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Thermodynamic properties of ferrimagnetic large spin systems

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Abstract

The application of cumulant expansion is generalized to a ferrimagnetic system of large spins. An effective Hamiltonian is obtained in terms of classical variables for a quantum spin chain to describe the thermodynamic properties at moderate and high temperature regimes. We have considered quantum corrections up to second order in the cumulant expansion where fairly good results are observed in the convergent regime, i.e. $T > Js$. (J is the exchange coupling between spins, s is the smaller spin (S_1, s_2) and T is temperature.) Our results show that the correlation of more than two neighbouring sites is negligible at moderate and high temperature. Thus the results of a single ferrimagnet molecule can be applied to the chain of interacting molecules for temperature greater than the classical energy scale, i.e. $T > JS_1s_2$.

1. Introduction

The low temperature properties of ferrimagnets can be described in different ways [2, 1]. For instance spin wave theory gives an explanation for low temperature physics of ferrimagnetic chains [2]. However, it is valid for temperature smaller than the classical energy scale ($T \lesssim JS_1s_2$). It is our task to obtain the physics of ferrimagnets systematically at moderate (where quantum corrections are important) and higher temperatures. Cumulant expansion (CE) [3–5] is our approach to this problem. Recently this method has been implemented to study finite temperature behaviour of homogeneous large spin systems [6, 7]. It has been observed that cumulant expansion converges in a region $T > Js$ which is wider than the corresponding one in the high temperature series expansion [9] and even overlaps the validity regime of spin wave theory for homogeneous spin systems.

We have generalized the application of cumulant expansion to ferrimagnetic models. In this approach we obtain a quasiclassical Hamiltonian for an (S_1, s_2) system. The effective Hamiltonian which is a function of classical variables takes into account the quantum corrections systematically in the order of (Js/T) where s is the smaller value of

(S_1, s_2) . For nearest neighbour interaction in the ferrimagnetic chain, the first order effective Hamiltonian is composed of two-site correlation. Consequently the results of CE for a single molecule of (S_1, s_2) are the same as for one-dimensional interacting molecules. We have also found that in the convergence region the gas model is a good representation of a chain of interacting molecules up to second order expansion.

2. Theory of cumulants and effective Hamiltonian

Thermodynamic properties of any quantum spin system with a Hamiltonian \hat{H} can be described by using the effective Hamiltonian \mathcal{H} which can be constructed using cumulant expansion and spin coherent state $|\mathbf{n}\rangle$ representation [4–8, 10]. The partition function (\mathcal{Z}) of a spin system can be expressed in the basis of coherent states which are overcomplete, so it contains all quantum states. The classical state of a spin is achieved in the limit $S \rightarrow \infty$. The partition function of a system of N spins defined by the Hamiltonian \hat{H} , is the following:

$$\mathcal{Z} = \int \prod_{i=1}^N \left(\frac{2S_i + 1}{4\pi} \right) d\mathbf{n}_i \langle \mathbf{n}_1 \dots \mathbf{n}_N | e^{-\beta \hat{H}} | \mathbf{n}_1 \dots \mathbf{n}_N \rangle \quad (1)$$

where $\beta = 1/T$ (choosing $k_B = 1$). If we define

$$e^{-\beta \mathcal{H}} \equiv \langle \mathbf{n}_1 \dots \mathbf{n}_N | e^{-\beta \hat{H}} | \mathbf{n}_1 \dots \mathbf{n}_N \rangle, \quad (2)$$

the partition function is the same as a classical one. The above equation can be expanded in the following form by a Taylor expansion.

$$\beta \mathcal{H} = \beta \langle \hat{H} \rangle^c - \frac{\beta^2}{2!} \langle \hat{H} \hat{H} \rangle^c + \frac{\beta^3}{3!} \langle \hat{H} \hat{H} \hat{H} \rangle^c + \dots = \beta (\mathcal{H}^{(0)} + \mathcal{H}^{(1)} + \mathcal{H}^{(2)} + \dots). \quad (3)$$

Thus the effective Hamiltonian (\mathcal{H}) can be considered as a systematic expansion in terms of cumulants of powers of \hat{H} . The function \mathcal{H} evidently depends on temperature, thus calculation of physical quantities should be done with care. The cumulant of N operators (A_i) [3, 4] is defined as

$$\langle A_1 \dots A_n \rangle^c = \frac{\partial}{\partial \lambda_1} \dots \frac{\partial}{\partial \lambda_n} \ln \langle e^{\lambda_1 A_1 + \lambda_2 A_2 + \dots} \rangle |_{\lambda_1 = \dots = \lambda_n = 0}. \quad (4)$$

Let us consider an interacting system of ferrimagnetic molecules where each molecule is composed of two spins (S_1, s_2) . The interaction between spins is either ferromagnetic (–) (F) or antiferromagnetic (+) (AF), given by the following Hamiltonian:

$$\hat{H} = \mp \sum_{i,j}^{\frac{N}{2}, \frac{N}{2}} J_{2i-1, 2j} \mathbf{S}_{2i-1} \cdot \mathbf{s}_{2j} \quad (5)$$

where \mathbf{S}_{2i-1} and \mathbf{s}_{2j} are spin operators of size S_1 and s_2 , respectively. To find an expansion for the physical quantities we should first calculate the semiclassical effective Hamiltonian defined in equation (3). We will consider the cumulant expansion up to second order corrections which comes from $\mathcal{H}^{(2)}$.

The first term of \mathcal{H} is $\mathcal{H}^{(0)} = \langle H \rangle^c = \mp \omega \sum_{i,j} \tilde{J}_{2i-1, 2j} \mathbf{n}_{2i-1} \cdot \mathbf{n}_{2j}$ where $\omega = \frac{S_1}{s_2} > 1$, $s \equiv \min\{S_1, s_2\} = s_2$ and $\tilde{J}_{2i-1, 2j} = J_{2i-1, 2j} s^2$. This term shows the classical contribution which simply represents the energy of coupled classical spins whose lengths are S_1 and s_2 . $\mathcal{H}^{(1)}$ and $\mathcal{H}^{(2)}$ are quantum corrections, where $\mathcal{H}^{(1)}$ is as follows,

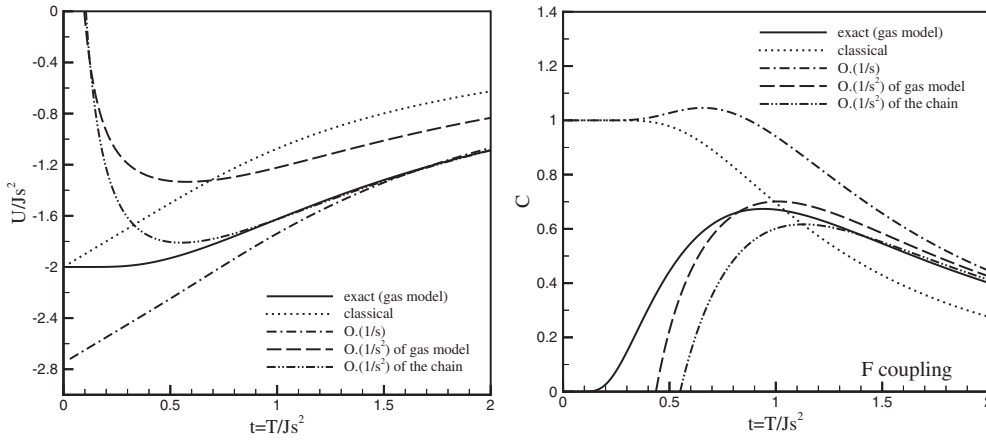


Figure 1. The internal energy per molecule and heat capacity of the ferrimagnetic ($S_1 = 4, s_2 = 2$) gas model with ferromagnetic (F) coupling. The exact result of the gas model (solid curve) and different orders of cumulant expansion for the gas model and a chain of interacting molecules.

$$\begin{aligned}
 \mathcal{H}^{(1)} = & \frac{-\beta\omega}{8s^2} \sum_{i,j} \tilde{J}_{2i-1,2j}^2 (1 - \mathbf{n}_{2i-1} \cdot \mathbf{n}_{2j})^2 - \frac{\beta\omega}{4s} \sum_{i,j,l} \tilde{J}_{2i-1,2j} \tilde{J}_{2i-1,2l} \\
 & \times (\mathbf{n}_{2j} \cdot \mathbf{n}_{2l} - (\mathbf{n}_{2i-1} \cdot \mathbf{n}_{2j})(\mathbf{n}_{2i-1} \cdot \mathbf{n}_{2l})) - \frac{\beta\omega^2}{4s} \sum_{i,j,l} \tilde{J}_{2i-1,2j} \tilde{J}_{2l-1,2j} \\
 & \times (\mathbf{n}_{2i-1} \cdot \mathbf{n}_{2l-1} - (\mathbf{n}_{2i-1} \cdot \mathbf{n}_{2j})(\mathbf{n}_{2l-1} \cdot \mathbf{n}_{2j})). \quad (6)
 \end{aligned}$$

As is obvious from equation (6), the second and third terms contain coupled interaction of three sites, i.e. $(\mathbf{n}_{2i-1} \cdot \mathbf{n}_{2j})(\mathbf{n}_{2i-1} \cdot \mathbf{n}_{2l})$ and $(\mathbf{n}_{2i-1} \cdot \mathbf{n}_{2j})(\mathbf{n}_{2l-1} \cdot \mathbf{n}_{2j})$. This is different from the two-site interaction of the original Hamiltonian, equation (5). It is the price of working with a classical Hamiltonian instead of the original quantum form. The last two terms in equation (6) give the information on three-site correlation. We will come back to this point later when comparing the results of the gas model with a chain of interacting ferrimagnetic molecules. It can be shown that the second order correction, $\mathcal{H}^{(2)}$, is in $(\frac{\beta\tilde{J}}{s})^2$ order (the detailed version of the paper can be found in [8]). Generally, in the cumulant expansion the n th order term, $\mathcal{H}^{(n)}$, can be considered of $\tilde{J}(\frac{\beta\tilde{J}}{s})^n$ order. It is clear that \mathcal{H} is an expansion in powers of $\beta\tilde{J}/s = Js/T$, so it is valid as long as $T > Js$. Quantum corrections have a non-Heisenberg form and their structure become more and more complex with increasing order. These corrections are important in the intermediate temperature where classical fluctuations are not strong enough to suppress quantum ones. At very high temperature the classical term is dominant.

3. Results and discussions

To obtain the physical quantities we consider the nearest neighbour case, $J_{ij} = J\delta_{i,j\pm 1}$, in a chain of ferrimagnets, because most synthesized materials behave as nearest neighbour interacting molecules; moreover, a comparison with other results is possible in this case.

Quasiclassical expansions for the internal energy per molecule (U) and heat capacity (C) of ($S_1 = 4, s_2 = 2$) ferrimagnetic chains ($-\cdots-\cdots-$ line) for F and AF coupling are shown in figures 1, 2, respectively.

The gas model is an ideal gas of ferrimagnetic molecules where each molecule is composed of two spins (S_1, s_2) and the interaction between spins in all molecules is given

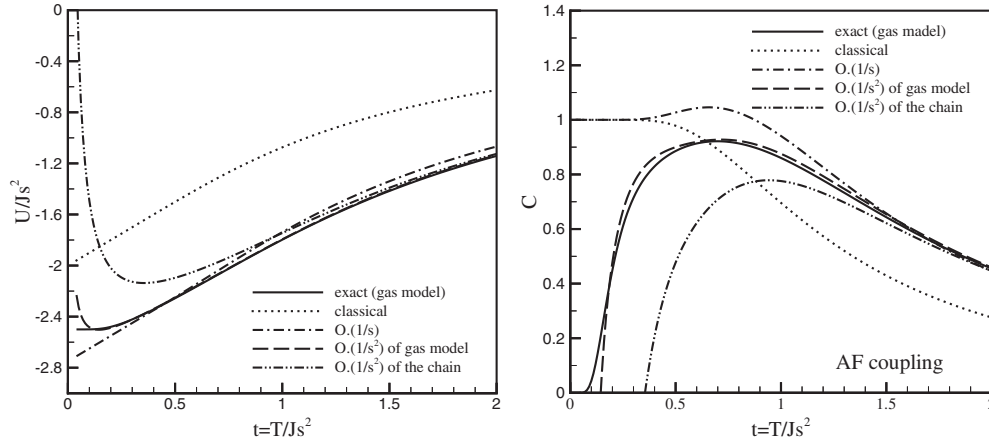


Figure 2. The internal energy per molecule and heat capacity of the ferrimagnetic ($S_1 = 4, s_2 = 2$) gas model with antiferromagnetic (AF) coupling. Exact result of the gas model (solid curve) and different orders of cumulant expansion for the gas model and a chain of interacting molecules.

by $\hat{H} = \mp JS_1 \cdot s_2$. The cumulant expansion of internal energy and heat capacity up to second order and the exact solution for this model have been also plotted in figures 1 and 2. It is surprising that the second order expansion of the AF gas model fits very well to the exact result even for very small values of t . The different behaviour of F and AF cases is related to the absolute value of ΔE for the lowest states. For F coupling, ΔE is larger than the AF one, i.e. $\Delta E^{(1)}(F) - \Delta E^{(1)}(AF) = J(2s_2 - 1)$. This means that cumulant expansion works better for the AF case. In this respect we should note that a cumulant expansion is similar to a high temperature series expansion which starts from $T \rightarrow \infty$, where the probabilities of all states are equal. Then different orders of expansion are responsible for recovering the non-equal probability of states. This is crucial when the low energy spectrum is not dense (like the F case) where a big difference exists between the occupation probability of lowest levels. Thus we expect to observe stronger quantum effects in the ferromagnetic case.

Meanwhile, we observe that the zero classical and first order expansion is the same for the gas model and the interacting chain, because the first order expansion ($\mathcal{H}^{(0)} + \mathcal{H}^{(1)}$) contains information on two-site correlation. This can be shown easily by the following equation:

$$\langle \mathcal{H}^{(1)} \rangle = \frac{-\beta\omega\tilde{J}^2}{8s^2} \sum_{i=1}^N \langle (1 - \mathbf{n}_i \cdot \mathbf{n}_{i+1})^2 \rangle - \frac{\beta\tilde{J}^3}{4s} \omega(\omega + 1) \sum_{i=1}^N \langle 1 - (\mathbf{n}_i \cdot \mathbf{n}_{i+1})^2 \rangle. \quad (7)$$

As far as the correlation of two sites is concerned the behaviour of a single molecule and a chain of molecules with nearest neighbour interaction is the same. The inset of different behaviour for chain and non-interacting molecules comes from the correlation of three sites. Such terms exist in $\mathcal{H}^{(2)}$ and contribute in the $1/s^2$ correction. The difference of the $1/s^2$ expansion is seen in figure 1. This deviation is clearer for the low temperature regime where quantum effects are important. But for temperature greater than the classical energy scale, $T > JS_1s_2$, there is a good agreement between the gas model and the ferrimagnetic chain. This shows that the correlation of more than two sites is important for low temperatures, $t < \omega$ ($t = T/JS^2$). In other words, at moderate and higher temperature an ideal gas of molecules represents a chain of ferrimagnets very well.

The heat capacity of both figures 1 and 2 decreases for high t which is the sign of antiferromagnetic behaviour in the upper part of spectrum of a ferrimagnetic chain. We also see

a Schottky-like peak in C which is the result of ferromagnetic to antiferromagnetic crossover. However, this peak is in the region where cumulant expansion is not necessarily convergent. Then our plots for $t \lesssim 1$ might not be reliable, although we know from other arguments [12, 13] that this peak exists.

The large S spin wave approximations [1, 11, 13] are imposed to an extra constraint to be applicable for moderate temperature, because it is valid for low temperature (by definition) where only the low energy spectrum has the dominant effect. Adding an extra constraint needs a self-consistent numerical solution to obtain thermodynamic properties. We have learnt about the recent high temperature expansion derived for the alternating spin chains [14]. As has been mentioned earlier the high temperature expansion is convergent for temperatures higher than our cumulant expansion. However, it is possible to implement a Padé approximation to connect the results of high temperature expansion to the low temperature regime. In this respect we need to know some information on the ground state energy of the system which is not generally available. In comparison, our results are analytic (in a closed form) and general in the sense that it can be applied to any ferrimagnetic system for arbitrary exchange couplings. The effective Hamiltonian derived here is in terms of arbitrary $J_{i,j}$ which covers long range interactions as well as any lattice structures.

As an outlook, this work can be generalized to ladder geometry. It is known that the zero-temperature behaviour of ferrimagnetic ladders [15] is different from homogeneous spin counterparts [16]. It is interesting to see the difference for finite temperature, for instance the evolution of magnetization plateaux [17, 18].

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