Curie Temperatures for Layer Structures

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The problem of calculating Curie (or Néel) temperatures for layer structures is discussed by considering a simple example—that of a simple-cubic lattice of spins in which the (Heisenberg) exchange interactions J (or $-J$) within a set of parallel planes is allowed to differ from the interactions K (or $-K$) between them. Ferromagnetism and antiferromagnetism are both considered, and particular attention is paid to the cases where $K/J \ll 1$. Most of the well-tried methods for obtaining transition temperatures are discussed, and it is shown that the molecular-field theory, the Opechowski high-temperature expansion method, the constant-coupling treatment, and the cluster methods of Oguchi and of Bethe-Peierls-Weiss are all unable to give results which are even qualitatively satisfactory for the weakly interacting layer problem, if we accept the spin-wave conditions for the existence or nonexistence of long-range order at low temperatures. The breakdown of these methods is shown to be particularly serious in the antiferromagnetic case. Finally, the method of Green functions is used and is shown to give acceptable approximations for both ferromagnetism and antiferromagnetism.

1. INTRODUCTION

THE problem of locating magnetic transition temperatures for Heisenberg ferromagnets and antiferromagnets is one which has received considerable attention from theoretical physicists ever since the model was first introduced by Heisenberg[1] in 1928. In spite of the large number of approximate treatments which have been given, only very recently has it been found possible to make a reliable estimate of the Curie temperature in even the simplest of three-dimensional lattice structures. We refer, of course, to recent work on the isotropic cubic ferromagnets with a single exchange parameter J (see, for example, Rushbrooke and Wood[2] and Domb and Sykes[3]) employing the exact series high-temperature expansion method introduced by Opechowski,[4] and more recently pursued by Brown and Luttinger.[5]

Despite this very considerable success, it is well to bear in mind some of the limitations of the Opechowski method as a tool for the evaluation of transition temperatures. Firstly, the price to be paid for obtaining these "accurate" Curie temperatures is the very large amount of arithmetical computation which is required to extend the series expansions to a sufficient number of terms. It seems unlikely that anyone would be willing to carry out such a detailed calculation for each experimentally important case, or even whether such a calculation would be feasible using a more realistic Hamiltonian with anisotropy and possibly more than one exchange parameter. Secondly, the method is far less suited for the calculation of antiferromagnetic transition temperatures (see, for example, Brown and Luttinger[5]), and this is a very serious restriction since the localized-spin model of Heisenberg is, in general, a far more realistic model for antiferromagnets than for ferromagnets. Thirdly, the Opechowski method seems to be particularly unsuited for dealing with two-dimensional lattice structures for which it predicts the onset of long-range order at nonzero temperatures although the spin-wave theory of Bloch[6] (see also Van Kranendonk and Van Vleck[7]) makes it clear that this should not be the case. Although the problem of two-dimensional lattices might be considered to be of relatively minor importance, we shall show that it is this particular weakness which makes the method unsuited for problems concerning the weakly interacting layer structures with which the present paper is mainly concerned.

In view of these limitations it is encouraging to find that the well-tried cluster treatments of Bethe-Peierls-Weiss[8,9] (the BPW approximation) and of Kasteleijn and Van Kranendonk[10] (the constant-coupling method), in spite of their relative simplicity, are both surprisingly good approximations. For the isotropic cubic ferromagnets they give Curie temperatures which are in general too high, but only by a little over 10%. Both of these methods may successfully be applied to the antiferromagnetic case, though they may be difficult to adapt for use with the more complicated types of Hamiltonian and of antiferromagnetic-spin patterns.

Like the Opechowski method, however, the cluster treatments are essentially high-temperature approximations, and they do tend to exhibit unphysical behaviour at low temperatures—the appearance of an anti-Curie point being a typical example. For the purpose of evaluating transition temperatures and magnetic properties in the critical region this might not, perhaps, appear to be a serious restriction. We shall show, however, that if we accept the spin-wave criterion for the

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[6] F. Bloch, Z. Physik, **74**, 295 (1932).
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existence of long-range order at low temperatures, there are cases of some physical importance for which all the cluster treatments (and also the Opechowski method) break down completely, and are unable to give even a qualitatively satisfactory estimate for the transition temperature. Such instances can occur both for ferromagnetism and for antiferromagnetism but they tend to be more serious in the latter case. The breakdowns occur in cooperative problems for which the lattice consists of weakly interacting layers or chains of spins, and for antiferromagnetism they may occur even when the "weak" interaction is considerable. Experimentally, the most important examples are possibly those of the face-centered cubic antiferromagnetic orders for the case of dominantly nearest-neighbor interactions [actual examples are MnS_2, $MnTe_2$, βMnS, K_2IrCl_6, and $(NH_4)_2IrCl_6$], but we may also cite the case of the hexagonal layer crystals $FeCl_2$, $CoCl_2$, and $NiCl_2$.

In the present paper we investigate what is possibly the simplest structure and Hamiltonian for which these difficulties can adequately be demonstrated. We consider a simple-cubic (sc) lattice with isotropic nearest-neighbor interactions, but we allow the exchange J (or $-J$) within a set of parallel planes to differ from the exchange K (or $-K$) between adjacent planes. Thus, for $K \ll J$, the lattice is one of weakly interacting layers, and for $K \gg J$ one of weakly interacting chains. In this work we concentrate on the former and show that there are cases for which none of the above methods is able to give a reasonable estimate for the transition temperature. After demonstrating the breakdown of these methods, we attack the same problem by the recently developed method using Green functions with the simple random-phase approximation. We show that this method, in contrast with the cluster techniques, is able to give a reasonable approximation for all values of K/J.

The breakdown of the other methods would seem to stem from their inability to point to the dimensionality as the criterion for the existence or nonexistence of long-range order at nonzero temperatures. The Green function method, like the spin-wave approximation, is able to do this, and has the additional advantage of being a valid approximation at all temperatures.

After the present introduction, we set out the detailed problem in Sec. 2, where we also discuss the molecular-field and high-temperature expansion theories. Section 3 deals with the BPW approximation, and Sec. 4 with the constant-coupling treatment. Finally, in Sec. 5, we discuss the method of Green functions.

2. THE MOLECULAR-FIELD THEORY

In this and each of the following sections, we investigate the cooperative problem of a sc lattice of interacting spins. Consider such a lattice with cubic axes x, y, z. Each spin has six nearest neighbors of which four are ocated in an xy plane and two along a z axis. We consider the case of nearest neighbor only isotropic Heisenberg interactions, and we associate an exchange parameter J with the interactions between the nearest neighbors in an xy plane, but allow for an exchange parameter K, possibly different from J, to exist between the z-axis neighbors. We take, therefore, the Hamiltonian

$$\mathcal{H} = -\sum_{xy} 2J\mathbf{S}_i\cdot\mathbf{S}_j - \sum_{z} -2K\mathbf{S}_i\cdot\mathbf{S}_j \qquad (2.1)$$

for the ferromagnetic case, and the Hamiltonian

$$\mathcal{H} = \sum_{xy} 2J\mathbf{S}_i\cdot\mathbf{S}_j + \sum_{z} 2K\mathbf{S}_i\cdot\mathbf{S}_j \qquad (2.2)$$

for the antiferromagnetic case, where $\sum_{xy}$ is the sum over all nearest-neighbor pairs $\mathbf{S}_i$ and $\mathbf{S}_j$ with connections in the x and y directions, and where $\sum_z$ is the sum over all nearest neighbors with connections in the z direction. We shall refer to this structure as a layer structure, reserving the term simple-cubic lattice for the case when $K=J$. In considering the problem for a range of values of $\gamma=K/J$ we are able to include as special cases the plane quadratic layer ($\gamma=0$) and the sc lattice ($\gamma=1$). Also, for a number of the approximations to be used, the results for the case $\gamma=2$ are either exactly, or very nearly, equivalent to those for the body-centered cubic (bcc) lattice.

The simplest of all the approximations which have been used to treat the magnetic cooperative problem is the molecular-field method,[11] and we may very easily apply it to the present problem. Let us first consider the ferromagnetic problem. We replace the Hamiltonian (2.1) by $\frac{1}{2}\sum_i \mathcal{H}_i$ where

$$\mathcal{H}_i = -8J\mathbf{S}_i\cdot\langle\mathbf{S}\rangle - 4K\mathbf{S}_i\cdot\langle\mathbf{S}\rangle, \qquad (2.3)$$

where $\langle\mathbf{S}\rangle$ is the time-average value of the spin on each lattice site. We thus replace each neighbor spin $\mathbf{S}_j$ by its time average value and the spin $\mathbf{S}_i$, therefore, behaves as if it were in an "effective" magnetic field $\mathbf{H}_{\text{eff}}$ given by

$$\mathbf{H}_{\text{eff}} = (8J+4K)\langle\mathbf{S}\rangle/g\beta, \qquad (2.4)$$

where g is the Landé factor and β the Bohr magneton.

The time-average value of a spin $\mathbf{S}_i$ in a field $\mathbf{H}_{\text{eff}}$ at a temperature T is well known from the theory of paramagnetism and is

$$\langle\mathbf{S}_i\rangle = \langle\mathbf{S}\rangle = SB_s[g\beta S\mathbf{H}_{\text{eff}}/kT], \qquad (2.5)$$

where S is the spin quantum number and B_s is the Brillouin function for spin S. Near the Curie temperature T_c, $\langle\mathbf{S}\rangle \to 0$ and hence $\mathbf{H}_{\text{eff}} \to 0$. In this limit Eq. (2.5) may be written

$$\langle\mathbf{S}\rangle = \frac{g\beta S(S+1)\mathbf{H}_{\text{eff}}}{3kT} = \frac{S(S+1)(8J+4K)\langle\mathbf{S}\rangle}{3kT}, \qquad (2.6)$$

[11] J. H. Van Vleck, J. Chem. Phys. **9**, 85 (1941).

and hence

$$kT_c=S(S+1)(8J+4K)/3. \quad (2.7)$$

For the antiferromagnetic case we proceed in an exactly similar manner, but this time the spins are separated into two sublattices with a spin $\mathbf{S}_i$ having all its nearest neighbors $\mathbf{S}_j$ on the opposite sublattice such that $\langle\mathbf{S}_i\rangle=\langle-\mathbf{S}_j\rangle=\langle-\mathbf{S}\rangle$. Using the Hamiltonian (2.2) we find

$$\mathbf{H}_{\text{eff}}=-(8J+4K)\langle\mathbf{S}\rangle/g\beta, \quad (2.8)$$

and in the limit of T going to the Néel point T_n we have

$$\langle\mathbf{S}_i\rangle=\langle-\mathbf{S}\rangle=\frac{g\beta S(S+1)\mathbf{H}_{\text{eff}}}{3kT}=\frac{-S(S+1)(8J+4K)\langle\mathbf{S}\rangle}{3kT}, \quad (2.9)$$

and hence

$$kT_n=S(S+1)(8J+4K)/3, \quad (2.10)$$

which is exactly the result obtained for kT_c from Hamiltonian (2.1).

In the present paper we shall concern ourselves primarily with the spin $\frac{1}{2}$ case; the above results reducing, for this case, to

$$kT_c/J=kT_n/J=2+K/J=2+\gamma. \quad (2.11)$$

As previously noted, the values $\gamma=0$ and $\gamma=1$ have a particular significance representing, respectively, the set of quadratic layers and the sc lattice. We may also easily verify that, in the molecular-field approximation, the result for $\gamma=2$ is exactly that which the method would predict for the bcc lattice. In other words, the present method is not able to distinguish between the $\gamma=2$ case and the bcc lattice.

The weakness of the molecular-field method for calculating Curie temperatures may now be observed by comparing the $\gamma=1$ and $\gamma=2$ values with those obtained by Rushbrooke and Wood[2] for the sc and bcc spin $\frac{1}{2}$ cases. We find that the molecular-field results are respectively factors of 1.76 and 1.54 too high. A fundamental weakness is also indicated by the $\gamma=0$ case for which spin-wave methods indicate that the correct values for T_c and T_n are almost certainly zero. It is evident that the molecular-field results are poor, and become progressively poorer as γ decreases until we get a complete breakdown in the limit of isolated layers.

It is clear that the problem of calculating transition temperatures for lattices of weakly interacting layers can only adequately be treated by a method which is at least able to predict the instability of long-range order in plane-layer lattices. This requirement immediately rules out the Opechowski method which would seem to indicate,[2,5] for the layer lattices, a normal series convergence behaviour leading to finite transition temperatures. A value $kT_c/J\doteqdot 1.0$ is reported[5] for the quadratic-layer lattice. Because of this unsatisfactory behaviour in the $\gamma=0$ limit, and bearing in mind the unsuitability of the method for locating antiferromagnetic transition temperatures in general, we conclude that the method is not suited for attacking the layer problem and we have not pursued it for the present problem.

3. THE BETHE-PEIERLS-WEISS METHOD

One well-tried approach to the magnetic cooperative problems which does correctly predict no long-range order for the hexagonal and the quadratic-layer lattices is the cluster method of Bethe-Peierls-Weiss. The method was applied to ferromagnetism by Weiss,[8] and to antiferromagnetism by Li[9], and has since been used by many authors. Although, as Anderson has pointed out,[12] the BPW criterion for the existence of long-range order is not, in any obvious way, connected with dimensionality (but only with certain topological conditions concerning near neighbors), the fact that it does give the correct results for the above-mentioned layer structures tempts us to consider it for our case.

In the BPW method one considers a cluster of spins in the effective field of the rest of the lattice. The cluster is usually taken to include a centre spin and all of its nearest neighbors. The exchange interactions within the cluster are treated in a correct quantum-mechanical manner, but the interactions of the cluster with the rest of the lattice are replaced by internal fields.

In the simple one-exchange parameter problems only one internal field is required and hence only one condition concerning it is needed to determine the problem. This condition, for ferromagnetism, is usually taken to be that the average component of spin on a centre site should be equal to that on a first-shell site, both of which can be written down in terms of the partition function for the cluster. This condition enables T_c to be located without further assumption concerning the internal field.

The layer problem of Sec. 2 can, in principle, be treated in an exactly similar way. Because of the two different exchange parameters it is necessary to introduce two different internal fields. One can, however, choose two conditions to determine the problem by equating the average moment of a center spin, in turn, to that of an xy-plane neighbor spin, and to that of a z-axis neighbor spin. The task of calculating the partition-function and inserting the consistency conditions, however, proves to be considerably more difficult than for the single-parameter case. Because of this, we have simplified the problem by reducing the size of the cluster to exclude the z-axis neighbors. This obviously necessitates a modification of one of the consistency conditions. We proceed as follows, treating firstly ferromagnetism and considering only the case of spin half and of zero external magnetic field.

Taking a cluster of spins consisting of a center spin s_0 and its four nearest-neighbor xy spins, we treat the interactions within the cluster in a proper quantum-

[12] P. W. Anderson, Phys. Rev. **80**, 922 (1950).

mechanical manner, but replace the interactions between the cluster and the rest of the lattice by effective internal fields. For the Hamiltonian of the cluster we write

$$\mathcal{H}_c=-2J\mathbf{s}_0\cdot\mathbf{S}_1-s_0{}^zH_0-S_1{}^zH_1, \tag{3.1}$$

where S_1 is the total spin of the four first-shell neighbors, and where H_0 and H_1 (we write H in place of $g\beta H$ for brevity) are the z components of the internal fields acting, respectively, upon the centre and first-shell spins. The problem of diagonalizing a Hamiltonian of the form (3.1) has been discussed, for the case of spin $\frac{1}{2}$, by Weiss[8], who gives the eigenvalues $E(S_1,S,S^z)$, where S is the total spin of the cluster, as a power series in $H=H_1-H_0$ of the form

$$E(S_1,S_1\pm\tfrac{1}{2},m)=E_\pm-mH_0+\sum_{n=1}^{\infty}e_n{}^\pm H^n, \tag{3.2}$$

where

$$E_+=-JS_1,\quad E_-=J(S_1+1), \tag{3.3}$$

$$e_1{}^\pm=-m\left[1\mp\frac{1}{2S_1+1}\right], \tag{3.4}$$

and

$$e_2{}^\pm=\frac{\mp1}{4J(2S_1+1)}\left[1-\frac{4m^2}{(2S_1+1)^2}\right]. \tag{3.5}$$

The partition function for the cluster may be written in the form

$$P(H_0,H_1,T)=\sum_{S_1}\omega(S_1)\sum_{\pm}\sum_{m=-S}^{S}\exp[-E(S_1,S,m)/kT], \tag{3.6}$$

where S_1 can take the values 0, 1, 2; where $\sum_\pm$ is over the values $S=S_1\pm\frac{1}{2}$, and where $\omega(0)=2$, $\omega(1)=3$, $\omega(2)=1$. The average values of spin on a centre and on a first-shell site are, respectively,

$$\bar{m}_c=g\beta kT\frac{\partial}{\partial H_0}\ln(P), \tag{3.7}$$

and

$$\bar{m}_s=\tfrac{1}{4}g\beta kT\frac{\partial}{\partial H_1}\ln(P), \tag{3.8}$$

and, requiring these to be equal as the condition for ferromagnetism, we look for a solution in the limit of the effective fields approaching zero ($T\rightarrow T_c$). Detailed calculation gives

$$\sum_{S_1}\omega(S_1)\sum_{\pm}\sum_{m}(a_0{}^\pm+a_1{}^\pm/kT)\exp[-E_\pm/kT]=0, \tag{3.9}$$

where

$$a_0{}^\pm=2e_2{}^\pm, \tag{3.10}$$

and

$$a_1{}^\pm=me_1{}^\pm(\lambda-4/5)+(4/5)m^2\lambda-(e_1{}^\pm)^2, \tag{3.11}$$

and where we have introduced the parameter

$$\lambda=H_0/(H_1-H_0). \tag{3.12}$$

To determine the problem, we need only specify the ratio H_0/H_1. Using the internal-field concept, we assume that we may write equations of the form

$$H_0=2CK,\qquad H_1=C(3J+2K), \tag{3.13}$$

where C is a temperature-dependent proportionality constant. We have, therefore,

$$\lambda=2K/3J=2\gamma/3. \tag{3.14}$$

A detailed examination of Eq. (3.9) shows that, for a general value of γ, it possesses either two real solutions (representing the Curie point and the well-known anti-Curie point) or no real solutions. In Fig. 1 we show a plot of Curie temperature versus γ which has been obtained by solving the Eq. (3.9) by machine. We observe that the BPW method indicates a sudden disappearance of ordered states for values of γ less than a limiting value which has been computed to be $\gamma=0.154$. At this point of minimum inter-plane interaction for the onset of long-range order, we find a value of $kT_c/J=0.623$.

For the larger values of γ we find that the anti-Curie temperature is many times smaller than T_c so that, for these cases, unphysical behaviour does not occur until the temperature is well below T_c. As γ decreases, however, T_c decreases and the anti-Curie temperature increases, so that the unphysical behaviour sets in at temperatures nearer and nearer to T_c. Finally, when $\gamma=0.154$, the two critical temperatures coincide, and for still smaller values of the interplane interactions, the unphysical behaviour sets in at temperatures above T_c in such a way that the method incorrectly predicts the absence of long-range order in the three-dimensional lattice. For $\gamma=1$, and for $\gamma=2$, the results obtained from the present method are very close to those obtained by Weiss[8] for the sc and the bcc lattices taking clusters of seven and nine spins, respectively. The values

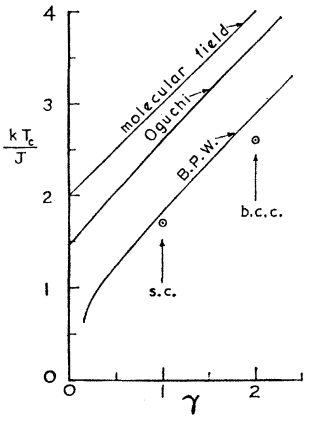

FIG. 1. The Curie temperature for spin $\frac{1}{2}$ calculated as a function of γ $(=K/J)$ by using the molecular field, Oguchi, and Bethe-Peierls-Weiss methods. The ringed points show the Rushbrooke and Wood[2] values for the sc and bcc cases.

TABLE I. Values for kT_c/J.

	sc	bcc
Rushbrooke and Wood[a]	1.7	2.60
Weiss[b]	1.85	2.91
This section	1.82	2.89

[a] See Ref. 2.
[b] See Ref. 8.

(see table 1) are also within ~10% of the Rushbrooke and Wood[2] results.

If we used a five-spin cluster to investigate the transition temperature for the bcc lattice, we should proceed exactly as in Eqs. (3.1) to (3.12), but we should relate the parameter λ to the exchange J by putting H_0 proportional to $4J$, and H_1 proportional to $7J$. This gives a value $\lambda=\frac{4}{3}$, and from (3.14) we see that this is just the value which is required for the $\gamma=2$ case in the layer problem. Thus, the five-spin cluster method is not able to distinguish the $\gamma=2$ case from the bcc problem. It is also very easy to show that, for the larger values of γ in the layer problem, the present method gives results which approach the molecular-field ones as γ increases.

For the antiferromagnetic case we may proceed in a very similar manner using the same cluster and the same notation as for the ferromagnetic problem, but reversing the sign of the exchange interactions. Once again we obtain a partition function in the form (3.6), and the equations (3.1) to (3.5) are still valid but with the opposite sign for J. We now distinguish two different types of lattice site, the "spin up" or α sites, and the "spin down" or β sites. For a cluster with a center α site and first-shell β sites, we write a partition-function

$$P_{\alpha\beta}=P(H_0^{\alpha},H_1^{\beta},T), \tag{3.15}$$

and for a cluster with centre β and first-shell α sites, we write

$$P_{\beta\alpha}=P(H_0^{\beta},H_1^{\alpha},T). \tag{3.16}$$

The average magnetic moments on the α and β centre sites may be expressed in terms of these partition functions in the form

$$\bar{m}_c^{\alpha}=g\beta kT\frac{\partial}{\partial H_0^{\alpha}}\ln(P_{\alpha\beta}),\quad \bar{m}_c^{\beta}=g\beta kT\frac{\partial}{\partial H_0^{\beta}}\ln(P_{\beta\alpha}), \tag{3.17}$$

and, in the same way, we have for the equivalent first-shell moments

$$\bar{m}_s^{\alpha}=\tfrac{1}{4}g\beta kT\frac{\partial}{\partial H_1^{\alpha}}\ln(P_{\beta\alpha}),\quad \bar{m}_s^{\beta}=\tfrac{1}{4}g\beta kT\frac{\partial}{\partial H_1^{\beta}}\ln(P_{\alpha\beta}). \tag{3.18}$$

If we write consistency conditions $\bar{m}_c^{\alpha}=\bar{m}_s^{\alpha}$, and $\bar{m}_c^{\beta}=\bar{m}_s^{\beta}$, we find that there exist solutions for which $H_0^{\alpha}=-H_0^{\beta}$, and $H_1^{\alpha}=-H_1^{\beta}$. For this case, in the limit of the internal fields going to zero ($T\rightarrow T_n$), we again obtain an equation of the form (3.9), but where now

$$a_0^{\pm}=2e_2^{\pm}, \tag{3.19}$$

$$a_1^{\pm}=-me_1^{\pm}(\lambda+\tfrac{4}{3})-(\tfrac{4}{3})m^2\lambda-(e_1^{\pm})^2, \tag{3.20}$$

and where this time the parameter λ is defined by

$$\lambda=H_0^{\alpha}/(H_1^{\alpha}+H_0^{\alpha})=H_0^{\beta}/(H_1^{\beta}+H_0^{\beta}). \tag{3.21}$$

Using Eq. (3.13), we have

$$\lambda=2K/(3J+4K),\qquad \gamma=3\lambda/(2-4\lambda). \tag{3.22}$$

Again we find that the consistency equation (3.9) has either two real solutions or none. The critical values of γ and Néel temperature below which no long-range order occurs are this time found to be $\gamma=0.765$ and $kT_n/J=1.439$. The situation concerning the Néel and anti-Néel temperatures is similar to that for the Curie and anti-Curie temperatures in the ferromagnetic case, but the detailed solution of (3.9) for T_n (shown in Fig. 2) shows clearly that the BPW method is considerably less suited for dealing with the antiferromagnetic problem. We see that there is a very large range of γ values for which the method breaks down completely, this range being almost five times the size of the equivalent ferromagnetic one.

If one goes, in the series expansion (3.2), to higher terms than those so far considered, it is possible to obtain expressions for the internal fields near to the transition temperature (when it exists), and to relate these fields to the average sublattice spin. We find that the relationship is not the simple molecular-field one of the type used in Sec. 2. It follows that such a simple molecular-field relationship between the cluster and the rest of the lattice would not satisfy the BPW consistency conditions. Nevertheless, an approximation is sometimes used (and was introduced by Oguchi[13]) which uses just this molecular-field criterion to obtain an approximate solution for the cluster problem. Thus,

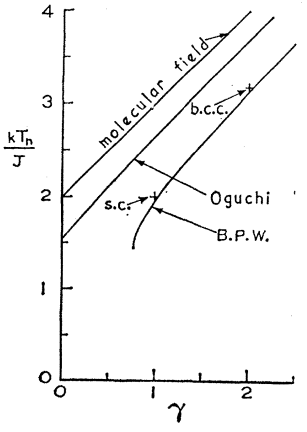

FIG. 2. The Néel temperature for spin $\frac{1}{2}$ calculated as a function of γ ($=K/J$) by using the molecular field, Oguchi, and Bethe-Peierls-Weiss methods. The crosses indicate the values obtained by Li[9] for the sc and bcc cases.

[13] T. Oguchi, Progr. Theoret. Phys. (Kyoto) 13, 148 (1955).

interactions within the cluster are treated correctly, but the interactions between the cluster and the rest of the lattice are replaced by effective fields which are related to the average lattice (or sub-lattice) magnetization by the molecular-field condition.

In the remaining part of this section we have applied the Oguchi method to the cluster used above for the BPW method. For the ferromagnetic case, using the equations (3.1) to (3.7), we write $\bar{m}_c=g\beta\bar{S}$ and obtain

$$\bar{S}=kT\partial/\partial H_0 \ln (P). \tag{3.23}$$

Replacing H_1-H_0 by its molecular-field value $6J\bar{S}$, we may again obtain an equation of the form (3.9) but where now

$$a_0^{\pm}=1-12Je_2^{\pm}, \tag{3.24}$$

and

$$a_1^{\pm}=-J(m+e_1^{\pm})(4m\gamma-6e_1^{\pm}). \tag{3.25}$$

This equation may be solved for T_c as a function of γ and the results are shown in Fig. 1. No anti-Curie temperature occurs, there being only one real solution for each value of γ. In general, however, we see that the results are considerably poorer than those obtained by the BPW method and, though better than the molecular-field values, they retain the latter's weakness of predicting long-range order in the quadratic layer.

Similar results are obtained by applying the Oguchi conditions to the antiferromagnetic case. Letting the spin on a centre α site have an average value $\bar{S}$, we have

$$\bar{S}=kT\frac{\partial}{\partial H_0{}^{\alpha}}\ln(P_{\alpha\beta}). \tag{3.26}$$

Replacing $H_0{}^{\alpha}$ by $4K\bar{S}$, and $H_1{}^{\beta}$ by $-(6J+4K)\bar{S}$, we may again obtain Eq. (3.9) but where

$$a_0^{\pm}=1+2e_2^{\pm}(6+8\gamma)J, \tag{3.27}$$

and

$$a_1^{\pm}=-J(m+e_1^{\pm})[4m\gamma+(6+8\gamma)e_1^{\pm}]. \tag{3.28}$$

Solving this equation for T_n as a function of γ we get just one solution for each γ, and the detailed results are shown in Fig. 2. Again we see that in the region for which both the BPW method and the Oguchi method give results, the BPW values are almost certainly the better. In the weakly interacting layer region where the BPW method breaks down, we can have little confidence in the Oguchi results because of the incorrect prediction of long-range order for the quadratic-layer ($\gamma=0$) case. We must conclude that neither of the cluster methods discussed in this section is able to give a reliable estimate of transition temperature for the case of weakly interacting layers.

Some improvement could certainly be brought about by performing a proper BPW approximation on the full seven-spin cluster (formed by a centre spin and its six nearest neighbors) instead of treating a smaller cluster as the work of the present section does. One obvious deficiency of the smaller cluster method which could be overcome in this way is its inability to distinguish between the $\gamma=2$ case and the bcc lattice. Since, however, the breakdown for small values of γ stems directly from the occurrence in the method of an antitransition temperature, and this unphysical behaviour exists also when the full cluster is used,[8,9] the method remains basically unsuited for dealing with problems concerning weakly interacting layers.

4. THE CONSTANT-COUPLING APPROXIMATION

A cluster method which is mathematically less complex than the BPW method and the results of which are, at least for the sc and the bcc lattices, almost identical with the latter, was developed by Kasteleijn and Van Kranendonk[10] and called by them the constant-coupling approximation. It deals with a simple pair cluster and has, for ferromagnetism, an advantage over BPW in that it does not exhibit unphysical behaviour at low temperatures. For antiferromagnetism the method is less satisfactory when an anti-Néel temperature does occur. For the spin $\frac{1}{2}$ case, which is our main concern, the method correctly predicts a disordered quadratic-layer lattice but, like the BPW method, its criterion for the existence of an ordered state has no obvious connection with dimensionality; and it is, for example, incorrect in predicting a long-range order for the hexagonal-layer lattice.

In this section, we treat the layer problem using the constant coupling method. We consider only the spin $\frac{1}{2}$ case for ferromagnetism and for antiferromagnetism. For both cases we have followed closely the arguments of Kasteleijn and Van Kranendonk,[10] but we have modified the work to allow an approximation to be made for use with Hamiltonians containing more than one exchange parameter. In the original work[10] the cooperative problem is reduced to the evaluation of E, the average value of energy of states of the N-spin system which have a given value S of the z component of total spin. For the one-parameter problem (nearest-neighbor isotropic exchange J) this energy E is shown to be of the form

$$E=\tfrac{1}{2}Nz\,\mathrm{Tr}(\rho_J\mathcal{H}_J), \tag{4.1}$$

where ρ_J is a density matrix of an ensemble of pairs of spins, where $\mathcal{H}_J$ is the Hamiltonian for a neighboring pair of spins, and where z is the number of nearest neighbors of any one spin. In general, ρ_J is an extremely complicated function of both S and temperature, and the problem is made tractable by approximating it by a particularly simple form (the constant-coupling approximation) which is correct in the limit of high temperatures.

In the two-parameter layer problem, as set out in Sec. 2, we again have isotropic nearest-neighbor interactions, and we may write the average energy E (defined above) in the form

$$E=\tfrac{1}{2}Nz_J\,\mathrm{Tr}(\rho_J\mathcal{H}_J)+\tfrac{1}{2}Nz_K\,\mathrm{Tr}(\rho_K\mathcal{H}_K), \tag{4.2}$$

where ρ_J and $\mathcal{H}_J$ have the same meanings as in Eq. (4.1), where ρ_K and $\mathcal{H}_K$ differ from these only in referring to pairs of spins with exchange interactions K instead of J, and where z_J and z_K are the number of nearest neighbors with exchange interactions J and K, respectively ($z_J=4$, $z_K=2$, for the problem in question). If we now approximate ρ_J as for the one parameter problem, and we make for ρ_K an exactly equivalent approximation (with K in place of J), we may go on to evaluate transition temperatures for both the ferromagnetic and antiferromagnetic cases. We omit the details of the calculation, which exactly parallels the original,[10] and give only the resulting equations which are, for ferromagnetism

$$2e^{-y}+e^{-\gamma y}=1, \tag{4.3}$$

where $y=2J/kT_c$, and for antiferromagnetism

$$10(e^{y}-1)(e^{\gamma y}-1)-2y(e^{y}+3)(e^{\gamma y}-1)-\gamma y(e^{\gamma y}+3)(e^{y}-1)=0, \tag{4.4}$$

where $y=2J/kT_n$.

Equation (4.3) has one real solution for T_c for every real value of γ and the detailed solution is plotted in Fig. 3. We see that the constant-coupling method gives acceptable results right down to the smallest values of interplane interaction where it correctly shows $T_c\to 0$ as $\gamma\to 0$. That this correct limiting behaviour is fortuitous is, however, immediately evident from the work of Kasteleijn and Van Kranendonk. They show that, in the simple single-exchange ferromagnetic problem for the case of spin $\frac{1}{2}$, the condition for ferromagnetism is just that the number of nearest neighbors should be greater than four. The method would therefore give incorrect results both for the hexagonal-layer lattice and also for the diamond structure. Also, for values of spin $S>\frac{1}{2}$ the method predicts a finite Curie temperature for the $\gamma=0$ case. Thus, although the results are acceptable for the present example and, as such, are the best which we have so far obtained, we nevertheless ought not to put too much trust in the quantitative manner of the approach of T_c to zero for the $\gamma\to 0$ limit. One pleasing result, however, is that the constant-coupling approximation is able to distinguish between the $\gamma=2$ case and the bcc lattice. For the latter case the equation for the Curie-temperature is [10]

$$e^{y}=2, \tag{4.5}$$

from which it follows that $kT_c/J=2.89$, and this may be compared with the result for $\gamma=2$ which, from Eq. (4.3), is $kT_c/J=2.27$. We have here a demonstration of the greater stability of ferromagnetism when the exchange interactions are symmetrically distributed in three dimensions.

Comparing Fig. 1 and Fig. 3 we see that the shapes of the BPW and constant-coupling curves are quite different. Although the methods are in good agreement at $\gamma=1$ (the sc case), we find that, in general, the latter method gives the smaller Curie temperatures when

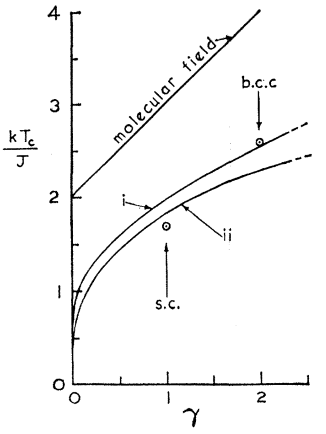

FIG. 3. The Curie temperature for spin $\frac{1}{2}$ calculated as a function of $\gamma(=K/J)$ by using the method of Green functions (curve i), and the constant-coupling approximation (curve ii). The ringed points show the Rushbrooke and Wood[2] values for the sc and bcc cases.

$\gamma>1$ and the larger Curie temperatures when $\gamma<1$. There is good reason to suppose that this difference in shape is largely due to the smallness of the cluster which was used in Sec. 3. We should almost certainly expect the full seven-spin cluster BPW treatment to be in a better agreement with the constant-coupling method, particularly so for the higher γ values where the method using the smaller five-spin cluster replaces the major interactions by effective fields.

As was the case for the BPW method, we again find that the approximation is less satisfactory for antiferromagnetism. The Eq. (4.4), like its BPW counterpart, has either two real solutions or none, thus providing us with both a Néel and an anti-Néel temperature. The detailed solution of (4.4) for the Néel temperature is plotted as kT_n/J against γ in Fig. 4, and the transition temperature becomes imaginary for values of γ below 0.307 (which may be compared with the value $\gamma=0.765$ for the BPW approximation). The Néel temperature, as calculated from (4.4), also becomes imaginary for large γ values. This is the breakdown for the weakly interacting chains problem, but we shall not consider it further in the present paper. At the limiting value $\gamma=0.307$, the Néel temperature is given by $kT_n/J=1.102$. We may again note the difference between the $\gamma=2$ case ($kT_n/J=2.37$) and the bcc case, for which the transition temperature may be calculated from[10]

$$7(e^{y}-1)-2y(e^{y}+3)=0, \tag{4.6}$$

and is $kT_n/J=3.16$.

We have now exhausted the well-tried cluster techniques for dealing with the magnetic cooperative problem and we have found that, with the possible exception of the constant-coupling approach for spin $\frac{1}{2}$ and ferromagnetic-interacting quadratic layers, there is no cluster method which can deal with the problem of weakly interacting layers.

It is clear that we must turn from the cluster techniques and look for a method for which the criterion for the existence of long-range order is one concerning dimensionality. One such method has recently been

investigated by Bogolyubov and Tyablikov,[14] and by Tahir-Kheli and ter Haar.[15] It is the method of Green functions. Another is the spherical model which has been investigated by Lax.[16] We shall discuss only the former, although the expressions for transition temperatures which we shall derive [Eqs. (5.18) and (5.33)] could also be obtained from the spherical approximation.

5. THE METHOD OF GREEN FUNCTIONS

Recently, a number of authors have used the properties of the double-time temperature-dependent Green functions in order to attack the magnetic-cooperative problem. They find, even using a simple random-phase decoupling approximation, that a formula may be obtained for magnetization which is valid over the entire range of temperature, and which is in reasonable agreement both at low temperatures with the spin-wave theories, and also at high temperatures with the exact expansion method. The spin-wave-like behavior at low temperatures means that the Green function method, in contrast with the methods previously discussed, is able to take into account the spatial positions of the spins; and to distinguish, for example, between the diamond lattice and the quadratic-layer lattice. We also find that the conditions for the existence of long-range order are the spin-wave conditions which are the best which have yet been obtained for Heisenberg ferromagnets and antiferromagnets.

The double-time temperature-dependent Green functions have been discussed at length by Zubarev.[17] The Green function method for magnetism requires only two of the relations concerning them, and we shall write these down without discussion referring the reader to Zubarev[17] or to Bonch-Bruevich and Tyablikov[18] for their derivation. We shall denote the Fourier transform of the Green function involving the Heisenberg operators $A(t)$ and $B(t')$ by $\langle\langle A\,;B\rangle\rangle$. The equation of motion for this function may be written

$$E\langle\langle A\,;B\rangle\rangle=\frac{1}{2\pi}\langle[A,B]_-\rangle+\langle\langle[A,\mathcal{H}]_-\,;B\rangle\rangle, \quad (5.1)$$

where the single pointed brackets indicate averages over a canonical ensemble, and where the square brackets indicate commutation relationships as follows

$$[A,B]_-\equiv AB-BA. \quad (5.2)$$

From the analytical properties of the Green functions, the transforms may be shown to be directly related to correlation functions in the following way

$$\langle B(t')A(t)\rangle=\lim_{\epsilon\to+0} i \int_{-\infty}^{\infty}\frac{\langle\langle A\,;B\rangle\rangle_{E=\omega+i\epsilon}-\langle\langle AB\rangle\rangle_{E=\omega-i\epsilon}}{e^{\omega/kT}-1}\times e^{-i\omega(t-t')}d\omega \quad (5.3)$$

Equations (5.1) and (5.3) are the only basic equations from the Green function theory which will be required for the present calculations.

We shall again investigate the layer problem of ferromagnetism and of antiferromagnetism using the same defining Hamiltonians as for the previous sections [that is, Eqs. (2.1) and (2.2)]. Let us first consider the ferromagnetic case. We shall investigate the motion of the function $\langle\langle S_g^+\,;S_h^-\rangle\rangle$ where S_g^+ stands for the operator S_x+iS_y for the spin at site g, and where S_h^- stands for the operator S_x-iS_y for the spin at site h. Using the Hamiltonian (2.1) together with the familiar commutation relationships for the components of spin, the equation of motion for $\langle\langle S_g^+\,;S_h^-\rangle\rangle$ is

$$E\langle\langle S_g^+\,;S_h^-\rangle\rangle=\frac{F\delta_{gh}}{2\pi}-\Big(\sum_{xy}{}^{j}2J+\sum_{z}{}^{j}2K\Big)\times\langle\langle(S_g^zS_j^+-S_g^+S_j^z)\,;S_h^-\rangle\rangle, \quad (5.4)$$

where $\sum_{xy}{}^{j}$ means the sum over the four nearest neighbors j of g with connections in an xy plane, where $\sum_{z}{}^{j}$ means the sum over the two nearest neighbors j of g with connections in the z direction, where δ_{gh} is the Kronecker delta, and where

$$F=\langle[S_g^+,S_g^-]_-\rangle. \quad (5.5)$$

Using the simplest random phase approximation, we 'decouple' this equation by writing

$$\langle\langle S_g^zS_j^+\,;S_h^-\rangle\rangle=\langle S_g^z\rangle\langle\langle S_j^+\,;S_h^-\rangle\rangle, \quad (5.6)$$

$$\langle\langle S_g^+S_j^z\,;S_h^-\rangle\rangle=\langle S_j^z\rangle\langle\langle S_g^+\,;S_h^-\rangle\rangle. \quad g\neq j \quad (5.7)$$

Putting $\langle S_g^z\rangle=\langle S_j^z\rangle=\bar{S}$, the equation of motion becomes

$$E\langle\langle S_g^+\,;S_h^-\rangle\rangle=\frac{F\delta_{gh}}{2\pi}-\bar{S}\Big(\sum_{xy}{}^{j}2J+\sum_{z}{}^{j}2K\Big)\times(\langle\langle S_j^+\,;S_h^-\rangle\rangle-\langle\langle S_g^+\,;S_h^-\rangle\rangle). \quad (5.8)$$

Using the translational invariance of the lattice we Fourier transform the Green functions with respect to the reciprocal lattice and define the function $G_{\mathbf{K}}$ by the equations

$$\langle\langle S_g^+\,;S_h^-\rangle\rangle=(1/N)\sum_{\mathbf{K}}G_{\mathbf{K}}e^{i\mathbf{K}\cdot(\mathbf{g}-\mathbf{h})}, \quad (5.9)$$

$$G_{\mathbf{K}}=\sum_{\mathbf{g}-\mathbf{h}}\langle\langle S_g^+\,;S_h^-\rangle\rangle e^{-i\mathbf{K}\cdot(\mathbf{g}-\mathbf{h})}, \quad (5.10)$$

[14] N. N. Bogolyubov and S. V. Tyablikov, Dokl. Acad. Nauk. SSSR **126**, 53 (1959) [translation: Soviet Phys.—Doklady 4, 589 (1959)].
[15] R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. **127**, 88 and 95 (1962).
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[18] V. L. Bonch-Bruevich and S. V. Tyablikov, *The Green Function Method in Statistical Mechanics* (North-Holland Publishing Company, Amsterdam, 1961).

where N is the number of spins in the lattice, where $\mathbf{g}$ and $\mathbf{h}$ are the position vectors of the spins g and h, and where $\mathbf{K}$ is a reciprocal lattice vector which may take on N values (allowed by periodic boundary conditions) in the first Brillouin zone of the reciprocal lattice. Expressed in terms of $G_{\mathbf{K}}$, the equation of motion becomes

$$G_{\mathbf{K}}=F/2\pi(E-E_0'), \tag{5.11}$$

where

$$E_0'=\bar{S}E_0=\bar{S}[8J+4K-2J\sum_{xy}{}^{j}e^{i\mathbf{K}\cdot(\mathbf{j}-\mathbf{g})} -2K\sum_{z}{}^{j}e^{i\mathbf{K}\cdot(\mathbf{j}-\mathbf{g})}]. \tag{5.12}$$

Using the identity

$$\lim_{\epsilon\to+0}\left[\frac{1}{\omega+i\epsilon-E}-\frac{1}{\omega-i\epsilon-E}\right]\equiv-2\pi i\delta(\omega-E), \tag{5.13}$$

together with (5.3), in the limit $t-t'\to 0$, we find

$$\langle S_g^- S_g^+\rangle=S(S+1)-\bar{S}-\langle S_z^2\rangle=\left\langle\frac{F}{e^{E_0'/kT}-1}\right\rangle_{\mathbf{K}}, \tag{5.14}$$

where $\langle\cdots\rangle_{\mathbf{K}}$ is the average value when $\mathbf{K}$ runs over its N allowed values in the first Brillouin zone.

For the case of spin $\frac{1}{2}$, we have $\langle S_z^2\rangle=\frac{1}{4}$, and we may use (5.14) to give us an expression for the average value of spin as a function of temperature. We obtain

$$\bar{S}=\tfrac{1}{2}(1+2\Phi), \tag{5.15}$$

where

$$\Phi=\langle 1/(e^{E_0'/kT}-1)\rangle_{\mathbf{K}}. \tag{5.16}$$

As $T\to T_c$ from below, $\bar{S}$ becomes vanishingly small and (5.15) reduces to

$$kT_c=1/\langle 4/E_0\rangle_{\mathbf{K}}, \tag{5.17}$$

which becomes, on using the familiar reciprocal lattice vectors for the simple-cubic structure,

$$J/kT_c=\left\langle\frac{1}{2-c_1-c_2+\gamma(1-c_3)}\right\rangle_{\mathbf{K}}, \tag{5.18}$$

where

$$c_1=\cos(K_x),\ c_2=\cos(K_y),\ c_3=\cos(K_z), \tag{5.19}$$

and where the average $\langle\cdots\rangle_{\mathbf{K}}$ is now over values of K_x, K_y, and K_z, each running between $-\pi$ and π. In the limit of very large N we replace the average by an equivalent integral which may be evaluated to a good approximation by computer. The resulting values for kT_c/J as a function of γ are shown in Fig. 3. We note that in the limit $\gamma\to 0$, then $T_c\to 0$ as demanded by spin-wave theory, the right-hand side of (5.18) diverging along the line $c_1=c_2=1$.

The antiferromagnetic case may be treated in a very similar manner. The equation of motion for the Green functions is again given by (5.8), but where now we must change the sign of the exchange parameters K and J. Decoupling as in (5.6) and (5.7) we now proceed by separating the lattice into two sublattices, the "up" and the "down" sublattices with average spin vaues $\bar{S}$ and $-\bar{S}$ on the respective sites. Transforming the Green functions with respect to the reciprocal, sublattice we define the functions $G_{1\mathbf{K}}$ and $G_{2\mathbf{K}}$ in the following way:

a) When g and h are both on the same sublattice,

$$\langle\langle S_g^+;S_h^-\rangle\rangle=(2/N)\sum_{\mathbf{K}}G_{1\mathbf{K}}e^{i\mathbf{K}\cdot(\mathbf{g}-\mathbf{h})}, \tag{5.20}$$

$$G_{1\mathbf{K}}=\sum_{\mathbf{g}-\mathbf{h}}\langle\langle S_g^+;S_h^-\rangle\rangle e^{-i\mathbf{K}\cdot(\mathbf{g}-\mathbf{h})}, \tag{5.21}$$

b) When g and h are on different sublattices,

$$\langle\langle S_g^+;S_h^-\rangle\rangle=(2/N)\sum_{\mathbf{K}}G_{2\mathbf{K}}e^{i\mathbf{K}\cdot(\mathbf{g}-\mathbf{h})} \tag{5.22}$$

$$G_{2\mathbf{K}}=\sum_{\mathbf{g}-\mathbf{h}}\langle\langle S_g^+;S_h^-\rangle\rangle e^{-i\mathbf{K}\cdot(\mathbf{g}-\mathbf{h})}, \tag{5.23}$$

where now $\mathbf{K}$ runs over $N/2$ allowed values in the first Brillouin zone of the reciprocal sublattice. The equation of motion may now be expressed in terms of $G_{1\mathbf{K}}$ and $G_{2\mathbf{K}}$, when we obtain

$$(E-\mu\bar{S})G_{1\mathbf{K}}=F/2\pi+\lambda\bar{S}G_{2\mathbf{K}} \tag{5.24}$$

$$(E+\mu\bar{S})G_{2\mathbf{K}}=-\lambda\bar{S}G_{1\mathbf{K}}, \tag{5.25}$$

where we have taken S_h to be on the "up" sublattice, and where

$$\lambda=2J\sum_{xy}{}^{j}e^{i\mathbf{K}\cdot(\mathbf{j}-\mathbf{g})}+2K\sum_{z}{}^{j}e^{i\mathbf{K}\cdot(\mathbf{j}-\mathbf{g})}, \tag{5.26}$$

and

$$\mu=8J+4K. \tag{5.27}$$

Solving these equations for $G_{1\mathbf{K}}$, we find

$$4\pi G_{1\mathbf{K}}=\frac{(1-A)F}{E+E_0'}+\frac{(1+A)F}{E-E_0'}, \tag{5.28}$$

where

$$A=\mu/(\mu^2-\lambda^2)^{1/2}, \tag{5.29}$$

and

$$E_0'=\bar{S}E_0=\bar{S}(\mu^2-\lambda^2)^{1/2}. \tag{5.30}$$

Using (5.13) and (5.3) and proceeding as for the ferromagnetic case we finally obtain for the sublattice spin

$$1/\bar{S}=2\langle A\coth(E_0'/2kT)\rangle_{\mathbf{K}}, \tag{5.31}$$

and for the Néel temperature

$$kT_n=1/\langle 4A/E_0\rangle_{\mathbf{K}}. \tag{5.32}$$

From (5.26), (5.27), (5.29), and (5.30), we find that the equation for the transition temperature takes the form

$$J/kT_n=\left\langle\frac{2+\gamma}{(2+\gamma)^2-(c_1+c_2+\gamma c_3)^2}\right\rangle_{\mathbf{K}}, \tag{5.33}$$

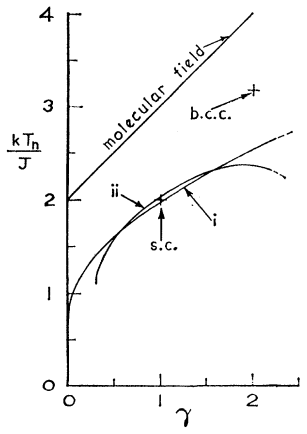

FIG. 4. The Néel temperature for spin $\frac{1}{2}$ calculated as a function of $\gamma(=K/J)$ by using the method of Green functions (curve i), and the constant-coupling approximation (curve ii). The crosses indicate values obtained by Li[9] for the sc and bcc cases.

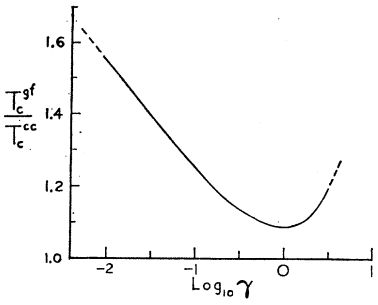

FIG. 5. The ratio of the Curie temperatures as calculated from the Green function and constant-coupling methods plotted as a function of $\log_{10}\gamma$ for the case of spin $\frac{1}{2}$.

where c_1, c_2, and c_3, are as defined in (5.19), and where $\langle\cdots\rangle_K$ is again an average value for K_x, K_y, and K_z each running between $-\pi$ and π. By separating the right hand side of (5.33) into two component fractions, we observe that the results (5.18) and (5.33), for T_c and T_n, respectively, are identical. Thus, like the molecular-field theory, but unlike the cluster methods, the Green function approach predicts that the transition temperatures for "equivalent" ferromagnetic and antiferromagnetic structures are equal. The Green function results for kT_n/J as a function of γ are therefore the same as those shown in Fig. 3 for kT_c/J, and we have reproduced these for the antiferromagnetic case in Fig. 4.

We find, therefore, that the method of Green functions is able to treat the problem of weakly interacting layer structures, and to give results which are qualitatively acceptable for both ferromagnetism and antiferromagnetism. For antiferromagnetism we see from Figs. 2 and 4 that it is the only method from amongst those considered which is even qualitatively satisfactory. Thus, although there is no reason to suppose that the Green function (or spherical model) estimate of Néel temperature for the sc lattice is any more accurate than, for example, the BPW or the constant-coupling approximations, there are very good reasons for preferring it whenever the dominant interactions in a lattice are confined to one or two dimensions. Even for ferromagnetism, where its approximation for the sc case (see Figs. 1 and 3) is certainly inferior to those of the BPW and constant-coupling methods, the Green function method is the only one which gives qualitatively satisfactory values for Curie temperatures in layer problems. The method also has an important additional advantage over the cluster techniques since it is easily able to cope with Hamiltonians containing several different exchange parameters, and it may readily be adapted for use with complicated antiferromagnetic orders and with problems containing anisotropy (see, for example, Lines[19]).

Finally, it is interesting to compare the results of the Green function and constant-coupling methods for the spin $\frac{1}{2}$ ferromagnetic-layer problem (Fig. 3) for which case the latter method fortuitously gives results which are qualitatively acceptable in the limit of weakly interacting layers. In Fig. 5, we plot a graph of the ratio of the Green function Curie temperature T_c^{gf} and the constant-coupling Curie temperature T_c^{cc} against $\log_{10}(\gamma)$. We see that the ratio T_c^{gf}/T_c^{cc} becomes progressively larger as the interplane exchange is reduced. This difference in shape between the curves in Fig. 3 might, perhaps, be explained by saying that the K interactions, when they are small, assume an extra importance, because of their spatial position (out of the xy planes), which the cluster method cannot take into account.

[19] M. E. Lines, Phys. Rev. **131**, 540 (1963).