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Theory of Spatially Random Heisenberg Ferromagnet

——Simplified Formulation——

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A simplified method is proposed to discuss a completely random dilute Heisenberg ferromagnet with isotropic nearest neighbor exchange. A formulation is given in the context of Takagi's cluster variation method in alliance with the Green's function method.

§1. Introduction

Phase transition of a dilute Heisenberg ferromagnet reveals certain oddities unlike the dense case. The most striking one lies in the relation between the transition temperature T_c and the magnetic ion concentration x. As dilution is advanced with magnetic ions replaced with nonmagnetic ions, the transition temperature T_c goes down monotonically as a function of the dilution. And ultimately there arises a certain nonzero critical concentration x_c of magnetic ions below which the transition temperature vanishes or the long range order does not occur.

This intrinsic peculiarity has been attracting the interest of theoretical physicist. Three sorts of fairly typical methods are identified so far to evaluate x_c, including the classical spin case; the further improvements on the conventional lines of Weiss' mean field approximation (or the cluster theories),1)–5) the direct expansions in the concentration (or temperature)6)–9) and the applications of Green's function.10)–13) Their estimates for x_c seem to lie rather close both in the classical spin case and in the Heisenberg spin case. It will do for the present to note that Tahir-Kheli's result13) obtained recently by his dynamical magnon scattering theory is 0.33(*sc*), 0.25(*bcc*) and 0.17 (*fcc*) for each regular cubic lattice.

However those theories never seem to be well established. In this work we present somewhat different approach. In §2, adopting Takagi's variation method14), 15) we work out a form of the reduced Hamiltonian which is linked to the Green's function method. In §3, the Green's function method is introduced to take the necessary formulation for the transition temperature and the spin pair correlation.

§ 2. Reduced Hamiltonian

We consider a system of N equivalent Heisenberg magnetic ions randomly distributed on L lattice sites ($N<L$). Hamiltonian is written as

$$\mathscr{H}=-\sum_{i<j}^{L}\sum^{L} t_i t_j J_{ij} \boldsymbol{s}_i \cdot \boldsymbol{s}_j - \gamma H \sum_{i}^{L} t_i s_i^Z, \tag{1}$$

where t_i is the occupation parameter which takes unity or zero according as the site i is occupied by a magnetic ion or not, satisfying the relation

$$\sum_{i=1}^{L} t_i = N = Lx. \tag{2}$$

The partition function is defined as

$$Z=\underset{\{t_i\}}{T_r}\ \underset{\{s_i\}}{T_r}\ e^{-\beta\mathscr{H}} \tag{3}$$

where β^{-1} stands for $k_B T$ and each trace is taken over all possible configurations for site sets $\{t_i\}$ or spin sets $\{s_i\}$. We somewhat drastically assume that each trace is to be taken quite independently on its own. In addition to this, the subsequent assumption of the nearest neighbor approximation

$$J_{ij}=\begin{cases} J & (i,\, j \text{ nearest neighbor}) \\ 0 & (\text{otherwise}). \end{cases} \tag{4}$$

enables us to take the following substitution and denotement,

$$t_i \longrightarrow \bar{t}_i = x, \tag{5,a}$$

$$t_i t_{i+\delta} \longrightarrow \overline{t_i t_{i+\delta}} = \alpha \bar{t}_i \bar{t}_{i+\delta} = \alpha x^2, \tag{5,b}$$

$$s_i^Z \longrightarrow \overline{s_i^Z} = \sigma, \tag{5,c}$$

$$\boldsymbol{s}_i \cdot \boldsymbol{s}_{i+\delta} \longrightarrow \overline{\boldsymbol{s}_i \cdot \boldsymbol{s}_{i+\delta}} = \chi_1, \tag{5,d}$$

where α is one variational parameter for the nearest neighbors. Then we have

$$Z=\sum_{\{t_i\}}\sum_{\{s_i\}} \exp\left\{\beta J \sum_{i,\delta} t_i t_{i+\delta} \boldsymbol{s}_i \cdot \boldsymbol{s}_{i+\delta} + \beta\gamma H \sum_i t_i s_i^Z \right\}$$

$$\cong \sum_{\{t_i\}}\sum_{\{s_i\}} \exp\left[L\beta\left(\frac{1}{2} J z_1 \alpha x^2 \chi_1 + \gamma H x \sigma\right)\right], \tag{6}$$

where z_1 is the number of the nearest neighbors. Consequently the summand of Eq. (6) is found to be independent of the sets $\{t_i\}$ and $\{s_i\}$.

Now let us denote the summations $\sum_{\{t_i\}} 1$ and $\sum_{\{s_i\}} 1$ by $g_t(\alpha)$ and g_s respectively and Helmholz free energy of this system will be given in the form

$$F=-\frac{1}{\beta}\ln Z=-\frac{1}{\beta}\left\{\ln g_t(\alpha)+\ln g_s\right\}-L\left\{\frac{1}{2}Jz_1\alpha x^2\chi_1+\gamma Hx\sigma\right\}. \tag{7}$$

Apparently $g_t(\alpha)$ means the total number of possible site configurations of N magnetic ions on L lattice sites. Therefore the version of Takagi's conbinatory method[14),15)] into the present case leads to

$$g_t(\alpha)\cong\frac{(z_1L/2)!}{N_{aa}!N_{ah}!N_{ha}!N_{hh}!}\left(\frac{N!(L-N)!}{L!}\right)^{z_1-1}, \tag{8}$$

where, the label, ah say, indicating magnetic-nonmagnetic ion pair,

$$N_{aa}=\frac{z_1L}{2}\overline{t_it_{i+\delta}}=\frac{z_1L}{2}\alpha x^2=\frac{z_1N}{2x}\alpha x^2, \tag{9,a}$$

$$N_{ah}=N_{ha}=\frac{z_1L}{2}\overline{t_i(1-t_{i+\delta})}=\frac{z_1L}{2}(x-\alpha x^2)=\frac{z_1N}{2x}(x-\alpha x^2), \tag{9,b}$$

$$N_{hh}=\frac{z_1L}{2}\overline{(1-t_i)(1-t_{i+\delta})}=\frac{z_1L}{2}(1-2x+\alpha x^2)=\frac{z_1N}{2x}(1-2x+\alpha x^2), \tag{9,c}$$

which satisfies

$$N_{aa}+N_{ah}=\frac{z_1N}{2},\ N_{ha}+N_{hh}=\frac{z_1(L-N)}{2}, \tag{10,a}$$

$$N_{aa}+N_{ah}+N_{ha}+N_{hh}=\frac{z_1L}{2}. \tag{10,b}$$

From Eqs. (7)–(10), it follows that

$$\begin{aligned}f\equiv\frac{\beta F}{N}=&-\frac{1}{N}\ln g_s-\frac{z_1-1}{x}\{x\ln x+(1-x)\ln(1-x)\}\\&+\frac{z_1}{2x}\{\alpha x^2\ln\alpha x^2+2(x-\alpha x^2)\ln(x-\alpha x^2)\\&+(1-2x+\alpha x^2)\ln(1-2x+\alpha x^2)\}-\frac{1}{2}\beta Jz_1\alpha x\chi_1-\beta\gamma H\sigma.\end{aligned} \tag{11}$$

We see that the parameter α is determined by the solution of the equation

$$\left(\frac{\partial f}{\partial\alpha}\right)_{T,N,L}=\frac{z_1x}{2}\ln\frac{\alpha x^2(1-2x+\alpha x^2)}{(x-\alpha x^2)^2}-\frac{1}{2}\beta Jz_1x\chi_1=0. \tag{12}$$

Re-forming Eq. (12) we find

$$\alpha=\frac{2(1-c_1)}{1-2xc_1+\sqrt{1-4x(1-x)c_1}}, \tag{13}$$

where

$$c_1\equiv 1-\exp(\beta J\chi_1). \tag{14}$$

Now that $\overline{t_i t_j}$ or α has been given as a function of χ_1 by Eq. (13), the Hamiltonian (1) may be reduced to

$$\mathscr{H}_s=\langle\mathscr{H}\rangle_t=-\sum_{i<i}^{N}\sum^{N}\overline{t_i t_j}J_{ij}\boldsymbol{s}_i\cdot\boldsymbol{s}_j-\gamma H\sum_{i}^{N}\bar{t}_i s_i^z$$

$$=-\sum_{i<j}^{N}\sum^{N}\mathscr{J}_{ij}\boldsymbol{s}_i\cdot\boldsymbol{s}_j-h\sum_{i}^{N}s_i^z, \tag{15}$$

where

$$\mathscr{J}_{ij}=\overline{t_i t_j}J_{ij}=\alpha x^2 J=\mathscr{J}\ \text{(nearest neighbor approx.)}, \tag{16}$$

$$h=\gamma H\bar{t}_i=\gamma H x. \tag{17}$$

§3. Green's Function of the Spin System

The application of Green's function method to the reduced Hamiltonian (15) in turn will allow us to represent the quantity χ_1 and T_c as functions of α. Following Tyabricov,[16] we introduce the double time Green's function

$$G_{lm}(t)=\langle\langle s_l^+(t);\, s_m^-\rangle\rangle=-i\theta(t)\langle[s_l^+,\, s_m^-]\rangle\,. \tag{18}$$

For the present, we content ourselves with the crudest approximation (RPA),

$$\langle\langle s_l^z(t)s_{l+\rho}^+(t);\, s_m^-\rangle\rangle\simeq\langle s_l^z\rangle\langle\langle s_{l+\rho}^+(t);\, s_m^-\rangle\rangle=\sigma G_{l+\rho,m}(t), \tag{19,a}$$

$$\langle\langle s_l^+(t)s_{l+\rho}^z(t);\, s_m^-\rangle\rangle\simeq\langle s_{l+\rho}^z\rangle\langle\langle s_l^+(t);\, s_m^-\rangle\rangle=\sigma G_{lm}(t)\,. \tag{19,b}$$

Then we have the equation of motion

$$i\frac{dG_{lm}(t)}{dt}=2\sigma\delta(t)\delta_{lm}-\mathscr{J}\sigma\sum_{\rho}[G_{lm}(t)-G_{l+\rho,\,m}(t)]+hG_{lm}(t)\,. \tag{20}$$

The Fourier transform of Eq. (19) yields

$$G_k(\omega)=\frac{\sigma}{\pi}\frac{1}{\omega-\varepsilon_k}, \tag{21}$$

$$\varepsilon_k=h+\mathscr{J}\sigma z_1(1-\gamma_k), \tag{22}$$

where

$$G_{lm}(\omega)=\frac{1}{N}\sum_{k}G_{k}(\omega)e^{i\boldsymbol{k}\cdot(\boldsymbol{l}-\boldsymbol{m})}\,, \tag{23}$$

$$\gamma_{k}=\frac{1}{z_1}\sum_{\delta}\mathrm{e}^{-\boldsymbol{k}\cdot\boldsymbol{\delta}}\,. \tag{24}$$

Meanwhile, from the spectrum theorem, a spin pair correlation $<s_m^- s_l^+(t)>$ is derived generally as follows

$$<s_m^- s_l^+(t)>=\frac{2\sigma}{N}\sum_{k}\frac{\mathrm{e}^{i\boldsymbol{k}\cdot(\boldsymbol{l}-\boldsymbol{m})-i\varepsilon_k t}}{\mathrm{e}^{\beta\varepsilon_k}-1}\,. \tag{25}$$

For the temperature region $T \gtrsim T_c$ and vanishingly small magnetic field ($h \simeq 0$), assuming reasonably

$$<s_i^z s_{i+\delta}^z>=\frac{1}{2}<s_i^- s_{i+\delta}^+>\text{(isotropic)}\,, \tag{26}$$

$$\sigma \cong 0\,, \tag{27}$$

we obtain

$$\begin{aligned}\chi_1&=\frac{3}{2}<s^-_i s^+_{i+\delta}(0)>=\frac{3\sigma}{N}\sum_{k}\frac{e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}}}{\mathrm{e}^{\beta\varepsilon_k}-1}\\ &\cong\frac{3\sigma}{N}\sum_{k}\frac{e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}}}{\beta\varepsilon_k}=\frac{3}{\beta N}\sum_{k}\frac{e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}}}{\mathscr{J}z_1(1-\gamma_k)+\dfrac{\mathscr{J}z_1}{\chi_{/\!/}}}\,,\end{aligned} \tag{28}$$

where

$$\chi_{/\!/}=\frac{\mathscr{J}z_1\sigma}{h}\,. \tag{29}$$

It is recast in the form

$$\chi_1=\frac{3k_BT}{\mathscr{J}z_1}W_1(\mu) \qquad (h\simeq 0,\ T\gtrsim T_c) \tag{30}$$

$$=\frac{3k_BT_c}{\mathscr{J}z_1}W_1(1) \qquad (h=0,\ \chi_{/\!/}=+\infty)\,, \tag{31}$$

where

$$W_1(\mu)=\frac{1}{N}\sum_{k}\frac{e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}}}{\mu-\gamma_k}\,, \tag{32}$$

$$\mu=1+\frac{1}{\chi_{/\!/}}\,. \tag{33}$$

Again from Eq. (25), we get

$$<s_l^- s_l^+(0)> = \frac{2\sigma}{N}\sum_k \frac{1}{e^{\beta\varepsilon_k}-1} \tag{34}$$

$$= s(s+1) - <s^z> - <(s^z)^2> \cong \frac{2}{3}s(s+1) - \sigma\,, \tag{35}$$

and hence

$$\frac{s(s+1)}{3\sigma} = \frac{1}{N}\sum_k \coth\frac{\beta\varepsilon_k}{2}\,. \tag{36}$$

In close analogy of Eq. (31), finally we come up to the formula for the temperature, that is,

$$\frac{\mathscr{J}}{k_B T} = \frac{3}{2z_1 s(s+1)} W(\mu) \qquad (T \gtrless T_c,\ h \simeq 0)\,, \tag{37}$$

where

$$W(\mu) = \frac{1}{N}\sum_k \frac{1}{\mu - \gamma_k}\,. \tag{38}$$

Particularly in the limit $\chi_{//}^{-1} \to 0$, we get

$$\frac{\mathscr{J}}{k_B T_c} = \frac{3}{2z_1 s(s+1)} W(1) \qquad (h=0)\,. \tag{39}$$

$W(1)$ is the so-called Watson integral which has the value

$$W(1) = 1.51638\,(sc);\ 1.39320\,(bcc);\ 1.34466\,(fcc)\,. \tag{40}$$

§ 4. Short Summary

Three simultaneous Eqs. (13), (31) and (39) derived above offer the relation between the concentration x and the critical temperature T_c. Thus we have established a self consistent formulation within the present scheme. Incidentally, on scrutiny in detail, we mention that room is yet left for further improvement. The assumption imposed to compute the partition function (3) seems a bit too drastic to be in very decent consideration of the connectivity between randomly diluted magnetic ions. It causes the reduced exchange integral $\mathscr{J}$ to be linearly proportioned to x near zero critical temperature, which is, we regret to say, responsible for MFA-like behaviour of our present formulation in the limit $T_c \to 0$.

Such being the case, research for more decent treatment is currently under way and the numerical calculation will be published in due course.

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