

## Thermodynamics of Random Spin Systems

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It is shown that a large class of Heisenberg systems with random exchange interactions can be solved in a systematic formulation for the bond-annealed case. As tractable examples, the critical concentrations of dilute ferromagnets and ferro-antiferro magnetic alloys are derived for the general spin magnitude  $S$ .

Recently, Thorp and Beeman<sup>1)</sup> proposed a general formulation for solving the effects of random exchange interactions on magnetic ordering in terms of Ising model with spin magnitude  $1/2$  on each lattice site. In this note, we show that a similar general formulation can be given in the case of Heisenberg systems with random exchange interactions and with spin magnitude  $S$  on each lattice site. With several different exchange integrals  $\{J_{ij}\}$  between the nearest neighbor pairs, Hamiltonian can be expressed in the form

$$H = \sum_{\langle ij \rangle} H_{ij}, \quad H_{12} = - \sum_l J_l t_l \mathbf{S}_1 \cdot \mathbf{S}_2 - \sum_l \mu_l t_l, \quad (1)$$

where  $\mu_l$  is the chemical potential per bond and the indicator parameter  $t_l$  equals unity if the exchange interaction of the  $\langle 12 \rangle$  pair is  $J_l$  and zero otherwise, satisfying the identity  $\sum_l t_l = 1$  so that only a single exchange interaction is associated with each bond. Let us now introduce the resultant states of two spins associated with the  $\langle 12 \rangle$  bond, as

$$|S_0 m_0\rangle = \sum_{m_1, m_2} \delta_{m_0, m_1 + m_2} C(m_1 m_2 | S_0 m_0) |S_1 m_1\rangle |S_2 m_2\rangle, \quad (2)$$

with the Clebsch-Gordan coefficient,  $C(m_1, m_2 | S_0 m_0)$  and the one-spin states  $|S_1 m_1\rangle$ ,  $|S_2 m_2\rangle$ , where the eigenvalues of the magnitude and z-component of each spin are, respectively, denoted by the labells  $S$  and  $m$ 's. Doing the partial traces over all possible  $\{t_l\}$  on one-bond density matrix, we get the expression of Fisher's manipulation<sup>2)</sup> innovated for Heisenberg bond model,

$$\sum_l \exp(\beta J_l \mathbf{S}_1 \cdot \mathbf{S}_2 + \beta \mu_l) = A \exp(G \mathbf{S}_1 \cdot \mathbf{S}_2), \quad (3)$$

$$A s_0^{\sum d(S_0)} = \prod_{s_0=0}^{2S} \left( \sum_l \exp[\beta J_l f(S_0) + \beta \mu_l] \right)^{d(S_0)}, \quad (4)$$

$$\exp \{ [f(S_0) - f(0)]G \} = \sum_I \exp [\beta J_I f(S_0) + \beta \mu_I] / \sum_I \exp [\beta J_I f(0) + \beta \mu_I], \quad (5)$$

with

$$\mathbf{S}_1 \cdot \mathbf{S}_2 |S_0 m_0\rangle = f(S_0) |S_0 m_0\rangle, \quad (6)$$

$$f(S_0) = S_0(S_0 + 1)/2 - S(S + 1),$$

where  $d(S_0) = 2S_0 + 1$  and  $\beta = 1/k_B T$ .

Then we may write the grand partition function as

$$\Xi \equiv \text{Tr}_{\{t_i\}} \text{Tr}_{\{S_i\}} \exp [-\beta H] = A^{z_1 N/2} \text{Tr}_{\{S_i\}} \exp (G \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j) = A^{z_1 N/2} Z(G), \quad (7)$$

where  $Z(G)$  is the partition function for a regular Heisenberg model with interaction parameter  $G$ , the coordination number  $z_1$  and the total bond number  $z_1 N/2$ . The bond occupation fractions  $\{t_i\}$  are obtained from

$$\beta \langle t_i \rangle = \frac{2}{z_1 N} \frac{\partial \ln \Xi}{\partial \mu_i} = \frac{\partial \ln A}{\partial \mu_i} + \varepsilon \cdot \frac{\partial G}{\partial \mu_i}, \quad (8)$$

where  $\varepsilon(G)$  denotes two-spin correlation function of the regular Heisenberg model,  $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ . Eliminating  $\exp [\beta \mu_i]$  from (4), (5) and (8), we obtain

$$\sum_I \langle t_i \rangle / Q(\beta J_I - G, \varepsilon) = 1, \quad (9)$$

where

$$Q(X, \varepsilon) \equiv \sum_{S_0} d(S_0) \exp [(f(S_0) - f(0))X] / \sum_{S_0} d(S_0) + \varepsilon \{ \exp [X] - 1 \}.$$

Replacing the temperature-independent discrete fraction  $\langle t_i \rangle$  by the probability distribution increment  $P(J)dJ$  for the continuous  $J$ , we obtain

$$\int dJ P(J) / Q(\beta J - G, \varepsilon) = 1. \quad (10)$$

From (10),  $G(\beta)$  is soluble for any  $P(J)$  and for any known  $\varepsilon(G)$  of the regular reference systems.

From (4), (5) and (10), we may calculate the thermodynamic quantities such as entropy, internal energy and specific heat, e.g., the internal energy per bond  $E$  can be found as follows:

$$\begin{aligned} E &= - \sum_I J_I \langle t_i \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle = - \frac{2}{z_1 N \beta} \sum_I J_I \frac{\partial \ln \Xi}{\partial J_I} \\ &= - \sum_I \langle t_i \rangle J_I \bar{f}(\beta J_I - G, \varepsilon), \end{aligned}$$

which, in the continuous case, reduces to

$$E = - \int dJ P(J) J \bar{f}(\beta J - G, \varepsilon), \quad (11)$$

where

$$\begin{aligned} \bar{f}(X, \varepsilon) Q(X, \varepsilon) &\equiv \sum_{S_0} d(S_0) f(S_0) \exp [(f(S_0) - f(0))X] / \sum_{S_0} d(S_0) \\ &+ \varepsilon \cdot \{f(1) \exp [X] - f(0)\}. \end{aligned}$$

As examples tractable comparatively, we have such as

$$P(J) = \begin{cases} p \delta(J - J_0) + (1 - p) \delta(J), & \text{(dilute ferro.)} \\ p \delta(J - J_0) + (1 - p) \delta(J + J_0), & \text{(ferro-antiferro.)} \\ [\theta(J - J_0 + \Delta) - \theta(J - J_0 - \Delta)] / 2\Delta, & \text{(rectangular)} \\ (3/2\pi\Delta^2)^{1/2} \exp [-3(J - J_0)^2 / 2\Delta^2], & \text{(Gaussian)} \end{cases} \quad (12)$$

where  $p$  is the ferromagnetic bond concentration and,  $\delta(J)$  and  $\theta(J)$  are the Dirac delta function and unit step function, respectively. For the dilute ferromagnets, from (10) and (12), the concentration  $p$  is given by

$$p = [1 - Q(-G, \varepsilon)] / [1 - Q(-G, \varepsilon) / Q(\beta J_0 - G, \varepsilon)]. \quad (13)$$

Putting  $\beta = \beta_c \rightarrow \infty$ ,  $G = G_c$  and  $\varepsilon = \varepsilon_c$ , we obtain the critical value  $p_c$  below which the ferromagnetic phase does not exist,

$$p_c = 1 - Q(-G_c, \varepsilon_c), \quad (14)$$

where  $G_c$  and  $\varepsilon_c$  are the values at the critical point of the regular system. The internal energy  $E$  and its critical value  $E_c$  are given from (11) and (12), respectively, by

$$E = -J_0 p \bar{f}(\beta J_0 - G, \varepsilon), \quad (15)$$

$$E_c = -S^2 J_0 p_c.$$

In the case of  $S = 1/2$ , (14) gives very simple expression for the critical concentration;

$$p_c = (\varepsilon_c + 3/4) [1 - \exp(-G_c)]. \quad (16)$$

So far no approximation has been made to deduce the critical concentration of the bonds  $p_c$  of (16). The Green function method or an improved molecular field method is exploitable for the treatment of the reference regular Heisenberg model to practice actual computation of  $G_c$  and  $\varepsilon_c$ . For instance, the constant-coupling approximant<sup>3,4)</sup> gives

$$f(G_c, 0) = \varepsilon_c \quad \text{and} \quad \varepsilon_c = S(S+1)/(z_1-1).^{3)} \quad (17)$$

This is the unprecedented and perfect generalization of the well known result for  $S=1/2$ ,  $G_c = \ln[z_1/(z_1-4)]$  and  $\varepsilon_c = 3/[4(z_1-1)]$ .<sup>4)</sup> By substituting the solution of (17) into (14),  $p_c$  can be evaluated for any  $S$ . In the case of  $S=1/2$ , we find a quite decent value of  $p_c = 3/(z_1-1)$  which concludes that there is a critical concentration only for  $z_1 > 4$ . For cubic lattices we have  $p_c = 0.600$  (SC), 0.429 (BCC) and 0.273 (FCC). Furthermore, for  $S=1$  we have  $p_c = 0.565$  (SC), 0.406 (BCC) and 0.261 (FCC), the ferromagnetic phase appearing into the circumstances of lower concentration.

For the ferro-antiferro-magnetic alloys,  $p$  and  $p_c$  are given, from (10) and (12), as follows:

$$p = [1 - Q(-\beta J_0 - G, \varepsilon)] / [1 - Q(-\beta J_0 - G, \varepsilon) / Q(\beta J_0 - G, \varepsilon)], \quad (18)$$

$$p_c = 4S(S+1)/(2S+1)^2 + \varepsilon_c. \quad (19)$$

In the case of  $S=1/2$ , (19) with (17) gives a critical concentration  $p_c = 3z_1/[4(z_1-1)]$  for  $z_1 > 4$ . The results (14) and (19) can be generalized into the omnibus expression for the fractions of ferro-magnetic bonds  $F$ , missing bonds  $D$  and antiferromagnetic bonds  $A$ . At the critical point these fractions satisfy the following equations:

$$D/Q(-G_c, \varepsilon_c) + A/[(2S+1)^{-2} - \varepsilon_c] = 1, \quad (20)$$

$$F + D + A = 1,$$

with

$$F = \int_{0+}^{\infty} P(J)dJ, \quad A = \int_{-\infty}^{0-} P(J)dJ, \quad D = \int_{-\varepsilon}^{+\varepsilon} P(J)dJ. \quad (21)$$

Actual computations for  $S > 1$  are currently underway. Finally we note that the present formulation can be generalized for the anisotropic case, including XY model and Ising model, and could also be extended to deal with a quenched case to investigate spin glasses.

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