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Thermodynamics of impure chains of classical spins with isotropic interactions

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Exact thermodynamic quantities are derived for a chain of classical spins of arbitrary dimension interacting via an arbitrary isotropic nearest-neighbor exchange in both the annealed and quenched limit, and for both bond and site randomness.

I. INTRODUCTION

Various thermodynamic quantities for an impure chain of classical spins interacting via nearest-neighbor isotropic exchange interactions have been computed exactly for both the annealed and quenched limit, and for both the bond and site models.[1,2] On the other hand, exact results are also available for a pure chain of classical spins of arbitrary dimensionality interacting through an arbitrary, isotropic nearest-neighbor interaction.[3,4] By combining the ideas and results of these earlier works *exact* thermodynamic quantities are derived here for a chain of spins of *arbitrary* dimension interacting via an *arbitrary* isotropic nearest-neighbor interaction in both the *annealed* and *quenched* limit, and for both *bond* and *site* randomness.

II. PURE CHAIN

First we recall some known results for the pure system consisting of a closed chain of N classical spins $\mathbf{S}_i$ of arbitrary dimensionality ν which interact via an isotropic nearest-neighbor interaction of the form[3,4]

$$H=-\sum_{i=1}^{N} f(\mathbf{S}_{i-1}\cdot\mathbf{S}_i)\ , \tag{2.1}$$

with the constraints that $|\mathbf{S}_i|=1$ and $\mathbf{S}_0=\mathbf{S}_N$. The function $f(x)$ is a completely arbitrary function which is bounded and continuous in $|x|\leq 1$. The static properties of such a system can be calculated with use of standard transfer-integral (TI) methods from the eigenvalues and eigenfunction of the integral equation

$$\int \frac{d\Omega_{i-1}}{\omega}\exp[\beta f(\mathbf{S}_{i-1}\cdot\mathbf{S}_i)]\psi_{nl}(\mathbf{S}_{i-1})=\lambda_{nl}\psi_{nl}(\mathbf{S}_i)\ , \tag{2.2}$$

where $d\Omega_i$ denotes an element of solid angle in the ν-dimensional spin space, and $\omega=2\pi^{\nu/2}/\Gamma(\frac{1}{2}\nu)$ is the area of the unit hypersphere, and $\beta=1/k_BT$. According to the Funk-Hecke theorem,[5] the eigenfunctions are the hypersurface harmonics of order $(\nu-2)/2$, with the corresponding eigenvalues

$$\lambda_{nl}=\lambda_n=\frac{(4\pi)^{(\nu-2)/2}(n!)\Gamma(\frac{1}{2}(\nu-2))}{\omega\Gamma(n+\nu-2)}\times\int_{-1}^{1}dx\, e^{\beta f(x)}[C_n^{(\nu-2)/2}(x)](1-x^2)^{(\nu-3)/2}\ , \tag{2.3}$$

where $C_n^{(\nu-2)/2}$ is the Gegenbauer polynomial. The kernel in Eq. (2.2) can be written in the form

$$l^{\beta f(\mathbf{S}_{i-1}\cdot\mathbf{S}_i)}=\sum_{n=0}^{\infty}\sum_{l=1}^{h(n,\nu-2)}\lambda_n\psi_{nl}^{*}(\mathbf{S}_{i-1})\psi_{nl}(\mathbf{S}_i)\ , \tag{2.4}$$

where

$$h(n,\nu-2)=(2n+\nu-2)[(n+\nu-3)!]/(n!)[(\nu-2)!]$$

is the degeneracy for λ_n. It then follows that, for $N\rightarrow\infty$, the partition function is given by

$$Z=\lambda_0^N\ , \tag{2.5}$$

and the thermal average of any function $A(\mathbf{S}_i,\mathbf{S}_{i+k})$ of two spins separated by a distance k can be expressed in the form

$$\langle A(\mathbf{S}_i,\mathbf{S}_{i+k})\rangle=\sum_{n=0}^{\infty}\sum_{l=1}^{h(n,\nu-2)}\left[\frac{\lambda_n}{\lambda_0}\right]^k I(0,1;n,l)\ , \tag{2.6}$$

where

$$I(n,l;n',l')=\int\frac{d\Omega_i}{\omega}\int\frac{d\Omega_j}{\omega}\psi_{nl}^{*}(\mathbf{S}_i)\Psi_{n'l'}(\mathbf{S}_i)A(\mathbf{S}_i,\mathbf{S}_j)\psi_{nl}(\mathbf{S}_j)\psi_{n'l'}(\mathbf{S}_j)\ . \tag{2.7}$$

In particular, the $2m$-uple spin correlation function

$$Y_m(k)=\langle C_m^{(\nu-2)/2}(\mathbf{S}_i\cdot\mathbf{S}_{i+k})\rangle/C_m^{(\nu-2)/2}(1) \tag{2.8}$$

is given by

$$Y_m(k)=\left[\frac{\lambda_m}{\lambda_0}\right]^k\ . \tag{2.9}$$

It is important to note that as long as the interaction between nearest-neighbor spins is isotropic the eigenfunctions of the TI equation are always given by the hyperspherical harmonics regardless of the particular functional form of the interaction. The eigenvalues, however, do depend on the form of the function f. Thus, by changing the eigenvalues appropriately the thermodynamical properties of various impure magnetic chains can be calculated. This observation has already been pointed out by Thorpe[6] for the $\nu=3$ case of a Heisenberg chain with site randomness in the quenched limit.

III. BOND MODEL

The Hamiltonian for a given configuration of two kinds of bonds, i.e., the host (H) and impurity (I) bonds, is assumed to have the form

$$\tilde{H} = -\sum_{i=1}^{N} [(1-p_{i-1,i}) f_H(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + p_{i-1,i} f_I(\mathbf{S}_{i-1}\cdot\mathbf{S}_i)] \ , \tag{3.1}$$

where $p_{i-1,i}$ is an occupation variable which has the value of 1 or 0 depending on whether the bond between the $(i-1)$th and ith sites is the I or H bond. We confine ourselves throughout this paper to the thermodynamic limit and neglect effects due to the boundary conditions, i.e., closed or open chain.

A. Annealed limit

The grand-canonical partition function Ξ of the system is given by

$$\Xi = \int \frac{d\Omega_0}{\omega} \left[\prod_{i=1}^{N} \left(\frac{d\Omega_i}{\omega} \sum_{p_{i-1,i}=0}^{1} \right) \right] \exp\left(\beta \sum_{i=1}^{N} [(1-p_{i-1,i}) f_H(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + p_{i-1,i} f_I(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + \ln(\lambda) p_{i-1,i}] \right) \ , \tag{3.2}$$

where λ is the fugacity of the I bond. After performing the trace over the bond occupation variable one sees that the kernel for the TI equation of the spin problem is

$$\exp[\beta f_H(\mathbf{S}_{i-1}\cdot\mathbf{S}_i)] + \lambda \exp[\beta f_I(\mathbf{S}_{i-1}\cdot\mathbf{S}_i)] \ , \tag{3.3}$$

which has eigenfunctions $\psi_{nl}(\mathbf{S})$ as for the pure case, but the eigenvalues are $\lambda_{nl} = \lambda_n = \lambda_n^H + \lambda\lambda_n^I$, where $\lambda_n^{H,I}$ are given as in Eq. (2.3) but with $f(x)$ replaced by $f^{H,I}(x)$. Thus, we take over the previous results and write $\Xi = \lambda_0^N$ and $\lambda = \rho\lambda_0^H/(1-\rho)\lambda_o^I$, where ρ is the concentration of the I bond. The total free energy is

$$F^{(a)} = -Nk_B T[\rho \ln\lambda_0^I + (1-\rho)\ln\lambda_0^H - (1-\rho)\ln(1-\rho) - \rho\ln\rho] \tag{3.4}$$

from which one obtains the total internal energy

$$U^{(a)} = N[\rho u^I + (1-\rho) u^H] \ , \tag{3.5}$$

where $u^{H,I} = k_B T^2 \partial \ln\lambda_0^{H,I}/\partial T$, and the specific heat

$$C^{(a)} = Nk_B[\rho C^I + (1-\rho) C^H] \ , \tag{3.6}$$

where $C^{H,I} = \partial u^{H,I}/\partial T$. The two-spin correlation function is given by

$$\langle A(\mathbf{S}_i\cdot\mathbf{S}_{i+k}) \rangle = \sum_{n'=0}^{\infty} \sum_{l'=1}^{h(n',\nu-2)} \left(\frac{\lambda_{n'}}{\lambda_0} \right)^k I(0,1;n',l') \ , \tag{3.7}$$

where $I(n,l;n',l')$ is as defined in Eq. (2.7). In particular $Y_m(k)$ has the form

$$Y_m(k) = \left(\frac{\lambda_m}{\lambda_0} \right)^k = \left((1-\rho)\frac{\lambda_m^H}{\lambda_0^H} + \rho\frac{\lambda_m^I}{\lambda_0^I} \right)^k = (u_m)^k \tag{3.8}$$

from which one obtains the static susceptibility

$$\chi_1^{(a)} = \frac{(g\mu_B)^2}{4k_B T} \sum_{i,j} \langle S_i^z S_j^z \rangle = \frac{(g\mu_B)^2}{4\nu k_B T} \left(2\sum_{k=0}^{\infty} Y_1(k) - 1 \right) = \frac{(g\mu_B)^2}{4\nu k_B T} \left(\frac{1+u_1}{1-u_1} \right) \ . \tag{3.9}$$

The wave-vector-dependent susceptibility, which is of interest to discussing neutron scattering from such random system, is given by

$$\chi_1^{(a)}(q,T) = \frac{(g\mu_B)^2}{4\nu k_B T} \left(\frac{1-u_1^2}{1-2u_1\cos(qa)+u_1^2} \right) \ , \tag{3.10}$$

where a is the distance between adjacent spins.

B. Quenched limit

The forms of the results in the quenched limit are essentially the same as those previously studied. For a given concentration of N_I ($N_I = N\rho$) I bonds the partition function is $\tilde{Z} = (\lambda_0^I)^{N_I}(\lambda_0^H)^{N-N_I}$, the free energy is larger than for the annealed case by a term which is due to the entropy of mixing, i.e.,

$$F^{(q)} = F^{(a)} - Nk_B T[(1-\rho)\ln(1-\rho) + \rho\ln\rho] \ ,$$

and consequently one has $U^{(q)} = U^{(a)}$ and $C^{(q)} = C^{(a)}$. The spin correlation functions $\langle\langle A(S_i,S_j)\rangle\rangle_{\text{conf}}$ and thus the spin susceptibility are also the same as in the annealed case.

IV. SITE MODEL

For a given distribution of the H and I spins the Hamiltonian $\tilde{H}$ in the site model may be written as

$$\tilde{H} = -\sum_{i=1}^{N} [(1-p_{i-1})(1-p_i) f_{HH}(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + (p_{i-1}-p_i)^2 f_{IH}(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + p_{i-1}p_i f_{II}(\mathbf{S}_{i-1}\cdot\mathbf{S}_i)] \ , \tag{4.1}$$

where the three functions $f_{HH}(x)$, $f_{HI}(x)$, and $f_{II}(x)$ represent, respectively, the interaction between the nearest-neighboring pairs of H and H, I and H, and I and I spins, and p_i is equal to 1 or 0 depending on whether the spin at the ith site is the I or H spin.

A. Annealed limit

The grand-canonical partition function has the form

$$\Xi = \left(\prod_{i=0}^{N} \int \frac{d\Omega_i}{\omega} \sum_{p_i=0}^{1} \right) \exp\left(\beta \sum_{i=1}^{N} [(1-p_{i-1})(1-p_i) f_{HH}(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + (p_{i-1}-p_i)^2 f_{IH}(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + p_{i-1}p_i f_{II}(\mathbf{S}_{i-1}\cdot\mathbf{S}_i) + (\ln\lambda) p_i] \right) \ , \tag{4.2}$$

where λ is the fugacity for the I magnetic ions. The trace over the I occupation variable p_i can be performed by introducing 2×2 transfer matrices. The resultant kernel can be expanded in terms of hypersurface harmonics in the form

$$\sum_{n=0}^{\infty}\sum_{l=1}^{h(n,\nu-2)}\psi_{nl}^{*}(\mathbf{S}_{i-1})\begin{pmatrix}\lambda_n^{HH} & \sqrt{\lambda}\lambda_n^{IH}\\ \sqrt{\lambda}\lambda_n & \lambda\lambda_n^{II}\end{pmatrix}\psi_{nl}(\mathbf{S}_i)\ , \tag{4.3}$$

where λ_n^{HH}, λ_n^{IH}, and λ_n^{II} are defined as in Eq. (2.3) but with the function f replaced, respectively, by f_{HH}, f_{IH}, and f_{II}. Thermodynamic quantities can then be read off from the corresponding results in TSP. Defining

$$a\pm=\tfrac{1}{2}\{\lambda\lambda_0^{II}+\lambda_0^{HH}\pm[(\lambda\lambda_{II}-\lambda_{HH})^2+4\lambda\lambda_{IH}^2]^{1/2}\}$$

we have $\Xi=a_+^N$ and

$$\lambda=\frac{\lambda_0^{HH}}{\lambda^{II}}+\frac{(1-2\rho)^2}{2\rho(1-\rho)}\left(\frac{\lambda_0^{IH}}{\lambda_0^{II}}\right)^2-\frac{(1-2\rho)}{2\rho(1-\rho)}\frac{\lambda_0^{IH}}{(\lambda_0^{II})^2}$$

$$\times[4\rho(1-\rho)\lambda_0^{II}\lambda_0^{HH}+(1-2\rho)^2(\lambda_0^{IH})^2]^{1/2}\ . \tag{4.4}$$

The internal energy $U^{(a)}$ is then given by

$$U^{(a)}=N\frac{\lambda_0^{II}u_{II}\gamma_{II}+\lambda_0^{IH}u_{IH}\gamma_{IH}+\lambda_0^{HH}u_{HH}\gamma_{HH}}{\lambda\lambda_0^{II}+\lambda_0^{HH}+[(\lambda\lambda_0^{II}-\lambda_0^{HH})^2+4\lambda(\lambda_0^{IH})^2]^{1/2}}\ , \tag{4.5}$$

where $u_a=k_BT^2\partial\ln\lambda_0^\alpha/\partial T$, for $\alpha=II$, IH, or HH, and

$$\gamma_{II}=\frac{\lambda}{2}\left(1+\frac{\lambda\lambda_0^{II}-\lambda_0^{HH}}{[(\lambda\lambda_0^{II}-\lambda_0^{HH})^2+4\lambda(\lambda_0^{IH})^2]^{1/2}}\right)\ ,$$

$$\gamma_{IH}=\frac{2\lambda\lambda_0^{IH}}{[(\lambda\lambda_0^{II}-\lambda_0^{HH})^2+4\lambda(\lambda_0^{IH})^2]^{1/2}}\ , \tag{4.6}$$

$$\gamma_{HH}=\frac{1}{2}\left(1-\frac{\lambda\lambda_0^{II}-\lambda_0^{HH}}{[(\lambda\lambda_0^{II}-\lambda_0^{HH})^2+4\lambda(\lambda_0^{IH})^2]^{1/2}}\right)\ .$$

The specific heat can then be obtained from the fact that $C^{(a)}=(\partial U^{(a)}/\partial T)_\rho$. The two-spin correlation function can be written in the form

$$\langle A(\mathbf{S}_i,\mathbf{S}_{i+k})\rangle=\frac{1}{\Xi}\sum_{n'=0}^{\infty}\sum_{l'=1}^{h(n',\nu-2)}I(0,1;n',l')$$
$$\times\mathrm{Tr}(\underline{A}_0\underline{A}^i\underline{B}_{n'}^k\underline{A}^{N-i-k})\ , \tag{4.7}$$

where $\underline{A}\equiv\underline{B}_0$ and

$$\underline{B}_n\equiv\begin{pmatrix}\lambda_n^{HH} & \sqrt{\lambda}\lambda_n^{IH}\\ \sqrt{\lambda}\lambda_n^{IH} & \lambda\lambda_n^{II}\end{pmatrix}\ . \tag{4.8}$$

In particular for the m-tuple correlation function one has

$$\langle Y_m(k)\rangle=\frac{1}{\Xi}\mathrm{Tr}(\underline{A}_0\underline{A}^i\underline{B}_m^k\underline{A}^{N-i-k})\ , \tag{4.9}$$

which, in the limit of $1\ll i$, $i+k\ll N$, becomes

$$\langle Y_m(k)\rangle=(\underline{P}_m^{-1}\underline{Q}_m)_{11}(\underline{Q}_m^{-1}\underline{P}_m)_{11}(b_m^+/a_+)^k+(\underline{P}_m^{-1}\underline{Q}_m)_{12}(\underline{Q}_m^{-1}\underline{P}_m)_{21}(b_m^-/a_+)^k\ , \tag{4.10}$$

where

$$b_m^\pm=\tfrac{1}{2}\{\lambda\lambda_m^{II}+\lambda_m^{HH}\pm[(\lambda\lambda_m^{II}-\lambda_m^{HH})^2+4\lambda(\lambda_m^{IH})^2]^{1/2}\}\ ,$$

$$\underline{P}=\begin{pmatrix}\sqrt{\lambda}\lambda_0^{IH} & \sqrt{\lambda}\lambda_0^{IH}\\ a_+-\lambda_0^{HH} & a_--\lambda_0^{HH}\end{pmatrix},\quad \underline{P}^{-1}=\frac{1}{\sqrt{\lambda}\lambda_0^{IH}(a_+-a_-)}\begin{pmatrix}-a_-+\lambda_0^{HH} & \sqrt{\lambda}\lambda_0^{IH}\\ a_+-\lambda_0^{HH} & -\sqrt{\lambda}\lambda_0^{IH}\end{pmatrix}\ ,$$

$$\underline{Q}=\begin{pmatrix}\sqrt{\lambda}\lambda_1^{IH} & \sqrt{\lambda}\lambda_1^{IH}\\ b_1^+-\lambda_1^{HH} & b_1^--\lambda_1^{HH}\end{pmatrix},\quad \underline{Q}^{-1}=\frac{1}{\sqrt{\lambda}\lambda_1^{IH}(b_1^+-b_1^-)}\begin{pmatrix}-b_1^-+\lambda_1^{HH} & \sqrt{\lambda}\lambda_1^{IH}\\ b^+-\lambda^{HH} & -\sqrt{\lambda}\lambda_1^{IH}\end{pmatrix}\ . \tag{4.11}$$

The static susceptibility has exactly the same form as that given by Eq. (14) of T.[2] Moreover, the impurity density-density correlation function $\psi_{i,i+k}^{(a)}\equiv\langle p_ip_{i+k}\rangle_g$ can be expressed, in the limit $1\ll i$, $i+k\ll N$, as

$$\psi_{i,i+h}^{(a)}=\begin{cases}(\underline{P}^{-1}\underline{A}'\underline{P})_{12}(\underline{P}^{-1}\underline{A}'\underline{P})_{21}a_\mp^{-2}(a_-/a_+)^{|k|-1}+\rho^2 & \text{for } k\neq 0\ ,\\ \rho \text{ for } k=0\ ,\end{cases} \tag{4.12}$$

where

$$\underline{A}'=\begin{pmatrix}0 & \sqrt{\lambda}\lambda_0^{IH}\\ 0 & \lambda\lambda_0^{II}\end{pmatrix}\ . \tag{4.13}$$

B. Quenched limit

Results in the quenched limit follow readily from those derived in TSP.[1] For a given configuration of the I and H sites the partition function can be written in the form

$$\tilde{F}=\prod_{i=1}^{N}(\lambda_0)_{i-1,i}\ , \tag{4.14}$$

where $(\lambda_0)_{i-1,1}$ is equal to λ_0^{II}, λ_0^{IH}, or λ_0^{HH} depending on the type of spins at the $(i-1)$th and ith sites. The Helmholtz free energy $F^{(q)}$ involves taking the arithmetic average over all the possible configurations of the I and H ions for given concentrations of these ions. The calculation in TSP again applies and the result is given by

$$F^{(q)}=-Nk_BT[\rho^2\ln\lambda_0^{II}+2\rho(1-\rho)\ln\lambda_0^{IH}+(1-\rho)^2\ln\lambda_0^{HH}]\ , \tag{4.15}$$

and from which one obtains the internal energy

$$U^{(q)}=N[\rho^2u_{II}+2\rho(1-\rho)u_{IH}+(1-\rho)^2u_{HH}] \tag{4.16}$$

and the specific heat

$$C^{(q)}=N[\rho^2C_{II}+2\rho(1-\rho)C_{IH}+(1-\rho)^2C_{HH}]\ , \tag{4.17}$$

where $C^{\alpha} \equiv \partial u^{\alpha}/\partial T$, for $\alpha = II$, IH, or HH. The calculations of the two-spin correlation function and the zero-field susceptibility follow almost exactly as in TSP and T, and are given by

$$\begin{aligned} Y_m^{(q)}(k) = {} & [\rho(\underline{R}_m)_{11} + (1-\rho)(\underline{R}_m)_{21}][(\underline{R}_m^{-1})_{11} + (\underline{R}_m^{-1})_{12}](d_m^{+})^k + [\rho(\underline{R}_m)_{12} + (1-\rho)(\underline{R}_m)_{22}] \\ & \times [(\underline{R}_m^{-1})_{21} + (\underline{R}_m^{-1})_{22}](d_m^{-})^k \end{aligned} \tag{4.18}$$

and

$$\chi(q) = (N\mu_B^2/12k_B\Delta_1^{1,1})[g_I^2\rho\Delta_1^{0,1} + 4g_Ig_H\rho(1-\rho)\eta_1^{IH} + g_H^2(1-\rho)\Delta_1^{1,0}] \ ,$$

where

$$d_m^{\pm} \equiv \tfrac{1}{2}(\rho\eta_m^{II} + (1-\rho)\eta_m^{HH} \pm \{[\rho\eta_m^{II} - (1-\rho)\eta_m^{HH}]^2 + 4\rho(1-\rho)(\eta_m^{IH})^2\}^{1/2}) \ , \tag{4.19}$$

$$\begin{aligned} \underline{R}_m &= \begin{pmatrix} (1-\rho)\eta_m^{IH} & (1-\rho)\eta_m^{IH} \\ d_m^{+} - \rho\eta_m^{II} & d_m^{-} - \rho\eta_m^{II} \end{pmatrix} \ , \\ (\underline{R}_m)^{-1} &= \frac{1}{(1-\rho)\eta_m^{IH}(d_m^{+} - d_m^{-})} \begin{pmatrix} -d_m^{-} + \rho\eta_m^{II} & (1-\rho)\eta_m^{IH} \\ d_m^{+} - \rho\eta_m^{II} & -(1-\rho)\eta_m^{IH} \end{pmatrix} \ , \end{aligned} \tag{4.20}$$

and

$$\Delta_i^{i,j} \equiv 1 + (-1)^i\rho\eta_1^{II} + (-1)^j(1-\rho)\eta_1^{HH} + (-1)^{i+j}\rho(1-\rho)[\eta_1^{II}\eta_1^{HH} - (\eta_1^{IH})^2] \ ,$$

with

$$\eta_m^{\alpha} \equiv \lambda_m^{\alpha}/\lambda_0^{\alpha}, \quad \alpha = II,\ IH, \text{ or } HH, \quad m = 1, 2, \ldots \ .$$

V. CONCLUDING REMARKS

In the presence of a competing biquadratic exchange interaction the pure Heisenberg spin chain exhibits disordered points of the second kind.[7,8] As pointed out by Rajagopal[9] this competition can be enhanced by the presence of randomness in the system. The possibility of including all forms of nearest-neighbor isotropic interactions here can be used to help sharpen the effect further.

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