Modified Callen decoupling in the Green's-function theory of Heisenberg antiferromagnets

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The modified Callen decoupling (MCD) is extended to Heisenberg antiferromagnets with exchange between more than nearest neighbors. The results are illustrated by application to type-I order in the three cubic lattices. It is shown that type-I order in simple cubic and body-centered-cubic lattices is especially simple—and therefore especially stable—due to several accidental symmetries. The more general case is typified by type-I order in a face-centered-cubic lattice, which shows an instability at intermediate temperatures for all values of the exchange constants when $S = 1/2$. The MCD results are compared to the Padé-approximant estimates of the Néel temperatures and are shown to explain the anomalous behavior found for spin 1/2.

I. INTRODUCTION

Most of the work devoted to the Heisenberg model has been restricted to nearest-neighbor exchange and the simplest types of magnetic order. It has usually been assumed that removal of these restrictions merely complicates the mathematics without producing any qualitative differences. This assumption has been supported by the molecular-field approximation (MFA)[1-4] and the random-phase approximation (RPA),[5-7] which fail to show qualitative changes when second-nearest-neighbor exchange is included. However, more careful consideration of these approximations has revealed that certain competing interactions can cancel, allowing small corrections to drive the usually assumed magnetic order unstable. On the basis of qualitative arguments, phase transitions to new types of magnetic order have been predicted and used to explain certain anomalies that occur in the Padé-approximant estimates of the critical temperatures.[8]

To further investigate the corrections to RPA for Heisenberg magnets that do not necessarily satisfy the usual simplifying assumptions, we have used a modification and extension of a Green's-function decoupling technique first introduced by Callen for the study of ferromagnets. The original Callen decoupling[9] (CD) introduced corrections to RPA that eliminated the spurious T^3-term in the low-temperature magnetization[10] and produced excellent agreement with the Padé-approximant Curie temperatures[11,12] in the high-spin limit. We have shown in a previous paper that a modified Callen decoupling[13] (MCD) is capable of retaining the advantages of CD and, in addition, producing very good agreement with the Padé-approximant Curie temperatures for all spins. We also extended MCD to include exchange between more distant neighbors in a ferromagnet and found the same agreement with the Padé results for first- and second-neighbor exchange in all three cubic lattices.[11-16]

All Green's-function theories involving Callen-type decouplings attribute the most significant corrections to the correlations between spins. The elimination of the restriction to nearest-neighbor exchange is therefore especially interesting since the spin-spin correlations lead to different renormalizations of different contributions to the magnon energies. The magnon-energy spectrum is determined by "effective-exchange constants," which depend on temperature, magnetization, and applied magnetic field. The wave-vector dependence of the magnon energies changes as these parameters are varied.

Antiferromagnetism proves to be more complex than ferromagnetism because the ith neighbors of a given site are generally not all on the same magnetic sublattice. Since the correlation functions depend on the relative orientation of the spins as well as their separation, the contributions to the magnon energies also have different renormalizations for different alignments. The resulting magnon energy spectrum not only has a k-dependent renormalization, but its shape can be qualitatively different than any RPA spectrum.

The dependence of the renormalization factors on the alignment of the spins results in a large qualitative difference between type-I order in sc (simple cubic) and bcc (body-centered-cubic) lattices on the one hand and the fcc (face-centered-cubic) lattice on the other. Indeed, MCD provides a semiquantitative foundation for the qualitative explanations of the anomalies in the Padé-approximant critical temperatures, which we proposed earlier.[8] MCD supports the assumption that the corrections to RPA are "regular."

We shall discuss the application of MCD to Heisenberg antiferromagnets, using type-I order in the cubic lattices to illustrate the results. Section II contains a description of the essential

steps in the MCD method and Sec. III describes the various techniques that are used to calculate the critical temperatures. Section IV discusses the relatively simple type-I order found in sc and bcc lattices and Sec. V describes the added complications found in other cubic antiferromagnets, using fcc type-I order as an example.

II. THE MCD METHOD

We consider the Heisenberg Hamiltonian

$$\mathcal{H} = -\sum_{f,g} J_{f,g}\, \vec{S}_f \cdot \vec{S}_g - \mu \sum_f h_f S_f^z \ , \tag{1}$$

where h_f is a staggered magnetic field. The exchange integrals, $J_{f,g}$, are assumed to have the symmetry of the lattice, but are not restricted to nearest neighbors.

We shall define the retarded, temperature-dependent Green's functions in the usual manner

$$G(f,l,t) = -i\Theta(t)\langle\langle [S_f^+(t), S_l^-(0)]\rangle\rangle = \langle\langle S_f^+; S_l^-\rangle\rangle \ , \tag{2}$$

and follow Callen[9] in terminating the infinite hierarchy of equations of motion in first order with a decoupling approximation of the form

$$\pm\langle\langle S_g^z S_f^+; S_l^-\rangle\rangle \to \sigma\langle\langle S_f^+; S_l^-\rangle\rangle - \alpha\langle S_g^- S_f^+\rangle\langle\langle S_g^+ S_l^-\rangle\rangle \ . \tag{3}$$

The plus (minus) sign is used when the site g is on the up (down) magnetic sublattice, and

$$\sigma = |\langle S_g^z\rangle| \ . \tag{4}$$

We shall use the MCD expression for the decoupling parameter

$$\alpha = \frac{1}{2S}\left(\frac{S-1}{S+1}\right)\left(\frac{\sigma}{S}\right) + \frac{1}{S(S+1)}\left(\frac{\sigma}{S}\right)^3 \ , \tag{5}$$

which is known to give good predictions for the properties of Heisenberg ferromagnets.[13] There are no further adjustable parameters.

The decoupled Green's function equations of motion can be Fourier transformed with respect to time and take the form

$$\begin{aligned}\omega G(f,l,\omega) = {} & 2\sigma\delta_{f,l} + \mu h_f G(f,l,\omega) \\ & - 2\sigma\sum_g J_{gf}[G(f,l,\omega) - G(g,l,\omega)] \\ & + 2\alpha\sum_g J_{gf}[\langle S_f^- S_g^+\rangle G(f,l,\omega) \\ & - \langle S_g^- S_f^+\rangle G(g,l,\omega)] \ . \end{aligned} \tag{6}$$

Because the antiferromagnetic state does not have full translational symmetry, it is necessary to divide the lattice into sublattices in order to solve Eq. (6) by spatial Fourier transformation.[17,18] We shall only present the solutions of the equations for cases in which the magnetic sublattices are translationally invariant. With this restriction, we only need two Green's functions: $G_s(f,l,\omega)$, for which f and l are on the same magnetic sublattice, and $G_d(f,l,\omega)$, for which f and l are on different magnetic sublattices. Since bcc type-II order and fcc type-III*A* order do not satisfy this restriction, our results are not directly applicable to them, although they can be treated by using four sublattices (and four Green's functions) without further approximation.

To simplify the equations of motion, we use a slight modification of a trick developed for the ferromagnetic case.[9,13,19] By restricting the summation to those members of the jth shell of neighbors that are on the same sublattice as the central site, we can define the partial Fourier transforms of the exchange constants

$$J_{sj}(\vec{k}) = J_j \sum_{\vec{\Delta}_j}^{s} e^{i\vec{k}\cdot\vec{\Delta}_j} \ , \tag{7}$$

where the $\vec{\Delta}_j$'s are the vectors connecting a central site with the members of the jth shell and the superscript s indicates that the sum is restricted to neighbors on the same magnetic sublattice. $J_{dj}(\vec{k})$ is defined by a similar restriction to neighbors that are on different sublattices. These restrictions are also used to define partial Fourier transforms of the correlation functions, so that

$$\Psi_s(\vec{k}) = \frac{1}{2\sigma}\sum_f^s e^{i(\vec{r}_f - \vec{r}_g)\cdot\vec{k}}\langle S_f^- S_g^+\rangle \ , \tag{8}$$

with a similar equation for $\psi_d(\vec{k})$.

By defining

$$f_{sj} = [1/NJ_{sj}(\vec{0})]\sum_{\vec{k}} J_{sj}(\vec{k})\psi_s(\vec{k}) \tag{9}$$

(and f_{dj} analogously), we can write the renormalized, or "effective," Fourier transformed exchange constants as

$$\tilde{J}_s(\vec{k}) = \sum_j \tilde{J}_{sj}(\vec{k}) = \sum_j J_{sj}(\vec{k})(1 + 2\alpha f_{sj}) \tag{10}$$

and

$$\tilde{J}_d(\vec{k}) = \sum_j \tilde{J}_{dj}(\vec{k}) = \sum_j J_{dj}(\vec{k})(1 - 2\alpha f_{dj}) \ . \tag{11}$$

The equations of motion can then be written as

$$\begin{aligned}\omega G_s(\vec{k},\omega) = {} & 2\sigma + [\mu h + 2\sigma\tilde{W}(\vec{k})]G_s(\vec{k},\omega) \\ & - 2\sigma\tilde{J}_d(\vec{k})G_d(\vec{k},\omega)\end{aligned} \tag{12}$$

and

$$\begin{aligned}\omega G_d(\vec{k},\omega) = {} & -[\mu h + 2\sigma\tilde{W}(\vec{k})]G_d(\vec{k},\omega) \\ & + 2\sigma\tilde{J}_d(\vec{k})G_s(\vec{k},\omega) \ ,\end{aligned} \tag{13}$$

where

$$\tilde{W}(\vec{k}) = \tilde{J}_s(\vec{0}) - \tilde{J}_s(\vec{k}) - \tilde{J}_d(\vec{0}) \ . \tag{14}$$

These equations have exactly the same form as the corresponding RPA equations and can be solved directly in the same way.[6,17,18] The essential difference is that $\tilde{J}_s(\vec{k})$ and $\tilde{J}_d(\vec{k})$ are now self-consistent functions of the temperature, magnetization, and magnetic field.

Using the usual techniques, we find that the magnon energies are given by

$$E^2(\vec{k}) = [2\sigma\tilde{W}(\vec{k})]^2 - [2\sigma\tilde{J}_d(\vec{k})]^2 , \tag{15}$$

and the f's are

$$f_{sj} = \frac{1}{2NJ_{sj}(\vec{0})} \sum_{\vec{k}} J_{sj}(\vec{k}) \left(\frac{2\sigma\tilde{W}(\vec{k})}{E(\vec{k})}\right) \coth[\tfrac{1}{2}\beta E(\vec{k})] \tag{16}$$

and

$$f_{dj} = \frac{1}{2NJ_{dj}(\vec{0})} \sum_{\vec{k}} J_{dj}(\vec{k}) \left(\frac{2\sigma\tilde{J}_d(\vec{k})}{E(\vec{k})}\right) \coth[\tfrac{1}{2}\beta E(\vec{k})] , \tag{17}$$

where $\beta = 1/k_B T$

As shown by Callen,[9] the magnetization is then

$$\sigma = \frac{(S-\Phi)(1+\Phi)^{2S+1} + (S+1+\Phi)\Phi^{2S+1}}{(1+\Phi)^{2S+1} - \Phi^{2S+1}} , \tag{18}$$

where

$$\Phi = \frac{1}{2N} \sum_{\vec{k}} \left[\left(\frac{2\sigma\tilde{W}(\vec{k})}{E(\vec{k})}\right) \coth[\tfrac{1}{2}\beta E(\vec{k})] - 1 \right] . \tag{19}$$

Equations (14)–(19) are then solved self-consistently to obtain the physical properties of the system.

III. THE CRITICAL TEMPERATURE(S)

There are several methods of obtaining the critical temperature now that we have solved the equations of motion. It has been tacitly assumed in most previous work that all methods lead to the same result. This is, of course, necessarily true for an exact treatment, but is only justified for a Green's-function approximation in the simplest cases. Since we need the results of all methods to discuss the fcc antiferromagnet, we shall present each one explicitly.

The basic idea is to consider a physical quantity of interest as a function of the temperature and look for some sort of singularity. Three possibilities for an antiferromagnet are:

(1) The spontaneous sublattice magnetization $\sigma \to 0$ $(h_f = 0)$;

(2) the staggered susceptibility $(h_f = \pm h \to 0)$;

(3) the physical susceptibility $(h_f = H \to 0)$.

Most Green's-function work uses just the first possibility, but in cases in which discrepancies arise, it is important to remember that the Padé approximant method involves only the analysis of the high-temperature susceptibilities.

The first method has been extensively discussed in literature. We shall give the results for completeness and as a foundation for later discussion. When $\sigma \to 0$, $\Phi \to \infty$ and we can expand Eq. (18) as

$$\sigma = \tfrac{1}{3} S(S+1)\Phi^{-1} + O(\Phi^{-2}) . \tag{20}$$

The hyperbolic cotangents in Eqs. (16), (17), and (19) can also be expanded, since the magnon energy goes to zero in this limit. The resultant equations for the Néel temperatures are

$$k_B T_N = \tfrac{2}{3} S(S+1) \left(\frac{1}{2N} \sum_{k} (D_{N-}^{-1} + D_{N+}^{-1})\right)^{-1} , \tag{21}$$

$$\lim_{\sigma \to 0}(2\sigma f_{sj}) = \frac{k_B T_N}{2NJ_{sj}(\vec{0})} \sum_{\vec{k}} J_{sj}(\vec{k})(D_{N-}^{-1} + D_{N+}^{-1}) , \tag{22}$$

and

$$\lim_{\sigma \to 0}(2\sigma f_{dj}) = \frac{k_B T_N}{2NJ_{dj}(\vec{0})} \sum_{\vec{k}} J_{dj}(\vec{k})(D_{N-}^{-1} - D_{N+}^{-1}) , \tag{23}$$

where

$$D_{N\pm} = \tilde{W}(\vec{k}) \pm \tilde{J}_d(\vec{k}) . \tag{24}$$

In computing the high-temperature staggered susceptibility,

$$\chi_S = \lim_{h \to 0}(\mu\sigma/h) , \tag{25}$$

we use Eq. (20) to obtain equations for the inverse temperature as a function of χ_S. These equations are then investigated to find the highest possible value of χ_S, which occurs when

$$\mu^2 \chi_S^{-1} = -2 \operatorname{Min}\{D_{N\pm}\} . \tag{26}$$

The resulting critical temperature, which we shall denote by T_S, is then found as the self-consistent solution of the equations

$$k_B T_S = \tfrac{2}{3} S(S+1) \left(\frac{1}{N} \sum_{\vec{k}} (D_{S-}^{-1} + D_{S+}^{-1})\right)^{-1} , \tag{27}$$

$$\lim_{\sigma \to 0}(2\sigma f_{sj}) = \frac{k_B T_S}{NJ_{sj}(\vec{0})} \sum_{\vec{k}} J_{sj}(\vec{k})(D_{S-}^{-1} + D_{S+}^{-1}) , \tag{28}$$

and

$$\lim_{\sigma \to 0}(2\sigma f_{dj}) = \frac{k_B T_S}{NJ_{dj}(\vec{0})} \sum_{\vec{k}} J_{dj}(\vec{k})(D_{S-}^{-1} - D_{S+}^{-1}) , \tag{29}$$

where

$$D_{S\pm} = \mu^2 \chi_S^{-1} + 2\tilde{W}(\vec{k}) \pm 2\tilde{J}_d(\vec{k}) . \tag{30}$$

In RPA, it is very easy to find T_S. Equations (28) and (29) are no longer relevant. We simply set $\chi_S^{-1} = 0$ and perform the integral in Eq. (27). This automatically gives $T_S = T_N$.

In MCD, the situation is considerably more complicated. In addition to the necessity of keeping Eqs. (28) and (29), the quantity $\tilde{W}(\vec{k})^2 - \tilde{J}_d(\vec{k})^2$ can be negative for some values of the wave vector. This means that the magnon energy then becomes imaginary if χ_S^{-1} goes to zero and the whole formalism breaks down. The formalism gives a critical

temperature for the finite value of the staggered susceptibility given by Eq. (26).

The third method is to apply a physical magnetic field ($h_f = H$) for high temperatures and look for singularities in the physical susceptibility.

$$\chi_P = \lim_{H \to 0}(\mu\sigma/H) \ . \tag{31}$$

Here, we no longer have antiferromagnetic order (for a nonzero field, $\langle S_f^z \rangle = \sigma$ for all sites) and we analyze the system by the methods used for ferromagnets. The necessary equations are discussed in Refs. 9 and 13 and can also be rederived from the equations in Sec. II by setting J_{di} equal to zero throughout. Since χ_P never diverges for an antiferromagnet, we find a finite critical susceptibility,

$$\mu^2\chi_P^{-1} = -2\,\mathrm{Min}[\tilde{J}(\vec{0}) - \tilde{J}(\vec{k})] \ , \tag{32}$$

and the critical temperature T_P is given by the equations

$$k_B T_P = \tfrac{2}{3} S(S+1)\left(\frac{2}{N}\sum_{\vec{k}} \{\mu^2\chi_P^{-1} + 2[\tilde{J}(\vec{0}) - \tilde{J}(\vec{k})]\}^{-1}\right)^{-1} \tag{33}$$

and

$$\lim(2\sigma f_j) = \frac{2k_B T_P}{N\tilde{J}_j(\vec{0})} \sum_{\vec{k}} \tilde{J}_j(\vec{k})\{\mu^2\chi_P^{-1} + 2[\tilde{J}(\vec{0}) - \tilde{J}(\vec{k})]\}^{-1} \ . \tag{34}$$

IV. TYPE-I ORDER IN sc AND bcc LATTICES

For both the sc and bcc lattices, type-I order is characterized by having all nearest neighbors antiparallel and all next-nearest neighbors parallel.[1-4,6] The simplicity of this type of order greatly reduces the work required and has resulted in the majority of calculations being limited to these two cases (usually with the further condition that $J_2 = 0$). The MCD equations also reflect this simplicity and exhibit several symmetries not generally present.

To begin with, only $\tilde{J}_{d1}(k)$ and $\tilde{J}_{s2}(k)$ are nonzero. This means that there are only two renormalization factors and the magnon energies can be expressed in terms of the RPA energies for some effective exchange constants $\tilde{J}_{d1}$ and $\tilde{J}_{s2}$. Since the RPA magnon energies always take on their minimum value of zero at $k = 0$, the staggered susceptibility diverges at T_N and we can set $\chi_S^{-1} = 0$ in Eqs. (26)–(29). Comparison with Eqs. (20)–(23) then shows that $T_S = T_N$.

We can further exploit the symmetry of this order by noting that if $\vec{K}$ is a nearest-neighbor vector of the reciprocal magnetic sublattice,

$$\tilde{J}_{s2}(\vec{k}) = \tilde{J}_{s2}(\vec{k} + \vec{K}) \tag{35}$$

and

$$\tilde{J}_{d1}(\vec{k}) = -\tilde{J}_{d1}(\vec{k} + \vec{K}) \ . \tag{36}$$

Consequently, the integrals over both terms in Eqs. (21)–(23) are identical and we need only calculate the first one in each equation. We can further simplify the calculation by noting that the normalized integral over the Brillouin zone of the whole lattice is equal to the integral over the Brillouin Zone of the magnetic sublattice. The former not only involves simpler boundary conditions, but is also identical to the corresponding integral for the ferromagnetic case if we associate $-J_{d1}(\vec{k})$ for the antiferromagnet with $J_1(\vec{k})$ for the ferromagnet. This gives us a symmetry between the Néel and Curie temperatures, which is only valid for the sc and bcc lattices.

$$T_N(-|J_1|, J_2) = T_c(|J_1|, J_2) \ . \tag{37}$$

The critical value of $\mu^2\chi_P^{-1}$ is just $-2\tilde{J}_1(0)$. When this is substituted in Eqs. (32) and (33) and all k values in the integral are shifted by $\frac{1}{2}\vec{k}$, we again obtain Eqs. (21)–(24) for the critical temperature.

To sum up, for type-I order in sc and bcc lattices,

$$T_N = T_S = T_P = T_C \ . \tag{38}$$

As a final exploitation of symmetry, we note that the Curie temperatures for the corresponding ferromagnetic cases have already been calculated as functions of the exchange constants. In particular, expansions of the critical temperatures in terms of the ratios of the exchange constants have been published.[13]

For the special case of $J_2 = 0$, an analytic solution for the critical temperature can be obtained.[9,13,17,18] We find

$$T_N^{\mathrm{MCD}} = T_N^{\mathrm{RPA}}\left[1 + \tfrac{1}{3}\left(1 - \frac{1}{S}\right)\left(1 - \frac{1}{F(-1)}\right)\right] \ . \tag{39}$$

where the RPA value for the Néel temperature is

$$T_N^{\mathrm{RPA}} = T_C^{\mathrm{RPA}} = T_C^{\mathrm{MFA}}/F(-1) \ , \tag{40}$$

and $F(-1)$ is the appropriate Watson integral,[20] for which

$$1/F(-1) = 0.659\,465, \ \mathrm{sc}; \ 0.717\,772, \ \mathrm{bcc}. \tag{41}$$

Equation (39) was first found for the ferromagnetic case by Tahir-Kheli,[21] who noted empirically, prior to MCD, that it provides an excellent fit to the Padé approximant estimates of the Curie temperatures in cubic crystals. Although the Páde results indicate that T_N is higher than the corresponding T_C,[14,15] the difference is usually small so that Eq. (39) also gives a good fit to the antiferromagnetic data.

To present the MCD Néel temperatures graphically, it is convenient to normalize them to the MFA values.

$$\tau_N = T_N/T_N^{\mathrm{MFA}} \ . \tag{42}$$

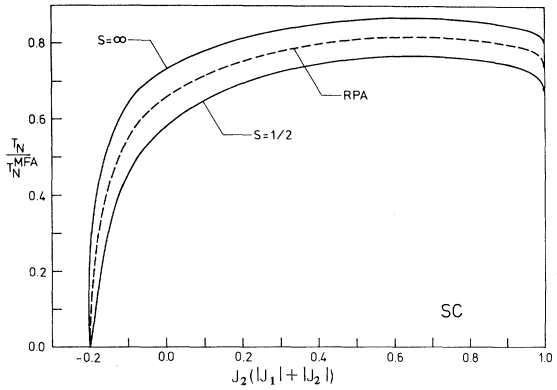

FIG. 1. MCD Néel temperatures for an sc type-I antiferromagnet. (Curves are identical to those for the sc ferromagnet.) The dashed curve gives the RPA results, which coincide with those of MCD for $S=1$.

This normalization has the advantages of removing the $S(S+1)$ spin dependence and expressing T_N in terms of a dimensionless number between zero and one.

We shall plot τ_N as a function of

$$\xi = J_2/(|J_1| + |J_2|) , \tag{43}$$

(which varies between -1 and 1), in order to include all possible ratios of the exchange constants in a single graph.

A plot of τ_N versus ξ for the sc lattice is shown in Fig. 1. The dashed curve represents the RPA results, which coincide with those of MCD for $S=1$. The lower curve gives the MCD results for $S=\frac{1}{2}$ and the upper curve gives the limit of $S=\infty$.

Unfortunately, the available Padé results for the simple cubic antiferromagnet are limited to nearest-neighbor interactions.[14,15] They lie slightly above the Pade results for ferromagnets, but, except for spin $\frac{1}{2}$, are still in good agreement with MCD. For spin $\frac{1}{2}$, the Padé values for T_N and T_C differ by about 13% ($T_N > T_C$), while MCD gives a single value that lies about halfway between them (all three are below the RPA value).

We can also make a comparison with the Padé results in the limit $J_1 \to 0 (\xi \to 1)$. In this limit, the system consists of two independent, ferromagnetic, fcc sublattices with a nearest-neighbor exchange J_2. As we have shown previously, the agreement between MCD and the Padé results is also very good in this case.[13]

The cusp that occurs at $\xi = 1$ ($J_1 = 0$) is found in all Green's function theories. It is associated with the magnons that rotate the magnetic sublattices with respect to each other and whose energy vanishes as $J_1 \to 0$. This effect has been discussed elsewhere in detail for the ferromagnetic case (for which it is essentially the same), but it has not yet been investigated by the Padé method.[13]

When J_2 is negative and sufficiently large, type-I order becomes unstable and a phase transition occurs. At $T=0$, this happens for an sc lattice at a critical ratio of $J_2/J_1 = \frac{1}{4}$. This value is also found in simple spin-wave theory, as well as MFA, RPA, and CD.

However, for higher temperatures, MCD predicts qualitative differences in the behavior of the system when the ratio of the exchange constants is near the critical ratio. As shown by the dashed curve in Fig. 1, RPA predicts that the critical temperature goes to zero as $|J_2/J_1 - \frac{1}{4}|^{1/2}$ and that type-I order is impossible when $J_2 < \frac{1}{4} J_1$. For $S=1$, the MCD curve coincides with the RPA curve.

For $S>1$, the correlations support the order and MCD predicts that the effective ratio of the exchange constants can be characteristic of type-I order for nonzero temperatures even when J_2 is slightly less than $\frac{1}{4}J_1$. This makes the curve for the Néel temperature bulge slightly to the left of the critical ratio as shown in Fig. 1 for $S=\infty$. This effect has been discussed elsewhere in connection with the corresponding ferromagnetic problem and can lead to phase transitions beyond those predicted by MFA and RPA.[13]

For $S=\frac{1}{2}$, MCD lowers the curve for T_N substantially and results in a linear approach to zero as $J_2 \to \frac{1}{4}J_1$. However, this prediction is not to be taken too seriously, since the magnetization becomes double valued when J_2 is too close to $\frac{1}{4}J_1$. This reflects an instability arising from a cancellation of the effective interactions between neighboring (100) planes.[8] The instability leads to a new type of magnetic order that probably involves some sort of canted alignment. As a result of the change in the magnetic order, the Néel temperatures for spin $\frac{1}{2}$ are expected to be anomalously high when J_2 is near $\frac{1}{4}J_1$. An analogous anomaly is present in the Padé approximant results for fcc and bcc ferromagnets, but the sc case has not yet been treated.

Figure 2 compares the MCD Néel temperatures with the Padé approximant calculations for bcc type-I order. The Padé results are indicated by x's for $S=\infty$ and 0's for $S=\frac{1}{2}$. The RPA curve is again given by a dashed line.

For infinite spin, the comparison shows that MCD correctly gives a positive correction to RPA, as is normal when $S>1$. The correction is also of approximately the right magnitude. The MCD curve lies slightly above the Padé values, but the Padé results are somewhat irregular. Again, the limit of $J_1 \to 0 (\xi \to 1)$ shows good agreement.

For $S=\frac{1}{2}$, MCD correctly gives a negative correction to RPA, but the quantitative agreement is less satisfactory. The MCD correction seems to be too large by about a factor of two, resulting in predicted values that are somewhat too low. In

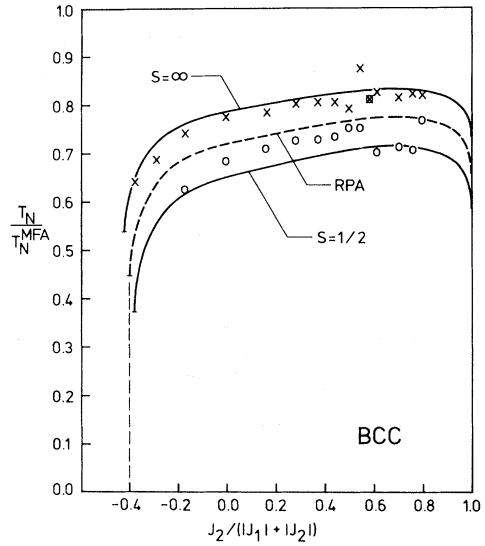

FIG. 2. MCD Néel temperatures for a bcc type-I antiferromagnet. (Curves are identical to those for the bcc ferromagnet.) The dashed curve gives the RPA results, which coincide with those of MCD for $S=1$. The ×'s ($S=\infty$) and the O's ($S=\frac{1}{2}$) give the Padé results from Ref. 11.

the $J_1 \to 0 (\xi \to 1)$ limit, the Padé approximant critical temperature has been shown to be strongly dependent on the number of terms included in the high-temperature expansion.[15] In this limit, the MCD values are slightly too high.

Note that the Néel temperature for bcc type-I order (Fig. 2) does not go to zero at the critical ratio of the exchange constants ($J_2/J_1 = \frac{2}{3}$). In contrast to the sc case, the bcc magnon energies only go to zero at a point (instead of a line) in k space when the critical ratio is reached and the integrals determining the Néel temperature do not diverge.[7] The boundary of the MCD solutions for temperatures below T_N can be found by looking for the line along which the effective ratio of the exchange constants takes on the critical value as a function of the temperature. For RPA, the effective ratio is always equal to the true ratio and the line is vertical. In MCD, the critical ratio is a function of the temperature, but has an infinite slope at $T=0$.

$$[\tilde{J}_{s2}(T)/\tilde{J}_{d1}(T) - J_2/J_1] \propto T^{3/2} \ . \tag{44}$$

Although the analysis of the MCD equations for bcc type-II order has not yet been completed, it is likely that type-II order is stable at zero temperature when $J_2 < \frac{2}{3}J_1$. If, however, J_2 is only slightly lower than the critical value and $S>1$, MCD predicts a first-order phase transition to type-I order as the temperature is raised and second-order transition to the paramagnetic state at a higher temperature. Due to the symmetry with the ferromagnetic state when the sign of J_1 is changed, MCD predicts an analogous antiferromagnetic-ferromagnetic-paramagnetic series of transitions when J_1 is positive and J_2 is slightly less than the critical value.

V. TYPE-I ORDER IN THE fcc LATTICE

Because nearest neighbors of a given site in an fcc lattice can be nearest neighbors of each other, the simple antiferromagnetic order found in the sc and bcc lattices is impossible. As a result, any antiferromagnetic order that does occur will be opposed by some of the interactions. This feature destroys the symmetry with the ferromagnetic ($T_N \neq T_C$) and leads to more complex behavior than found in the sc and bcc type-I antiferromagnets.[8]

Despite the added complexity, fcc type-I order ($J_1<0$; $J_2>0$) is easy to visualize. It consists of ferromagnetic (100) planes which are antiferromagnetically aligned with the neighboring (100) planes.[2-4,6] This structure can be derived on the basis of two assumptions:

(1) All next-nearest neighbors are parallel (as they must be in the limit $J_1 \to 0$, $J_2>0$), and

(2) any two spins are either parallel or antiparallel.

[The four nearest neighbors within the ferromagnetic (100) plane oppose the order, while all other interactions support it.]

If we consider a (010) plane, we see that the four nearest neighbors within this plane are antiparallel and the four next-nearest neighbors are parallel, so that all intraplanar exchange supports the order. The other two next-nearest neighbors are on next-nearest-neighbor (010) planes and are also parallel. The remaining eight nearest-neighbor interactions connect adjacent (010) planes. However, four of the spins are parallel and four are antiparallel. Consequently, in MFA, there is no net interaction between adjacent (010) planes; the two (010) sublattices can rotate freely with respect to each other without the restoring forces needed to support the assumption that the spins are either parallel or antiparallel.

In RPA, we have a similar situation. The magnon energy goes to zero on the Brillouin-zone boundary at $\vec{k}^* = (0, 2\pi/a, 0)$, which corresponds to those magnons that rotate the (010) sublattices with respect to each other.

In both MFA and RPA, the exact cancellation of the interaction between (010) sublattices is a consequence of the assumption that the energy contributions from all exchange interactions renormalize identically. In MCD, the renormalization of the energy contributions is affected by correlations between spins and leads to substantial changes in the physical properties of the system due to the behavior of the magnons near $\vec{k}^*$.

The MCD equations are more complicated for

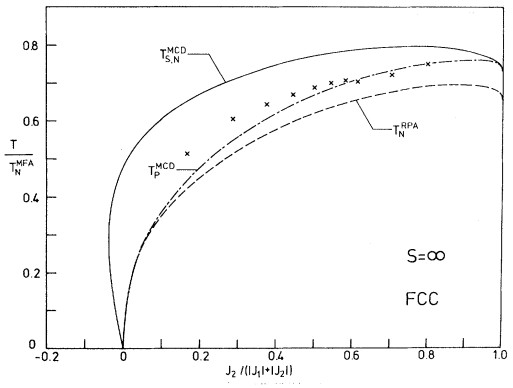

FIG. 3. MCD critical temperatures for fcc type-I antiferromagnets when $S=\infty$. The dashed curve gives the RPA results, which coincide with those of MCD for $S=1$. The ×'s show the Padé results from Ref. 11.

type-I order in fcc lattice than they were for the sc and bcc lattices, due to the nearest-neighbor interactions that oppose the order. We now have three effective exchange constants (two arising from J_1 and one from J_2). This has the effect of making the MCD magnon-energy spectra qualitatively different from the RPA spectra. In particular, $E(\vec{k}^*)$ is no longer zero; the usual magnetic order can be either stabilized or driven unstable by the effect of correlations on the energy renormalization.

Let us first consider $S>1$. The renormalization turns out to be positive $[E(\vec{k}^*)>0]$ for all temperatures, stabilizing the type-I antiferromagnetic order. Because the lowest magnon energy now occurs at $k=0$, $T_S=T_N$.

The numerical results for infinite spin are given in Fig. 3. The dashed line gives the RPA results and the crosses are the Padé values calculated by Pirnie *et al.*[11] Although MCD gives the right qualitative prediction that the correction to RPA is positive and fairly uniform, the MCD curve for T_N is definitely too high. Only in the limit $J_1 \to 0$ ($\xi \to 1$) does MCD agree quantitatively with the Padé results. However, the qualitative agreement does suggest that the MCD prediction[22] that an fcc antiferromagnet with spin greater than one and only nearest-neighbor exchange has a nonzero Néel temperature is correct, although T_N appears to be lower than the MCD value. The relatively good agreement between T_P^{MCD} and the Padé values is interesting, but probably not significant.

For spin $\frac{1}{2}$, the situation is quite different. At low temperatures, the renormalization is again positive, stabilizing type-I order. However, as the temperature is raised, the renormalization becomes negative, driving the system unstable at a nonzero value of the magnetization (in MCD, this occurs at $\sigma=\frac{1}{2}S=\frac{1}{4}$). [The new magnetic order probably involves perpendicular alignment of spins on neighboring (010) planes.[8]] Since σ does not approach zero continuously, the derivation of Eqs. (21)–(24) is not valid and T_N, which we have defined as the solution of these equations, does not exist.

The temperature at which type-I order becomes unstable (which we shall call T_I) can be calculated directly and, as shown in Fig. 4, lies below the RPA Néel temperature for all values of the exchange constants. This is, as we have seen, the normal behavior of the Néel temperature for spin $\frac{1}{2}$. However, as mentioned in Sec. III, we should compare the Padé approximant results[11,12,15] with our calculation of T_S. Such a comparison is shown in Fig. 4. Both MCD and the Padé values show similar anomalous corrections to RPA. For values of ξ greater than about 0.6 ($J_2/|J_1|>1.5$), the correction is negative as usual, but for ξ less than 0.6, the correction is positive. Although the quantitative agreement between MCD and the Padé values is not especially good, the qualitative prediction of the anomalous behavior, including the correct point at which the RPA curve is crossed, certainly suggests that the MCD picture of a breakdown of type-I order at intermediate temperatures is correct.

A qualitative difference still exists for the limit $J_2 \to 0$. Here, the Padé method predicts a nonzero critical temperature ($T_P^{\text{Padé}}=0.52\ T_N^{MFA}$),[11] while MCD fails to predict a phase transition. On the other hand, MCD "almost" predicts a nonzero transition temperature in the limit $J_2 \to 0$; as J_2 is lowered, T_S approaches zero very slowly (as $|\log J_2|^{-1}$).

The MCD prediction of a finite staggered susceptibility at T_S for spin $\frac{1}{2}$ is, of course, incorrect.

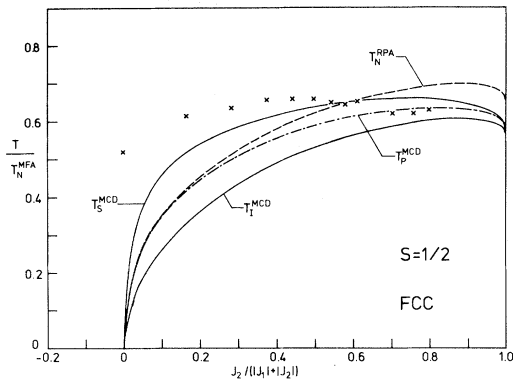

FIG. 4. MCD critical temperatures for fcc type-I antiferromagnets when $S=\frac{1}{2}$. The dashed curve gives the RPA results, which coincide with those of MCD for $S=1$. The ×'s show the Padé results from Ref. 11.

The staggered field associated with the new order at intermediate temperatures [which probably involves rotated (010) sublattices[8]] is just a linear combination of staggered fields with the symmetry of type-I order, so that the staggered susceptibility must diverge at T_S. The difficulty lies in the Green's-function formalism, which is not capable of showing a divergence at this level of approximation unless the transition is to the usual type of order.

A final point of interest is the discrepancy between T_P and T_S for the spin-$\frac{1}{2}$ fcc antiferromagnet. This is, of course, a defect in the MCD method, but it raises the question of whether other methods suffer from the same defect. In particular, the Padé approximant method has been applied to both the physical[15] and the staggered susceptibilities[11] for bcc and fcc, spin-$\frac{1}{2}$, nearest-neighbor antiferromagnets. For the bcc lattice, the Padé method found the same critical temperature from both susceptibilities ($T_P = T_S$). This agrees with the MCD predictions. On the other hand, for the fcc lattice, the Padé work gave $T_p = 0$ and $T_s = 0.52\ T_N^{\mathrm{MFA}}$. This also agrees with the (incorrect) MCD prediction that $T_S \gg T_P$ for very small values of J_2. It would be interesting to see if the Padé method follows the MCD behavior in that the two estimates of the critical temperatures are again equal when $S > 1$.

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