

Susceptibility and magnetisation of a random Ising model*

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Abstract. The susceptibility of a bond disordered Ising model is calculated by configurationally averaging an Ornstein-Zernike type of equation for the two spin correlation function. The equation for the correlation function is derived using a diagrammatic method due to Englert. The averaging is performed using bond CPA. The magnetisation is also calculated by averaging in a similar manner a linearised molecular field equation.

Keywords. Susceptibility; magnetisation; Ising model; spin correlation function.

1. Introduction

Recently, considerable attention, both theoretical as well as experimental, has been given to the study of thermodynamic properties of magnetic alloys (Montgomery *et al* 1969; Oguchi and Obokata 1969; Lagendijk and Huiskamp 1972; Breed *et al* 1973). Experimental studies on several systems have indicated that the data can be quite successfully interpreted in terms of disordered Heisenberg and Ising models. Lagendijk and Huiskamp (1972) have obtained good agreement of their data on specific heat and susceptibility of $\text{CO}_x\text{Zn}_{1-x}\text{Cs}_3\text{Cl}_5$ with theoretical considerations based on Ising antiferromagnet. Breed *et al* (1973), who have made measurements on $\text{KMn}_x\text{Mg}_{1-x}\text{F}_3$, $\text{K}_2\text{Mn}_x\text{Mg}_{1-x}\text{F}_4$ and $\text{K}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$, find again that their data on specific heat and susceptibility can be well interpreted in terms of a diluted antiferromagnetic Heisenberg model. The theoretical approximation employed in the above cases is the two spin cluster theory of Oguchi and Obokata (1969) or similar generalisations of the constant coupling method (Bhargava and Kumar 1976).

Given the fact that there exist magnetic systems which correspond closely to the idealised models it is of obvious interest to improve theoretical approximations to see if the comparison between the experiments and theoretical predictions is indeed justified. Further, there exists a class of experiments like elastic neutron scattering, which measure wave vector dependent properties. Such properties provide even more detailed tests of the theoretical features of the model. For calculation of wavevector dependent properties one has to go beyond the cluster approximations employed for calculation of free energy and other bulk thermodynamic quantities.

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The purpose of this paper is to present a calculation of the two site spin correlation function and hence the wavevector dependent susceptibility. We have also employed our method to calculate magnetisation for the Ising model in a new approximation. Our calculation is based on the recently developed techniques of multiple scattering theory (see Elliot *et al* 1974 for review). Here we first derive an Ornstein-Zernike type of equation for the two site correlation function in the random system using the diagrammatic procedure of Englert (1963). The approximation involved in the above derivation is the well known random phase approximation or the high density approximation of Brout (1965). This equation being linear in correlation function can be cast in the same form as the Green's function equation of motion for a particle propagating in the random potential. On comparing the RPA equation with the particle problem we find that the randomness occurs in the off-diagonal matrix elements, which in the Ising problem are simply the direct correlation functions βJ_{ij} 's. The energy-like parameter of the corresponding particle problem is simply unity (in appropriate units), while the potential depends upon temperature. The configuration averaging is performed using a 'bond CPA' type approximation. This gives us the averaged correlation functions and susceptibility. Some earlier workers (Montgomery *et al* 1969, Tahir-Kheli 1972, Harris *et al* 1974) have discussed random Heisenberg model in spin wave approximation. The single spin wave problem corresponds exactly to the particle problem and the use of multiple scattering formalism is quite straightforward there. The spin-wave theory is valid only at low temperatures, whereas the work described here is valid at temperatures above Curie temperature.

The random phase approximation employed here has an obvious disadvantage. For the pure system, its predictions for the thermodynamic quantities like uniform susceptibility and Curie temperature are same as those of molecular field theory. For the disordered system too, we find that our results for the above mentioned quantities are not too far from the predictions of molecular field theory. It should, however, be noted that the wave-vector dependent generalisations of molecular field theory for the disordered system are not as straightforward as they are for ordered systems. The reason is that the disordered systems are inhomogeneous and the various operators are not diagonal in the q -space, as they are for the ordered systems. For this reason, the linear RPA equation prove the most suitable one. The RPA equation and its generalisations have much theoretical value and have given a good qualitative account of the neutron scattering experiments. It, thus, seems worthwhile to see the predictions of such an approach for the disordered systems.

For calculating the averaged magnetisation, we linearise the molecular field equation for magnetisation. The linearised equation can again be configurationally averaged using the Green's function method mentioned above. Unlike earlier calculations (Tahir-Kheli 1972), our approximation takes explicit account of the fact that the magnetisation in a disordered system is inhomogeneous. Our approximation is not expected to be valid near Curie temperature, where the linearised molecular field equation fails in an obvious way.

We study here bond-disorder problem, which is not really appropriate to discuss the above mentioned materials, as disorder in these materials is more properly described as site-disorder. The site disorder problem is mathematically more difficult to treat, as the impurity potential extends to at least $Z+1$ sites, where Z is the coordination number of the lattice. The method described here can be generalized to

deal with this more complex problem by using the cluster generalizations of single bond CPA. Such a project is not undertaken here, as we intend to use a more realistic Hamiltonian for this problem.

2. Calculation of susceptibility

We consider a random Ising model defined by the Hamiltonian

$$H = - \sum_{\langle ij \rangle} J_{ij} \sigma(i) \sigma(j) \quad (1)$$

where σ 's are the spin variables taking values ± 1 the sum $\langle ij \rangle$ is over all the nearest neighbour sites, and the exchange integrals are taken to be random variables. Our explicit calculations are done for the following probability distribution: $J_{ij} = J_1$ with probability x and $J_{ij} = J_0$ with probability $1-x$.

We are interested in calculating the spin-correlation function C_{ij} defined as

$$C_{ij} = \langle \sigma(i) \sigma(j) \rangle. \quad (2)$$

This calculation will be performed by deriving an Ornstein-Zernike type equation for the correlation function. This equation for the random system may be derived using the diagrammatic method developed by Englert (1963). Referring the reader to the paper of Englert for terminology and proofs, we have merely outlined the procedure. To calculate the correlation function C_{ij} , we draw all possible linked, irreducible graphs joining the points i and j . This means that the graphs have no disconnected parts and there are no vertices where the graph can be split into different pieces by a line passing through a vertex that does not cut any bond. The rules for calculating the contribution of these graphs are: (i) to each line $i-j$, we attribute a factor βJ_{ij} , (ii) to each vertex, we attribute a renormalised semi-invariant $M_n(i)$, where n is the number of lines arriving at the vertex, (iii) we divide each graph by an appropriate symmetry factor and finally (iv) we sum over all the internal sites. The renormalised semi-invariants are obtained as follows

$$M_n(i) = \exp \left[\sum_k G_k(i) \frac{\partial^k}{\partial x^k} \right] M_n^0(x) \Big|_{x=0} \quad (3)$$

where

$$M_n^0(x) = \frac{d^n}{dx^n} \ln \cosh x \quad (4)$$

and $G_k(i)$ are the renormalized self-energy parts terminating with k -lines. The typical diagrams for the correlation functions and self-energy parts are shown in figures 1 and 2.

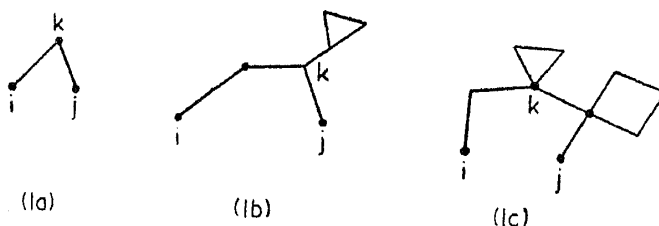


Figure 1. Some diagrams for the correlation function C_{ij} . (1a) irreducible, (1b) reducible at vertex k and (1c) reducible at vertices k and l .

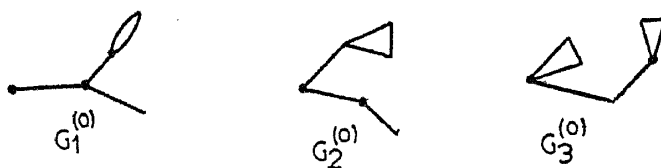


Figure 2. Some typical diagrams for self-energy parts $G_1^{(0)}$, $G_2^{(0)}$, and $G_3^{(0)}$. G_k is obtained by renormalising the vertices occurring in diagrams of G_k^0 .

Following Brout (1965), we make the high density approximation of retaining only those diagrams in which the two sites i and j are connected by only one path. This approximation gives the leading order terms in the parameter Z^{-1} where Z denotes the number of sites connected by J_{ij} . For the homogenous Ising model, this approximation is equivalent to the RPA approximation. Furthermore, we renormalise the semi-invariant at each vertex by first order self-energy G_1 . In this approximation we have

$$C_{ij} = M_1(i)M_1(j) + \beta J_{ij}M_2(i)M_2(j) + \beta^2 \sum'_p J_{ip}J_{pj}M_2(i)M_2(p)M_2(j) \\ + \beta^3 \sum'_{p,q} J_{ip}J_{pq}J_{qj}M_2(i)M_2(p)M_2(q)M_2(j) + \dots \quad (5)$$

where the prime on summation implies that the internal sites p, q , etc. occur just once in these terms. The corresponding diagrams are shown in figure 1. The semi-invariants M_1 and M_2 are given as

$$M_1(i) = \tanh [\beta \sum_j J_{ij}M_1(j)] \quad (6)$$

and

$$M_2(i) = 1 - M_1^2(i). \quad (7)$$

We now define a more convenient variable g_{ij} as

$$g_{ij} = \frac{C_{ij} - M_1(i)M_1(j)}{M_2(j)}. \quad (8)$$

In terms of g_{ij} , we can write eq. (5) as

$$g_{ij} = \beta J_{ij}M_2(i) + \beta^2 \sum'_p M_2(i)J_{ip}M_2(p)J_{pj} + \dots \quad (9)$$

This is a simple iterative equation except for the summation restrictions. We can convert it into an approximate integral equation, if we either ignore the summation restrictions altogether and multiply the kernel by K/Z , where K is the connectivity constant of the lattice. K^n gives approximately the number of self-avoiding walks of n steps, for large values of n . Thus we write

$$g_{ij} = v_{ij} + \alpha \sum'_p v_{ip}g_{pj} \quad (10)$$

where $v_{ij} = \beta M_2(i)J_{ij}$, and $\alpha = K/Z$. For the pure system, this procedure leads to the familiar random phase approximation—when we put $\alpha = 1$. The use of α , improves slightly the value of the critical temperature. It should be mentioned further that

this procedure is consistent to the leading order in Z^{-1} . Equation (10) can be brought into a more suitable form by defining

$$\mathbf{G} = [1 - \alpha \mathbf{v}]^{-1} \quad (11)$$

where we have used an obvious matrix notation. Then

$$\mathbf{g} = \mathbf{G} \mathbf{v} \quad (12)$$

If we split \mathbf{v} into a translationally invariant part and a random perturbation as

$$\alpha v_{ij} = v_{ij}^0 + \tilde{v}_{ij} \quad (13)$$

we can write eq. (11) as follows

$$G_{ij} = G_{ij}^0 + \sum_{p,l} G_{ip}^0 \tilde{v}_{pl} G_{lj} \quad (14)$$

where G_{ij}^0 obeys the equation

$$G_{ij}^0 = \delta_{ij} + \sum_l v_{il}^0 G_{lj}^0 \quad (15)$$

Equation (14) is a familiar random equation that occurs in the theory of electronic states in random crystals. The CPA type approximation is obtained by defining a medium characterised by a self-energy matrix ζ of the form

$$\zeta_{ij} = \zeta_0 \delta_{ij} + \zeta_1 \Delta_{ij} \quad (16)$$

where δ_{ij} is the usual Kronecker delta function, and Δ_{ij} is unity when i and j are nearest neighbours and is zero otherwise. We now write eq. (14) in terms of the medium Green functions G_{ij}^m which are defined as

$$\mathbf{G}^m = \mathbf{G}^0 + \mathbf{G}^0 \zeta \mathbf{G}^m \quad (17)$$

Equation (14) is then

$$G_{ij} = G_{ij}^m + \sum_{p,l} G_{il}^m V_{lp} G_{pj} \quad (18)$$

where
$$V_{pl} = \tilde{v}_{pl} - \zeta_{pl}. \quad (19)$$

Equation (19) is now expanded in a perturbation series in terms of potential \mathbf{V} . The terms of this series are arranged in a familiar \mathbf{T} matrix expansion in which the successive terms involve coherent scattering from one bond, two bonds, three bonds, etc., respectively. In CPA method (Sovan 1967 Elliott *et al* 1974) the self-energy is self-consistently determined by requiring that the averaged \mathbf{T} -matrix of the single bond scattering be set equal to zero. This requirement gives

$$\langle \mathbf{T}^{ij} \rangle = \langle \mathbf{V}^{ij} [1 - \mathbf{G}^{mij} \mathbf{V}^{ij}]^{-1} \rangle = 0 \quad (20)$$

where \mathbf{T}^{ij} , \mathbf{V}^{ij} , \mathbf{G}^{ij} are 2×2 matrices of the form

$$\mathbf{V}^{ij} = \begin{vmatrix} -\zeta_0 & \alpha v_{ij} - \zeta_1 \\ \alpha v_{ij} - \zeta_1 & -\zeta_0 \end{vmatrix} \quad (21)$$

$$\mathbf{G}^{mij} = \begin{vmatrix} G_{ii}^m & G_{ij}^m \\ G_{ji}^m & G_{jj}^m \end{vmatrix} \quad (22)$$

Equation (20) thus provides a set of two equations from which the quantities ζ_0 and ζ_1 can be evaluated as a function of temperature.

However averaging over V 's presents problems in this case, since v involves the factor of $M_2(i)$. Above Curie temperature $M_2=1$, so we simply have $v_{ij}=\beta J_{ij}-v_{ij}^0$. Below Curie temperature, we must obtain a probability distribution of $M_2(i)$ in terms of the given probability distribution of J_{ij} 's, by solving eqs (6) and (7). Equation (6) couples all the random variables $M_1(i)$'s and is too complicated to solve without further approximations. So we replace eq. (6) by

$$M_1 = \tanh [\beta \langle \sum_j J_{ij} \rangle M_1] \quad (23)$$

$$= \tanh \left[\frac{T_c}{T} M_1 \right]$$

where T_c will be determined by the divergence of high temperature susceptibility. By making this approximation, we neglect the segttering that arises due to fluctuations in magnetisation. This approximation does not seem so serious as we expect magnetisation to be more or less uniform except in cases of extreme disorder i.e. when J_1-J_2 is large.

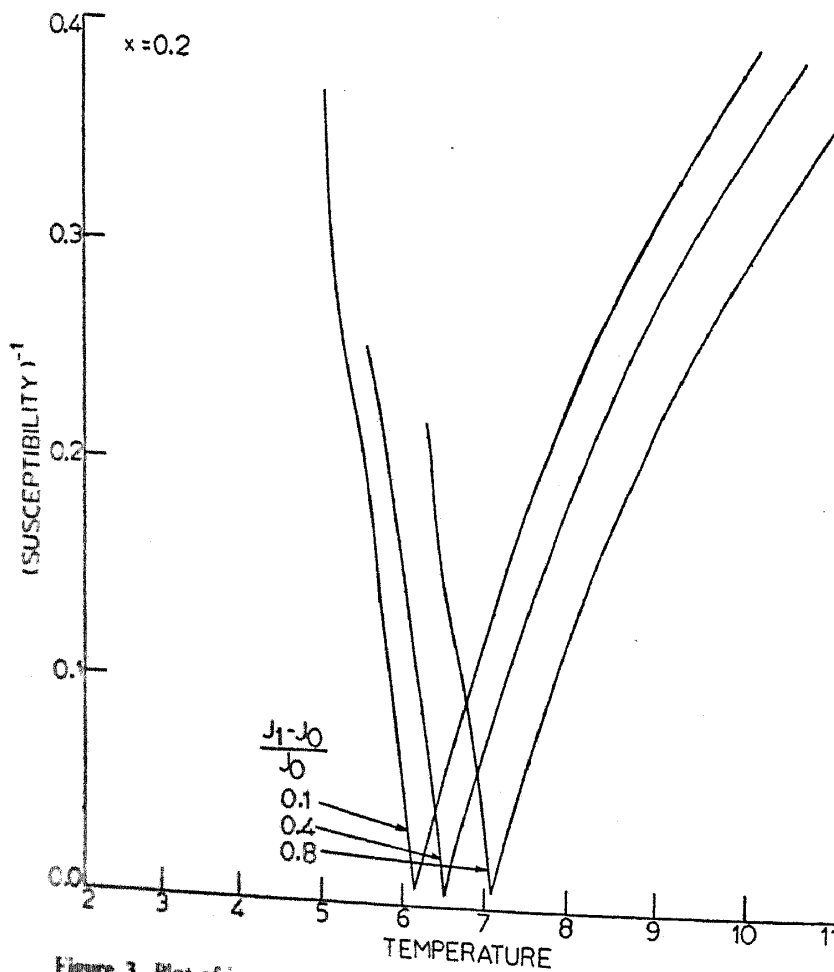


Figure 3. Plot of inverse susceptibility versus temperature for $x=0.2$.

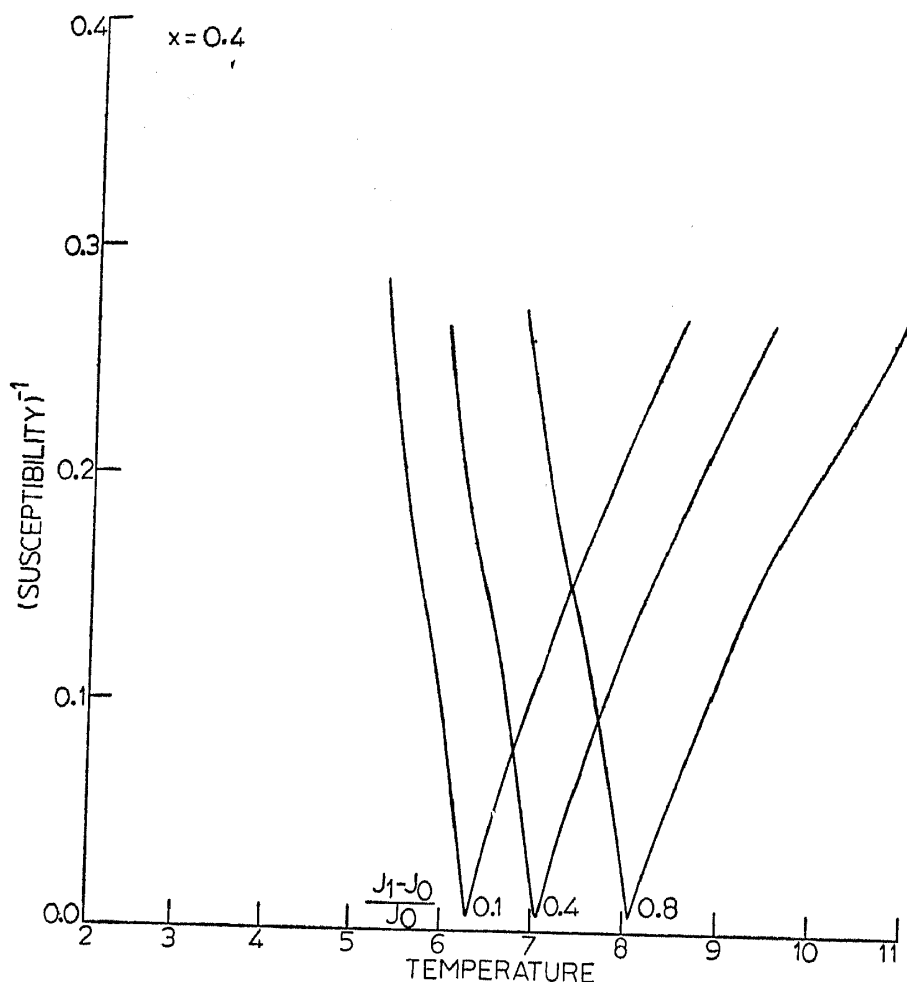


Figure 4. Plot of inverse susceptibility versus temperature for $x=0.4$.

The susceptibility may be calculated from the average Green's function $\langle G \rangle = G^m$. The expression for the wave-vector dependent susceptibility is

$$\begin{aligned} \chi(q, T) &= M_2 \langle g(q, T) \rangle \\ &= \frac{M_2}{1 - \zeta_0 - (v_0 + \zeta_1)s(q)} \end{aligned} \quad (24)$$

where $s(q) = \sum_{\delta} e^{iq \cdot \delta}$, where δ 's denote nearest neighbour lattice vectors; $v_0 = \beta J_0$. Equation (24) is also of the *OZ* form, ζ_0 and ζ_1 denote temperature dependent corrections due to disorder. We have solved eq. (18) numerically for various ratios of $(J_1 - J_0)/J_0$ and concentrations x . The results for static susceptibility $\chi(0)$ are shown in figures 3-5. The plots of χ^{-1} vs T show only slight deviations from linearity. The Curie temperature is determined by the equation

$$1 - \zeta_0(\beta_c) - Z(\beta_c J_0 + \zeta_1(\beta_c)) = 0. \quad (25)$$

A plot of Curie temperature vs concentration is shown in figure 6 for some ratios of $(J_1 - J_0)/J_0$.

The values of Curie temperature obtained in this approximation are close to the molecular field values. The reason is that the Curie temperature in the *OZ* approxi-

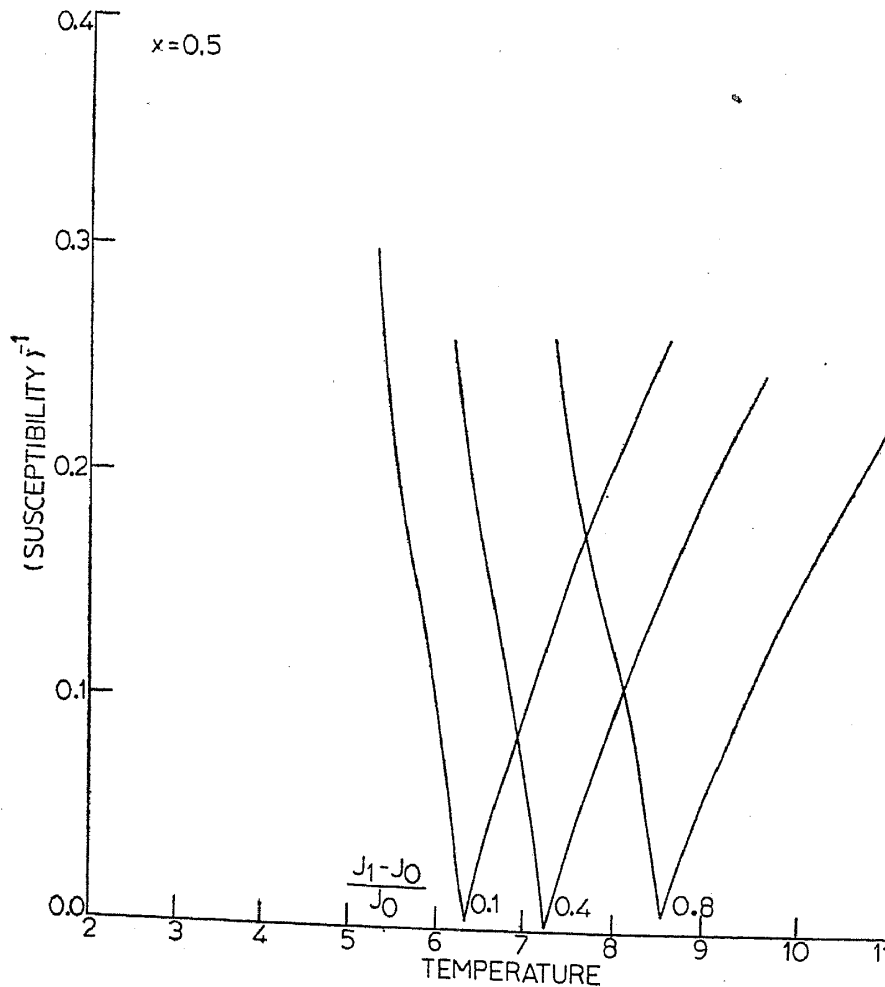


Figure 5. Plot of inverse susceptibility versus temperature for $x=0.5$.

mation for the pure system is the same as that in the molecular field approximation. The present theory being a self-consistent perturbation theory over the molecular field approximation of the random system, cannot give significant quantitative corrections for the Curie temperatures. The main advantage of the present theory is that we have obtained q -dependent susceptibility and spin correlation functions for the random system. Moreover this calculation demonstrates the use of multiple scattering theory in a novel context. An obvious way to improve the calculation of Curie temperature is to start with the Bethe-Peirels-Weiss approximation in the form developed by one of the authors (Kumar 1976). There again, one can obtain an equation for the correlation functions, which may be treated by multiple scattering theory. The Curie temperatures obtained in this manner may be comparable to those obtained by two spin clusters approximations (Bhargava and Kumar 1976).

3. Calculation of magnetisation

In this section we consider the calculation of magnetisation using eq. (6). For this purpose we also assume that an external uniform magnetic field H is present. We first linearise eq. (6) about a mean value m and then average the resulting linear

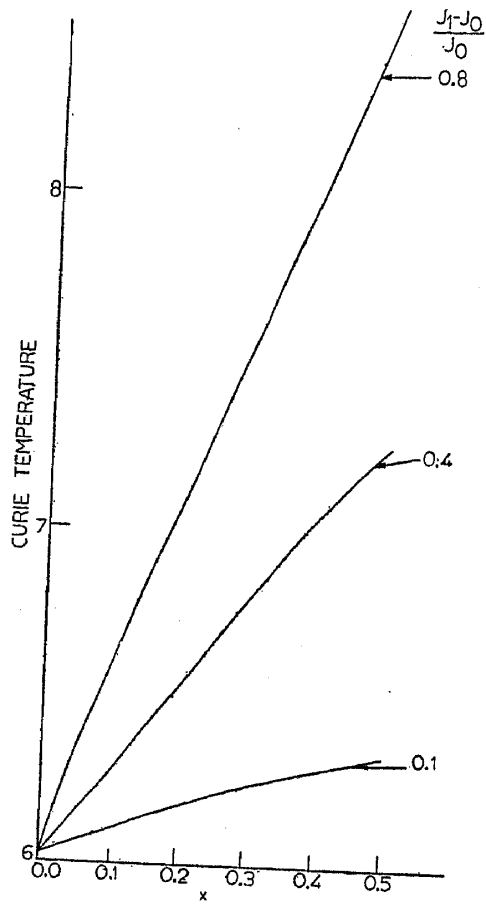


Figure 6. Variation of critical temperature with concentration.

equation in the manner described in the last section.

$$M_1(i) = m + \delta m_i \tag{25a}$$

where m is determined by eq. (21), i.e.

$$m = \tanh \left(g\mu\beta H + \frac{T_c}{T} m \right). \tag{26}$$

Then for δm_i , we have

$$\delta m_i = \tanh \beta [J_i(0)m + g\mu H + \sum_j J_{ij}\delta m_j - \tanh \beta [\bar{J}(0) + g\mu H]] \tag{27}$$

where $J_i(0) = \sum J_{ij}$ and $\bar{J}(0) = \langle J_i(0) \rangle$. The linearisation of eq. (27) yields

$$\sum_j (\delta_{ij} - v_{ij}) \delta m_j = X_i \tag{28}$$

where

$$X = m(1 - m^2) \beta (J_i(0) - \bar{J}(0)). \tag{29}$$

Thus

$$\delta m_j = \sum_j G_{ij} X_j. \tag{30}$$

The averaged magnetisation is

$$\begin{aligned} \langle \delta m \rangle &= \sum_j \langle G_{ij} X_j \rangle \\ &= \sum_J G_{ij}^m \langle X_j \rangle + \sum_{j, l, m} \langle G_{il}^m T_{lp} G_{pj}^m K_j \rangle. \end{aligned} \quad (31)$$

The first term vanishes. For the second term we use the Born approximation i.e. replace T_{lp} by V_{lp} . This may not be a bad approximation since G^m already contains self-consistent self-energies. From eq. (31), we obtain

$$\langle \delta m \rangle = Z\beta^2 m(1-m^2)^2 (\langle J^2 \rangle - \langle J \rangle^2) G(\mathbf{q}=0) [G_{00} + G_{0\delta}]. \quad (32)$$

From this expression it is clear that the corrections from average values are small at small temperatures. They become larger with the increasing temperature due to factor $G(\mathbf{q}=0)$. In fact they diverge as $(T_c - T)^{-1/2}$. This is an obvious contradiction to our assumption that average magnetisation becomes zero at the critical temperature. This should be taken to mean that the process of linearisation about the mean progressively fails as one approaches the critical point.

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