

Equivalence of the Krieger-James approximation and the constant-coupling approximation in magnetism

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Abstract. We have applied the Krieger-James approximation to a description of short range order effects in both ferro- and antiferromagnetism. In this method, a mechanical consistency condition is imposed on the probability distribution of a pair of near neighbours in a cluster of $(Z+1)$ ions. The resulting theory is analytically equivalent to the familiar constant coupling approximation, thus ensuring thermodynamic equilibrium of the system.

Keywords. Krieger-James approximation; constant coupling approximation; magnetism

1. Introduction

There have been a number of theoretical discussions of (nearest neighbour) short range order effects in ferro- and antiferromagnets assuming clusters of the Bethe type (Bethe 1935); each cluster consists of a central ion surrounded by Z nearest neighbour ions, no two nearest neighbours being nearest neighbours of each other. In the constant coupling (CC) approximation (Kasteleijn and Kranendonk 1956; Strieb *et al* 1963), the effective Hamiltonian (H_e) of a pair of coupled ions is written in terms of the direct exchange interaction between the pair and an effective field acting on each of them due to the external medium. The effective field, assumed to be a function of the average magnetization and temperature, is derived by minimizing the free energy of the system. The other well known methods are those of Weiss (1948) [i.e., the BPW method (Strieb *et al* 1963)] and of Oguchi (1955), which make use of mechanical consistency conditions, but it has been proved that they do not lead to thermodynamic equilibrium of the system (Strieb *et al* 1963). In the present paper we show that the mechanical consistency condition of Krieger and James (1954), proposed by them in their treatment of rotational phase transition in solids, when applied to magnetism results in a theory that is analytically equivalent to the CC approximation. The KJ solutions fulfil the thermodynamic equilibrium condition and moreover provide a simple physical picture underlying the CC approximation. An equivalent result has been derived by Elliot and Marshall (1958) in the case of ferromagnetism.

2. Krieger-James approximation

In the present context, the KJ consistency condition may be stated as follows: the

relative probability of a pair of ions '0' and '1' taking the angular coordinates Ω_0 and Ω_1 respectively should be the same regardless of which of them is considered as the central one, i.e.,

$$\chi(\Omega_0, \Omega_1) = \chi(\Omega_1, \Omega_0) \quad (1)$$

where $\chi(\Omega_0, \Omega_1)$ and $\chi(\Omega_1, \Omega_0)$ are the probabilities when '0' and '1' are respectively the central ions in the cluster. This is also equivalent to Chang's (1937) consistency relation which states that the probability of any ion taking an angular coordinate Ω should be the same, whether it is treated as a central or an outer ion.

Following Krieger and James, we can write

$$\begin{aligned} \chi(\Omega_0, \Omega_1) &= \frac{1}{P^{(0)}} \exp[-\beta \{V(\Omega_0, \Omega_1) + g_1(\Omega_1) + h_0(\Omega_0)\}] \\ &\times \prod_{j=2}^Z \sum_{\Omega_j} F(\Omega_0, \Omega_j) \end{aligned} \quad (2)$$

where $F(\Omega_0, \Omega_j) = \exp[-\beta \{V(\Omega_0, \Omega_j) + g_j(\Omega_j)\}]$, $V(\Omega_0, \Omega_j)$ is the potential energy of the interaction between the ions 0 and j , $g_j(\Omega_j)$ is the potential energy due to the effective field acting on an ion j considered as an *outer* ion, the field including the external field and the interactions of j with all the ions in the medium except the central ion of the cluster, $h_0(\Omega_0)$ is the potential energy of the ion 0 considered as a central ion due to the external field, $\beta = 1/k_B T$ and $P^{(0)}$ is the normalizing constant when the central ion has the label 0 and is given by

$$P^{(0)} = \sum_{\Omega_0} \exp[-\beta h_0(\Omega_0)] \prod_{j=1}^Z F(\Omega_0, \Omega_j) \quad (3)$$

Using (1), we write

$$P^{(0)} \frac{\exp\{\beta[-g_0(\Omega_0) + h_0(\Omega_0)]\}}{\prod_{j=2}^Z \sum_{\Omega_j} F(\Omega_0, \Omega_j)} = P^{(1)} \frac{\exp\{\beta[-g_1(\Omega_1) + h_1(\Omega_1)]\}}{\prod_{j=2}^Z \sum_{\Omega_j} F(\Omega_1, \Omega_j)} \quad (4)$$

3. Antiferromagnetism

3.1. External field parallel to the easy axis

As an illustration of the application of the KJ method to magnetism, we consider first an antiferromagnet. Assuming only nearest neighbour interactions, the total Hamiltonian of the system can be expressed as

$$H = \sum_{(i,j)} H^{(2)} = \sum_{(i,j)} 2J \mathbf{S}_i \cdot \mathbf{S}_j - \frac{2\mu B}{Z} (S_{1z} + S_{2z}) \quad (5)$$

where $\Sigma_{(i,j)}$ implies a summation over nearest neighbours only and B is the external magnetic field acting along z . As usual, we divide the medium into two equivalent interpenetrating sub-lattices A and B such that all the nearest neighbours of an A -ions are B -ions and *vice versa*. The spontaneous magnetization of the A -sublattice say parallel to z is opposite in direction to that of the B -sublattice. To apply the KJ approximation, we write

$$\begin{aligned} V(\Omega_0, \Omega_1) &= 2J S_0 \cdot S_1, \\ g_0(\Omega_0) &= -2\mu (A_3 + A_4) S_{0z}, \\ g_1(\Omega_1) &= -2\mu (A_3 - A_4) S_{1z} \text{ and} \\ h_j(\Omega_j) &= -2\mu B S_{jz} \end{aligned} \quad (6)$$

where S_k is the vector spin operator in units of \hbar of the k -th spin taking only the allowed values of Ω_k . J is the coupling constant due to the exchange interaction and is positive. We assume that the ions 0 and 1 are in the A and B sublattices respectively. A_3 and A_4 are the effective fields acting on an outer ion, A_4 being a 'staggered' field with reference to A and B sublattices as is evident from eq. (6). A_3 arises entirely because of the external field B , and includes the effect of the induced magnetisation of the medium due to B . In the spirit of the KJ model, we assume that the coupling between the central ion and any one of the outer ions is independent of the other outer ions of the cluster. Then (4) is reduced to

$$\begin{aligned} & \frac{P^{(0)} \exp [2\mu\beta \{(A_3 + A_4) - B\} S_{0z}]}{\left[\sum_{S_j} \exp [2\beta \{-J S_0 \cdot S_j + \mu (A_3 - A_4) S_{jz}\}] \right]^{Z-1}} \\ &= \frac{P^{(1)} \exp [2\mu\beta \{(A_3 - A_4) - B\} S_{1z}]}{\left[\sum_{S_j} \exp [2\beta \{-J S_1 \cdot S_j + \mu (A_3 + A_4) S_{jz}\}] \right]^{Z-1}} \end{aligned} \quad (7)$$

where the summation is over all possible orientations of the j -th spin,

$$P^{(0)} = \sum_{S_0} \exp (2\mu\beta B S_{0z}) \left[\sum_{S_j} \exp [2\beta \{-J S_0 \cdot S_j + \mu (A_3 - A_4) S_{jz}\}] \right]^Z$$

and

$$P^{(1)} = \sum_{S_1} \exp (2\mu\beta B S_{1z}) \left[\sum_{S_j} \exp [2\beta \{-J S_1 \cdot S_j + \mu (A_3 + A_4) S_{jz}\}] \right]^Z. \quad (8)$$

We see from eqs (8) that $P^{(0)} \neq P^{(1)}$. Multiplying the numerator and denominator of the LHS of eq. (7) by $\exp [2(Z-1)\mu\beta(A_3 + A_4)S_{0z}]$, it can be seen that the denominator can be expressed in terms of an effective Hamiltonian for a coupled pair, viz.,

$$H_e = 2J S_0 \cdot S_j - 2\mu (A_3 + A_4) S_{0z} - 2\mu (A_3 - A_4) S_{jz}. \quad (9)$$

Similarly, $P^{(1)}$ as well as the RHS of eq. (7) can be expressed in terms of H_e . As observed by Kasteleijn and Kranendonk H_e is a non-diagonal matrix in the $H^{(2)}$ representation. The eigen values of H_e for spin $\frac{1}{2}$ particles and the non-vanishing elements of the density matrix are given by Kasteleijn and Kranendonk (1956).

Using these values in (7-9) we get

$$\begin{aligned} \frac{[U^Z W^{Z-1} Y^{-1}]^{1/2}}{[X_+ + U]^{Z-1}} R^{(0)} &= \frac{[U^{-Z} W^{-(Z-1)} Y]^{1/2}}{[X_- + 1/U]^{Z-1}} R^{(0)}, \\ &= \frac{[U^Z W^{-(Z-1)} Y^{-1}]^{1/2}}{[X_- + U]^{Z-1}} R^{(1)} = \frac{[U^{-Z} W^{(Z-1)} Y]^{1/2}}{[X_+ + 1/U]^{Z-1}} R^{(1)} \end{aligned} \quad (10)$$

where

$$\begin{aligned} R^{(0)} &= \frac{P^{(0)}}{\{\text{Tr} [\exp (-\beta H_e)]\}^Z} = [U^{-Z} W^{-(Z-1)} Y]^{1/2} (X_+ + U)^Z \\ &\quad + [U^Z W^{(Z-1)} Y^{-1}]^{1/2} (X_- + 1/U)^Z = Q_+^{(0)} + Q_-^{(0)} \text{ (say),} \end{aligned}$$

and

$$\begin{aligned} R^{(1)} &= \frac{P^{(1)}}{\{\text{Tr} [\exp (-\beta H_e)]\}^Z} = [U^{-Z} W^{(Z-1)} Y]^{1/2} (X_- + U)^Z \\ &\quad + [U^Z W^{-(Z-1)} Y^{-1}]^{1/2} (X_+ + 1/U)^Z = Q_+^{(1)} + Q_-^{(1)} \text{ (say),} \end{aligned}$$

$$\begin{aligned} X_{\pm} &= \frac{1}{2} \{ (1 \pm \sin \omega) \exp \{ \beta [J + (J^2 + 4\mu^2 A_4^2)^{1/2}] \} \\ &\quad + (1 \mp \sin \omega) \exp \{ (\beta [J - (J^2 + 4\mu^2 A_4^2)^{1/2}]) \} \}, \end{aligned}$$

$$\sin \omega = 2\mu A_4 / (J^2 + 4\mu^2 A_4^2)^{1/2},$$

$$U = \exp (2\beta \mu A_3), \quad Y = \exp (2\beta \mu B), \quad W = \exp [2\beta \mu Z A_4 / (Z-1)],$$

and $Q_+^{(0)}$ is the probability for a spin on the A -sublattice to have a positive projection along the z axis, etc. It turns out that only two of the eqs (10) are independent, which can be conveniently written in the form

$$Y = U \left[\frac{W(1 + UX_-)}{(X_+ + U)} \right]^{Z-1} = U \left[\frac{(1 + UX_+)}{W(X_- + U)} \right]^{Z-1}. \quad (11)$$

The long range order parameter of the spins on the A -sublattice is given by

$$2\langle S_{0z} \rangle = \zeta + s = \frac{Q_+^{(0)} - Q_-^{(0)}}{R^{(0)}}.$$

Similarly $2\langle S_{1z} \rangle = \zeta - s$ represents the order parameter for the spins on the B -sublattice. Using eqs (11) and some algebra, we get

$$\zeta \pm s = \tanh \beta \left[\mu \beta - \frac{Z}{2} J \left\{ \Phi(\zeta, s, \beta) \mp \varphi(\zeta, s, \beta) \right\} \right] \quad (12)$$

where

$$\Phi(\zeta, s, \beta) = \frac{1}{\beta J} \left[\frac{1}{2} \ln \left\{ \frac{(1-\zeta-s)(1-\zeta+s)}{(1+\zeta+s)(1+\zeta-s)} \right\} + 2\beta\mu A_3 \right]$$

$$\varphi(\zeta, s, \beta) = \frac{1}{\beta J} \left[\frac{1}{2} \ln \left\{ \frac{(1+\zeta+s)(1-\zeta+s)}{(1-\zeta-s)(1+\zeta-s)} \right\} - 2\beta\mu A_4 \right].$$

Equations (11) and (12) which determine the effective field parameters A_3 and A_4 as well as the long range order parameters ($\zeta \pm s$) are the basic equations of the KJ model and are identical with the results of the constant coupling approximation (Kasteleijn and Kranendonk 1956). In fact it can be easily shown that the (nearest neighbour) short range order parameter $4(S_0 \cdot S_1)$, the Neel temperature T_N , etc. are also exactly identical with the results of the CC theory. As in the latter theory, we also get the unphysical result of an 'anti-Neel' temperature (see, for instance, the discussion by Smart 1966).

3.2. External field perpendicular to the easy axis

Let B be a vanishingly small external field applied in the direction \mathbf{x} . The Hamiltonian of the spin system is now given by

$$H = \sum_{(i,j)} H^{(2)} = \sum_{(i,j)} 2J \mathbf{S}_i \cdot \mathbf{S}_j - \frac{2\mu B}{Z} (S_{ix} + S_{jx}).$$

The potential energies describing the model are given by

$$\begin{aligned} V(\Omega_0, \Omega_1) &= 2J \mathbf{S}_0 \cdot \mathbf{S}_1, \\ g_0(\Omega_0) &= -2\mu (A_3 S_{0x} + A_4 S_{0z}), \\ g_1(\Omega_1) &= -2\mu (A_3 S_{1x} - A_4 S_{1z}) \text{ and} \\ h_j(\Omega_j) &= -2\mu B S_{jx}. \end{aligned} \tag{13}$$

The net field, acting on say the A -sublattice, is now inclined at a small angle to the z axis. S_{ix} and S_{iz} are the projections of the spin i along the \mathbf{x} and z directions respectively. Each of them can take two values S_{ix}^+ , S_{ix}^- and S_{iz}^+ , S_{iz}^- corresponding to the spin i being in the $+\frac{1}{2}$ or $-\frac{1}{2}$ states respectively. A_4 is the staggered field and A_3 , as in the previous case is the field arising from the external field.

Using the potential energies given by eqs (13) in the Krieger-James approximation, i.e., eq. (4), we can write two independent consistency conditions corresponding to the ions 0 and 1 belonging to A and B sublattices as follows:

$$\frac{\exp \left[2\beta\mu \left\{ (A_3 - B) S_{0x}^+ + A_4 S_{0z}^+ \right\} \right]}{\left[W_A^+ \right]^{Z-1}} = \frac{\exp \left[2\beta\mu \left\{ (A_3 - B) S_{0x}^- + A_4 S_{0z}^- \right\} \right]}{\left[W_A^- \right]^{Z-1}} \tag{14a}$$

$$\frac{\exp \left[2\beta\mu \left\{ (A_3 - B) S_{1x}^+ - A_4 S_{1z}^+ \right\} \right]}{\left[W_B^+ \right]^{Z-1}} = \frac{\exp \left[2\beta\mu \left\{ (A_3 - B) S_{1x}^- - A_4 S_{1z}^- \right\} \right]}{\left[W_B^- \right]^{Z-1}} \tag{14b}$$

$$\zeta_x = \left(\frac{A_3}{A_4}\right) \left[\frac{1-\cos \omega}{1+\cos \omega} \exp(-\beta\lambda_0) + \frac{1+\cos \omega}{1-\cos \omega} \exp(-\beta\lambda_2) \right. \\ \left. + \frac{4 \cos \omega}{\sin^2 \omega} \exp(-\beta\lambda_3) \right] \frac{\sin \omega}{\sum_{\nu=0}^3 \exp(-\beta\lambda_\nu)} \tag{16a}$$

and

$$s_z = \frac{[\exp(-\beta \lambda_0) - \exp(\beta \lambda_2)] \sin \omega}{\sum_{\nu=0}^3 \exp(-\beta \lambda_\nu)} . \tag{16b}$$

In order to eliminate A_3 and A_4 from eqs (16), it is convenient to rewrite eqs (15) for ζ_x and s_z in a somewhat different form. For this purpose, we use the expressions for $W_A^{+,-}$ and $W_B^{+,-}$ given after eqs (4) and get

$$\zeta_x = \frac{S_{0x}^+ \exp(2\beta\mu BS_{0x}^+) [W_A^+]^Z + S_{0x}^- \exp(2\beta\mu BS_{0x}^-) [W_A^-]^Z}{\exp(2\beta\mu BS_{0x}^+) [W_A^+]^Z + \exp(2\beta\mu BS_{0x}^-) [W_A^-]^Z} \\ + \frac{S_{1x}^+ \exp(2\beta\mu BS_{1x}^+) [W_B^+]^Z + S_{1x}^- \exp(2\beta\mu BS_{1x}^-) [W_B^-]^Z}{\exp(2\beta\mu BS_{1x}^+) [W_B^+]^Z + \exp(2\beta\mu BS_{1x}^-) [W_B^-]^Z} .$$

Using the Krieger-James consistency conditions (14), W_A , W_B can be eliminated to get

$$\zeta_x = \sum_{S_0} \sum_{S_1} (S_{0x} + S_{1x}) \exp(-\beta H_0) / \sum_{S_0} \sum_{S_1} \exp(-\beta H_0) \tag{17a}$$

and similarly

$$s_z = \sum_{S_0} \sum_{S_1} (S_{0z} - S_{1z}) \exp(-\beta H_0) / \sum_{S_0} \sum_{S_1} \exp(-\beta H_0) \tag{17b}$$

where

$$H_0 = -\frac{2\mu}{Z-1} [(ZA_3 - B)(S_{0x} + S_{1x}) + ZA_4(S_{0z} - S_{1z})].$$

The exact eigen values of H_0 are given by

$$\lambda_0^0 = -\frac{2\mu ZA_4}{Z-1} \left[1 + \left(\frac{ZA_3 - B}{ZA_4} \right)^2 \right]^{\frac{1}{2}}, \\ \lambda_1^0 = 0, \\ \lambda_2^0 = \frac{2\mu ZA_4}{Z-1} \left[1 + \left(\frac{ZA_3 - B}{ZA_4} \right)^2 \right]^{\frac{1}{2}} \text{ and} \\ \lambda_3^0 = 0. \tag{18}$$

Using eqs (17) and (18), doing some algebra, we get

$$(Z-1) \frac{\zeta_x}{\zeta} \ln \left[\frac{1-\zeta}{1+\zeta} \right] - 2\mu\beta[Z A_3 - B] = 0 \quad (19a)$$

$$(Z-1) \frac{s_z}{\zeta} \ln \left[\frac{1-\zeta}{1+\zeta} \right] - 2\mu\beta Z A_4 = 0 \quad (19b)$$

$$\text{where } \zeta^2 = \zeta_x^2 + s_z^2.$$

Equations (16) and (19) are exactly identical with the results of the CC approximation. They can be used to eliminate A_3 , A_4 and s_z to get the perpendicular susceptibility (Kasteleijn and Kranendonk 1956).

The theory of the Heisenberg model of *ferromagnetism* is obtained by replacing J by $-J$ and equating A_4 to 0 in equation (6). In the Ising case the proof is trivial. In these cases equivalent results have been obtained by Elliot and Marshall (1958) and hence will not be discussed here.

Thus the constant coupling approximation implies a simple physically consistent description of the (nearest neighbour) short range order in the medium; viz., that the probability for the occurrence of any particular configuration of a near-neighbour pair should be independent of the choice of the central ion between them. It also means that unlike the models which employed other consistency conditions for the description of magnetism (Weiss 1948; Oguchi 1955), the KJ method leads to solutions ensuring thermodynamic equilibrium of the medium (see also Strieb *et al* 1963).

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