

Green Function Theory of Random Ferromagnets with Large Exchange Anisotropy

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Recebido em 9 de Fevereiro de 1977

Spin 1/2 systems which are coupled with an Ising-like Hamiltonian with fluctuating exchange are discussed by the use of thermodynamic Green functions in four different approximations. The first is equivalent to a local mean field approximation and the second is an approximation to the first in which the local mean field is assumed to be proportional to the overall magnetization and to the sum of the neighboring exchange "bonds". The third is also a special case of the first, but the local mean field is approximated in such a way as to be appropriate for the discussion of spin glasses. The fourth is a coherent potential - like approximation (CPA). The second approximation can be shown to imply no reduction of the Curie temperature (T_c) due to exchange fluctuations, while the CPA does result in a lowering of T_c (assuming small fluctuations about an average positive exchange in both cases). However, the CPA that was used is also an approximation to the second method. Hence, even though the CPA does result in a lowering of T_c (which is generally conceded to be correct for the original model), this fact cannot be used as an argument for the validity of the CPA. Our calculation thus emphasizes the necessity of critically examining any CPA-like calculation before accepting its predictions as valid.

Sistemas de spin 1/2 com acoplamento do tipo Ising, no qual a interação de intercâmbio é variável, são estudados através de funções de Green termodinâmicas em quatro diferentes aproximações. A primeira é equivalente à aproximação de campo médio local, enquanto na segunda - uma aproximação da anterior - o campo médio é assumido proporcional à mag-

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netização total e a soma das constantes de interação com os vizinhos. A terceira é também um caso especial da primeira, mas aqui o campo médio local \bar{e} é aproximado de maneira a representar um vidro de spin. A quarta é uma aproximação de potencial coerente (CPA). Resulta a segunda aproximação em uma não diminuição da temperatura de Curie (T_c) devido às flutuações da interação de troca enquanto que a CPA resulta em uma diminuição de T_c (supondo-se pequenas flutuações em torno de uma interação média positiva em ambos os casos). Todavia, a CPA utilizada é também uma aproximação do segundo método. Em consequência, apesar do uso da CPA resultar em uma redução de T (o que é geralmente reconhecido como sendo correto para o modelo original), esse fato não pode ser usado como argumento para a validade da CPA. Nosso cálculo, assim, enfatiza a necessidade de se realizar um exame crítico de qualquer cálculo do tipo CPA, antes de se aceitar suas previsões.

1. INTRODUCTION

Since the classic paper of Zubarev¹, the use of thermodynamic Green functions in solid state physics has increasingly grown. In particular, the magnetic properties of solids have been extensively studied by Green functions. In recent years the study of magnetism in amorphous materials has been spurred by the large technological progress that has been made in connection with metallic glasses²⁻⁵. At the same time the theoretical techniques for handling non-crystalline solids have been improving⁶, and it has been found that thermodynamic Green functions in connection with 'coherent potential approximations' can be very useful for calculating the properties of solids with some degree of randomness. The existence of a new type of phase (the spin glass) has even apparently been found (both experimentally and theoretically) in certain kinds of random systems at sufficiently low temperatures⁷. In Section 2 of this paper we define our model and set up the basic equations for the thermodynamic Green functions which we will use. In Section 3, we solve these equations in a simple random phase approximation, and show that our results are equivalent to a local mean field theory. In Section 4, we make two further approximations, one of which will be used

again in Section 5 and the other will allow us to make a short discussion of spin glasses. In Section 5, we solve for the magnetization of our system within a random phase coherent potential-like approximation. Despite the plausibility of this CPA method and the reasonableness of its results, our use of a coherent potential-like approximation has certain difficulties. It is suggested that this calculation can serve as a warning against the uncritical acceptance of the predictions of such approximations.

2. MODEL AND GREEN FUNCTION FORMALISM

Let $G_E(A,B)$ be the energy (E) dependent Fourier time transform of the double time - temperature dependent thermodynamic Green function (retarded if $\text{Im}(E) > 0$, advanced if $\text{Im}(E) < 0$) associated with operators A and B, Refs. 1,8. Then, as is well known, the Green functions satisfy

$$EG_E(A,B) = \frac{1}{2\pi} \langle [A,B] \rangle + G_E([A,\hat{H}],B) , \quad (1)$$

where $\langle \dots \rangle$ refers to a quantum statistical average, H is the Hamiltonian, and $[A,B] = AB - BA$. A common way of solving Eq.(1) is to write $G_E([A,\hat{H}],B)$ in terms of $G_E(A,B)$ by some decoupling approximation^{1,8}. Assuming Eq. (1) can be solved for $G_E(A,B)$, from the general theory of Green functions we have for equal time correlation functions^{1,8},

$$\langle BA \rangle = i \int_{-\infty}^{\infty} \frac{G_{E+i\epsilon}(A,B) - G_{E-i\epsilon}(A,B)}{e^{\beta E} - 1} dE , \quad (2)$$

where the $\epsilon \rightarrow 0$ limit is to be taken after the integral is performed and $\beta = 1/(kT)$, where k is Boltzmann's constant and T is the temperature. Eqs. (1) and (2) are the basic Green functions equations that we will need.

The Hamiltonian describing the interacting spins, each with spin $S = 1/2$, is assumed to be

$$H = \frac{1}{2} \sum_{i,j} J_{i,j} S_{iz} S_{jz} - B_0 \sum_i S_{iz} , \quad (3)$$

where the \bar{i} and j label different lattice sites and $J_{i,j} = J_{ji}$ is zero unless i and j refer to nearest neighbors (n.n.) sites. The $J_{i,j}$ will be allowed to fluctuate from n.n. pair to n.n. pair and except for the discussion of spin glasses we will assume $\bar{J} > 0$ and for \bar{i} and j n.n. pairs $(|J_{i,j} - \bar{J}|/J) \ll 1$, where \bar{J} refers to the average of all $J_{i,j}$ (which are not zero). For simplicity, we will assume the number of n.n.'s (Z) is 6 (consistent with a simple cubic structure). The second term on the right of Eq. (3) is the Zeeman term, where B_0 is the magnetic field in suitable units. We will also use units with $\hbar = 1$. The Hamiltonian (3) can be viewed as an Ising-like Hamiltonian which results from a Heisenberg Hamiltonian in an appropriate limit of extreme exchange anisotropy. If we let

$$A = S_j^+ \equiv S_{jx} + iS_{jy} \quad \text{and} \quad B = S_i^- \equiv S_{ix} - iS_{iy} ,$$

Eq. (1) becomes

$$E G_E(S_j^+, S_i^-) = \frac{1}{\pi} \langle S_{iz} \rangle \delta_{i,j} + G_E((F_j + B_0) S_j^+, S_i^-) , \quad (4)$$

where

$$F_j = \sum_{j'} J_{jj'} S_{j'z} . \quad (5)$$

Note that $\langle F_j \rangle$ is the local molecular field associated with site j . It is also worth noticing for $S = 1/2$ and $i = j$ that

$$\langle BA \rangle = \langle S_j^- S_j^+ \rangle = \frac{1}{2} - \langle S_{jz} \rangle . \quad (6)$$

3. RANDOM PHASE APPROXIMATION

The random phase approximation consists simply in assuming

$$G_E(F_j S_j^+, S_i^-) \approx \langle F_j \rangle G_E(S_j^+, S_i^-) \quad (7)$$

Using Eqs. (7) and (4) we then find

$$G_E(S_j^+, S_i^-) = \frac{1}{\pi} \frac{\langle S_{iz} \rangle \delta_{ij}}{\tilde{E} - \langle F_j \rangle} \quad (8)$$

where $\tilde{E} = E - B$. Using that the Dirac delta function $\delta(x)$ can be represented as

$$(2\pi i) \delta(E-K) = \lim_{\epsilon \rightarrow 0} \left[\frac{1}{E-i\epsilon-K} - \frac{1}{E+i\epsilon-K} \right] \quad (9)$$

we then obtain by Eqs. (2), (6), (8) and (9) with $\bar{\mu} = j$

$$\frac{1}{2} - \langle S_{iz} \rangle = 2 \langle S_{iz} \rangle \left[\frac{1}{e^{\beta(\langle F_i \rangle + B_0)} - 1} \right] \quad (10)$$

Eq. (10) can be recast into the more familiar form

$$\langle S_{iz} \rangle = \frac{1}{2} \tanh \left[\frac{1}{2} \beta(\langle F_i \rangle + B_0) \right] \quad (11)$$

If J_{ij} did not fluctuate, this would be exactly the result of Weiss mean field theory for a crystalline spin 1/2 ferromagnet. However since it does, the $\langle S_{iz} \rangle$ vary from site to site and so does the local mean field $\langle F_i \rangle$. Thus, we see that the random phase approximation just gives us the same result as would be obtained by a local mean field theory in the case of an Ising-like interaction. It is quite possible that Eq.(11) is not a very good representation of the system. This is because iterative computer studies on Eq. (11) with $B_0 = 0$ have led to the prediction⁹ that T_C is increased by fluctuations in J_{ij} ; whereas more realistic

studies⁹ as well as rigorous results¹⁰ suggest that a fluctuation in J_{ij} will decrease T_c with respect to the corresponding crystalline (c) system in which $T_c^{(c)}$ is determined by $J_{ij}^{(c)} = \bar{J}$ for i and j n.n.'s. A simple calculation based on Eq.(11) and on the correlation idea of Richards¹¹, also resulted in an increase in T_c , Ref. 12.

4. SPIN GLASSES AND OTHER DECOUPLING APPROXIMATIONS

We now make an even cruder decoupling approximation which is consistent with the type of decoupling that has been used by Tahir-Kheli in a discussion of a random bond Heisenberg ferromagnet¹³. We assume

$$G_E(F_j S_j^+, S_i^-) \approx M \sum_j F_j J_{ij} G_E(S_j^+, S_i^-) \quad (12)$$

where $M = \overline{\langle S_{jz} \rangle}$ is the overall magnetization for a quenched system¹⁴. The bar refers to averaging over the J_{ij} 's where each J_{ij} (from n.n. pairs) is randomly determined by some probability distribution which is the same for each J_{ij} . Going through the same sort of manipulation which led to Eq.(11), we now clearly obtain

$$\langle S_{iz} \rangle = \frac{1}{2} \tanh \left[\frac{1}{2} \beta (M \sum_j J_{ij} J_{jz} + B_0) \right] . \quad (13)$$

To find the Curie temperature, we assume M is small (and $B_0 = 0$) so

$$\langle S_{iz} \rangle = \frac{\beta_c}{4} M \sum_j J_{ij} J_{jz}, \quad M \rightarrow 0 . \quad (14)$$

If $Z = 6$, we find since $\bar{J}_{ij} J_{ij} = \bar{J}$, $M = \overline{\langle S_{jz} \rangle}$,

$$k T_c = \frac{1}{\beta_c} = \frac{3\bar{J}}{2} . \quad (15)$$

In this approximation there is no lowering of Curie temperature due to fluctuating the J_{ij} 's in the random system. That is, $T_c^{(R)} = T_c^{(c)}$ (with $J_{ij}^{n.n.} = \bar{J}$ for $T_c^{(c)}$), where $T_c^{(R)}$ and $T_c^{(c)}$ refer to the Curie temperature

of the random and crystalline systems, respectively. Note that after the approximation of Eq. (12), we have made no further approximations in arriving at the result of Eq. (15). We will return to this later.

Let us now apply our results to situations where spin glass behavior may occur. We assume J_{ij} can now be either positive or negative but $\bar{J} \geq 0$. We also assume the J_{ij} 's are distributed in such a fashion that ferromagnetism is the only possibility for long range order (LRO). For our situation, we can then define spin glasses by requiring two conditions (with $B_0 = 0$):

$$M = \langle \overline{S_{iz}} \rangle = 0, \quad (16a)$$

$$q = \langle \overline{S_{iz}} \rangle^2 \neq 0. \quad (16b)$$

Eq. (16a) implies there is no LRO. The parameter q is also an order parameter, but it measures local order. For example, if each spin were 'frozen' in position at low temperature in such a way that it had a preferred direction which was randomly distributed along either $\pm z$ from site to site, then $M = 0$, $q \neq 0$ and we have a spin glass state.

Clearly the decoupling represented by Eq. (12) is not appropriate for the discussion of spin glasses, for this decoupling implies if $B = 0$, $M = 0$, then $\langle S_{iz} \rangle = 0$ and hence $q = 0$. We need somehow to incorporate the idea that the molecular field $\langle F_i \rangle$ can be locally non zero even though $M = 0$. In particular we might expect that if $q \neq 0$, then $\langle F_i \rangle \neq 0$, since q , in some sense, measures local order. To formalize this, we use a decoupling like Eq. (7), but we assume that the $\langle F_j \rangle$ in Eq. (7) can be replaced by

$$\langle F_j \rangle \equiv 2M\bar{J} + Z(J_j - \bar{J})\sqrt{q}, \quad (17)$$

where

$$J_j = \frac{1}{Z} \sum_j J_{j^*j}. \quad (18)$$

The first term is exactly the molecular field that we would have for a crystalline system with $J_{ij} = \bar{J}$ for all n.n. pairs. The second term vanishes if $J_j = \bar{J}$, as it should, and it is also proportional to \sqrt{q} as we would expect on dimensional considerations alone ($q = M^2$ for a crystalline ferromagnetic system). The Z factor comes in because we would expect the ZM in the first term to be analogous to the factor which multiplies $(J_j - \bar{J})$ in the second. In fact, a method formally equivalent to assumption (17) has been developed by Klein¹⁵. Thus for the details of developing our assumptions in a more mathematical way, reference can be made to Klein's paper. Using Eqs. (7) and (17), and the same ideas that led to Eq. (11), we now obtain (with $B_0 = 0$),

$$\langle S_{jz} \rangle = \frac{1}{2} \tanh \left[\frac{1}{2} \beta (ZM\bar{J} + Z(J_j - \bar{J}) \sqrt{q}) \right]. \quad (19)$$

If we assume the J_j 's are independently distributed by a Gaussian distribution with width ΔJ , then so are the J_j 's but with width $\Delta J/\sqrt{Z}$. If we use for a probability density

$$P(J_j) = \frac{\sqrt{Z}}{\sqrt{2\pi} \Delta J} \exp \left[- \frac{Z(J_j - \bar{J})^2}{2(\Delta J)^2} \right], \quad (20)$$

then

$$M = \int_{-\infty}^{\infty} \langle S_{iz} \rangle P(J_j) dJ_j, \quad (21)$$

and

$$q = \int_{-\infty}^{\infty} \langle S_{iz} \rangle^2 P(J_j) dJ_j. \quad (22)$$

We do not carry this calculation any further, because Eqs. (21) and (22) (with Eqs. (19) and (20)) are formally the same Eqs. as derived by Sherrington and Kirkpatrick^{16,17} and in a rigorous fashion (for $Z \rightarrow \infty$) by Morita and Horiguchi¹⁸. Klein shows, however, that the same type of procedure leads to a slightly different specific heat from that of Sherrington and Kirkpatrick. For low enough temperatures and $\Delta J/(\bar{J})$

large enough Eqs. (21) and (22) predict $^{16,17}M = 0$ and $q \neq 0$, i.e., the spin glass state. Actually there are still some difficulties in present theories of the spin glass state when they are compared with all available experimental results¹⁹. Eqs. (21), and (22) are surely not the final and best solution for M and q. Mention here should also be made of the random site spin glass calculation of Luttinger²⁰, and of the random bond - random site Green function CPA calculation of Tahir-Kheli²¹. Both authors obtain the spin glass state for appropriate values of the parameters governing their systems. It is also worthwhile to note that annealed systems (in which the thermodynamics is determined by averaging (over the J_{ij} 's) the partition function rather than averaging the thermodynamic functions directly) are not expected to show spin glass behavior at all²².

5. A COHERENT POTENTIAL APPROXIMATION

We now rewrite Eq. (8) with $i = j$ and use the decoupling approximation (12) to obtain with $G_{\vec{r}}(S_i^+, S_i^-) \equiv G_j$,

$$G_j = \frac{1}{\pi} \frac{\langle S_{i,z} \rangle}{\tilde{E} - \Sigma_j} \quad (23)$$

where

$$\Sigma_j \equiv (\Sigma_j, J_{jj})M. \quad (24)$$

We now evaluate \overline{G}_j (and hence $M = \overline{\langle S_{i,z} \rangle}$) by Eqs. (2) and (6) following a coherent potential-like approximation which is somewhat similar to a calculation of Tahir-Kheli¹³. Let

$$A_j = \frac{1}{\pi} \langle S_{i,z} \rangle. \quad (25)$$

Then, we can write Eq. (23) as

$$(\tilde{E} - \Sigma_j)G_j = A_j. \quad (26)$$

We introduce a site independent C , to be determined later, and define

$$g^{-1} = \tilde{E} - \Sigma \quad (27a)$$

and

$$V_j = \Sigma_j - \Sigma. \quad (27b)$$

Then Eq. (26) can be written as

$$G_j = g A_j + g V_j G_j. \quad (28)$$

We introduce a T_j defined so

$$V_j G_j = T_j g A_j, \quad (29)$$

and thus Eq. (28) becomes

$$G_j = g A_j + g T_j A_j g. \quad (30)$$

We determine C so that

$$\overline{T_i A_i} = 0, \quad (31)$$

and hence the desired average Green function is

$$\overline{G_j} = g \overline{A_j} = \frac{1}{\pi} \frac{M}{\tilde{E} - \Sigma}. \quad (32)$$

The problem is solved, once we evaluate C from Eq. (31). We approximate¹³ $\overline{T_i A_i}$ by $\overline{T_i} \overline{A_i}$ and thus we determine C by

$$\overline{T_i} = 0. \quad (33)$$

Note that $\overline{T_i A_i} \approx \overline{T_i} \overline{A_i}$ is the only further approximation we have made beyond Eq. (12). Thus any deviation of our results from Eq. (15), which follows exactly from Eq. (12), must be due to this further approximation. In order to apply Eq. (33), we need a convenient expression for T_i . By Eq. (30), we have

$$V_j G_j = g V_j A_j + V_j g T_j A_j$$

or

$$T_j g A_j = V_j g A_j + g V_j T_j g A_j \quad \text{by Eq. (29),}$$

or

$$T_j = \frac{V_j}{1 - g V_j}$$

Using Eq. (27b), Eq. (33) becomes

$$\left[\frac{\Sigma_j - \Sigma}{1 - g(\Sigma_j - \Sigma)} \right] = 0, \quad (34)$$

where $g = (\tilde{E} - \Sigma)^{-1}$. Eqs. (32) and (34) are the basic equation of our coherent potential approximation.

To go further, we must assume a probability distribution for the J_{ij} 's. We assume, for $Z = 6$ and for n.n. interactions, that

$$J_{ij} = \bar{J} + \delta \quad \text{with } p = 1/2, \quad (35a)$$

and

$$J_{ij} = \bar{J} - \delta \quad \text{with } p = 1/2, \quad (35b)$$

where p is the probability. Also, we have in mind that $\bar{J} > 0$ and later

$$\Delta \equiv \delta/\bar{J} \ll 1, \quad (36)$$

will be assumed. Equation (34) then becomes

$$\frac{1}{64} \sum_{\ell=0}^6 \frac{6!}{\ell!(6-\ell)!} \left[\frac{M[\ell\bar{J}(1+\Delta) + (6-\ell)\bar{J}(1-\Delta)] - \Sigma}{1 - g[M(\ell\bar{J}(1+\Delta) + (6-\ell)\bar{J}(1-\Delta)) - \Sigma]} \right] = 0. \quad (37)$$

Although the details are not particularly edifying, it turns out that

Eq. (37) is not too difficult to solve for g (where $C = \tilde{E} - g^{-1}$); \bar{g}_3 can then be obtained from Eq. (32). Then using Eqs. (2), (6), and (9) and solving for $M = \langle \bar{S}_{jz} \rangle$, we finally obtain (with $B_0 = 0$):

$$\begin{aligned}
 M^{-1} = & \frac{1}{32} \{ \coth[\beta M(3\bar{J} - 3\delta)] + \coth[\beta M(3\bar{J} + 3\delta)] \\
 & + 6 (\coth[\beta M(3\bar{J} - 2\delta)] + \coth[\beta M(3\bar{J} + 2\delta)]) \\
 & + 15(\coth[\beta M(3\bar{J} - \delta)] + \coth[\beta M(3\bar{J} + \delta)]) \\
 & + 20 \coth(3\beta M\bar{J}) \} .
 \end{aligned} \tag{38}$$

Assuming M is small (T near $T_c^{(R)}$) and $A \equiv \delta/\bar{J} \ll 1$, we obtain from Eq. (38)

$$M \approx \left[\frac{k(T_c^{(R)} - T)}{2\bar{J}} \right]^{1/2}, \tag{39}$$

where

$$T_c^{(R)} \equiv T_c^{(e)} \left[1 - \frac{1}{6} (\delta/\bar{J})^2 \right] \tag{40}$$

and where

$$T_c^{(e)} = 3\bar{J}/(2k)$$

Eq. (40) should be an approximation to Eq. (15). It is not, in the sense that it gives the apparently pleasing result that small $\Delta > 0$ implies the Curie temperature is lowered. Thus only if we believe that the simultaneous applications of the approximation of Eq. (12) and the approximation $\overline{T_{iA_i}} \equiv \overline{T_{iA_i}}$ is somehow better than the approximation of Eq. (12) alone, can we accept Eqs. (39) and (40) as reliable predictions. More likely, the pleasing result of Eq. (40) results from a fortuitous cancellation of errors. In fact, approximations^{2,3} similar to those we have made (and which also result in a lowering of the Curie temperature) are known to be unsatisfactory in other respects^{2,4}. Our demonstration of inconsistency appears, however, to be particularly clear. We are not

claiming the CPA in general is not useful. We are only noting that its straightforward application can lead to inconsistent predictions. To improve our calculation we would have to improve on our decoupling approximation and/or try to improve on the approximation (Eq.(33)) used for determining C. This may lead to very deep waters, but unfortunately when one looks critically, the simplest, most straightforward calculation, even when it leads to reasonable results, may not be accurate to the order necessary to predict these results with confidence.

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