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Specific heat and magnetic susceptibility of ferromagnetic mixed-spin chain systems

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Abstract

Using a combination of high-temperature series expansion, exact diagonalization and Quantum-Monte-Carlo, we perform a complementary analysis of the thermodynamic properties of one-dimensional mixed-spin systems with alternating magnetic moments. In particular, we focus on the difference between ferromagnetic (FM) and antiferromagnetic (AFM) exchange. For the specific heat we find a multi-peak structure in the FM case which is strongly enhanced as compared to that known for the AFM case. We also report the behavior of the susceptibility in the corresponding temperature range.

Key words: mixed-spin chain, susceptibility, Quantum Monte Carlo, high-temperature series expansion, exact diagonalization

In this paper, we focus on a chain with two kinds of spin species, i.e., S and s, arranged alternatingly and coupled by a nearest-neighbor Heisenberg exchange. Namely, the Hamiltonian reads

$$\mathcal{H} = \sum_{i=1}^{N} \left[-J(\mathbf{S}_i \cdot \mathbf{s}_i + \mathbf{s}_i \cdot \mathbf{S}_{i+1}) - \mathbf{h} \cdot (\mathbf{S}_i + \mathbf{s}_i) \right], \quad (1)$$

where $\mathbf{h}$ is a magnetic field and we consider the limit of $h \to 0$. The subscript $i = 1 \ldots N$ refers to the unit cells, and we always use periodic boundary conditions. Typically, quasi-1D mixed-spin (MS) compounds display *antiferromagnetic* (AFM) intra-chain exchange [1], which has stimulated theoretical investigations of AFM MS models [2]. Interestingly, since a unit cell of the MS chain comprises of two different magnetic moments, the spectrum will allow for excitations of "acoustic" as well as "optical" nature [2]. While not identified unambiguously in present day experiments, the character of these excitations should appear in thermodynamic and other observable properties as two *independent energy scales*. On the other hand, what remains less well studied are MS chains with *ferromagnetic* (FM) intra-chain exchange, which arise in materials of recent interest such as $MnNi(NO_2)_4(en)_2$ with en = ethylenediamine [3].

In a recent paper [4], we have studied the mixed-spin chain systems using a combination of high-temperature series expansion (HTSE), exact diagonalization (ED), and Quantum-Monte-Carlo (QMC) techniques. We have contrasted the cases of FM and AFM intra-chain exchange. As a result, it is found that not only the AFM but also the FM case displays a double-peak-like structure in the specific heat. For comparison between FM and AFM exchange coupling, we plot in Fig. 1 the specific heat per unit cell for both cases with $S = 1$ and $s = 1/2$ calculated by HTSE and QMC. The AFM case agrees with former results by Yamamoto et al. [2], and shows a large peak at $T \sim 0.7J$ and a small shoulder at $T \sim 0.1J$. In contrast, the structure of the FM case seems to be a sum of two peaks of equal size at $T \sim 0.3J$ and $T \sim 0.7J$. In addition, there might

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be another small peak at $T \sim 0.1J$. Motivated by the difference of these two curves, it is the purpose of this paper to perform a detailed analysis of the susceptibility of MS chains using HTSE, ED, and QMC.

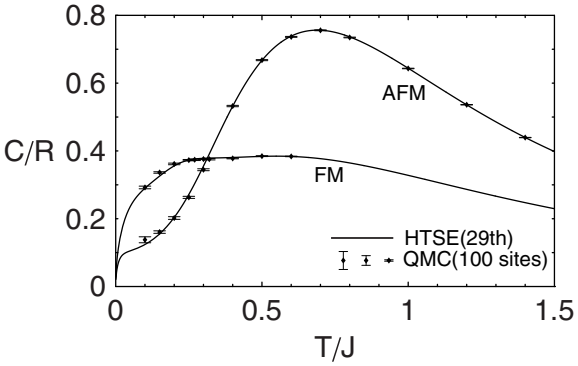

Fig. 1. The specific heat per unit cell for the MS chain with $s = 1/2$ and $S = 1$, calculated by HTSE and QMC.

Figure 2 shows temperature times susceptibility per unit cell, in the case of $S = 1$ and $s = 1/2$ both for FM and AFM coupling. For the HTSE, we have obtained the series coefficients up to 14th order in βJ. The series coefficients up to seventh order are consistent with those in the literature (see comments in Ref. [4]). The obtained series is extrapolated using Padé approximations. Our QMC simulations are based on stochastic series expansion [5], and performed for chains of 128 sites. We regard these results to represent the thermodynamic limit. We also include in Fig. 2 the ED results for finite chains. The full spectrum has been calculated for finite chains with seven unit-cells (14 sites). In the finite systems, the zero-temperature limit of $T\chi$ is reduced to $S_{\rm tot}(S_{\rm tot}+1)/(3N)$, where $S_{\rm tot}$ is the total spin in the ground state. With $N = 7$, we find $S_{\rm tot} = 21/2$ for the FM case, and $S_{\rm tot} = 7/2$ for the AFM case. We find overall good agreement between the QMC and the HTSE data, both for the AFM and the FM case. The ED result for 14 sites is already very close to the thermodynamic limit down to temperatures of $T \sim 0.3J$ for both the FM and the AFM case. All the data in Fig. 2 will be provided on request.

The susceptibility of the AFM case is consistent with former results [2]. It is also qualitatively similar to that reported for the "half-classical" limit $S \to \infty$ which is exactly solvable [6,7]. That is, as temperature decreases, antiferromagnetic short-range correlation develops and $T\chi$ decreases; at even lower temperature it increases because the ground state has macroscopic uniform magnetization. These two different correlations, i.e., high-temperature AFM and low-temperature FM correlation, should be related to the two structure of the specific heat [2]. However, in the FM case, the corresponding high- and low-

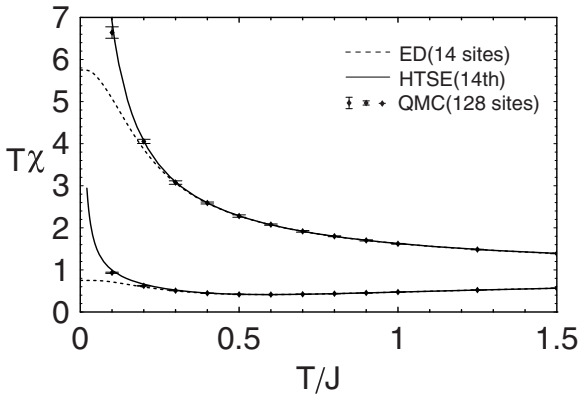

Fig. 2. Temperature times susceptibility per unit cell for the MS chain with $s = 1/2$ and $S = 1$, calculated by HTSE, QMC, and ED.

temperature correlations are both ferromagnetic. This might be a reason why a structure corresponding to that of the specific heat does not appear in the susceptibility of the FM case from the present analysis. The nature of structures of the specific heat might appear more clearly in other quantities such as the staggered susceptibility, which are left for future investigations.

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