Magnetic susceptibility of frustrated spin-$s$ $J_1$-$J_2$ quantum Heisenberg magnets: High-temperature expansion and exact diagonalization data

J Richter$^1$, A Lohmann$^1$, H-J Schmidt$^2$, D C Johnston$^3$

$^1$Institut für Theoretische Physik, Otto-von-Guericke-Universität Magdeburg, PF 4120, D - 39016 Magdeburg, Germany
$^2$Universität Osnabrück, Fachbereich Physik, Barbarastr. 7, D - 49069 Osnabrück, Germany
$^3$Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011

E-mail: johannes.richter@physik.uni-magdeburg.de

Abstract. Motivated by recent experiments on low-dimensional frustrated quantum magnets with competing nearest-neighbor exchange coupling $J_1$ and next nearest-neighbor exchange coupling $J_2$ we investigate the magnetic susceptibility of two-dimensional $J_1$-$J_2$ Heisenberg models with arbitrary spin quantum number $s$. We use exact diagonalization and high-temperature expansion up to order 10 to analyze the influence of the frustration strength $J_2/J_1$ and the spin quantum number $s$ on the position and the height of the maximum of the susceptibility. The derived theoretical data can be used to get information on the ratio $J_2/J_1$ by comparing with susceptibility measurements on corresponding magnetic compounds.

1. Introduction

The investigation of frustrated magnetic systems is currently a field of active theoretical and experimental research [1, 2]. Systems with competing nearest-neighbor (NN) exchange coupling $J_1$ and next nearest-neighbor (NNN) exchange coupling $J_2$ can serve as model systems to study the interplay of quantum effects, thermal fluctuations and frustration. The quantum $J_1$-$J_2$ Heisenberg models on the square-lattice exhibit several ground-state phases including non-classical non-magnetic ground states, see, e.g., [3]. The corresponding Hamiltonian reads

$$H = J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{[i,j]} S_i \cdot S_j,$$

where $(S_i)^2 = s(s + 1)$, and $\langle i,j \rangle$ denotes NN and $[i,j]$ denotes NNN bonds. For antiferromagnetic NNN bonds, $J_2 > 0$, the spin system is frustrated irrespective of the sign of $J_1$. Due to frustration the theoretical treatment of this model is challenging.

The numerous theoretical studies of the ground state phase diagram so far did not lead to a consensus on the nature of the quantum ground state and on the nature of the quantum phase transitions present in the model, see, e.g., [4] and references therein. Interestingly there are also various compounds well described by square-lattice $J_1$-$J_2$ Heisenberg models.
such as oxovanadates [5] and iron pnictides [6]. In experiments, typically temperature-dependent quantities are reported. Hence reliable (and flexible) tools are desirable to calculate thermodynamic quantities such as the uniform magnetic susceptibility $\chi$. In this paper we present two methods, namely the full exact diagonalization, see, e.g., [7], and the high-temperature expansion [8, 9, 10] to calculate the temperature dependence of the magnetic susceptibility for the square-lattice $J_1$-$J_2$ spin-$s$ Heisenberg model with both ferromagnetic (FM) and antiferromagnetic (AFM) NN coupling $J_1$ and AFM NNN bonds $J_2$ for arbitrary spin quantum number $s$. In particular, we analyze the position and the height of the maximum in the susceptibility in dependence on $J_1$, $J_2$ and $s$.

![Figure 1](image_url)

**Figure 1.** Uniform susceptibility $\chi$ as a function of renormalized temperature $T/s(s+1)$ for NNN exchange $J_2 = 1$ and three values of the spin quantum number $s = 1/2, 1, \text{and } 7/2$. (a) Numerical exact data for a finite square lattice of $N = 8$ sites and antiferromagnetic $J_1 = 1$. (b) Numerical exact data for a finite square lattice of $N = 8$ sites and ferromagnetic $J_1 = -1$. (c) [6,4] Padé approximant of the 10th order HTE series for an infinite square lattice and ferromagnetic $J_1 = -1$. (d) [6,4] Padé approximant of the 10th order HTE series for an infinite square lattice and antiferromagnetic $J_1 = 1$. The order of labeling in each legend is the same as the order of the plots (top to bottom).
2. Methods

The full exact diagonalization (ED) yields numerical exact results at arbitrary temperature \( T \), but it is typically limited to about \( N = 22 \) sites for \( s = 1/2 \) models. For larger spin quantum numbers \( s \) the system size \( N \) accessible for ED shrinks significantly. Hence, ED is used preferably for \( s = 1/2 \) and \( s = 1 \). In the present study we exploit the special symmetry properties of the finite square-lattice of \( N = 8 \) sites and perform full ED for the \( J_1-J_2 \) model for \( s = 1/2, 1, \ldots, 9/2 \), thus allowing to study the role of the spin quantum number. Since the ED approach suffers from the finite-size effect, the ED calculations do not yield quantitatively correct results for the thermodynamic limit. Nevertheless, they will give insight into the qualitative behavior of the susceptibility. The high-temperature expansion (HTE) for the \( J_1-J_2 \) model up to 10th order was presented in [9], however, restricted to \( s = 1/2, 1, \ldots, 9/2 \). The scheme is encoded in a simple C++-program and can be downloaded [11] and freely used by interested researchers.

Very recently the present authors have extended this general HTE scheme up to 10th order [12]. Here we use this 10th order HTE as an alternative method to the ED. We use here three different subsequent Padé approximants, namely Padé \([4,6],[5,5],[6,4]\), see e.g. [8, 10]. Such a Padé approximant extends the region of validity of the HTE series down to lower temperatures. Since the HTE approach is designed for infinite systems the HTE data for the susceptibility maximum, in principle, can be quantitatively correct, if the maximum is not located at too low temperatures. Indeed, it was found [10] that for the unfrustrated \((J_2 = 0)\) square-lattice spin-1/2 Heisenberg antiferromagnet the Padé \([4,4]\) approximant of the 8th order HTE series yields correct data for the susceptibility maximum located at \( T \approx 0.94|J_1| \). However, it may happen that a certain Padé approximant does not work for some particular values of \( J_1, J_2, \) and \( s \), since Padé approximants may exhibit unphysical poles for temperatures in the region of interest. Hence we show in the next section only those Padé data not influenced by poles.

3. Results

First we present the temperature dependence of the susceptibility \( \chi \) in Fig. 1 for a particular value of \( J_2 \) and both FM and AFM \( J_1 \). In this paper the symbol \( \chi \) means \( \chi |J_1|/Ng^2\mu_B^2 \), where \( N \) is the number of spins and \( \mu_B \) is the Bohr magneton. The temperature is measured in terms of \( |J_1| \), i.e. the symbol \( T \) means \( T/|J_1| \). The qualitative behavior of \( \chi(T) \) shown in Figs. 1(a-d) is similar, there is the broad maximum in \( \chi(T) \) that is typical for a two-dimensional antiferromagnet (note that for \( J_2/|J_1| = 1 \) the system is in the AFM ground state irrespective of the sign of \( J_1 \)). The various \( \chi(T) \) curves give an impression on the finite-size effects, the effect of the sign of the NN exchange \( J_1 \), and the influence of spin quantum number \( s \). The height, \( \chi_{\text{max}} \), and the position, \( T_{\text{max}} \), of the maximum in the \( \chi(T) \) curve are interesting features for the comparison with experimental data, in particular to get information on the ratio \( J_2/|J_1| \) from susceptibility measurements, see e.g. [14]. Therefore we will discuss \( \chi_{\text{max}} \) and the \( T_{\text{max}} \) now in more detail.

We present our data for the susceptibility maximum for both FM and AFM NN exchange \( J_1 \) in Figs. 2 (ED data) and 3 (HTE and ED data). For FM \( J_1 \), \( \chi_{\text{max}} \) \( (T_{\text{max}}) \) becomes larger (smaller) upon lowering \( J_2 \). Finally, when approaching the critical value \( J_2^c \), where the transition to the ferromagnetic ground state takes place, \( \chi_{\text{max}} \) diverges and \( T_{\text{max}} \) goes to zero. The critical point for \( s = 1/2 \) is \( J_2^c = 0.333|J_1| \) for \( N = 8 \) (but it is \( J_2^c \approx 0.4|J_1| \) for \( N \to \infty \) [13]). It increases with growing \( s \) and becomes \( J_2^c = 0.5|J_1| \) for \( s \to \infty \). The data for \( N = 8 \) and \( N \to \infty \) are in qualitative agreement. Although the finite-size effects are obviously large, the general features of \( \chi_{\text{max}} \) and \( T_{\text{max}} \) as functions of \( J_2 \) and \( s \) are quite similar. Naturally the HTE fails when approaching \( J_2^c \), since in this limit low temperatures become relevant. Note that the HTE data for FM \( J_1 \) and \( s = 1/2 \) are also in qualitative agreement with recently reported data calculated
Figure 2. Position $T_{\text{max}}$ (a and c) and height $\chi_{\text{max}}$ (b and d) of $\chi(T)$ for the finite $N = 8$ square-lattice $J_1$-$J_2$ model (left panels FM $J_1 = -1$, right panels AFM $J_1 = +1$).

by second-order Green’s function approach [14]. We discuss now the case of AFM $J_1$ (right panels in Figs. 2 and 3). For large $J_2$ the behavior of $\chi_{\text{max}}$ and $T_{\text{max}}$ is very similar to that for
Figure 3. Position $T_{max}$ (a and c) and height $\chi_{max}$ (b and d) of $\chi(T)$ for an infinite square-lattice $J_1$--$J_2$ model obtained by 10th order HTE (left panels FM $J_1 = -1$, right panels AFM $J_1 = +1$). For comparison we show the ED data for $N = 8$.

FM $J_1$, i.e. the sign of $J_1$ becomes irrelevant, cf. Ref. [13]. On the other hand, for smaller values of $J_2$ naturally both cases behave completely different, since $J_1$ dominates the physics. We find...
a well pronounced minimum in $T_{\text{max}}$ in the region of strongest frustration around $J_2 = 0.5$. For the finite system $\chi_{\text{max}}$ exhibits a maximum in this region, whereas for the infinite system $\chi_{\text{max}}$ is almost constant in the region $0 \leq J_2 \leq 0.5$.

To take a closer look on the role of the spin quantum number $s$ we present in Fig. 4 the quantities $\chi_{\text{max}}$ and $T_{\text{max}}$ as a function of $1/s$ for particular values of $J_2$. Obviously, there is monotonous increase (decrease) of $\chi_{\text{max}}$ ($T_{\text{max}}/s(s+1)$) with growing $s$. For FM $J_1 = -1$ the increase of $\chi_{\text{max}}$ is particular strong for $J_2 = 0.7$ (see the insets in panels a and c), since for large $s$ this value of $J_2$ becomes quite close to the transition point to the FM ground state. From Figs. 4(a-d) it is also seen that the position $T_{\text{max}}$ of the maximum for $J_2 \gtrsim 0.7 |J_1|$ is almost independent of the sign of $J_1$, whereas the height $\chi_{\text{max}}$ strongly depends on the sign of the NN coupling. Let us finally mention the special $s$-dependence of the maximum in $\chi(T)$ for $J_1 = 1$ and $J_2 = 0.5$, where the classical ground state exhibits a large non-trivial degeneracy. The position $T_{\text{max}}/s(s+1)$ of the maximum shifts to zero in the limit $s \to \infty$, whereas the height remains finite. This behavior is quite similar to that found for the pyrochlore AFM [10, 15, 16], where the classical ground state is also highly degenerate.
4. Summary
Using high-temperature expansion and full exact diagonalization we have calculated the uniform susceptibility $\chi$ of the spin-$s$ $J_1-J_2$ square-lattice Heisenberg magnet in a wide parameter regime of FM and AFM $J_1$ and frustrating AFM $J_2$. Especially, we have studied the height and the position of the maximum in the $\chi(T)$ curve as functions of $J_2/J_1$ and the spin quantum number $s$. These data can be used to get information on the ratio $J_2/|J_1|$ from susceptibility measurements, e.g. on oxovanadates which are well described by the square-lattice $J_1$-$J_2$ model.

Acknowledgments
The work at Ames Laboratory was supported by the U.S. Department of Energy under Contract No. DE-AC02-07CH11358.

References
[1] Quantum Magnetism 2004 ed Schollwöck U, Richter J, Farnell D J J, and Bishop R F, Lecture Notes in Physics 645 (Berlin: Springer)
[2] Frustrated Spin Systems 2005 ed Diep H T (Singapore: World Scientific)
[3] Schmalfuß D, Darradi R, Richter J, Schulenburg J, and Ihle D 2006 Phys. Rev. Lett. 97 157201; Jiang H-C, Yao H, and Balents L 2012 Phys. Rev. B 86 024424
[4] Sirker J, Weihong Z, Sushkov O P, and Oitmaa J 2006 Phys. Rev. B 73 184420; Darradi R, Derzhko O, Zinke R, Schulenburg J, Krüger S E, and Richter J 2008 Phys. Rev. B 78 214415; Richter J and Schulenburg J 2010 Eur. Phys. J. B 73 117; Wang L, Poilblanc D, Gu Z-C, Wen X-G, and Verstraete F 2013 Phys. Rev. Lett. 111 037202; Hu W J, Becca F, Parola A, and Sorella S 2013 Phys. Rev. B 88 060402; Ren Y Z, Tong N H, Xie X C 2014 J. Phys. Cond. Matter 26 115601
[5] Nath R, Tsirlin A A, Rosner H, and Geibel C 2008 Phys. Rev. B 78 064422; Bossoni L, Carretta P, Nath R, Moscardini M, Baenitz M, and Geibel C 2011 Phys. Rev B 83 014412; Roy B, Furukawa Y, Nath R, and Johnston D C 2011 J. Phys. Conf. Series 320 012048
[6] Si Q and Abrahams E 2008 Phys. Rev. Lett. 101 076401; Johnston D C 2010 Adv. Phys. 59 803
[7] Hättel M, Richter J, Ihle D, and Drechsler S-L 2008 Phys. Rev. B 78 174412
[8] Oitmaa J, Hamer C J, and Zheng W H 2006 Series Expansion Methods (Cambridge: Cambridge University Press)
[9] Rosner H, Singh R R P, Zheng W H, Oitmaa J, and Pickett W E 2003 Phys. Rev. B 67 014416
[10] Schmidt H J, A. Lohmann A, and Richter J 2011 Phys. Rev. B 84 104443
[11] see http://www.uni-magdeburg.de/jspalten/HTE/
[12] Lohmann H, Schmidt H J, and Richter J 2014 Phys. Rev. B 89 014415
[13] Shannon N, Momoi T, and Sindzingre P 2006 Phys. Rev. Lett. 96 027213; Richter J, Darradi R, Schulenburg J, Farnell D J J, and Rosner H 2010 Phys. Rev. B 81 174429
[14] Hättel M, Richter J, Götz O, Ihle D, and Drechsler S-L 2013 Phys. Rev. B 87 054412
[15] Moessner R and Berlinsky A J 1999 Phys. Rev. Lett. 83 3293
[16] Garcia-Adeva A J and Huber D L 2001 Phys. Rev. B 63 140404