Onsager reaction-field theory for magnetic models on diamond and hcp lattices

G. M. Wysin

Department of Physics, Kansas State University, Manhattan, KS 66506-2601

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I. INTRODUCTION

The Onsager reaction-field (ORF) theory is extended so that it applies to any three-dimensional Bravais lattice with a basis. The ORF calculation is used to predict the critical temperature for classical Ising, XY, and Heisenberg magnetic models, in particular, on diamond and hexagonal close packed lattices. Results are compared with series extrapolations and other theoretical approaches where available. For the hcp lattice the ORF calculation is seen to be exactly equivalent to a Green’s function approach.

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II. ONSAGER REACTION FIELD CORRECTION: BRAVAIN LATTICES

In the ORF calculation (see Ref. for more details), a spin at a chosen site interacts with the mean-field reduced by a “reaction field” that depends on the spin at that site. For completeness we summarize key aspects of this
calculation here to see how the extension to a non-Bravais lattice is accomplished.

To effect the reaction term, in the real space Hamiltonian an extra self-interaction term is added:

$$\mathcal{H}_{\text{eff}} = \lambda \sum_{n} S_n S_n$$

(2)

This is equivalently a delta-function exchange term of strength $\lambda$. The constant $\lambda$ is the reaction-field, which is determined self-consistently in the calculation, by a constraint on the magnetic susceptibility, below.

The Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{eff}}$, transformed into wavevector space is,

$$\mathcal{H} = -\frac{1}{2} \sum_{\mathbf{q}} [S_{-\mathbf{q}}(J_\mathbf{q} - \lambda) S_\mathbf{q} + (H_{-\mathbf{q}} S_\mathbf{q} + H_{\mathbf{q}} S_{-\mathbf{q}})]$$

(3)

where the Fourier-space quantities derive from

$$S_n = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} S_\mathbf{q} e^{i\mathbf{q} \cdot \mathbf{n}}.$$  \hspace{1cm} (4)

$$J_{n,m} = \frac{1}{N} \sum_{\mathbf{q}} J_\mathbf{q} e^{i\mathbf{q} \cdot (n-m)}.$$  \hspace{1cm} (5)

together with a similar definition for the field $H_\mathbf{q} = H_{-\mathbf{q}}$, for real applied field $H_\mathbf{q}$, $N$ is the number of lattice sites.

The sum in Eq. $\mathcal{H}_{\text{eff}}$ is over all $\mathbf{q}$ in the appropriate Brillouin zone. The Fourier-transformed exchange interaction is

$$J_\mathbf{q} = \sum_{\mathbf{r}} J_r e^{i\mathbf{q} \cdot \mathbf{r}},$$  \hspace{1cm} (6)

e.g., a sum over displacements to nearest neighbors, $\mathbf{r} \equiv \mathbf{m} - \mathbf{n}$.

The $\mathbf{q}$-dependent magnetization and zero-field susceptibility have the usual definitions,

$$M_\mathbf{q} = \langle S_\mathbf{q} \rangle,$$

(7a)

$$\chi_\mathbf{q} = \frac{\partial M_\mathbf{q}}{\partial H_\mathbf{q}} = \beta \langle S_{-\mathbf{q}} S_\mathbf{q} \rangle.$$  \hspace{1cm} (7b)

Then for an arbitrary Hamiltonian, there results the constraint (used to determine the reaction field $\lambda$)

$$\frac{1}{N} \sum_{\mathbf{q}} \chi_\mathbf{q} = \frac{\beta}{n} S^2 \equiv \chi_0$$

(8)

The number of spin components $n$ enters here, in the expression for $\chi_0$, when models other than the Ising model are considered.

Now we consider the mean-field approximation in $q$-space, i.e., using the random phase approximation (RPA), magnetization components at different wavevectors are assumed to be independent. The mean-field Hamiltonian for the interaction of a negative $q$-component can be written

$$\mathcal{H} = -\sum_{\mathbf{q}} H^{\text{eff}}_\mathbf{q} S_{-\mathbf{q}}$$

(9)

where the effective (or mean-field) magnetic field is

$$H^{\text{eff}}_\mathbf{q} = H_\mathbf{q} + (J_\mathbf{q} - \lambda) \langle S_\mathbf{q} \rangle$$

(10)

From the definition (7a), and the relation,

$$\chi_\mathbf{q} = \frac{\partial M_\mathbf{q}}{\partial H^{\text{eff}}_\mathbf{q}} \frac{\partial H^{\text{eff}}_\mathbf{q}}{\partial H_\mathbf{q}}$$

(11)

the mean-field Hamiltonian gives the well-known expression,

$$\chi_\mathbf{q} = \frac{\chi_0}{1 - \chi_0(J_\mathbf{q} - \lambda)}$$

(12)

The reaction field $\lambda$ is determined by forcing this expression for $\chi_\mathbf{q}$ to satisfy the constraint (8). The critical temperature $T_c$ is determined as the temperature at which $\chi_\mathbf{q} = 0$ diverges. For lower temperatures the ORF calculation gives a negative susceptibility at $q = 0$, signifying the presence of the ordered state. In a continuum limit of the constraint (8), a short manipulation leads to an expression for the critical temperature,

$$k_B T_c = \frac{J_0 S^2}{n I}$$

(13)

The constant $I$ is given from a $q$-space integral over the appropriate Brillouin zone:

$$I = \frac{V}{N} \int_{BZ} \frac{d^3q}{(2\pi)^3} \frac{J_0}{J_0 - J_\mathbf{q}}$$

(14)

and $V/N$ is the specific volume per lattice site (for example, $V/N = a^3, 1/2 a^3, 1/a^3$ for sc, bcc and fcc lattices, respectively, where $a^3$ is the cubic unit cell volume). At $q = 0$ we have $J_\mathbf{q} = J$, which gives the energy scale in the mean-field approximation. Then the dimensionless integral $I$ gives the correction to the mean-field critical temperature. From integration over the appropriate Brillouin zones, the values of $I$ are 1.517, 1.393 and 1.343 for sc, bcc and fcc lattices, respectively. When applied to the 3D Ising model ($n = 1$) one gets $k_B T_c/JS^2 = z/I = 3.955, 5.743, 8.932$ for sc, bcc and fcc lattices, considerable improvements over the standard mean-field results given from $k_B T_c/JS^2 = z$. They compare favorably with the exact Ising model results from series: $k_B T_c/JS^2 = 4.5103, 6.3508, 9.794$, respectively. For the Heisenberg model ($n = 3$), the ORF predictions would be $k_B T_c/JS^2 = 1.318, 1.914$, for sc and bcc lattices, whereas precise Monte Carlo estimates give $k_B T_c/JS^2 = 1.443, 2.054$, respectively. Note, however, that the ORF estimates are all below the exact results.
III. BRAVAIS LATTICES WITH A BASIS

Now suppose the lattice has an underlying set of $N$ Bravais lattice points $n$, each of which has a two-atom basis $\{0,d\}$. (The generalization to a larger basis is straightforward.) Therefore at each site $n$ we suppose there is a two component field $w_n$ written as a column vector:

$$w_n = \left( \begin{array}{c} S_n \\ S_{n+d} \end{array} \right) \quad (15)$$

It is useful to employ a sublattice notation, $S_n \equiv S_n^A, S_{n+d} \equiv S_n^B$. The exchange interaction occurs between neighboring Bravais sites, via a 2×2 matrix, $G_{nm}$:

$$H_{\text{ex}} = -\frac{1}{2} \sum_n \sum_m w_n^T \cdot G_{nm} \cdot w_m. \quad (16)$$

where it is stressed that the sums are over all Bravais sites; the factor of 1/2 avoids double counting, and

$$G_{nm} = \begin{pmatrix} G_{AA}^{nm} & G_{AB}^{nm} \\ G_{BA}^{nm} & G_{BB}^{nm} \end{pmatrix} = \begin{pmatrix} J_{nm} & J_{n+d,m} \\ J_{n,m+d} & J_{n+d,m+d} \end{pmatrix} \quad (17)$$

In fact, the matrix $G_{nm}$ is taken as zero unless $n-m$ is a near neighbor displacement. The details of the specific lattice will determine which components of $G$ are nonzero.

The reaction terms, two for each site $n$, can be written with a 2×2 unit matrix,

$$H_{\text{orf}} = \frac{1}{2} \sum_n \sum_m \left[ w_n^T \cdot \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \right] w_m \quad (18)$$

which is equivalent to expression (23), and essentially shifts the original exchange matrix by $-2\lambda \delta_{n,m} I$, where $I$ is the 2×2 unit matrix.

Finally, for the purpose of the calculation, there is a separate applied field for each sublattice, so at a given site $n$, we have fields $H_n^A$ (acting on $S_n$) and $H_n^B$ (acting on $S_{n+d}$). These compose a column vector field,

$$h_n = \left( \begin{array}{c} H_n^A \\ H_n^B \end{array} \right) \quad (19)$$

Then we choose to write the q-space Hamiltonian, including the reaction term and applied fields, as

$$H = -\frac{1}{2} \sum_q \left[ w_{-q}^T \cdot (G_q - 2\lambda I) \cdot w_q \right] \quad (20)$$

The Fourier transforms needed here obviously are related to those already defined for $n, m, S_n$, etc. The most significant difference from Eq. (3) is the presence of "2" on $\lambda$, due to the two-atom basis. This Hamiltonian is exact. Now we define the sublattice magnetizations (where $i = A, B$),

$$M_i^q = \langle S_i^q \rangle \quad (21)$$

and related susceptibilities (where also $j = A, B$),

$$\chi_{ij}^q = \frac{\partial M_i^q}{\partial H_j^q} = \beta \langle S_i^q S_j^{-q} \rangle, \quad (22)$$

In the RPA, from the point of view of the negative q-components, the Hamiltonian is approximated as

$$H = -\sum_q \left[ w_{-q}^T \cdot h_q + w_{-q}^T \cdot K_q \cdot \langle w_q \rangle \right] \quad (23)$$

where we use the shorthand notation for the shifted exchange interaction,

$$K_q \equiv G_q - 2\lambda I \quad (24)$$

The above Hamiltonian can alternatively be written in terms of effective fields in components,

$$H = -\sum_q \left[ H_q^A \text{eff} S_q^A + H_q^B \text{eff} S_q^B \right] \quad (25)$$

$$H_q^A \text{eff} = H_q^A + K_q^A \langle S_q^A \rangle + K_q^{AB} \langle S_q^B \rangle \quad (26a)$$

$$H_q^B \text{eff} = H_q^B + K_q^B \langle S_q^A \rangle + K_q^{BB} \langle S_q^B \rangle \quad (26b)$$

Then in the limit of zero applied field, using this RPA Hamiltonian, the susceptibility definitions (22) become

$$\chi_{ij}^q = \frac{\partial M_i^q}{\partial H_j^q \text{eff}} \quad (27)$$

and we get equations for the susceptibility components,

$$\chi_{AA}^q = \beta \langle S_q^A S_q^{-A} \rangle_0 \left[ 1 + K_q^{AA} \chi_q^A + K_q^{AB} \chi_q^B \right] \quad (28a)$$

$$\chi_{AB}^q = \beta \langle S_q^A S_q^{-A} \rangle_0 \left[ K_q^{AA} \chi_q^A + K_q^{AB} \chi_q^B \right] \quad (28b)$$

$$\chi_{BB}^q = \beta \langle S_q^B S_q^{-B} \rangle_0 \left[ 1 + K_q^{BB} \chi_q^A + K_q^{AB} \chi_q^A \right] \quad (28c)$$

$$\chi_{BA}^q = \beta \langle S_q^B S_q^{-B} \rangle_0 \left[ K_q^{BB} \chi_q^A + K_q^{BA} \chi_q^A \right] \quad (28d)$$

where $\langle \cdot \rangle_0$ means the expectation value using the RPA Hamiltonian. In the high-temperature limit, these expectations are

$$\beta \langle S_q^A S_q^{-A} \rangle_0 = \beta \langle S_q^B S_q^{-B} \rangle_0 = \frac{\beta}{n} S^2 \equiv \chi_0. \quad (29)$$
The equations (28) can be solved in the general case for all four susceptibility components. We get

$$\chi_{q}^{AA} = \chi_0 \frac{1 - \chi_0 K_{BB}}{(1 - \chi_0 K_{q}^{AA})(1 - \chi_0 K_{q}^{BB}) - \chi_0^2 K_{q}^{AB} K_{q}^{BA}}$$

(30a)

$$\chi_{q}^{BB} = \chi_0 \frac{K_{q}^{BA} \chi_{q}^{AA}}{1 - \chi_0 K_{q}^{BB}}$$

(30b)

and similar equations for $\chi_{q}^{AB}$ and $\chi_{q}^{BA}$ by appropriately interchanging the AB indices. For the lattices considered in this paper, however, there are the symmetries, $K_{q}^{AA} = K_{q}^{BB}$ and $K_{q}^{AB} = (K_{q}^{BA})^*$. Therefore the solutions are seen to satisfy symmetries $\chi_{q}^{AA} = \chi_{q}^{BB}$, $\chi_{q}^{AB} = (\chi_{q}^{BA})^*$. Now consider how to determine the reaction field $\lambda$, and subsequently, $T_c$. A little consideration using the definitions of $\chi_{q}^{AA}$ and $S_{q}$ shows that there is still the constraint,

$$\frac{1}{N} \sum_{q} \chi_{q}^{AA} = \frac{\beta}{n} S^2 \equiv \chi_0$$

(31)

Converting to the continuum limit,

$$\left( \frac{V}{N} \right) \left( \frac{1}{2\pi} \right)^3 \int d^3 q \chi_{q}^{AA} = \chi_0$$

(32)

This equation implicitly determines the reaction coupling $\lambda$ for any $T > T_c$. It is not clear how to get an explicit solution for $\lambda$ from it; a solution for $\lambda(T)$ for a given lattice can be found numerically (below).

Now, just as described for the Bravais lattice systems, the critical temperature $T_c$ is the temperature at which any of the susceptibility components, at $q = 0$, diverges. (The $\chi_q^2$ are well defined on the high temperature side of $T_c$.) So $T_c$ is determined as the point at which the denominator of Eq. (30a) goes to zero, leading to a relation between the critical temperature (via $\chi_0$) and the critical reaction field,

$$\chi_0^{-1} + 2\lambda = G_{0}^{AA} + \sqrt{G_{0}^{AB} G_{0}^{BA}}$$

(33)

Using this in the constraint equation (32) together with the result (30a) for $\chi_{q}^{AA}$ gives the general result when the symmetry $G_{q}^{AA} = G_{q}^{BB}$ holds,

$$k_B T_c = \frac{J_0 S^2}{n I}$$

(34a)

$$I = \frac{V}{N} \int_{BZ} d^3 q \frac{J_0 (J_0 - G_{q}^{AA})}{(J_0 - G_{q}^{AA})^2 - G_{q}^{AB} G_{q}^{BA}}$$

(34b)

where $J_0$ is the effective $q = 0$ exchange strength.

$$J_0 \equiv G_{0}^{AA} + \sqrt{G_{0}^{AB} G_{0}^{BA}}$$

(35)

The integral $I$ defined in this way again gives the correction to the mean-field prediction for $T_c$. Thus the determination of $T_c$ has been reduced to evaluating this integral over the Brillouin zone.

Below we will use Eq. (34) to estimate the critical temperature for diamond and hcp lattices. But first we can verify that the result is correct by using it for a bcc lattice, considered as a simple-cubic lattice with a basis, where we already know the standard ORF result for $T_c$.

IV. BCC LATTICE AS SC WITH BASIS

The bcc lattice points can be generated from the simple cubic primitive vectors, $a_1 = a\hat{x}$, $a_2 = a\hat{y}$, and $a_3 = a\hat{z}$, where $a$ is the cubic cell lattice constant, together with the basis, $\{0, d\}$, where $d = \frac{a}{2}(\hat{x} + \hat{y} + \hat{z})$ is the displacement to the body-centered point. The lattice can be thought of as a pair of interpenetrating sc lattices (A, B sublattices) with displacement $d$. The near neighbors of an ‘A’ site are all ‘B’ sites, and vice-versa, with the result that the $G_{q}^{AA}$ and $G_{q}^{BB}$ couplings are all zero. The nonzero $G_{q,n,m}^{ij} = G_{q,r}^{ij}$ couplings depend only on the near neighbor displacement, $r \equiv m - n$, as follows:

$$G_{q,r}^{BA} = J, \quad r = 0, a_1, a_2, a_3, a_1 + a_2 + a_3, a_1 + a_2, a_2 + a_3, a_3 + a_1$$

(36a)

$$G_{q,r}^{AB} = J, \quad r = 0, -a_1, -a_2, -a_3, -a_1 - a_2 - a_3, -a_1 - a_2, -a_2 - a_3, -a_3 - a_1$$

(36b)

where the terms for $r = 0$ correspond to the coupling within the two-atom basis. The fact that $G_{q,n,m}^{AA} = G_{q,n,m}^{BB} = 0$ simplifies the determination of $T_c$ considerably, as we only need to know the product, $G_{q}^{AB}$.

The Fourier-transformed interactions are found from sums over all the nonzero $G_{q}^{ij}$ (15 possible terms):

$$G_{q}^{AB} = \sum_{r} G_{q,r}^{AB} e^{i q \cdot r} = (G_{q}^{ij})^*$$

(37)

$$G_{q}^{AB} = J \{ 1 + e^{-i q_x} + e^{-i q_y} + e^{-i q_z}$$

$$+ e^{-i (q_x + q_y)} + e^{-i (q_y + q_z)} + e^{-i (q_z + q_x)}$$

$$+ e^{-i (q_x + q_z)} \}$$

(38)

In this and the following equations, $q_x, q_y, q_z$ are in units of $1/a$. For the underlying sc lattice, the density of points is 1 for every volume $a^3$, i.e., $V/N = a^3$. After a short calculation, there results

$$G_{q}^{AB} G_{q}^{BA} = 8 J^2 \{ 1 + \cos q_x + \cos q_y + \cos q_z$$

$$+ \cos q_x \cos q_y + \cos q_y \cos q_z + \cos q_x \cos q_z$$

$$+ \cos q_x \cos q_y \cos q_z \}$$

(39)
and the determination of $T_c$ relies on evaluation of the simplified integral,

$$I = \int \frac{d^3q}{(2\pi)^3} \left[ \frac{J_0^2}{J_0^2 - G^{AB}_q G^{BA}_q} \right]$$

where $J_0 = G^{AB}_0 = G^{BA}_0 = 8J = zJ$ and the factor $V/N = a^3$ was absorbed into the dimensionless $q$.

$$I = \int_{-\pi}^{\pi} \frac{dq_x}{2\pi} \int_{-\pi}^{\pi} \frac{dq_y}{2\pi} \int_{-\pi}^{\pi} \frac{dq_z}{2\pi} \left\{ 1 - \frac{1}{8} \left[ 1 + \cos q_x + \cos q_y + \cos q_z + \cos q_x \cos q_y + \cos q_y \cos q_z + \cos q_z \cos q_x + \cos q_x \cos q_y \cos q_z \right] \right\}^{-1}$$

The integral $I$ was evaluated numerically by sampling $q_x, q_y, q_z$ uniformly with a constant increment, and then using an extrapolation of the results in the limit that the increment goes to zero. Essentially, we let $dq_x = dq_y = dq_z = 2\pi/N_x$, where $N_x$ is some integer number of divisions of the axes, and then generated a simple cubic lattice of sampling points from these. The integral $I$ was then estimated as a sum over the resulting cubic grid of sampling points in the BZ in $q$-space. A plot of $I$ versus $1/N_x$ results in a straight line whose extrapolation to $1/N_x \rightarrow 0$ gives a very accurate estimate of the integral. (Errors in the integral estimate clearly go as $1/N_x$.) In this way we found $I = 1.39321$. Therefore, the estimate of critical temperature for the bcc system obtained viewing it as sc with a basis is

$$k_B T_c = \frac{8JS^2}{n} \frac{1}{1.39321} = \frac{5.74213}{n} JS^2.$$

For comparison with the standard ORF procedure, when the original bcc lattice is used, the specific volume is $V/N = \frac{1}{2}a^3$. The Fourier transformed exchange (Eq. (3)) is

$$J_q = 8J \cos \frac{q_x}{2} \cos \frac{q_y}{2} \cos \frac{q_z}{2}$$

The integral needed for Eq. (39) is

$$I = \frac{1}{2} \int_{BZ} \frac{d^3q}{(2\pi)^3} \left[ 1 - \frac{1}{8} \left[ 1 + \cos q_x \cos q_y \cos q_z \right] \right]^{-1}$$

$$= \frac{1}{2} \times 2.786 = 1.393,$$

where the integral is over the Brillouin zone for the bcc lattice (an fcc Wigner-Seitz cell), and was evaluated by the sampling technique already described. The integral is exactly twice that given by expression (11), but because $V/N = \frac{1}{2}a^3$ is half as large for the bcc lattice as for the sc lattice, the result for $T_c$ from Eq. (14) is that given by Eq. (42). Thus this approach using the two-atom basis, for the bcc system, is equivalent to the standard ORF procedure, and should be reliable for application to the diamond and hcp lattices.

As an interesting mathematical note, the integral $I$ in (14) can be rewritten using the periodicity and symmetry in $q$-space to be over the same region as in (11). A cubic cell $-2\pi \leq q_x \leq 2\pi, -2\pi \leq q_z \leq 2\pi, -2\pi \leq q_z \leq 2\pi$, contains 4 copies of the Brillouin zone of the bcc lattice. Thus we can integrate over this cubic cell and divide by 4; also, shifting the variables of integration to $q'_x = q_x/2$, etc., leads to an additional factor of $2^3$ and gives

$$I = \frac{1}{2} \times \frac{1}{4} \times 2^3 \int_{-\pi}^{\pi} \frac{dq'_x}{2\pi} \int_{-\pi}^{\pi} \frac{dq'_y}{2\pi} \int_{-\pi}^{\pi} \frac{dq'_z}{2\pi} \left[ 1 - \cos q'_x \cos q'_y \cos q'_z \right]^{-1}$$

Overall, the prefactor on the integral is one. Thus the integral here must be the same as the integral in (11). The integrands, however, are not equivalent; there is no way to transform one into the other.

Finally we also note that it is easier and more precise to evaluate (15) by a uniform cubic grid of sampling points than expression (44), because a cubic grid of sampling points easily fits to the cubic integration boundaries. On the other hand, for expression (14), there is always more difficulty to fit any grid of points smoothly to all of the integration boundaries of the fcc Wigner-Seitz cell in $q$-space, leading to greater discretization errors. Removing these errors is important for producing a smooth extrapolation to $N_x \rightarrow \infty$.

V. DIAMOND LATTICE

The diamond lattice can be considered as an fcc lattice with a two atom basis. The fcc primitive vectors are $a_1 = \frac{2}{3}(\hat{x} + \hat{y}), a_2 = \frac{\sqrt{3}}{2}(\hat{y} + \hat{z}), a_2 = \frac{2}{3}(\hat{z} + \hat{x})$, where $a$ is the standard cubic cell lattice constant, and the basis is $\{0, d\}$, where $d = \frac{\sqrt{2}}{3}(\hat{x} + \hat{y} + \hat{z})$. The nearest neighbors of a site $n + d$ (a ‘B-site’) are $\{n, n + a_1, n + a_2, n + a_3\}$ (all ‘A-sites’). The nearest neighbors of a site $n$ (an ‘A-site’) are at $\{n + d, n + d - a_1, n + d - a_2, n + d - a_3\}$ (all ‘B-sites’). As a result, only $G^{AB}$ and $G^{BA}$ are nonzero. In terms of the neighbor displacements $r = m - n$, the only nonzero exchange couplings are

$$G^{BA}_r = J, \ r = \{0, a_1, a_2, a_3\}$$

$$G^{AB}_r = J, \ r = \{0, -a_1, -a_2, -a_3\}$$

Then it is straightforward to evaluate

$$G^{AB}_q = \sum_r G^{AB}_r e^{-iqr} = (G^{BA}_q)^*$$

$$= J \left[ 1 + e^{-iqa_1} + e^{-iqa_2} + e^{-iqa_3} \right]$$

and what is needed for the $T_c$ integral:
where \( q \)'s are in units of \( 1/a \). Clearly we also have once again, \( J_0 = G_{0q}^A = G_{0q}^B = 4J = zJ \). The specific volume per lattice point on the underlying fcc lattice is \( V/N = \frac{1}{4} a^3 \). Then \( T_c \) will be evaluated using Eq. (53) and the simplified integral like (40), which becomes

\[
I = \frac{1}{4} \int_{\text{BZ}} \frac{d^3q}{(2\pi)^3} \left\{ 1 - \frac{1}{4} \left[ 1 + \cos \frac{q_x}{2} \cos \frac{q_y}{2} + \cos \frac{q_y}{2} \cos \frac{q_z}{2} + \cos \frac{q_z}{2} \cos \frac{q_x}{2} \right] \right\}^{-1}
\]

(49)

The BZ for the fcc lattice is a bcc Wigner-Seitz cell, with lattice constant \( 4\sqrt{2}/a \). The easiest and most precise way to evaluate this integral is to apply the same procedure we noted for the bcc system: Change the integration region to the cube, \( -2\pi \leq q_x \leq 2\pi, -2\pi \leq q_y \leq 2\pi, -2\pi \leq q_z \leq 2\pi \), which contains 2 copies of the BZ, therefore include a factor of \( \frac{1}{2} \), and sum over points on a cubic grid. Also make the variable change, \( q' = q/2 \), which leads to a factor of \( 2^3 \), giving

\[
I = \frac{1}{4} \times \frac{1}{2} \times 2^3 \int_{-\pi}^{\pi} \frac{dq_x}{2\pi} \int_{-\pi}^{\pi} \frac{dq_y}{2\pi} \int_{-\pi}^{\pi} \frac{dq_z}{2\pi} \left\{ 1 - \frac{1}{4} \left[ 1 + \cos q'_x \cos q'_y + \cos q'_y \cos q'_z + \cos q'_z \cos q'_x \right] \right\}^{-1}
\]

(50)

Using a cubic grid spacing \( dq_x' = dq_y' = dq_z' = 2\pi/N_x \), we evaluated the integral as a sum over points within the cubical integration region, for \( N_x \) ranging from 100 to 2000. A plot of \( I \) versus \( 1/N_x \) gives a straight line (Fig. 4), and its extrapolation to \( 1/N_x \to 0 \) gives \( I = 1.79288 \). Thus the critical temperature is estimated as

\[
k_B T_c = \frac{4JS^2}{n} \frac{1}{1.79288} = \frac{2.23105}{n} JS^2
\]

(51)

The exact result for \( T_c \) (Ising model, \( n = 1 \)) as estimated from series expansions, is known to be \( k_B T_c = 2.7040JS^2 \). Thus the ORF calculation, as is usual, underestimates \( T_c \), but is a considerable improvement over the simple mean-field result, \( k_B T_c = 4JS^2 \).

VI. SIMPLE HEXAGONAL BRAVAIS LATTICES AND HCP LATTICES

Another example of a lattice with a basis is the hcp system, which can be considered as interpenetrating simple hexagonal Bravais lattices (i.e., stacked triangular nets). The primitive vectors of the simple hexagonal Bravais lattice can be taken as \( a_1 = a\hat{x}, a_2 = a(\frac{1}{2}\hat{x} + \frac{\sqrt{3}}{2}\hat{y}), a_3 = c\hat{z} \), where \( a \) and \( c \) are the lattice constants. For the hcp system, a two-atom basis of \( \{0, d\} \) is used, where \( d = \frac{1}{2}(a_1 + a_2) + \frac{1}{2}a_3 \), and one triangular net is stacked on top of the previous one, but shifted to be over the centers of one set of the triangular cells below, in what is usually referred to as the ABAB... packing. For the lattice constant ratio \( c/a = \sqrt{8/3} \), the highest density packing is obtained, however, for the calculation here this ratio does not directly enter, and need not be specified. Instead, it is interesting to consider that the near neighbor exchange interactions within the triangular nets (xy-plane) have one strength, \( J_{xy} \), while there is a different strength, \( J_z \), for the bonds between the planes. In general we can consider the calculation of \( T_c \) as a function of the ratio, \( \Delta \equiv J_z/J_{xy} \).

We present first the calculation of \( T_c(\Delta) \) for the simple hexagonal Bravais lattice, using the standard ORF theory in Sec. 5 which acts as an introduction to the corresponding calculation for the hcp system, because they both rely on the same information concerning the Brillouin zone.

A. Simple Hexagonal Bravais Lattice

Here there are 6 neighbor displacements from some arbitrary site to neighbors in the same plane \( \{\pm a_1, \pm a_2, \pm (a_1 - a_2)\} \), with exchange strength, \( J_{xy} \). The remaining two neighbors, with displacements, \( \pm a_3 \), have exchange strengths, \( J_z \). A short calculation shows that the q-space exchange (Eq. 3) is

\[
J_q = 2J_{xy} \left[ \cos q \cdot a_1 + \cos q \cdot a_2 + \cos q \cdot (a_1 - a_2) \right] + 2J_z \cos q \cdot a_3
\]

(52a)

\[
= 2J_{xy} \left[ \cos q_x a + 2 \cos \frac{1}{2} q_x a \cos \frac{\sqrt{3}}{2} q_y a \right] + 2J_z \cos q_z c
\]

(52b)

and \( J_0 = 6J_{xy} + 2J_z = 2(3 + \Delta)J_{xy} \) will determine the mean-field critical temperature.

The area of one triangle in the net is \( \frac{1}{2} a_1 \times \frac{\sqrt{3}}{2} a_2 = \frac{\sqrt{3}}{4} a^2 \), and there is \( \frac{1}{2} \) site per triangle per layer. Thus the specific volume per site is \( V/N = \frac{2\sqrt{3}}{9a^2} \). The primitive vectors of the reciprocal space are

\[
b_1 = \frac{4\pi}{\sqrt{3}a} \left( \frac{\sqrt{3}}{2} \hat{x} - \frac{1}{2} \hat{y} \right)
\]

(53a)

\[
b_2 = \frac{4\pi}{\sqrt{3}a} \hat{y}
\]

(53b)

\[
b_3 = \frac{2\pi}{c} \hat{z}
\]

(53c)

The reciprocal space is another simple hexagonal lattice, with lattice constants \( \frac{\sqrt{3}}{\sqrt{3}a} \) in the xy-plane and \( \frac{2\pi}{c} \) in the
z-direction, rotated by 30° from the real space lattice. The Brillouin zone Wigner-Seitz cell is a hexagonal cylinder, however, for the purpose of the integral needed here (Eq. [4]) it is more convenient to do the summation inside a cell bounded in the xy-plane by a rhombus formed by $b_1$ and $b_2$ (See Fig. [4]). The hexagonal cylinder and rhombus cylinders have equal areas and are equivalent to each other by appropriate symmetry operations. This rhombic cylinder cell is very convenient for evaluation of the integral $I$, especially with the variable change on $q$:

$$q = x b_1 + y b_2 + z b_3$$  \hspace{1cm} (54)

where the dimensionless parameters $x, y, z$ all range from 0 to 1, mapping out the entire cell. This leads to

$$d^3q = b_1 \cdot (b_2 \times b_3) \, dx \, dy \, dz = (2\pi)^3 \frac{N}{V} \, dx \, dy \, dz$$  \hspace{1cm} (55)

and the integral is simplified to,

$$I = \int_0^1 \! \! dx \int_0^1 \! \! dy \int_0^1 \! \! dz \left\{ 1 - \frac{1}{3 + \Delta} \left[ \cos 2\pi x + \cos 2\pi y + \cos 2\pi(x - y) + \Delta \cos 2\pi z \right] \right\}^{-1}$$  \hspace{1cm} (56)

The integral gives the correction to the mean-field prediction, i.e.,

$$k_B T_c = \frac{k_B T^{MF}_c}{\frac{I}{\int}}, \quad k_B T^{MF}_c = \frac{2(3 + \Delta) J_{xy} S^2}{n}.$$  \hspace{1cm} (57)

The integral $I$ was evaluated by the numerical techniques described above (Sec. [VIA]), for a range of anisotropy parameter $0 < \Delta \leq 2$, see Fig. [5]. At the isotropic limit, $\Delta = 1$, we get $I = 1.44930$ and $nk_B T_c = 5.5199 J S^2$. In the limit $\Delta \to 0$, the system becomes two-dimensional, the integral $I$ diverges logarithmically due to small-$q$ contributions, and ORF is not applicable.

**B. Hexagonal Close Packed Lattices**

Again there are 6 neighbor displacements from an arbitrary site to neighbors in the same plane \{±$a_1$, ±$a_2$, ±($a_1 - a_2$)\}, with exchange strength, $J_{xy}$. The difference from the simple hexagonal lattice, is that there are 3 neighbors in a layer above and 3 neighbors in a layer below the one being considered, with exchange couplings $J_z$, giving 12 neighbors in all. However, to evaluate the matrix elements $G^{ij}_r$, we need to consider these couplings from the point of view of the simple hexagonal Bravais lattice with a basis (see Fig. [5]). Thus, an arbitrary A-site, has the 6 neighbor displacements to other A-sites in the same xy-plane: \{±$a_1$, ±$a_2$, ±($a_1 - a_2$)\}, which will give nonzero $G^{AA}$ coupling terms. The neighbors in adjacent planes are B-sites, leading to nonzero $G^{AB}$ terms. Further consideration leads to the nonzero coupling elements,

$$G^{AA}_r = G^{BB}_r = J_{xy}, \quad \mathbf{r} = \pm a_1, \pm a_2, \pm(a_1 - a_2)$$  \hspace{1cm} (58a)

$$G^{AB}_r = G^{BA}_r = J_z, \quad \mathbf{r} = 0, a_1, a_2, a_3 + a_1, a_3 + a_2 \quad \Delta < 0.1$$  \hspace{1cm} (58b)

It is notable that it is the first example where the diagonal elements are nonzero. The terms where $\mathbf{r} = 0$ are the coupling within the basis. The $q$-space couplings (Eq. [37]) are found to be:

$$G^{AA}_q = G^{BB}_q = 2 J_{xy} \left[ \cos \mathbf{q} \cdot \mathbf{a}_1 + \cos \mathbf{q} \cdot \mathbf{a}_2 + \cos \mathbf{q} \cdot (\mathbf{a}_1 - \mathbf{a}_2) \right]$$  \hspace{1cm} (59)

$$G^{AB}_q = (G^{BA}_q)^* = J_z \left[ 1 + e^{-i \mathbf{q} \cdot \mathbf{a}_1} + e^{-i \mathbf{q} \cdot \mathbf{a}_2} + e^{-i \mathbf{q} \cdot (a_3 + a_1)} + e^{-i \mathbf{q} \cdot (a_3 + a_2)} \right]$$  \hspace{1cm} (60)

and what is needed for evaluation of $T_c$:

$$G^{AB}_q G^{BA}_q = J_z^2 \left[ 6 + 4 \left\{ \cos \mathbf{q} \cdot \mathbf{a}_1 + \cos \mathbf{q} \cdot \mathbf{a}_2 + \cos \mathbf{q} \cdot (\mathbf{a}_1 - \mathbf{a}_2) \right\} \times [1 + \cos \mathbf{q} \cdot \mathbf{a}_3] \right]$$  \hspace{1cm} (61)

The $q=0$ exchange strength (Eq. [35]) is seen to be $J_0 = 6(J_{xy} + J_z) = 6 J_{xy}(1 + \Delta)$ and determines the $\Delta$-dependent mean-field critical temperature. The specific volume per site (for the underlying simple hexagonal Bravais lattice) is $V/N = \sqrt{3} a^2 c$. The reciprocal space is that of the simple hexagonal Bravais lattice as described in Sec. [VIA]. Therefore, using the rhombic cylinder Brillouin zone cell, the integral $I$ of Eq. [3] can be transformed using Eqs. [4] and [5] into

$$I = \int_0^1 \! \! dx \int_0^1 \! \! dy \int_0^1 \! \! dz \frac{J_0 (J_0 - G^{AA}_q)^2 - G^{AB}_q G^{BA}_q}{(J_0 - G^{AA}_q)^2}$$  \hspace{1cm} (62)

where we have the transformed quantities

$$G^{AA}_q = 2 J_{xy} \left[ \cos 2\pi x + \cos 2\pi y + \cos 2\pi(x - y) \right]$$  \hspace{1cm} (63a)

$$G^{AB}_q G^{BA}_q = 2 J_{xy} \Delta \left[ 1 + \cos 2\pi z \right] \times \left[ 3 + 2 \left\{ \cos 2\pi x + \cos 2\pi y + \cos 2\pi(x - y) \right\} \right]$$  \hspace{1cm} (63b)

The correction to the mean-field prediction is

$$k_B T_c = \frac{k_B T^{MF}_c}{\frac{I}{\int}}, \quad k_B T^{MF}_c = \frac{6(1 + \Delta) J_{xy} S^2}{n}.\quad \left(64\right)$$

$I$ was evaluated by the numerical techniques described above (Sec. [VIA]), including the $N_z \to \infty$ extrapolation. For example, at the isotropic limit, $\Delta = 1$, we get $I = 1.34466$, and $nk_B T_c = 8.92418 J S^2$. It is interesting to note that the same value of $I$ results for the fcc lattice
(Eq. (4)), when evaluated to the same precision. Thus the ORF corrections to the mean-field $T_c$ for fcc and hcp lattices, both with 12 nearest neighbors, are the same. Some other hcp results in a limited range of anisotropy $\Delta$ are shown in Fig. 3. Once again, in the limit $\Delta \to 0$ there is a weak divergence of the integral as the system crosses over into a two-dimensional one, with $T_c \to 0$ over a very narrow range of $\Delta$. ORF is not applicable in this limit; $T_c$ should pass over to the finite value for the 2D triangular lattice model.[4]

There are a few theoretical results to compare with for the hcp system. In a series of papers, Domany, Gubernatis and Auerbach[7, 8] analyzed a Lifshitz tricritical point for the hcp Ising model, which occurs at a negative value of $\Delta$. As part of their analysis they applied Monte Carlo calculations[8] to determine the phase diagram; very roughly for $\Delta = 1$ they obtained $k_B T_c \approx 10 J S^2$.

Values of $T_c$ at other anisotropies also can be estimated from their Fig. 1 but with poor precision. However, it does appear that the Onsager results fall below the Monte Carlo estimates of $T_c$, as expected.

For the hcp Heisenberg model, Adler[3] estimated $T_c$ by a Green’s function approach together with a random phase approximation. It is surprising to see that at $\Delta = 1$ Alder found the correction to the mean-field $T_c$ to be by the factor $F(-1) = 1.34 \pm 0.005$, where $F(-1)$ is a certain sum over the Brillouin zone. Making a more accurate evaluation of $F(-1)$ by the techniques described here, we get $F(-1) = 1.34466$, i.e., a value exactly equal to the correction integral $I$ obtained from the ORF procedure. In fact, it is easy to show that the expression for $F(-1)$ given by Adler is exactly equivalent to our expression for $I$, including the anisotropic case, $\Delta \neq 1$. Therefore, the Green’s function approach used there is exactly equivalent to the ORF procedure presented here; they are different approaches to impose the random phase approximation.

Furthermore, in this level of approximation, the question posed by Domb and Sykes[2] and investigated by Adler is answered: $T_c$ for fcc and hcp Ising models are the same, even though the hcp lattice is more densely packed and might be expected to have a higher $T_c$. Apparently a more precise procedure is needed to determine whether there is a true difference in their critical temperatures.

VII. REACTION FIELD AND THERMODYNAMIC QUANTITIES AT $T > T_c$

It is clear that any quantities such as specific heat, magnetization, etc, can be evaluated via the RPA Hamiltonian for temperatures away from $T_c$, provided that the reaction field, $\lambda$, has been determined. Thus we take a few sentences to examine how $\lambda$ can be calculated.

At the critical temperature, the reaction field as determined from Eq. (33) is seen to be

$$\lambda_c \equiv \lambda(T_c) = \frac{1}{2} (J_0 - \chi_0^{-1}) = \frac{1}{2} (J_0 - n k_B T_c/S^2)$$  (65)

For higher temperatures, the constraining equation (22) to determine $\lambda$ is equivalent to

$$\frac{V}{N} \int_{BZ} d^3q \left[ \frac{1 - \chi_0(G_{AA}^A - 2\lambda)}{[1 - \chi_0(G_{AA}^A - 2\lambda)]^2 - \lambda^2 G_{AB} G_{BA}^A} \right] = 1$$  (66)

Considering the left hand side as a function of $\lambda$, one can apply Newton’s method to search for the $\lambda$ at which the function passes through 1. This search can be aided by the requirement that the denominator of this integrand must be positive everywhere in the BZ, including at $q = 0$. This leads to the inequality for $T > T_c$,

$$\lambda(T) > \frac{1}{2} (J_0 - \chi_0^{-1}) = \frac{1}{2} (J_0 - n k_B T/S^2) < \lambda_c$$  (67)

Indeed, we have found as well that $\lambda(T) < \lambda_c$.

Results for $\lambda(T)$ for the diamond lattice and hcp lattice (at $\Delta = 1$) are shown in Fig. 4. The hcp lattice, which has higher coordination number, also has the stronger reaction field at $T_c$. As the temperature is increased, the reaction fields diminish and become of comparable sizes for $T > 2T_c$. It is also expected that the slope relates to the specific heat. The graph then reasonably demonstrates a larger specific heat for hcp compared to diamond near $T_c$, in contrast to more similar specific heats at higher temperatures.

VIII. CONCLUSIONS

We have reviewed the standard Onsager reaction field approximation for estimating $T_c$, and have shown how it can be extended to apply to a Bravais lattice with a basis. The bcc lattice was used as a test case because it can be calculated either by the standard approach or the new method, when considered as sc with a two-atom basis. We used the new method to get $T_c$ for diamond and anisotropic hcp lattice systems, however, it certainly can be extended to more complex systems with a greater number of atoms per unit cell. For the hcp lattice system, the ORF procedure used here was found to be exactly equivalent to a Green’s function (plus RPA) approach used by Adler.[4] While it is an approximate method, it does give reasonable estimates of $T_c$ and other quantities where other methods may be more cumbersome or time-consuming to apply.

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FIG. 3. ORF results for $T_c$ on the simple hexagonal Bravais lattice (hex) and the hexagonal close packed lattice (hcp), as functions of the exchange anisotropy $\Delta = J_z/J_{xy}$.

FIG. 4. XY projection of some nearest-neighbor bonds between A-sites (solid circles) and B-sites (open circles) in an hcp lattice. Double solid lines connect A, B sites at the same Bravais lattice point. Solid lines show AA bonds (within the planes), dotted lines show AB bonds.

FIG. 5. The reaction field $\lambda$ for $T > T_c$ for the diamond lattice and isotropic hexagonal close packed lattice ($\Delta = 1$).