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Theory of A Magnetically Dilute System

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We deal with a randomly dilute ferromagnet with the magnitude of spin 1/2 by using the Constant-Coupling Method, in new and reasonable consideration of the short range order of the nearest neighbors proposed in the previous paper. The result is that the critical concentration, $x_c=2/z_2$ (Ising); $4/z_2$ (Heisenberg).

§1. Introduction

In the previous paper,[1] we proposed a formulation for a randomly dilute ferromagnet. However, so simple a manner of linking our formulation to the Green function has not us win what should be, otherwise, better than the simplest MFA-like result. Prior to this, Oguchi and Obokata[2] used a rather intuitively effective Hamiltonian method which seems consequently correspondent to the Constant-Coupling Method,[3] to find the dilution effect. In the present paper, we shall re-examine the very Constant-Coupling Method[3] with due consideration for the short range order in site-occupation configurations of magnetical atoms, and dilution effect will be explored in a new light.

In §2, first, in terms of Takagi's combinatory cluster variation method,[4],[5] we re-formulate the expression of the short range order parameter of the nearest neighbors in the previous paper,[1] into the form available for both Ising and Heisenberg spins of each magnitude 1/2. Next, we proceed to extend to our system the Constant-Coupling Method[3] which has gained success in the ordinary Heisenberg ferromagnet.

§2. Eormulation

We consider a dilute ferromagnetic system with a cubic Bravais structure and L lattice sites occupied quite randomly by N magnetic and $(L-N)$ nonmagnetic atoms. The vanishingly small external magnetic field $\boldsymbol{H}$ is applied chiefly to stabilize the spin arrays along a preferred axis z. For simplicity, we assume the magnitude of spin is 1/2 and omit the effect of the second nearest and further neighbor interactions. The system Hamiltonian can be written in the form

$$\mathscr{H}=-J\sum_i^L t_i t_{i+\delta}\{s_i^z s_{i+\delta}^z+u(s_i^x s_{i+\delta}^x+s_i^y s_{i+\delta}^y)\}-\gamma H\sum_i^L t_i s_i^z, \tag{1}$$

where the vectors $\boldsymbol{\delta}$ connect the i-th atom with its nearest neighbors; J is the positive exchange integral; u is the anisotropy parameter of exchange interaction which is reduced to the limiting value, zero or unity for Ising or Heisenberg ferromagnet, respectively; t_i, the occupation parameter takes unity or zero according as the i-th site is occupied by a magnetic atom or not.

We follow the previous paper[1] to summarize what we need and finally to derive α, the short range order parameter of the first neighbors in the occupation configurations, $\{t_i\}$. The reduced free energy is as follows,

$$\begin{aligned} f(\alpha, u) = \beta F/N = &-(1/N)\ln g_s - ((z_1-1)/x)\{x\ln x + (1-x)\ln(1-x)\} \\ &+ (z_1/2x)\{\alpha\ln\alpha + 2(x-\alpha)\ln(x-\alpha) + (1-2x+\alpha)\ln(1-2x+\alpha)\} \\ &-(1/2x)\beta J z_1 \alpha \chi_1(u) - \beta\gamma H < s_z > . \end{aligned} \tag{2}$$

where α and $\chi_1(u)$ are in the following mean values,

$$\alpha = \overline{t_i t_{i+\delta}}, \quad \chi_1(u) = < s_i^z s_{i+\delta}^z + u(s_i^x s_{i+\delta}^x + s_i^y s_{i+\delta}^y) > . \tag{3, 4}$$

As defined in the previous paper,[1] x is the molecular fraction N/L, g_s is the total number of spin configurations and β is $1/k_B T$.

Solving the minimum free energy condition,

$$[\partial f(\alpha, u)/\partial\alpha]_{T=T_c} = 0, \tag{5}$$

We get easily

$$\alpha_{T=T_c} = 2(1-C_1)/\{1-2xC_1 + \sqrt{[1-4x(1-x)C_1]}\}, \tag{6}$$

the value of which lies in the region

$$2x-1 \leqq \alpha \leqq x \ (1/2 \leqq x \leqq 1), \qquad 0 \leqq \alpha \leqq x \ (0 \leqq x \leqq 1/2), \tag{7}$$

where

$$C_1 = 1 - \exp(\beta_c J \chi_1(u)) . \tag{8}$$

It is to be noted that

$$\alpha_{T=T_c} \longrightarrow x_c \quad \text{as} \quad T_c \longrightarrow 0 \ (\beta_c \longrightarrow \infty) \tag{9}$$

where x_c is the value of x in the limit $T_c = 0$, namely, the critical concentration.

On the other hand, we may effectively identify our dilute system with the fully occupied system with the number of the magnetical nearest neighbors αz_1, to be denoted by z_1^*, in the nature of the short range order parameter. Thus, we can make use of the Constant-Coupling Method[3] which has gained good success in the fully occupied Heisenberg case, to evaluate T_c, $\chi_1(u)$ and so on.

We assume that the effective Hamiltonian for two atoms located on the i-th and its nearest neighboring $(i+\delta)$-th site would be

$$\mathscr{H}_{\mathrm{II}}=-J\{s_i^z s_{i+\delta}^z+u(s_i^x s_{i+\delta}^x+s_i^y s_{i+\delta}^y)\}-h(s_i^z+s_{i+\delta}^z), \tag{10}$$

where h is the variational parameter while J is the constant coupling coefficient as a matter of course. The total energy of our system is

$$\begin{aligned} &E(\beta,\ <s^z>)\\ &=<\textstyle\sum_i\sum_\delta\{-J[s_i^z s_{i+\delta}^z+u(s_i^x s_{i+\delta}^x+s_i^y s_{i+\delta}^y)]-\gamma H(s_i^z+s_{i+\delta}^z)/z_1^*\}>\\ &=-(1/2)Nz_1^*\chi_1(u)-N\gamma H<s^z>\\ &=-(1/2)Nz_1^*\{(\partial/\partial\beta)[\beta J\chi_1(u)-\textstyle\sum_\nu f_\nu \ln f_\nu]\}_{<s^z>}-N\gamma H<s^z>, \end{aligned} \tag{11}$$

where ε_ν and f_ν are eigenvalues of $\mathscr{H}_{\mathrm{II}}$ and $\exp(-\beta\mathscr{H}_{\mathrm{II}})/Z_{\mathrm{II}}$; $Z_{\mathrm{II}}=\mathrm{Tr}\exp(-\beta\mathscr{H}_{\mathrm{II}})$, respectively;

$$f_\nu=\exp(-\beta\varepsilon_\nu)/\textstyle\sum_{\nu=0}^{3}\exp(-\beta\varepsilon_\nu) \qquad (\nu=0,\ 1,\ 2,\ 3), \tag{12}$$

$$\varepsilon_0=(1+2u)J/4 \qquad \text{(singlet)}, \tag{13-1}$$

$$\varepsilon_1=-J/4-h \qquad \text{(triplet)}, \tag{13-2}$$

$$\varepsilon_2=(1-2u)J/4 \qquad (\quad '' \quad), \tag{13-3}$$

$$\varepsilon_3=-J/4+h \qquad (\quad '' \quad). \tag{13-4}$$

According to eqs. (4) and (12), $\chi_1(u)$ is given by

$$\begin{aligned} \chi_1(u)&=\mathrm{Tr}\,[\exp(-\beta\mathscr{H}_{\mathrm{II}})\{s_i^z s_{i+\delta}^z+u(s_i^x s_{i+\delta}^x+s_i^y s_{i+\delta}^y)\}]/Z_{\mathrm{II}}\\ &=(1/4)\{f_1+f_3+(2u-1)f_2-(2u+1)f_0\}\\ &=(1/4)\frac{\exp[\beta(J/4+h)]+\exp[\beta(J/4-h)]}{\exp[\beta(J/4+h)]+\exp[\beta(J/4-h)]}\\ &\qquad\frac{+(2u-1)\exp[-(1-2u)\beta J/4]-(2u+1)\exp[-(1+2u)\beta J/4]}{+\exp[-(1-2u)\beta J/4]+\exp[-(1+2u)\beta J/4]}; \end{aligned} \tag{14}$$

$$\chi_{1\ T=T_c}(0)=(1/4)\tanh(\beta_c J/2) \qquad \text{(Ising)}, \tag{15-1}$$

$$\chi_{1\ T=T_c}(1)=(3/4)[1-\exp(-\beta_c J)]/[3+\exp(-\beta_c J)] \qquad \text{(Heisenberg)}. \tag{15-2}$$

Integrating eq. (11) with respect to β, we obtain, for the partition function $Z(\beta,\ <s^z>)$,

$$\ln Z(\beta,\ <s^z>)=\beta N(\gamma H-z_1^* h)<s^z>+(z_1^* N/2)\ln Z_{\mathrm{II}}-(z_1^*-1)\ln Z(0,\ <s^z>), \tag{16}$$

where

$$Z(0, <s^z>) = N!/[N(1+2<s^z>)/2]![N(1-2<s^z>/2]!$$
$$= -(1/2)\,N[\textstyle\sum_{\nu=0}^{\nu=3} f_\nu \ln f_\nu]_{\beta=0}. \tag{17}$$

From the condition,

$$\partial/\partial <s^z> \ln(\beta, <s^z>)_\beta = 0, \tag{18}$$

we get

$$<s^z> = (1/2)\tanh\{[(1/2)\ln[(1+2<s^z>)/(1-2<s^z>)]]\}$$
$$= (1/2)\tanh\{\gamma\beta H/2 + (z_1^*/2)\ln[(1+2<s^z>)/(1-2<s^z>)]$$
$$-\beta z_1^* h/2\}. \tag{19}$$

Meanwhile, we have

$$<s^z> = (f_1 - f_3)/2$$
$$= \frac{1/2[\exp(\beta h) - \exp(-\beta h)]}{\exp(\beta h) + \exp(-\beta h) + \exp[-\beta J(1-u)/2] + \exp[-\beta J(1+u)/2]}. \tag{20}$$

From eqs. (19) and (20), the following relation can be derived

$$\exp(\beta\gamma H) = \exp(\beta H)\{[\exp(\beta h) + X(u)]/[1 + X(u)\exp(\beta h)]\}^{z_1^*-1}, \tag{21}$$

where

$$X(u) = 2/\{\exp[-\beta J(1-u)/2] + \exp[-\beta J(1+u)/2]\}; \tag{22}$$

$$X_{T=T_c}(0) = \exp(\beta_c J/2) \qquad \text{(Ising)}, \tag{23–1}$$

$$X_{T=T_c}(1) = 2/[1 + \exp(-\beta_c J)] \qquad \text{(Heisenberg)}. \tag{23–2}$$

For zero external field, $H=0$, and vanishingly small nonzero h, eq. (21) is reduced to

$$X_{T=T_c}(u) = z_1^*/(z_1^* - 2) \tag{24}$$

From eqs. (23–1, 2) and (24), we obtain, for the Curie temperature,

$$k_B T_c/J = 1/\ln[z_1^*/(z_1^* - 2)] \qquad (u=0;\ \text{Ising}); \tag{25–1}$$

$$2/\ln[z_1^*/(z_1^* - 4)] \qquad (u=1;\ \text{Heisenberg}). \tag{25–2}$$

It follows from eqs. (9) and (25–1, 2) that the critical concentration,

$$x_c=2/z_1 \quad (u=0;\ \text{Ising}); \qquad 4/z_1\ (u=1;\ \text{Heisenberg}). \qquad (26\text{–}1,\ 2)$$

§3. Summary

In the foregoing section, we have derived the coupled self-consistent relations (6), (8), (15–1, 2) and (25–1, 2) between x and T_c. And the present method has succeeded to give the critical concentration, $x_c=2/z_1$ (Ising); $4/z_1$ (Heisenberg), while Oguchi and Obokata's[2] gave the critical concentration $x'_c=1/(z_1-1)$ (Ising); $3/(z_1-1)$ (Heisenberg), with the value x'_c substituted by $x_c\{1-(1-x_c)^{z_1}\}$. The table I shows the calculated value of the critical concentration x_c for various values of z_1 in each case of the Ising and Heisenberg model, the present result compared with Oguchi and Obokata's.

Table I. The calculated value of the critical concentration x_c for various values of z_1.

	z_1		12	8	6	4
x_c	Ising	P	0.17	0.25	0.33	0.50
		O–O	0.12	0.18	0.25	0.39
	Heisenberg	P	0.33	0.50	0.67	1.00
		O–O	0.28	0.43	0.60	1.00

P: present theory, O–O: Oguchi and Obokata

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