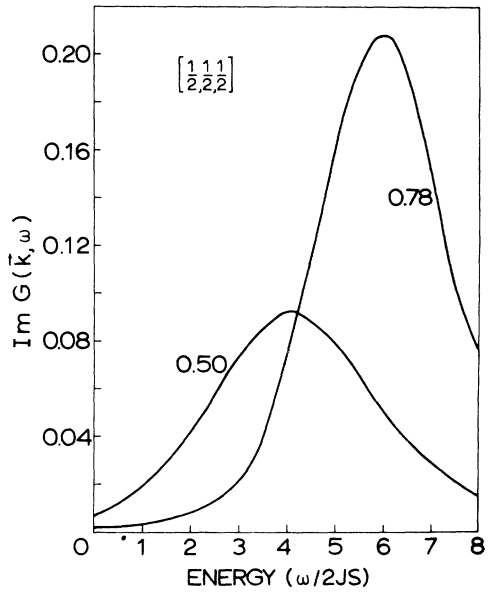

FIG. 5. Same quantities as shown in Fig. 4, but calculated by the method of Buyers, Pepper, and Elliott (Ref. 4) for $Mn_mZn_{1-m}F_2$.

The unsatisfactory aspects of the results of T-K's model stem from the failure to sum coherently all the scattering from one defect. In an attempt to improve upon the spin-wave calculation of the critical concentration, we have used the CPA in a way which includes this effect.[10] The calculation involves replacing $\underline{v}$ of (3) by the full scattering matrix for a nonmagnetic defect, which for the simple cubic lattice is[11]

$$\underline{v}' = 2JS \begin{pmatrix} -6 & 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & -1 & 0 & 0 & 0 & 0 & 0 \\ 1 & 0 & -1 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & -1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & -1 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 & -1 & 0 \\ 1 & 0 & 0 & 0 & 0 & 0 & -1 \end{pmatrix}. \quad (14)$$

$\underline{w}$ and $\underline{G}_0$ are now 7×7 matrices; $\underline{w}$ is again given in the CPA by (5). In order to solve for the elements of $\underline{w}$, we use the symmetry of the lattice to transform to block-diagonal form.[11] There are two s-like modes with symmetric eigenvectors so $\underline{w}_s$ is a 2×2 matrix

$$\underline{w}_s = \begin{bmatrix} w_s^{00} & w_s^{01} \\ w_s^{10} & w_s^{11} \end{bmatrix} . \tag{15}$$

There are three identical p-like modes with w_p and two d-like modes with w_d. The same transformation is used on $\underline{v}'$ and $\underline{G}_0$. The components of $\underline{w}$ are then given by

$$\begin{aligned} \underline{w}_s &= c\underline{v}_s'[\underline{1} - \underline{G}_s(\underline{v}_s' - \underline{w}_s)]^{-1} , \\ w_p &= -c[1 + (1 + w_p)(G_{00} - G_{12})]^{-1} , \\ w_d &= -c[1 + (1 + w_d)(G_{00} + G_{12} - 2G_{13})]^{-1} , \end{aligned} \tag{16}$$

where G_{12} links a pair of opposite neighbors of the central atom, G_{13} links nearest neighbors on the first coordination sphere, and

$$\underline{v}_s' = 2JS \begin{bmatrix} -6 & \sqrt{6} \\ \sqrt{6} & -1 \end{bmatrix} , \tag{17}$$

$$\underline{G}_s = \begin{bmatrix} G_{00} & \sqrt{6}G_{01} \\ \sqrt{6}G_{01} & G_{00} + G_{12} + 4G_{13} \end{bmatrix} .$$

As in (7), the form of $\underline{v}_s'$ ensures that we can write

$$\underline{w}_s = w_s \underline{v}_s' , \tag{18}$$

where now

$$w_s = -c[1 + (1 + w_s)(7G_{00} - 12G_{01} + G_{12} + 4G_{13})]^{-1} . \tag{19}$$

The Fourier transform of $\underline{w}$ is related to the components by

$$\begin{aligned} w(\vec{k}) = w_s 6(1 - \gamma_{\vec{k}})^2 + w_p 3(1 - \gamma_{2\vec{k}}) \\ + w_d 3(1 + \gamma_{2\vec{k}} - 2\gamma_{\vec{k}}^2) . \end{aligned} \tag{20}$$

When $k^{-1} \gg a$, the lattice spacing, (20) becomes approximately

$$w(\vec{k}) \simeq \tfrac{1}{6} w_s (ka)^4 + w_p 2(ka)^2 + O(ka)^4 . \tag{21}$$

If we consider (19) in the limit of low c, by neglecting w_s on the right-hand side, and replacing the Green's functions by their values in the pure crystal, we find that w_s contains a factor ω^{-1}; thus the first term of (21) makes a contribution to $O(ka)^2$. There is no such divergence in either w_p or w_d. The ω^{-1} factor in w_s is an essential result of the model. In defining G in (2a) to satisfy the Dyson equation (2b), spin-wave creation and destruction operators have been used and $S_l^{-1/2}$ appears in the definition. In the case of a nonmagnetic impurity $S' = 0$, and this leads to an infinity in G. However, the defect matrix (14) is independent of S', and since it cuts all bonds from an impurity site, a theory using $S' \neq 0$ should give the correct treatment for the magnetic atoms, together with free spins at the impurity sites. The Green's function at these sites will then be proportional to $1/\omega$ since no energy is needed to reverse a spin there. However, the averaging process in both the low-c and CPA theories does not adequately maintain the difference between magnetic and nonmagnetic sites. The resonance in w_s is shifted away from $\omega = 0$ and mixed into G for the magnetic sites. As a result an unphysical resonance appears at low ω in the calculated quantities.

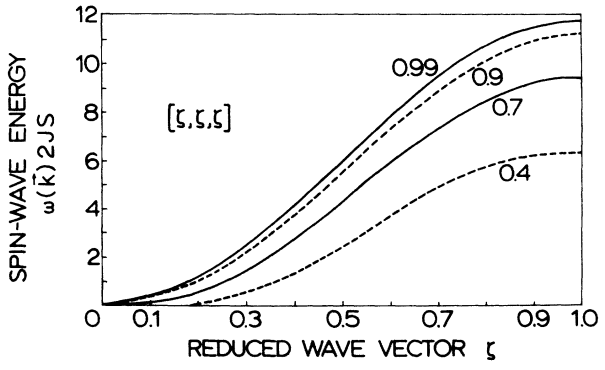

FIG. 6. Spin-wave dispersion curves for the simple cubic ferromagnet with the values of m indicated calculated from the CPA with nonmagnetic sites.

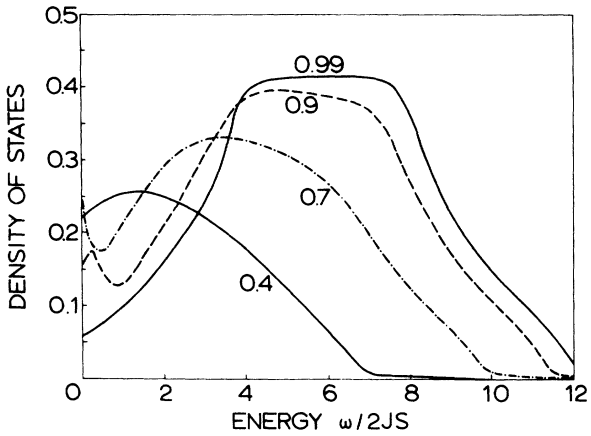

FIG. 7. Densities of states for the simple cubic ferromagnet with the values of m shown calculated using the CPA with nonmagnetic sites.

The first term in (20) does not contribute to $O(ka)^2$, and the long-wavelength spin-wave energy is determined purely by w_p. While this is not important for large ω, it means that we cannot properly describe the spin waves with small k. It follows from (20) that if w_s does not contain a factor ω^{-1} the coefficient of $(ka)^2$ in the spin-wave energy becomes negative when the real part of w_p is less than -0.5. Numerical calculations confirm that this is true when $m < 0.5$. Figure 6 shows the dispersion curves for several values of m, and it is seen that for $m = 0.4$ the curve passes through zero energy when k is finite. Similarly, the density of states in this concentration region extends

to negative energy, as in Fig. 7. The effect of the low-energy resonance can be seen in the curves for $m = 0.9$ and 0.7, which have peaks near $\omega = 0$.

We have attempted to remove this low-frequency resonance by various tricks, which are described in detail in Ref. 10. In one, we attempted to force G at the defect sites to retain its ω^{-1} form, but this led to computational difficulties. In another, we introduced a large field at the impurity site to remove the resonance to very high frequency. None of these attempts produced results which were physically reasonable at low ω.

We conclude that neither of these applications of the CPA yields a useful estimate of the critical concentration. The site method starts from a proper model for the dilute magnet and might be expected to give satisfactory results, but will not in fact do so unless the excitations can be excluded from the nonmagnetic atoms throughout the configurational averaging. The bond approach of Tahir-Kheli represents an interesting model in itself but is not really appropriate for a detailed description of the dilute magnet. Some of its predictions are seen here to be at variance with the properties of dilute magnets which we would expect on physical grounds.

[*Note added in proof.* A more satisfactory CPA treatment of the site problem for the dilute magnet has now been developed [Harris, Leath, Elliott, and Pepper (to be published)].

We are indebted to Dr. W. J. L. Buyers, Dr. A. Brooks Harris, and Dr. P. Leath for illuminating discussions of this problem.

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