

Partition functions of classical Heisenberg spin chains with arbitrary and different exchange

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Abstract

The classical Heisenberg model has been effective in modelling exchange interactions in molecular magnets. In this model, the partition function is important as it allows the calculation of the magnetization and susceptibility. For an ensemble of N -spin sites, this typically involves integrals in $2N$ dimensions. Here, for two-, three- and four-spin nearest neighbour open linear Heisenberg chains these integrals are reduced to sums of known functions, using a result due to Gegenbauer. For the case of the three- and four-spin chains, the sums are equivalent in form to the results of Joyce. The general result for an N -spin chain is also obtained.

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1. Introduction

From a theoretical point of view, magnetic systems where the exchange interaction dominates are of interest. Physically, such systems are realized within molecular magnets and experimentally large advances are being made in the synthesis of these compounds [1–4]. From a technological viewpoint these compounds offer possibilities as novel materials, in such applications as quantum computing [5]. Theoretically, where the spin quantum number is large, the classical Heisenberg model has been used to model such systems. Central to the classical thermodynamic description of these systems is the partition function from which, through differentiation with respect to the applied field, the magnetization and susceptibility can be obtained. For an ensemble of N -spin sites, these partition functions typically involve integrals in $2N$ dimensions. Joyce in his 1967 paper [6] analytically treated the partition functions of classical Heisenberg systems where through the application of graph theory, he

obtained some closed form expressions for partition functions. Subsequently Blume *et al* performed numerical solutions [7] and more recently Ciftja has discussed the difficulties in solving the partition functions for complex geometries, whilst treating an irregular tetrahedron [8]. Here, we consider a linear chain of spin sites with the nearest neighbour exchange interaction. We include an external field (Zeeman) and isotropic Heisenberg exchange energy terms in the Hamiltonian and we show how through the use of a result due to Gegenbauer [9], the classical partition function can be reduced in a simple manner and expressed in terms of known analytic functions. For two spins it can be expressed exactly, as a sum modified spherical Bessel functions of the first kind. For the three-spin open chain with two different exchange constants, a triple series is derived which is of the same form as that of Joyce for a cluster. The result of Joyce for four spins, with different exchange is re-obtained. The general result for an N -spin chain is also obtained.

2. Classical Heisenberg model for chains

The Hamiltonian for a N -spin chain contains the Zeeman (external field) and isotropic classical Heisenberg exchange energy terms [10]

$$\mathcal{H}_{N\text{-spin}} = -\mu_0 m \vec{H} \cdot \sum_{k=1}^N \vec{e}_k - \sum_{k=1}^{N-1} J_{C_{k,k+1}} \vec{e}_k \cdot \vec{e}_{k+1}, \quad (1)$$

where μ_0 is the permeability of free space, \vec{H} is the external magnetic field vector, \vec{e}_k represents the unit vector of each classical spin, with the subscript k indicating the position of the classical spin along the chain. The classical values for the exchange parameter $J_{C_{k,k+1}}$, between neighbouring spins k and $k + 1$, and classical magnetic moment m are taken as [10–12]

$$J_{C_{k,k+1}} = J_{k,k+1} s(s + 1), \quad m = g \mu_B \sqrt{s(s + 1)}. \quad (2)$$

Here $J_{k,k+1}$ is the exchange constant between nearest neighbour spins, where $J_{k,k+1} > 0$ for the ferromagnetic and $J_{k,k+1} < 0$ for the anti-ferromagnetic case, g is the Landé spectroscopic splitting factor, μ_B is the Bohr magneton and s is the spin quantum number. The classical partition function can be obtained by integrating for each classical spin over the solid angle of the sphere [6, 10]

$$Z_{N\text{-spin}} = \frac{1}{(4\pi)^N} \int_{\Omega_1} \cdots \int_{\Omega_N} \exp(-\mathcal{H}_{N\text{-spin}}/k_B T) \prod_{i=1}^N d\Omega_i, \quad (3)$$

where k_B is Boltzmann's constant and T is the absolute temperature. Using the spherical polar co-ordinates ($r = 1$) $e_x = \sin \vartheta \cos \phi$, $e_y = \sin \vartheta \sin \phi$ and $e_z = \cos \vartheta$ and noting the trigonometric identity $\cos \phi_1 \cos \phi_2 + \sin \phi_1 \sin \phi_2 = \cos(\phi_1 - \phi_2)$ we can write the N -spin Hamiltonian as

$$\begin{aligned} \mathcal{H}_{N\text{-spin}} = & -\mu_0 m H \sum_{i=1}^N \cos \vartheta_i - \sum_{k=1}^{N-1} J_{C_{k,k+1}} (\sin \vartheta_k \sin \vartheta_{k+1} \cos(\phi_k - \phi_{k+1}) \\ & + \cos \vartheta_k \cos \vartheta_{k+1}), \end{aligned} \quad (4)$$

where the external field is taken along the Cartesian z direction, which defines the polar axis and H is the magnitude of \vec{H} . The classical partition function can be then obtained by integrating over both polar and azimuthal angles for each spin site [10], namely

$$Z_{N\text{-spin}} = \frac{1}{(4\pi)^N} \int_0^{2\pi} \int_0^\pi \cdots \int_0^{2\pi} \int_0^\pi \exp(-\mathcal{H}_{N\text{-spin}}/k_B T) \prod_{i=1}^N \sin \vartheta_i d\vartheta_i d\phi_i. \quad (5)$$

3. Reduction of the partition functions

3.1. The partition function for the two-spin case

Use of the two-spin Hamiltonian from equation (1) allows us to obtain the two-spin partition function from equation (5). Rearranging the order of integration allows us to write

$$Z_{2\text{-spin}} = \frac{1}{(4\pi)^2} \int_0^\pi \int_0^\pi \exp(K \cos \vartheta_1 \cos \vartheta_2 + \xi(\cos \vartheta_1 + \cos \vartheta_2)) \\ \times \int_0^{2\pi} \int_0^{2\pi} \exp(K \sin \vartheta_1 \sin \vartheta_2 \cos(\phi_1 - \phi_2)) d\phi_1 d\phi_2 \sin \vartheta_1 d\vartheta_1 \sin \vartheta_2 d\vartheta_2, \quad (6)$$

where $\xi = \mu_0 m H / k_B T$ is a dimensionless field parameter and $K = J_{c_{1,2}} / k_B T$ is a dimensionless exchange parameter. Using equation (A.1) the azimuthal integral can be expressed in terms of modified Bessel functions $I_0(x)$ and so the partition function can be reduced to the double integral

$$Z_{2\text{-spin}} = \frac{1}{4} \int_0^\pi \int_0^\pi \exp(K \cos \vartheta_1 \cos \vartheta_2 + \xi(\cos \vartheta_1 + \cos \vartheta_2)) \\ \times I_0(K \sin \vartheta_1 \sin \vartheta_2) \sin \vartheta_1 d\vartheta_1 \sin \vartheta_2 d\vartheta_2. \quad (7)$$

We can now replace all terms in the integrand using equations (A.2) and (A.4) allowing us to express the partition as the triple series

$$Z_{2\text{-spin}} = \frac{1}{4} \sum_{l=0}^{\infty} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} (2n+1)(2m+1)(2l+1) i_n(K) i_m(\xi) i_l(\xi) \\ \times \int_0^\pi P_n(\cos \vartheta_1) P_m(\cos \vartheta_1) \sin \vartheta_1 d\vartheta_1 \int_0^\pi P_n(\cos \vartheta_2) P_l(\cos \vartheta_2) \sin \vartheta_2 d\vartheta_2, \quad (8)$$

where the functions $i_n(x)$ are the modified spherical Bessel functions of the first kind [13–15] as given in appendix A. Noting the orthogonality properties of the Legendre polynomials given by (A.5) this reduces to a single series

$$Z_{2\text{-spin}}(\xi, K) = \sum_{n=0}^{\infty} (2n+1) i_n(\xi)^2 i_n(K). \quad (9)$$

Since $i_n(0) = 0$ except for $i_0(0) = 1$, for zero field ($\xi = 0$) this reduces to

$$Z_{2\text{-spin}}(K) = i_0(K) = \frac{\sinh(K)}{K}, \quad (10)$$

which agrees with Fisher, and Stanley [16–18] for a two-spin chain.

3.2. The partition function for the three-spin chain

The same approach leads to

$$Z_{3\text{-spin}} = \frac{1}{(4\pi)^3} \int_0^\pi \int_0^\pi \int_0^\pi \exp(K_{1,2} \cos \vartheta_1 \cos \vartheta_2 + K_{2,3} \cos \vartheta_2 \cos \vartheta_3) \\ \times \exp(\xi(\cos \vartheta_1 + \cos \vartheta_2 + \cos \vartheta_3)) \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \exp(K_{1,2} \sin \vartheta_1 \sin \vartheta_2 \cos(\phi_1 - \phi_2)) \\ \times \exp(K_{2,3} \sin \vartheta_2 \sin \vartheta_3 \cos(\phi_2 - \phi_3)) d\phi_1 d\phi_2 d\phi_3 \sin \vartheta_1 \sin \vartheta_2 \sin \vartheta_3 d\vartheta_1 d\vartheta_2 d\vartheta_3, \quad (11)$$

where $K_{k,k+1} = J_{c_{k,k+1}}/k_B T$ are the dimensionless exchange parameters. The integrals in the azimuthal angles can be expressed as before, in terms of $I_0(x)$, so that

$$Z_{3\text{-spin}} = \frac{1}{8} \int_0^\pi \int_0^\pi \int_0^\pi \exp(K_{1,2} \cos \vartheta_1 \cos \vartheta_2 + K_{2,3} \cos \vartheta_2 \cos \vartheta_3) \times \exp(\xi(\cos \vartheta_1 + \cos \vartheta_2 + \cos \vartheta_3)) I_0(K_{1,2} \sin \vartheta_1 \sin \vartheta_2) I_0(K_{2,3} \sin \vartheta_2 \sin \vartheta_3) \times \sin \vartheta_1 \sin \vartheta_2 \sin \vartheta_3 \, d\vartheta_1 \, d\vartheta_2 \, d\vartheta_3. \quad (12)$$

Expanding the terms in the integrand through equations (A.2), (A.4) and (A.5) leads to the triple series

$$Z_{3\text{-spin}} = \frac{1}{2} \sum_{l=0}^\infty \sum_{m=0}^\infty \sum_{n=0}^\infty (2n+1)(2m+1)(2l+1) i_n(K_{1,2}) i_m(K_{2,3}) \times i_n(\xi) i_m(\xi) i_l(\xi) \int_0^\pi P_n(\cos \vartheta_2) P_m(\cos \vartheta_2) P_l(\cos \vartheta_2) \sin \vartheta_2 \, d\vartheta_2, \quad (13)$$

so that from equation (A.6) we can write in terms of the Wigner $3j$ symbol (see appendices),

$$Z_{3\text{-spin}} = \sum_{l=0}^\infty \sum_{m=0}^\infty \sum_{n=0}^\infty (2n+1)(2m+1)(2l+1) \begin{pmatrix} n & m & l \\ 0 & 0 & 0 \end{pmatrix}^2 \times i_n(\xi) i_m(\xi) i_l(\xi) i_n(K_{1,2}) i_m(K_{2,3}). \quad (14)$$

We note this result is of the same form as equation (4.21) of Joyce [6] for a one-dimensional cluster of spins with a centre spin and two nearest-neighbour interactions and where the remaining spins contribute to an internal field assumed parallel to the external field—the Bethe–Peierls–Weiss approximation [19] of Brown and Luttinger [20]—where here the ‘internal’ field equals the external field.

3.3. The partition function for the four-spin chain

Replacing the azimuthal dependence as before through equation (A.1) the four-spin partition function can be written

$$Z_{4\text{-spin}} = \frac{1}{2^4} \int_0^\pi \int_0^\pi \int_0^\pi \int_0^\pi \prod_{k=1}^3 I_0(K_{k,k+1} \sin \vartheta_k \sin \vartheta_{k+1}) \exp(K_{k,k+1} \cos \vartheta_k \cos \vartheta_{k+1}) \times \prod_{i=1}^4 \exp(\xi \cos \vartheta_i) \sin \vartheta_i \, d\vartheta_i. \quad (15)$$

Expanding the integrands as before using equations (A.2) and (A.4) allows us to write

$$Z_{4\text{-spin}} = \frac{1}{2^4} \int_0^\pi \int_0^\pi \int_0^\pi \int_0^\pi \prod_{k=1}^3 \sum_{n_k=0}^\infty (2n_k+1) i_{n_k}(K_{k,k+1}) P_{n_k}(\cos \vartheta_k) P_{n_k}(\cos \vartheta_{k+1}) \times \prod_{i=1}^4 \sum_{m_i=0}^\infty (2m_i+1) i_{m_i}(\xi) P_{m_i}(\cos \vartheta_i) \sin \vartheta_i \, d\vartheta_i. \quad (16)$$

The integral now contains only the Legendre polynomials, and the integrals in the angles ϑ_1 and ϑ_4 can be evaluated from the orthogonality properties of the Legendre polynomials, allowing two pairs of series to reduce to one pair where $n_1 = m_1$ and $n_3 = m_3$ leaving two

integrals each resulting in a Wigner $3j$ symbol. On reassigning the indices, where the sum for each of the five indices l_1, \dots, l_5 is from zero to infinity, we can write

$$Z_{4\text{-spin}} = \sum_{l_1, \dots, l_5} i_{l_1}(K_{1,2}) i_{l_4}(K_{3,4}) i_{l_5}(K_{2,3}) \times \prod_{i=1}^4 i_{l_i}(\xi) \prod_{j=1}^5 (2l_j + 1) \begin{pmatrix} l_1 & l_2 & l_5 \\ 0 & 0 & 0 \end{pmatrix}^2 \begin{pmatrix} l_3 & l_4 & l_5 \\ 0 & 0 & 0 \end{pmatrix}^2, \quad (17)$$

which is equivalent to equation (4.11) of Joyce [6] for the partition function of a chain of four spins in a field.

3.4. The partition function for an open N -spin chain

We can generalize for N spins and as before the azimuthal integrals can be replaced by modified Bessel functions so that

$$Z_{N\text{-spin}} = \frac{1}{2^N} \int_0^\pi \cdots \int_0^\pi \prod_{k=1}^{N-1} I_0(K_{k,k+1} \sin \vartheta_k \sin \vartheta_{k+1}) \exp(K_{k,k+1} \cos \vartheta_k \cos \vartheta_{k+1}) \times \prod_{i=1}^N \exp(\xi \cos \vartheta_i) \sin \vartheta_i \, d\vartheta_i. \quad (18)$$

Expanding the integrand terms through equations (A.2) and (A.4) we can write

$$Z_{N\text{-spin}} = \frac{1}{2^N} \prod_{k=1}^{N-1} \sum_{n_k=0}^\infty (2n_k + 1) i_{n_k}(K_{k,k+1}) \prod_{i=1}^N \sum_{n_i=0}^\infty (2n_i + 1) i_{n_i}(\xi) \times \int_0^\pi \cdots \int_0^\pi P_{n_k}(\cos \vartheta_k) P_{n_k}(\cos \vartheta_{k+1}) P_{n_i}(\cos \vartheta_i) \sin \vartheta_i \, d\vartheta_i. \quad (19)$$

Here, the integrals in the end angles ϑ_1 and ϑ_N reduce through the orthogonality properties and the remaining integrals can be calculated using Wigner $3j$ coefficients. In this case, for zero-field we obtain

$$Z_{N\text{-spin}} = \prod_{k=1}^{N-1} i_0(K_{k,k+1}), \quad (20)$$

which for equal exchange clearly reduces to Fisher's oft-quoted result [16–18, 21]

$$Z_{N\text{-spin}} = i_0(K)^{N-1} = \left(\frac{\sinh(K)}{K} \right)^{N-1} \quad (21)$$

for an open chain of length N .

4. Conclusions

Through a useful result of Gegenbauer, some results of Joyce for the classical partition functions for chains are re-derived in a simpler way and some new results obtained. These results should be of interest to those treating Heisenberg chains, and have allowed the present authors to obtain new expressions for the linear [22] and nonlinear susceptibility [23, 24] of Heisenberg chains.

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Appendix A. Equations for reduction of the integrals

Firstly, on expressing the partition function in spherical polar co-ordinates, the resulting azimuthal (ϕ) integrals, which are exponentials of cosines, can be expressed as modified Bessel functions of zero order $I_0(x)$ [13, 25, 26]

$$I_0(x) = \frac{1}{\pi} \int_0^\pi \exp(x \cos \phi) d\phi. \quad (\text{A.1})$$

Secondly an identity, stemming from Gegenbauer, allows exponentials of cosines times modified Bessel functions of zero order to be expressed as an infinite series of modified spherical Bessel functions of the first kind and decoupled products of Legendre polynomials [9]

$$I_0(R \sin \vartheta \sin \phi) \exp(R \cos \vartheta \cos \phi) = \sum_{n=0}^{\infty} (2n+1) i_n(R) P_n(\cos \vartheta) P_n(\cos \phi), \quad (\text{A.2})$$

where the functions $i_n(x)$ are the modified spherical Bessel functions of the first kind [13–15]

$$i_n(x) = \sqrt{\frac{\pi}{2x}} I_{n+\frac{1}{2}}(x) \quad (\text{A.3})$$

and where $I_{n+\frac{1}{2}}(x)$ are the (fractional order) modified Bessel functions of the first kind. For either angle equal to zero this reduces to

$$\exp(R \cos \vartheta) = \sum_{n=0}^{\infty} (2n+1) i_n(R) P_n(\cos \vartheta), \quad (\text{A.4})$$

which was used by Joyce in his 1967 treatment of classical Heisenberg chains [6]. Thirdly, the orthogonality properties of the resulting Legendre polynomials, namely [27],

$$\int_0^\pi P_m(\cos \vartheta) P_n(\cos \vartheta) \sin \vartheta d\vartheta = \frac{2}{2n+1} \delta_{m,n}, \quad (\text{A.5})$$

where $\delta_{m,n}$ is the Kronecker delta function, allow the remaining (polar) integrals to be solved for two Legendre polynomials whereby two of the three summations are replaced by a unique term. For three (or more) spins the orthogonality properties reduce the summations leaving integral(s) of three Legendre polynomials. These integrals occur in angular momentum calculations in quantum mechanics and can be expressed in terms of Clebsch–Gordon coefficients or in terms of the Wigner $3j$ symbol [28–37]. We use the Wigner $3j$ notation, owing to its useful symmetry properties (see appendix B). In this notation

$$\frac{1}{2} \int_0^\pi P_n(\cos \vartheta) P_m(\cos \vartheta) P_l(\cos \vartheta) \sin \vartheta d\vartheta = \begin{pmatrix} n & m & l \\ 0 & 0 & 0 \end{pmatrix}^2. \quad (\text{A.6})$$

For small values these can be calculated algebraically without difficulty, and are also available in packages such as Mathematica. For larger values, schemes exist to optimize their calculation [36, 38]. The values required in the magnetization for terms to the third order in the applied field required for the linear [22] and nonlinear susceptibility [23, 24] are given in appendix C.

Appendix B. Useful relations for the Wigner $3j$ symbol

We note the symmetry relations for the Wigner $3j$ symbol considered here, where the three lower indices are zero, whereby the order of n , m and l is interchangeable. The following relation is also useful:

$$\begin{pmatrix} l+1 & l & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 = \begin{pmatrix} l & l+1 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{l+1}{(2l+1)(2l+3)}. \quad (\text{B.1})$$

Furthermore values for which one index is unity not conforming to this pattern are zero [6, 21].

Appendix C. Values of Wigner $3j$ symbol

We follow the standard approach and consider the sum of the indices $n + m + l$. We note that for $n + m + l = \text{odd}$, the Wigner $3j$ symbols of the form considered are zero. Non-zero values for $n + m + l$ up to 8 are given below:

$n + m + l$

$$\begin{array}{l} 0 \quad \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}^2 = 1 \\ 2 \quad \begin{pmatrix} 1 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{1}{3} \\ 4 \quad \begin{pmatrix} 2 & 2 & 0 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{1}{5}; \quad \begin{pmatrix} 2 & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{2}{15} \\ 6 \quad \begin{pmatrix} 3 & 3 & 0 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{1}{7}; \quad \begin{pmatrix} 3 & 2 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{3}{35}; \quad \begin{pmatrix} 2 & 2 & 2 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{2}{35} \\ 8 \quad \begin{pmatrix} 4 & 4 & 0 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{1}{9}; \quad \begin{pmatrix} 4 & 3 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{4}{63}; \quad \begin{pmatrix} 4 & 2 & 2 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{2}{35}; \\ \quad \begin{pmatrix} 3 & 3 & 2 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{4}{105}. \end{array}$$

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