10th order high-temperature expansion for spin systems: Susceptibility and specific heat for spin-s Heisenberg models with arbitrary exchange patterns

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We present the high-temperature expansion (HTE) up to 10th order of the specific heat C and the uniform susceptibility χ for Heisenberg models with arbitrary exchange patterns and arbitrary spin quantum number s. We encode the algorithm in a C++ program which allows to get explicitly the HTE series for concrete Heisenberg models. We apply our algorithm to pyrochlore ferromagnets and kagome antiferromagnets using several Padé approximants for the HTE series. For the pyrochlore ferromagnet we use the HTE data for χ to estimate the Curie temperature T_c as a function of the spin quantum number s. We find that T_c is smaller than that for the simple cubic lattice, although both lattices have the same coordination number. For the kagome antiferromagnet the influence of the spin quantum number s on the susceptibility as a function of renormalized temperature $T/s(s+1)$ is rather weak for temperatures down to $T/s(s+1) \sim 0.3$. On the other hand, the specific heat as a function of $T/s(s+1)$ noticeably depends on s. The characteristic maximum in $C(T)$ is monotonously shifted to lower values of $T/s(s+1)$ when increasing s.

I. INTRODUCTION

Magnetic systems described by the Heisenberg Hamiltonian

$$H = \sum_{\mu<\nu} J_{\mu\nu} \mathbf{s}_\mu \cdot \mathbf{s}_\nu \tag{1}$$

are an active field of theoretical and experimental research.[1] The accurate description of these quantum many-body systems is a basic aim of theoretical investigations. The comparison with experimental studies typically requires the calculation of the temperature dependence of physical properties, such as the susceptibility χ and the specific heat C. For unfrustrated spin systems the quantum Monte Carlo technique is a suitable tool to provide precise data, but it is not applicable due to the sign problem for frustrated quantum spin models.[2]

A universal straightforward approach to calculate thermodynamic quantities is the high-temperature expansion (HTE). For Heisenberg models this method was introduced in an early work by W. Opechowski[3] based on a method of approximate evaluation of the partition function developed by H.A. Kramers. In the 1950ies and 1960ies the method was further developed and widely applied to various Heisenberg systems, see e.g. Refs. 4–8.

The HTE method is now well-established and its application to magnetic systems is a basic tool in theoretical physics, see Refs. 9, 10 and references therein. For the Heisenberg model with nearest-neighbor interaction on standard lattices now typically the HTE is known up to high-orders, see for example Refs. 11 and 12, where the HTE up to 14th order for the triangular lattice and up to 16th order for the kagome were published. On the other hand, often magnetic compounds and corresponding spin models are of interest, where two, three or even more exchange constants are relevant. Typical examples are magnetic systems with nearest-neighbor, next-nearest-neighbor, and 3rd-nearest-neighbor couplings, see, e.g., Ref. 13. Moreover, in most of the quasi-low-dimensional magnetic compounds interchain or interlayer couplings play a role. Note further that available high-order HTE are often restricted to spin quantum number $s = 1/2$, see again Refs. 11 and 12 as an example. As a rule, for such more complex exchange geometries and/or higher spin quantum numbers $s > 1/2$ relevant for the interpretation of experimental data the HTE is not available in higher orders. An earlier attempt to bridge this gap was published in Ref. 14, where general analytical HTE expressions were given for arbitrary s and arbitrary Heisenberg exchange couplings up to order three. Very recently the present authors have published a significant extension of this work using computer algebraic tools.[15] In that paper the HTE algorithm for general spin-s Heisenberg models up 8th order was presented. This algorithm was encoded as a C++ program. The download (URL `http://www.uni-magdeburg.de/jschulen/HTE/`) and use are free. Thus this algorithm provides a flexible tool for the community to compute the HTE for the susceptibility and the specific heat, which can be used to analyze the thermodynamics of spin models, to check approximations, and, last but not least, to compare experimental data with model predictions.

In our previous work[15] we considered several models. Our results demonstrated that the 8th order HTE with a subsequent Padé approximation is (i) able to describe correctly the maximum of the susceptibility of a square-

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lattice s=1/2 Heisenberg antiferromagnet, (ii) can yield better results for spin systems in dimension $d > 1$ than full exact diagonalization, and (iii) gives good agreement with Monte Carlo data for the classical pyrochlore antiferromagnet down to temperatures of about 40% of the exchange coupling.

In this paper we extend our approach up to 10th order for general Heisenberg Hamiltonians. Again we will provide this new extendended tool as a freely accessible C++ program, see Appendix A. After briefly explaining our method (Sect II) we apply it in Sect III to the pyrochlore ferromagnet and the kagome antiferromagnet. In the Appendices B and C we present explicitly the HTE series for the pyrochlore and the kagome spin-s Heisenberg magnets.

II. BRIEF EXPLANATION OF THE METHOD

We consider the HTE expansion of extensive quantities f, e. g., susceptibility χ or specific heat C, of the form

$$f^{\Sigma}(\beta) = \sum_{n=0}^{\infty} c_n^{\Sigma,f}\, \beta^n \ . \tag{2}$$

Here β is the inverse dimensionless temperature $\beta = \frac{J}{k_B T}$, where J is a typical exchange energy, and the index Σ indicates the dependence on the spin system Σ which is given by the Hamiltonian (1) and the value of the spin quantum number s. A further dependence on the magnetic field is possible but neglected in this paper. As mentioned in the Introduction we do not consider special systems Σ but rather look for a general HTE expansion valid for arbitrary Heisenberg systems.

Interestingly, the coefficients $c_n^{\Sigma,f}$ in (2) can be written in the form of scalar products between two vectors Q and p such that the first vector Q only depends on the spin system Σ, but not on s, and the second one p only on the considered quantity f and the spin quantum number s. The index set of the vectors p and Q can be identified with finite sets G_n^f of multigraphs, see Ref. 15 for the details. Thus the scalar product of Q and p is a sum over multigraphs $\mathcal{G} \in G_n^f$:

$$c_n^{\Sigma,f} = \sum_{\mathcal{G}\in G_n^f} Q^{\Sigma}(\mathcal{G})\, p^f(\mathcal{G}) \ . \tag{3}$$

To give an elementary example, consider $f = \chi$, the zero-field uniform susceptibility. Simplifying the notation a bit we may write

$$\begin{aligned}\chi(\beta) \ = \ & Q(\mathcal{G}_0)\, p_0\, \beta + Q(\mathcal{G}_1)\, p_1\, \beta^2 + \\ & (Q(\mathcal{G}_2)\, p_2 + Q(\mathcal{G}_3)\, p_3)\ \beta^3 + \mathcal{O}(\beta^4) \ . \end{aligned} \tag{4}$$

Here

$$p_0 \ = \ \frac{1}{3} r, \quad r \equiv s(s+1) \tag{5}$$

$$p_1 \ = \ -\frac{2}{9} r^2,\ p_2 = -\frac{1}{18} r^2,\ p_3 = \frac{2}{27} r^3, \tag{6}$$

$\mathcal{G}_0$ = •, $\mathcal{G}_1$ = , $\mathcal{G}_2$ = , $\mathcal{G}_3$ = (7)

$$Q(\mathcal{G}_0) \ = \ N \quad \text{(number of spins)}, \tag{8}$$

$$Q(\mathcal{G}_1) \ = \ \sum_{\mu<\nu} J_{\mu\nu},\ Q(\mathcal{G}_2) = \sum_{\mu<\nu} J_{\mu\nu}^2, \tag{9}$$

$$Q(\mathcal{G}_3) \ = \ \sum_{\lambda<\mu<\nu} J_{\lambda\mu} J_{\mu\nu} \ . \tag{10}$$

In this example the $p^f(\mathcal{G})$ are polynomials in the variable $r \equiv s(s+1)$ of the form $p^f(\mathcal{G}) = \sum_{\nu=0}^{n} a_\nu\, r^\nu$, where n is the order of HTE in (3). This holds in general. Also generally the $Q^{\Sigma}(\mathcal{G})$ are polynomials in the coupling constants $J_{\mu\nu}$ that can be calculated by considering the various ways of embedding the graph $\mathcal{G}$ into the spin system.

For example, each mapping of the 3-chain onto three spins with numbers $\lambda < \mu < \nu$ gives rise to a term $J_{\lambda\mu} J_{\mu\nu}$ in $Q^{\Sigma}(\mathcal{G})$. The condition $\lambda < \mu < \nu$ guarantees that different embeddings resulting from symmetries of $\mathcal{G}$ are counted only once. Also this is typical for the general situation.

If the spin system is an infinite lattice, the $Q^{\Sigma}(\mathcal{G})$ have to be redefined by first considering finite realizations of Σ, and then dividing by the number of spins N and considering the thermodynamic limit $N \longrightarrow \infty$. If $\mathcal{G}$ is not connected then $Q^{\Sigma}(\mathcal{G})$ would scale with N^c, where c is the number of connected components of $\mathcal{G}$. Hence, for sake of consistency, the sets G_n^f must consist of connected graphs only. Keeping this in mind, the coefficients $c_n^{\Sigma,f}$ obtained represent rigorous results on infinite spin lattices that are notoriously rare.

For the determination of $Q^{\Sigma}(\mathcal{G})$ there exist effective computer programs. On the other hand, we have determined the "universal" polynomials $p^f(\mathcal{G})$ for $f = \chi, C$ and $\gamma(\mathcal{G}) \leq 10$. The method used has been explained to some details in Ref. 15 and will only be sketched here. The crucial auxiliary data are the polynomials $p^{(t)}(\mathcal{G})(r)$ resulting from the moments of the Heisenberg Hamiltonian (1) via

$$\tilde{t}_n \equiv \mathrm{Tr}\, H^n = (2s+1)^n \sum_{\mathcal{G}\in G_n^{(t)}} Q^{\Sigma}(\mathcal{G})\, p^{(t)}(\mathcal{G}) \ . \tag{11}$$

In this case the polynomials can be shown to be of the form $p^{(t)}(\mathcal{G}) = \sum_{\nu=g}^{\gamma} a_\nu\, r^\nu$, where $g = g(\mathcal{G})$ is the number of vertices of $\mathcal{G}$ and $\gamma = \gamma(\mathcal{G})$ the number of edges. These polynomials have been essentially determined by numerically calculating $\mathrm{Tr}\, H^n$ and $Q^{\Sigma}(\mathcal{G})$ for a suitable number of randomly chosen spin systems and then solving the linear system of equations (11) for the $p^{(t)}(\mathcal{G}$. This has to be repeated for different values of $s = 1/2,\ 1,\ 3/2, \ldots$ in order to estimate the rational coefficients of the polynomials $p^{(t)}(\mathcal{G})(r),\ r = s(s+1)$. Additionally, partial analytical results from Refs. 9 and 15 have been used and various cross checks have been performed.

As described in Ref. 15, one deduces from the $p^{(t)}(\mathcal{G})$ the "magnetic moments"

$$\tilde{\mu}_n \equiv \mathrm{Tr}\left(S_3^2\, H^n\right) = (2s+1)^n \sum_{\mathcal{G}\in G_n^{(m)}} Q^{\Sigma}(\mathcal{G})\, p^{(m)}(\mathcal{G})\ , \tag{12}$$

and from the $\tilde{t}_n$ and $\tilde{\mu}_n$ the HTE series for $C(\beta)$ and $\chi(\beta)$. In this last step there will occur products of $Q^{\Sigma}(\mathcal{G}_\mu)$ as well as contributions from disconnected graphs. In order to obtain manifestly extensive quantities, these various non-extensive terms have to cancel by means of certain "product rules" of the form

$$Q^{\Sigma}(\mathcal{G}_\mu)\, Q^{\Sigma}(\mathcal{G}_\nu) = \sum_{\lambda} c_{\mu\nu}^{\lambda}\, Q^{\Sigma}(\mathcal{G}_\lambda)\ . \tag{13}$$

All calculations described in this section involve a total number of 7355 graphs. Especially those steps leading from the moments to the HTE series have been performed with the aid of the computer algebra system MATHEMATICA 8.0.

III. APPLICATIONS

The region of validity of the HTE can be extended by Padé approximants[16] (see also Refs. 9 and 10). The Padé approximants are ratios of two polynomials $[m,n] = P_m(x)/R_n(x)$ of degree m and n and they provide an analytic continuation of a function $f(x)$ given by a power series. As a rule approximants with $m \sim n$ provide best results. Since we have a power series up to 10th order, we use here the corresponding [4,6], [5,5], and [6,4] Padé approximants. As in our previous paper[15] we will present the temperature dependence of physical quantities using a renormalized temperature $T/s(s+1)$.

A. The pyrochlore Heisenberg ferromagnet

The pyrochlore antiferromagnet has attracted much attention over the last years as an example of a highly frustrated three-dimensional (3D) magnetic system, see, e.g. Refs. 17–19 and references therein. In our previous paper[15] we have already presented the analytical expressions for χ and C up to order 8 as well as the temperature dependence of the susceptibility for the pyrochlore Heisenberg antiferromagnet, where a very good agreement of the [4,4] Padé approximant with classical Monte Carlo results down to temperatures of about 40% of the exchange coupling was found. The new terms of HTE in orders 9 and 10 can be found in Appendix B.

Here we consider the ferromagnetic case. We consider only nearest neighbor bonds and set $J_{\mu\nu} = J = -1$ for neighboring sites μ and ν. We want to demonstrate that the 10th order HTE is an appropriate tool to determine the critical (Curie) temperature T_c for 3D ferromagnets. To the best of our knowledge so far no data for T_c of the pyrochlore ferromagnet were reported in the literature. In Fig. 1 we show the Padé approximant [4,6] of the inverse susceptibility $1/\chi$. We see the typical behavior of 3D ferromagnet. The extreme quantum case $s = 1/2$ is somewhat separated from the other curves, but for $s > 1$ the curves are very close to each other. The zeros of $1/\chi(T)$ curves can be understood as an estimate of the critical temperature. More sophisticated methods exploit the behavior of the expansion coefficients c_n, see Eq. (2), of the susceptibility to determine T_c, see, e.g. Refs. 4, 6, 20. One variant is to analyze

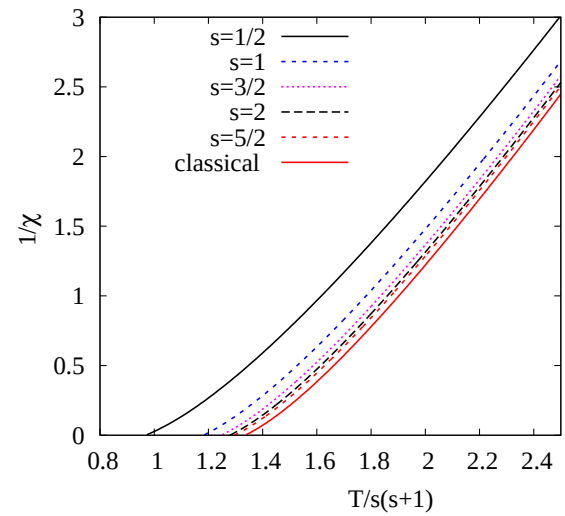

FIG. 1: (Color online) Padé approximant [4,6] of the inverse susceptibility $1/\chi$ of the pyrochlore Heisenberg ferromagnet for various spin quantum numbers s.

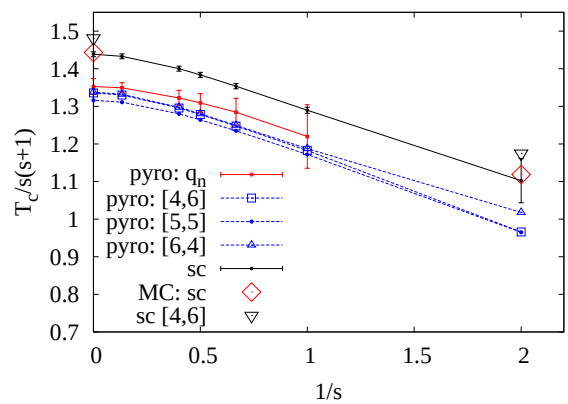

FIG. 2: (Color online) Curie temperature T_c in dependence on the inverse spin quantum number $1/s$ of the pyrochlore Heisenberg ferromagnet for $s = 1/2$, 1, 3/2, 2, 5/2, 15/2 and $s \to \infty$. For comparison we show also the T_c values for the simple-cubic Heisenberg ferromagnet. The Monte Carlo (MC) data for $s = 1/2$ and $s \to \infty$ are taken from Ref. 21 and from Ref. 22, respectively.

the quotient $q_n = c_n/c_{n-1}$. If the critical behavior of χ is given by $\chi(T) \propto (T - T_c)^{-\lambda}$, $T \to T_c + 0$, in the limit $n \to \infty$ this quotient depends linearly on $1/n$ according to $q_n = kT_c + (\lambda - 1)kT_c/n$. Hence we get T_c by $\lim_{n\to\infty} q_n = T_c$.

We made a linear fit of our HTE data for q_n including data points for $n = 5, \ldots, 10$ to get an approximate value for T_c. Our results for T_c are shown in Fig. 2. For comparison we also show T_c data for the simple-cubic ferromagnet, where precise Monte Carlo data are available for $s = 1/2$ (Ref. 21) and for $s \to \infty$ (Ref. 22), which yield an impression of the accuracy of the HTE estimate of T_c. Except for the $s = 1/2$-pyrochlore case the q_n data follow reasonably well a straight line (see also the error bars in Fig. 2). For the $s = 1/2$-pyrochlore ferromagnet the linear fit of the q_n data due to extremely large fluctuations of the data fails.[23] The comparison with the Monte Carlo data for the simple-cubic ferromagnet demonstrates that indeed the HTE series up to order 10 for the susceptibility may yield accurate values for T_c. Already the poles in the Padé approximants provide reasonable results (we have about 14% deviation from Monte Carlo data for $s = 1/2$ and about 9% for $s \to \infty$). The linear fit of q_n is even very close to the Monte Carlo results.

Comparing the pyrochlore and simple cubic lattices we find that T_c is significantly lower for the pyrochlore lattice. (Note that a simple molecular field approximation would lead to identical values of T_c, since both lattices have the same coordination number.) A similar finding was reported in Ref. 24, where the Curie temperatures of stacked square and a stacked kagome ferromagnets were compared. In analogy to the discussion in Ref. 24 we may attribute the lower T_c values of the pyrochlore lattice to geometric frustration. For the ferromagnetic ground state frustration is irrelevant, i.e. the ground-state energies are identical for the pyrochlore and simple-cubic ferromagnets. However, due to frustration the upper bound of the eigenenergies (related to the absolute value of ground-state energy of the corresponding antiferromagnet) is much lower for the pyrochlore ferromagnet than that for the simple-cubic lattice. Hence, one can expect that excited states with antiferromagnetic spin correlations have lower energy for the pyrochlore ferromagnet resulting in a larger contribution to the partition function at a certain finite temperature T in comparison with the simple-cubic ferromagnet.

B. The kagome Heisenberg antiferromagnet

The two-dimensional (2D) kagome antiferromagnet is one of the most interesting and challenging spin models. There are numerous papers investigating the ground state of the $s = 1/2$ case, see, e.g., Refs. 25–37 and references therein, but so far no conclusive answer on the nature of the ground state and the existence of a spin gap has been found. The finite-temperature properties are also widely discussed for the spin-1/2 model, including the analysis of HTE series.[12, 38–44] On the other hand, there are several kagome compounds with spin quantum number $s > 1/2$. We mention, the jarosite compounds with $s = 5/2$ (see, e.g., Refs. 45, 46), the magnetic compounds $KCr_3(OD)_6(SO_4)_2$ (Ref. 47) and $SrCr_{9p}Ga_{12-9p}O_{19}$ (Ref. 48) with $s = 3/2$, and the recently synthesized $BaNi3(OH)_2(VO4)_2$ (Ref. 49) compound with $s = 1$. For the $s = 5/2$ one may expect that a classical Monte-Carlo approach[50–54] might be reasonable, but for $s = 1$ and for $s = 3/2$ certainly quantum effects are important. However, we will see that at least for the specific heat the classical Monte-Carlo data significantly deviate from the data for $s = 5/2$, see below.

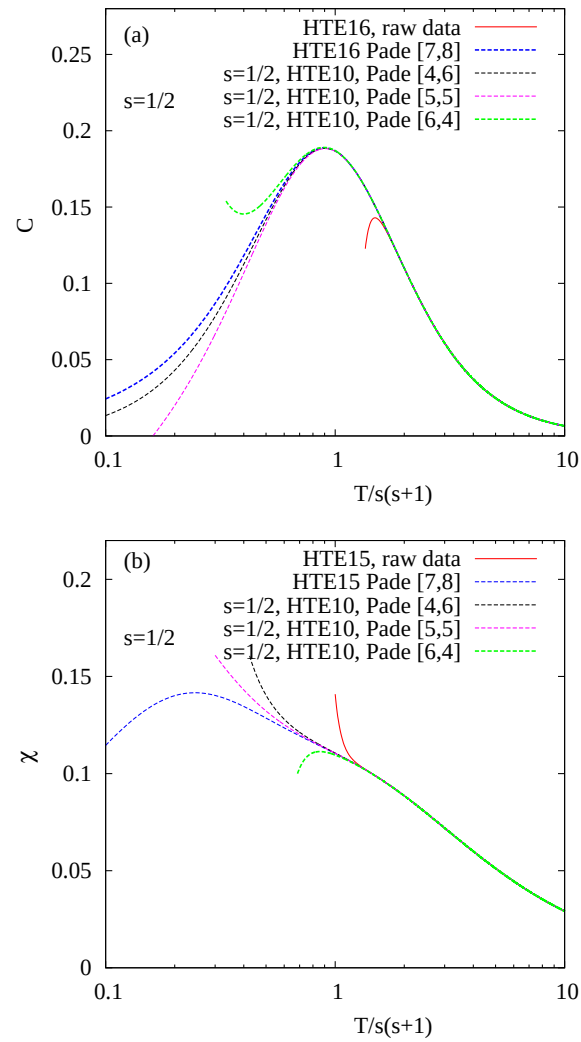

FIG. 3: (Color online) Specific heat C (a) and susceptibility χ (b) of the $s = 1/2$ kagome Heisenberg antiferromagnet. For comparison we show the raw data of the 15th/16th order HTE and the corresponding Padé [7,8] approximant taken from Ref. 12.

We present the HTE series for χ and C up to order

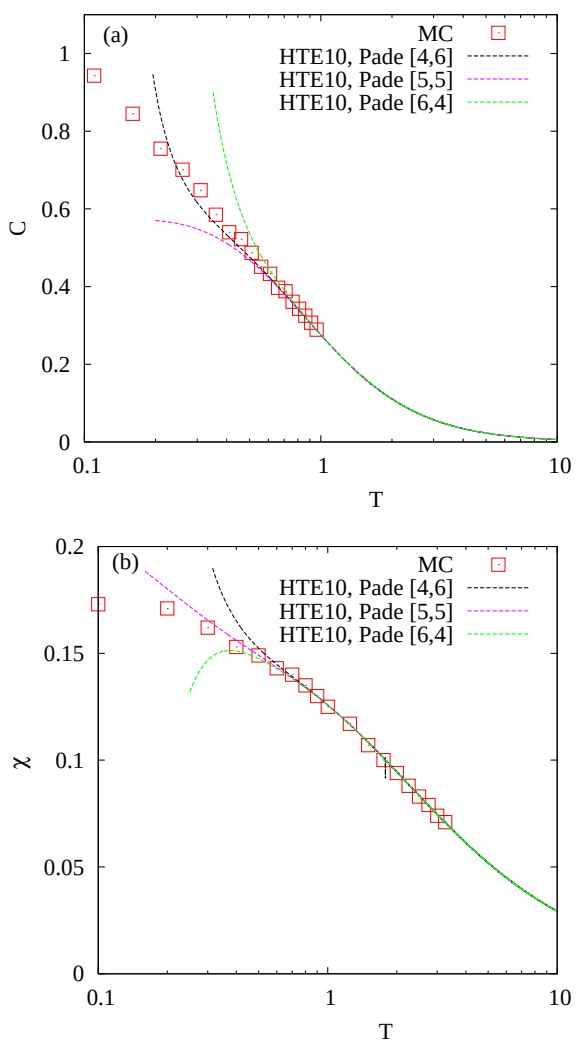

FIG. 4: (Color online) Specific heat C (a) and susceptibility χ (b) of the classical kagome Heisenberg antiferromagnet. For comparison we show the Monte Carlo data taken from Ref. 52.

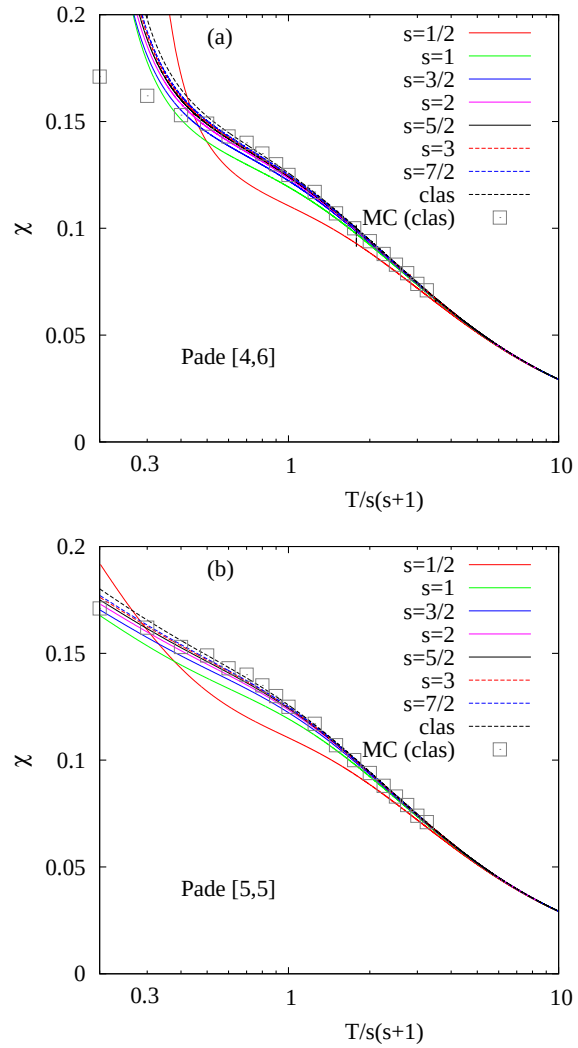

FIG. 5: (Color online) HTE data for the susceptibility χ of the spin-s kagome Heisenberg antiferromagnet, (a) Padé [4,6], (b) Padé [5,5]. For comparison we show the Monte Carlo data taken from Ref. 52.

10 and for arbitrary s in Appendix C. Remember that in Ref. 12 the HTE series for $s = 1/2$ for χ (C) was given up to order 15 (16).[55] As a benchmark test we first compare in Figs. 3 and 4 our HTE-Padé data with available data for $s = 1/2$ model[12] and for the classical model.[52] This comparison leads to the conclusion that (i) the [4,6] and [5,5] Padé approximants are favorable and that (ii) our HTE-Padé data are quite accurate down to temperatures $T/s(s+1) \sim 0.5$ ($T \sim 0.4$) for $s = 1/2$ ($s \to \infty$). In particular, the maximum in C present for the $s = 1/2$ model at $T/s(s+1) \sim 0.9$, cf. Refs. 12, 38–43, is correctly described by our Padé approximants. Note, however, that for $s = 1/2$ there are indications for a second low-temperature maximum in $C(T)$ below $T/s(s+1) = 0.1$, see Refs. 12, 39, 40, 42, which is not covered by our HTE approach. Another characteristic features is the shoulder present in $\chi(T)$ for $s = 1/2$ at about $T/s(s+1) = 1$, which is also well described by our Padé approximants.

In Figs. 5 and 6 we compare the $\chi(T)$ (Fig. 5) and $C(T)$ data (Fig. 6) for spin quantum numbers $s = 1/2, 1, \ldots, 7/2, \infty$. The susceptibility data clearly show that all curves for $s > 1/2$ form a narrow bundle in the temperature range accessible by our approach. Only for $s = 1/2$ the $\chi(T)$ curve is out of this bundle. Hence, one can argue that for $s > 1/2$ quantum effects in χ are almost negligible at normalized temperatures $T/s(s+1) \gtrsim 0.4$. The situation is quite different for the specific heat C, cf. Fig. 6. The maximum in $C(T)$, already mentioned above for $s = 1/2$, is evidently dependent on the spin quantum number s: Its position $T_{max}/s(s+1)$ is shifted to lower normalized temperatures and its height C_{max} in-

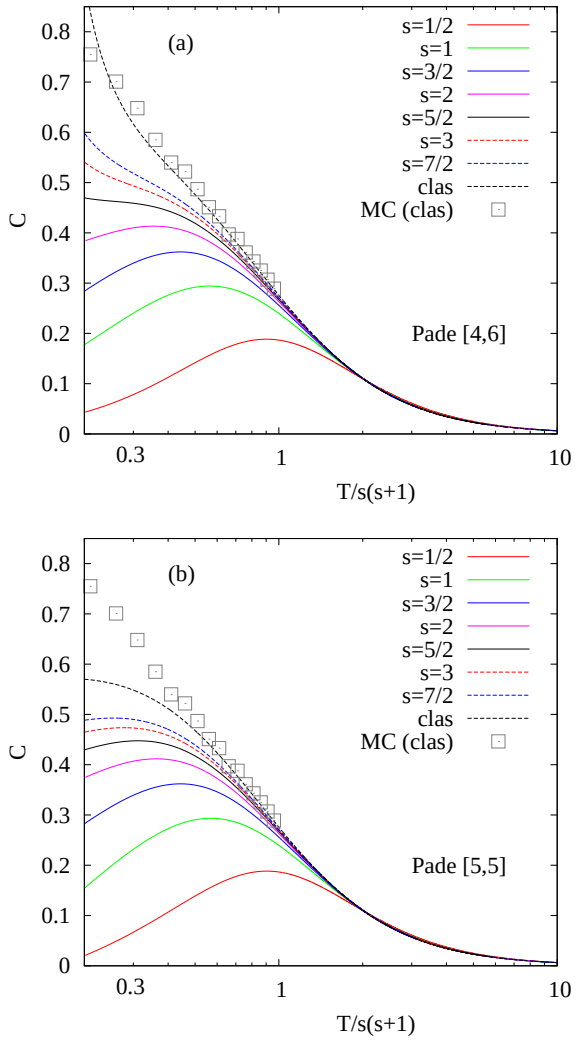

FIG. 6: (Color online) Specific heat C of the spin-s kagome Heisenberg antiferromagnet, (a) Padé [4,6], (b) Padé [5,5]. For comparison we show the Monte Carlo data taken from Ref. 52.

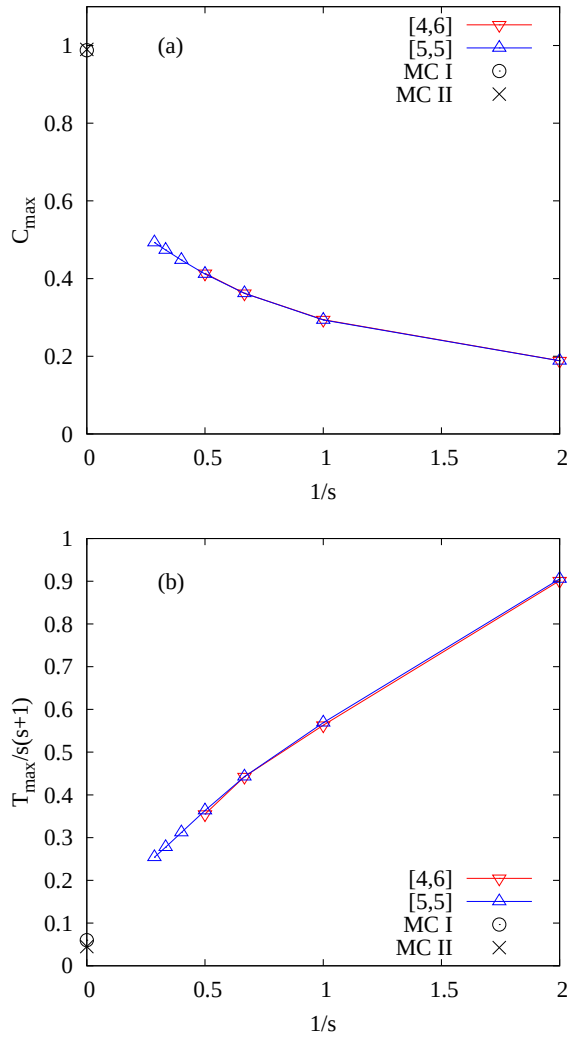

FIG. 7: (Color online) Height C_{max} (a) and position T_{max} (b) of the maximum in the specific heat C in dependence on the inverse spin quantum number $1/s$ of the kagome Heisenberg antiferromagnet. The Monte Carlo results for $s \to \infty$ are taken from Ref. 52 (MC I) and Ref. 53 (MC II). Note, however, that the maximum in $C(T)$ for the classical model is not well-pronounced, rather there is a fairly broad region of high C values.[53]

creases with growing s. Hence, the quantum effects seem to be important also for quite large values of s. The basic difference in the influence of s on $\chi(T)$ and $C(T)$ can be attributed to an exceptional density of low-lying singlet excitations, see e.g. Ref. 28. These nonmagnetic excitations are irrelevant for χ but important for C. Hence, our HTE-Padé data for $C(T)$ can be understood as an indirect indication for the existence of a unusual large density of low-lying singlet excitation also for $s > 1/2$. We show the position $T_{max}/s(s+1)$ and the height C_{max} as a function of $1/s$ in Fig. 7. From Fig. 6a it is obvious that for the [4,6]-Padé approximant a maximum is exists only for $s < 5/2$. The tendency of how the classical limit is approached is clearly visible from our HTE data. There is indeed a remarkably strong dependence on the spin quantum number. The slope of the corresponding curves shown in Fig. 7 is even increasing when approaching the classical limit $1/s = 0$.

IV. CONCLUSIONS

In this paper we present the HTE series up to 10th order of the specific heat C and the uniform susceptibility χ for Heisenberg models with arbitrary exchange patterns $J_{\mu\nu}$ and spin quantum number s. Our HTE scheme is encoded in a C++ program using as input the exchange matrix $J_{\mu\nu}$ and spin quantum number s. Using

Padé approximants for the HTE series the scheme can be used to discuss thermodynamic properties of general Heisenberg systems down to moderate temperatures of about $T/s(s+1) \sim 0.4 \ldots 0.5$ and thus for the interpretation of experimental data in rather wide temperature range, especially if other precise methods such as the quantum Monte Carlo method or the finite-temperature density matrix renormalization group approach are not applicable. We apply our scheme to the 3D pyrochlore ferromagnet to calculate the Curie temperature T_c in dependence on the spin quantum number s. Comparing T_c of the pyrochlore ferromagnet with corresponding values for the simple-cubic ferromagnet we find that the triangular configuration of bonds present in the pyrochlore lattices leads to a noticeable lowering of T_c. Using our HTE scheme for the kagome antiferromagnet we discuss the influence of s on the temperature dependence of C and χ. While the effect of s on χ in the accessible temperature range is rather weak, there is a well-pronounced shift of the maximum in the temperature dependence of specific heat to lower renormalized temperatures $T/s(s+1)$ when increasing s.

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Appendix A: Brief explanation how to explicitly get the HTE series for concrete Heisenberg models using the provided C++ program

As mentioned in the main text we encoded our HTE algorithm in a C++ program which allows to get explicitly the HTE series for concrete Heisenberg models. The C++ program code will be available at `http://www.uni-magdeburg.de/jschulen/HTE_II/` after publication of this manuscript. Then the code can be downloaded and freely used, e.g., on a state-of-the-art personal computer. To use the program the knowledge of C++ programming is not required. Until the paper is published we refer the interested user to our 8th order code available at `http://www.uni-magdeburg.de/jschulen/HTE/`. Let us briefly illustrate how to use the program.

1. General information

The C++ program calculates the coefficients of the HTE series of the susceptibility χ up to 10th order, the internal energy U up to 9th order and the specific heat C up to 10th order for a general Heisenberg model with up to four different exchange parameters labeled by j1, j2, j3, j4. There are two output modes. In the first one analytical expressions for the coefficients of the HTE as polynomials of j1, j2, j3 and j4 are obtained. In the second one the temperature dependence of χ, U and C for specific values of j1, j2, j3 and j4 are calculated numerically. Except the 'raw' HTE data in the output file also the Padé approximants (Padé [4,6], [5,5], [6,4] for $\chi(T)$ and $C(T)$, Padé [4,5], [5,4] for $U(T)$) for the HTE series are given. To use the second mode the -g parameter must be set.

The first step is to compile the C++ source code. There is a **Makefile** included in the package designed for a LINUX computer with a gnu-C++ compiler (version 4.5 or higher, C++11 Language Features are needed) to create an executable file **hte10** just by running 'make' in the directory with the source code files. By using the program on a computer with another operating system the compilation has to be adapted accordingly.

Before running the executable file you have to write a definition file defining your concrete Heisenberg system you want to investigate. This file contains the non-zero exchange integrals in the interaction matrix of the considered Heisenberg system, i.e. information on the interacting spin pairs (i, j) as well as the corresponding exchange parameters J_{ij}. Note that finite systems also can be considered, thus allowing the comparison with exact diagonalization data. Then the set of interacting spin pairs (i, j) is per se finite. In case you want to consider infinite lattice systems in principle this set is also infinite, but a finite-lattice representative can be used to calculate the HTE series. However, this finite-lattice representative must be large enough to avoid boundary effects. Since the 10th order terms contain lattice paths along the J_{ij} bonds with up to 10 steps the representative must allow more than 10 steps for any combination of exchange paths without reaching the boundaries when starting from a unit cell located in the center of the finite-lattice representative (which we will call *central unit cell* in what follows). Note that a unit cell here means the magnetic unit cell matching to the symmetry of the exchange matrix. When using a finite-lattice representative with periodic boundary conditions, of course, any unit cell can serve as the central unit cell. The definition file has to be copied to that directory which contains the HTE code and the corresponding executable file **hte10**.

Below we give more detailed information on the definition file. Moreover, some examples for definition files are also included in this package, see below.

2. Structure of the definition file

In the program package three examples for a definition file are included, namely (i) the square lattice J_1-J_2 model (definition file 'square.def'), (ii) the pyrochlore-lattice Heisenberg model (definition file 'pyrochlore.def'), and (iii) the Heisenberg model on the kagome lattice (definition file 'kagome.def'). The first two examples are discussed in our previuos paper, see Ref. 15. The kagome as well as again the pyrochlore lattices are discussed in the present paper.

The definition file shall contain information on the

number of sites in the system (respectively on the corresponding finite-lattice representative), the number of non-zero exchange bonds, the number of sites in the unit cell, the labels of the spins (site numbers) in the central unit cell, and the exchange matrix itself.
The following lines show a simple example of a definition file for an elementary square with nearest-neighbor exchange j1 and next nearest-neighbor exchange j2.

```
# 4-spin-system
# with 2 different exchange parameters j1  (NN) and j2 (diagonal NNN)
#
#  0    1
#  *----*
#  | \/ |
#  | /\ |
#  *----*
#  3    2
#
# Number of sites | Number of bonds | Number of sites in the unit cell
4 6 1
# site in the central unit cell (for this example: any site)
0
# Bond s1 s2 J
0 0 1 j1
1 0 2 j2
2 0 3 j1
3 1 2 j1
4 1 3 j2
5 2 3 j1
# end of file
```

At the beginning of the file an arbitrary number of comment lines (starting with #) are allowed to describe the system. In the line below the comment line
`# Number of sites | Number of bonds | Number of sites in the unit cell`
the total number of sites, the number of bonds and the number of sites in the unit cell are given. Next the numbers of the different sites in the central unit cell have to be given (one site per line), in the above example it is site 0 (because of the symmetry it could be any site). However, for the kagome lattice with three sites per unit cell in the definition file 'kagome.def' it reads for example

```
# site numbers in the central unit cell
0
2
4
```

This means that the sites in the central unit cell carry the numbers 0, 2 and 4. Below a further comment line
`# Bond s1 s2 J`
all bonds are listed, for example a particular line may read:
`2 0 3 j1`
Here is the
1st column: number of the bond, here bond number 2
2nd column: number of the first spin of the bond number 2, here 0
3rd column: number of the second spin of the bond number 2, here 3
4th column: symbol for the bond strength between spins 0 and 3, here j1.

3. Start of the program, input and output parameters

The following line and the table show how to run the executable file as well as all parameters of the program and its explanations.

```
 ./hte10 -i def [-mt] [-s spin] [-o out] [-d] [-g] [-tm Tm] [-dt dT] [-j1 J1] [-j2 J2] [-j3 J3]
[-j4 J4]
```

parameter	description	default value
def	name of the definition file	-
spin	spin length of the Heisenberg system, i.e. for s=3/2, set 1.5	0.5
out	name of the output file	out.dat

```
-mt         | use of multi threads                                          |   not set
-d          | detailed results for every graph are given in out.dat         |   not set
-g          | susceptibility and specific heat for the  temperature         |   not set
            | interval 0...Tm in steps of dT are calculated                 |
Tm          | -                                                             |   100
dT          | -                                                             |   0.05
J1,J2,J3,J4 | set the bonds 'Jn' to the specified value if -g is set        |   0
```

Note that there is a special option to consider classical unit vector spins replacing [-s spin] by [-cl]. Typically it takes only a few seconds to get the result. For 3D models it may take a few minutes or in exceptional cases some hours. To be more specific we give two examples how to start the executeable file.
Example (i)

```
./hte10 -i kagome.def -s 2.0 -o kagome_s_2.0.out
```

Here in the output file 'kagome_s_2.0.out' the coefficients of the HTE series as analytical expressions in j1 for the kagome lattice (definition file 'kagome.def') are given. The data in 'kagome_s_2.0.out' correspond to Eqs. (C1) and (C2) by setting $s = 2$.
Example (ii)

```
./hte10 -i square.def -s 0.5 -o data_square_s_0.5.out -j1 1 -j2 0.4 -g -tm 50 -dt 0.01
```

Here in the output file 'data_square_s_0.5.out' numerical data for the HTE series (raw data and Padé approximants) for the temperature dependence of the susceptibility, the internal energy and the specific heat for specific values of j1 and j2 (i.e. $J_1 = 1$, $J_2 = 0.4$) are given for the square lattice J_1-J_2 model (definition file 'square.def').

Appendix B: The high-temperature expansion for the susceptibility and the specific heat for the Heisenberg model on the pyrochlore lattice

The general formulas for the susceptibility and the specific heat for the Heisenberg model on the pyrochlore lattice with NN exchange constant J up to 8th order can be found in Ref. 15. For the sake of consistency with this reference we have set $\beta = \frac{1}{k_B T}$ in appendix A and B which is slightly different from the definition in section II. The formulas for the 9th and 10h order read for the susceptibility

$$\chi(\beta) = \frac{N}{J}\sum_{n=1}^{\infty} c_n (J\beta)^n \tag{B1}$$

$$c_9 = \frac{1}{5143824000} r^2(-2710665 + 142840908r - 2195288001r^2 + 14497581366r^3 - 45972407664r^4 + 77794619872r^5 - 82650432896r^6 + 46730617088r^7)$$

$$c_{10} = -\frac{1}{169746192000} r^2(51519240 - 2994073848r + 51386055291r^2 - 396940170060r^3 + 1579391570694r^4 - 3442568263344r^5 + 4692701814464r^6 - 4374573206272r^7 + 2124654831616r^8).$$

and for the specific heat

$$C(\beta) = Nk\sum_{n=2}^{\infty} d_n (J\beta)^n \tag{B2}$$

$$d_9 = -\frac{1}{285768000} r^2(-1807110 + 91861560r - 1255862151r^2 + 6268644864r^3$$
$$-8882615472r^4 - 1691186688r^5 - 21317760r^6 + 1042017280r^7)$$
$$d_{10} = -\frac{1}{6286896000} r^2(-25759620 + 1451298330r - 22610800701r^2$$
$$+142189820847r^3 - 349296723134r^4 + 154955752848r^5$$
$$+102919717624r^6 + 82927576960r^7 - 11100907520r^8),$$

where $r = s(s+1)$.

Appendix C: The high-temperature expansion for the susceptibility and the specific heat for the Heisenberg model on the kagome lattice

The general formulas for the Heisenberg model on the kagome lattice with NN exchange constant J read for the susceptibility

$$\chi(\beta) = \frac{N}{J} \sum_{n=1}^{\infty} c_n (J\beta)^n \tag{C1}$$
$$c_1 = \frac{1}{3} r$$
$$c_2 = -\frac{4}{9} r^2$$
$$c_3 = \frac{1}{9} r^2(-1 + 4r)$$
$$c_4 = -\frac{4}{405} r^2(3 - 28r + 37r^2)$$
$$c_5 = \frac{1}{4860} r^2(-45 + 702r - 1892r^2 + 1328r^3)$$
$$c_6 = -\frac{1}{510300} r^2(1728 - 35946r + 164289r^2 - 207896r^3 + 102576r^4)$$
$$c_7 = \frac{1}{6123600} r^2(-8694 + 218916r - 1401381r^2 + 2888772r^3 - 2251248r^4 + 909184r^5)$$
$$c_8 = -\frac{1}{22963500} r^2(15390 - 446256r + 3538764r^2 - 10535337r^3 + 12202552r^4 - 7318640r^5 + 2416640r^6)$$
$$c_9 = \frac{1}{7715736000} r^2(-2710665 + 87954822r - 807482331r^2 + 3091042674r^3 - 5118502560r^4 + 4009481184r^5$$
$$-2113197952r^6 + 518354176r^7)$$
$$c_{10} = -\frac{1}{1273096440000} r^2(257596200 - 9180862110r + 93799827171r^2 - 426255134022r^3 + 931126345494r^4$$
$$-977085756168r^5 + 621427831616r^6 - 280517703040r^7 + 48779713280r^8)$$

and for the specific heat

$$C(\beta) = Nk \sum_{n=2}^{\infty} d_n (J\beta)^n \tag{C2}$$
$$d_2 = \frac{2}{3} r^2$$
$$d_3 = -\frac{1}{9} r^2(-3 + 4r)$$

$$d_4 = -\frac{2}{45} r^2(-3 + 23r + 3r^2)$$
$$d_5 = \frac{1}{162} r^2(9 - 126r + 116r^2 + 48r^3)$$
$$d_6 = -\frac{1}{68040} r^2(-1728 + 33426r - 102969r^2 - 19464r^3 + 2144r^4)$$
$$d_7 = -\frac{1}{97200} r^2(-1242 + 29556r - 150039r^2 + 96676r^3 + 64544r^4 + 20992r^5)$$
$$d_8 = \frac{1}{1093500} r^2(7695 - 213084r + 1435806r^2 - 2537523r^3 - 539132r^4 + 58400r^5 + 186680r^6)$$
$$d_9 = -\frac{1}{214326000} r^2(-903555 + 28196370r - 227949579r^2 + 44035529526r^3 - 98661584568r^4 + 65112518418r^5$$
$$-16209976080r^6 + 1271065760r^7)$$
$$new:$$
$$d_9 = \frac{1}{214326000} r^2(903555 - 28196370r + 227949579r^2 - 634514526r^3 + 285950568r^4 + 230120832r^5$$
$$+135526080r^6 + 14890240r^7)$$
$$d_{10} = -\frac{1}{18860688000} r^2(-51519240 + 1775187630r - 16326321219r^2 + 59250202038r^3 - 69170925596r^4$$
$$-15707506528r^5 - 728311984r^6 + 9196378240r^7 + 3884989440r^8).$$

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Tenth-order high-temperature expansion for the susceptibility and the specific heat of spin-s Heisenberg models with arbitrary exchange patterns: Application to pyrochlore and kagome magnets

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We present the high-temperature expansion (HTE) up to 10th order of the specific heat C and the uniform susceptibility χ for Heisenberg models with arbitrary exchange patterns and arbitrary spin quantum number s. We encode the algorithm in a C++ program provided in the supplementary material and available at `http://www.uni-magdeburg.de/jschulen/HTE10/` which allows to get explicitly the HTE series for concrete Heisenberg models. We apply our algorithm to pyrochlore ferromagnets and kagome antiferromagnets using several Padé approximants for the HTE series. For the pyrochlore ferromagnet we use the HTE data for χ to estimate the Curie temperature T_c as a function of the spin quantum number s. We find that T_c is smaller than that for the simple cubic lattice, although both lattices have the same coordination number. For the kagome antiferromagnet the influence of the spin quantum number s on the susceptibility as a function of renormalized temperature $T/s(s+1)$ is rather weak for temperatures down to $T/s(s+1) \sim 0.3$. On the other hand, the specific heat as a function of $T/s(s+1)$ noticeably depends on s. The characteristic maximum in $C(T)$ is monotonously shifted to lower values of $T/s(s+1)$ when increasing s.

I. INTRODUCTION

Magnetic systems described by the Heisenberg Hamiltonian

$$H = \sum_{\mu<\nu} J_{\mu\nu} \mathbf{s}_\mu \cdot \mathbf{s}_\nu \tag{1}$$

are an active field of theoretical and experimental research.[1] The accurate description of these quantum many-body systems is a basic aim of theoretical investigations. The comparison with experimental studies typically requires the calculation of the temperature dependence of physical properties, such as the susceptibility χ and the specific heat C. For unfrustrated spin systems the quantum Monte Carlo technique is a suitable tool to provide precise data, but it is not applicable due to the sign problem for frustrated quantum spin models.[2] Hence, reliable results for strongly frustrated quantum spin models are notoriously rare. Since there is a very active research in the field of frustrated quantum magnetism, see e.g. Refs. 1, 3, 4 and references therein, theoretical methods to calculate thermodynamic quantities in the presence of frustration are highly desirable. One of the most interesting systems is the kagome Heisenberg antiferromagnet which we will consider in Sec. III B. This highly frustrated magnetic system has been extensively investigated theoretically for spin quantum number $s = 1/2$, see, e.g., Refs. 5–17, and in the classical limit $s \to \infty$, see, e.g., Refs. 18–22. On the experimental side, several kagome compounds have $s > 1/2$, however, the theoretical study of quantum models with $s > 1/2$ lags behind.

A universal straightforward approach to calculate thermodynamic quantities for unfrustrated as well as frustrated magnetic systems is the high-temperature expansion (HTE). For Heisenberg models this method was introduced in an early work by W. Opechowski[23] based on a method of approximate evaluation of the partition function developed by H.A. Kramers. In the 1950ies and 1960ies the method was further developed and widely applied to various Heisenberg systems, see e.g. Refs. 24–28.

The HTE method is now well-established and its application to magnetic systems is a basic tool in theoretical physics, see Refs. 29, 30 and references therein. For the Heisenberg model with nearest-neighbor interaction on standard lattices now typically the HTE is known up to high-orders, see for example Refs. 31, 32, and 33 where the HTE up to 14th order for the triangular and up to 16th order for the kagome and for the hyperkagome lattices were published. On the other hand, often magnetic compounds and corresponding spin models are of interest, where two, three or even more exchange constants are relevant. Typical examples are magnetic systems with nearest-neighbor, next-nearest-neighbor, and 3rd-nearest-neighbor couplings, see, e.g., Ref. 34. Moreover, in most of the quasi-low-dimensional magnetic compounds interchain or interlayer couplings play a role, see, e.g. Ref. 35. Note further that available high-order HTE are often restricted to spin quantum number $s = 1/2$, see, e.g., Refs. 31, 32, 33, 36 and 37 as an example. As a rule, for such more complex exchange geometries and/or higher spin quantum numbers $s > 1/2$ relevant for the interpretation of ex-

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perimental data the HTE is not available in higher orders. An earlier attempt to bridge this gap was published in Ref. 38, where general analytical HTE expressions were given for arbitrary s and arbitrary Heisenberg exchange couplings up to order three. Very recently the present authors have published a significant extension of this work using computer algebraic tools.[39] In that paper the HTE algorithm for general spin-s Heisenberg models up to 8th order was presented. This algorithm was encoded as a C++ program. The download (URL `http://www.uni-magdeburg.de/jschulen/HTE/`) and use are free. Thus this algorithm provides a flexible tool for the community to compute the HTE for the susceptibility and the specific heat, which can be used to analyze the thermodynamics of spin models, to check approximations, and, last but not least, to compare experimental data with model predictions.

In our previous work[39] we considered several models. Our results demonstrated that the 8th order HTE with a subsequent Padé approximation is (i) able to describe correctly the maximum of the susceptibility of a square-lattice s=1/2 Heisenberg antiferromagnet, (ii) can yield better results for spin systems in dimension $d > 1$ than full exact diagonalization, and (iii) gives good agreement with Monte Carlo data for the classical pyrochlore antiferromagnet down to temperatures of about 40% of the exchange coupling.

The aim of the present paper is twofold. On the one hand, we will extend our earlier approach[39] up to 10th order for general Heisenberg Hamiltonians (Sec II). Again we provide this new extended tool as a freely accessible C++ program, see supplementary material[40] and also `http://www.uni-magdeburg.de/jschulen/HTE10/`, were except the code a manual on how to use the code can also be found. We recommend to use `http://www.uni-magdeburg.de/jschulen/HTE10/`, since there will be provided available updates of the code (fixing of bugs, improvement of the code, etc.). On the other hand, results for the susceptibility and the specific heat for the spin-s Heisenberg model on the pyrochlore and the kagome lattices are provided (Sec III), which we will use to discuss the influence of the spin quantum number on the thermodynamics of these models. In the Appendices A and B we present explicitly the HTE series for the pyrochlore and the kagome spin-s Heisenberg magnets. In the supplementary part[40] we present the detailed results for the 10th order HTE of specific heat and susceptibility and the auxiliary quantities to be introduced in Sect II.

II. BRIEF EXPLANATION OF THE METHOD

We consider the HTE expansion of extensive quantities f, e. g., susceptibility χ or specific heat C, of the form

$$f^{\Sigma}(\beta) = \sum_{n=0}^{\infty} c_n^{\Sigma,f}\, \beta^n \ . \tag{2}$$

Here β is the inverse dimensionless temperature $\beta = \frac{J}{k_B T}$, where J is a typical exchange energy, and the index Σ indicates the dependence on the spin system Σ which is given by the Hamiltonian (1) and the value of the spin quantum number s. A further dependence on the magnetic field is possible but neglected in this paper. As mentioned in the Introduction we do not consider special systems Σ but rather look for a general HTE expansion valid for arbitrary Heisenberg systems.

Interestingly, the coefficients $c_n^{\Sigma,f}$ in (2) can be written in the form of scalar products between two vectors Q and p such that the first vector Q only depends on the spin system Σ, but not on s, and the second one p only on the considered quantity f and the spin quantum number s. The index set of the vectors p and Q can be identified with finite sets G_n^f of multigraphs, see Ref. 39 for the details. Thus the scalar product of Q and p is a sum over multigraphs $\mathcal{G} \in G_n^f$:

$$c_n^{\Sigma,f} = \sum_{\mathcal{G}\in G_n^f} Q^{\Sigma}(\mathcal{G})\, p^f(\mathcal{G}) \ . \tag{3}$$

To give an elementary example, consider $f = \chi$, the zero-field uniform susceptibility. Simplifying the notation a bit we may write

$$\begin{aligned}\chi(\beta) \ &= \ Q(\mathcal{G}_0)\, p_0\, \beta + Q(\mathcal{G}_1)\, p_1\, \beta^2 + \\ &\quad (Q(\mathcal{G}_2)\, p_2 + Q(\mathcal{G}_3)\, p_3)\ \beta^3 + \mathcal{O}(\beta^4) \ . \end{aligned} \tag{4}$$

Here

$$p_0 \ = \ \frac{1}{3} r, \quad r \equiv s(s+1) \tag{5}$$

$$p_1 \ = \ -\frac{2}{9} r^2,\ p_2 = -\frac{1}{18} r^2,\ p_3 = \frac{2}{27} r^3, \tag{6}$$

$\mathcal{G}_0 =$, $\mathcal{G}_1 =$, $\mathcal{G}_2 =$, $\mathcal{G}_3 =$ (7)

$$Q(\mathcal{G}_0) \ = \ N \quad \text{(number of spins)}, \tag{8}$$

$$Q(\mathcal{G}_1) \ = \ \sum_{\mu<\nu} J_{\mu\nu},\ Q(\mathcal{G}_2) = \sum_{\mu<\nu} J_{\mu\nu}^2, \tag{9}$$

$$Q(\mathcal{G}_3) \ = \ \sum_{\lambda<\mu<\nu} J_{\lambda\mu} J_{\mu\nu} \ . \tag{10}$$

In this example the $p^f(\mathcal{G})$ are polynomials in the variable $r \equiv s(s+1)$ of the form $p^f(\mathcal{G}) = \sum_{\nu=0}^{n} a_\nu\, r^\nu$, where n is the order of HTE in (3). This holds in general. Also generally the $Q^{\Sigma}(\mathcal{G})$ are polynomials in the coupling constants $J_{\mu\nu}$ that can be calculated by considering the various ways of embedding the graph $\mathcal{G}$ into the spin system.

For example, each mapping of the 3-chain onto three spins with numbers $\lambda < \mu < \nu$ gives rise to a term $J_{\lambda\mu} J_{\mu\nu}$ in $Q^{\Sigma}(\mathcal{G})$. The condition $\lambda < \mu < \nu$ guarantees that different embeddings resulting from symmetries of $\mathcal{G}$ are counted only once. Also this is typical for the general situation.

If the spin system is an infinite lattice, the $Q^\Sigma(\mathcal{G})$ have to be redefined by first considering finite realizations of Σ, and then dividing by the number of spins N and considering the thermodynamic limit $N \longrightarrow \infty$. If $\mathcal{G}$ is not connected then $Q^\Sigma(\mathcal{G})$ would scale with N^c, where c is the number of connected components of $\mathcal{G}$. Hence, for sake of consistency, the sets G_n^f must consist of connected graphs only. Keeping this in mind, the coefficients $c_n^{\Sigma,f}$ obtained represent rigorous results on infinite spin lattices that are notoriously rare.

For the determination of $Q^\Sigma(\mathcal{G})$ there exist effective computer programs. On the other hand, we have determined the "universal" polynomials $p^f(\mathcal{G})$ for $f = \chi, C$ and $\gamma(\mathcal{G}) \leq 10$, see Ref. 40. The method used has been explained to some details in Ref. 39 and will only be sketched here. The crucial auxiliary data are the polynomials $p^{(t)}(\mathcal{G})(r)$ resulting from the moments of the Heisenberg Hamiltonian (1) via

$$\tilde{t}_n \equiv \mathrm{Tr}\, H^n = (2s+1)^n \sum_{\mathcal{G}\in G_n^{(t)}} Q^\Sigma(\mathcal{G})\, p^{(t)}(\mathcal{G}) \,. \qquad (11)$$

In this case the polynomials can be shown to be of the form $p^{(t)}(\mathcal{G}) = \sum_{\nu=g}^{\gamma} a_\nu\, r^\nu$, where $g = g(\mathcal{G})$ is the number of vertices of $\mathcal{G}$ and $\gamma = \gamma(\mathcal{G})$ the number of edges. These polynomials have been essentially determined by numerically calculating $\mathrm{Tr}\, H^n$ and $Q^\Sigma(\mathcal{G})$ for a suitable number of randomly chosen spin systems and then solving the linear system of equations (11) for the $p^{(t)}(\mathcal{G})$. This has to be repeated for different values of $s = 1/2,\, 1,\, 3/2, \ldots$ in order to estimate the rational coefficients of the polynomials $p^{(t)}(\mathcal{G})(r)$, $r = s(s+1)$. Additionally, partial analytical results from Refs. 29 and 39 have been used and various cross checks have been performed.

As described in Ref. 39, one deduces from the $p^{(t)}(\mathcal{G})$ the "magnetic moments"

$$\tilde{\mu}_n \equiv \mathrm{Tr}\left(S_3^2\, H^n\right) = (2s+1)^n \sum_{\mathcal{G}\in G_n^{(m)}} Q^\Sigma(\mathcal{G})\, p^{(m)}(\mathcal{G}) \,, \qquad (12)$$

and from the $\tilde{t}_n$ and $\tilde{\mu}_n$ the HTE series for $C(\beta)$ and $\chi(\beta)$. In this last step there will occur products of $Q^\Sigma(\mathcal{G}_\mu)$ as well as contributions from disconnected graphs. In order to obtain manifestly extensive quantities, these various non-extensive terms have to cancel by means of certain "product rules" of the form

$$Q^\Sigma(\mathcal{G}_\mu)\, Q^\Sigma(\mathcal{G}_\nu) = \sum_\lambda c_{\mu\nu}^\lambda\, Q^\Sigma(\mathcal{G}_\lambda) \,. \qquad (13)$$

All calculations described in this section involve a total number of 7355 graphs. Especially those steps leading from the moments to the HTE series have been performed with the aid of the computer algebra system MATHEMATICA 8.0.

III. APPLICATIONS

The region of validity of the HTE can be extended by Padé approximants[41] (see also Refs. 29 and 30). The Padé approximants are ratios of two polynomials $[m,n] = P_m(x)/R_n(x)$ of degree m and n and they provide an analytic continuation of a function $f(x)$ given by a power series. As a rule approximants with $m \sim n$ provide best results. Since we have a power series up to 10th order, we use here the corresponding [4,6], [5,5], and [6,4] Padé approximants. As in our previous paper[39] we will present the temperature dependence of physical quantities using a renormalized temperature $T/s(s+1)$.

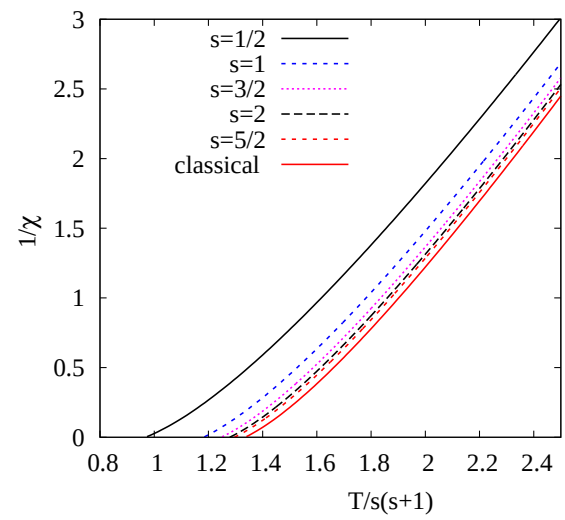

FIG. 1: (Color online) Padé approximant [4,6] of the inverse susceptibility $1/\chi$ of the pyrochlore Heisenberg ferromagnet for various spin quantum numbers s.

A. The pyrochlore Heisenberg ferromagnet

The pyrochlore antiferromagnet has attracted much attention over the last years as an example of a highly frustrated three-dimensional (3D) magnetic system, see, e.g. Refs. 42–44 and references therein. In our previous paper[39] we have already presented the analytical expressions for χ and C up to order 8 as well as the temperature dependence of the susceptibility for the pyrochlore Heisenberg antiferromagnet, where a very good agreement of the [4,4] Padé approximant with classical Monte Carlo results down to temperatures of about 40% of the exchange coupling was found. The new terms of HTE in orders 9 and 10 can be found in Appendix A.

Here we consider the ferromagnetic case. We consider only nearest neighbor bonds and set $J_{\mu\nu} = J = -1$ for neighboring sites μ and ν. We want to demonstrate that the 10th order HTE is an appropriate tool to determine the critical (Curie) temperature T_c for 3D ferromagnets.

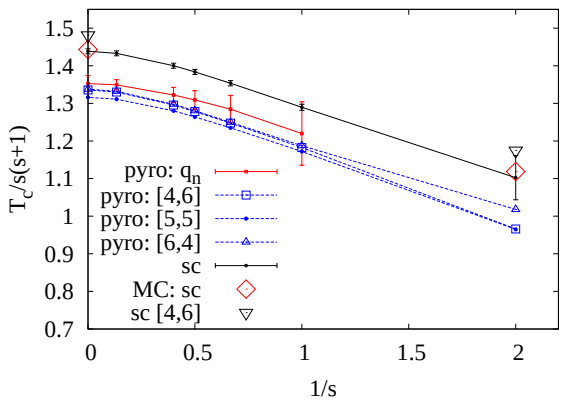

FIG. 2: (Color online) Curie temperature T_c in dependence on the inverse spin quantum number $1/s$ of the pyrochlore Heisenberg ferromagnet for $s = 1/2$, 1, 3/2, 2, 5/2, 15/2 and $s \to \infty$. For comparison we show also the T_c values for the simple-cubic Heisenberg ferromagnet. The Monte Carlo data (MC) for $s = 1/2$ and $s \to \infty$ are taken from Ref. 46 and from Ref. 47, respectively.

To the best of our knowledge so far no data for T_c of the pyrochlore ferromagnet were reported in the literature. In Fig. 1 we show the Padé approximant [4,6] of the inverse susceptibility $1/\chi$. We see the typical behavior of 3D ferromagnet. The extreme quantum case $s = 1/2$ is somewhat separated from the other curves, but for $s > 1$ the curves are very close to each other. The zeros of $1/\chi(T)$ curves can be understood as an estimate of the critical temperature. More sophisticated methods exploit the behavior of the expansion coefficients c_n, see Eq. (2), of the susceptibility to determine T_c, see, e.g. Refs. 24, 26, 45. One variant is to analyze the quotient $q_n = c_n/c_{n-1}$. If the critical behavior of χ is given by $\chi(T) \propto (T - T_c)^{-\lambda}$, $T \to T_c + 0$, in the limit $n \to \infty$ this quotient depends linearly on $1/n$ according to $q_n = \frac{kT_c}{J} + (\lambda - 1)\frac{kT_c}{Jn}$. Hence we get $\frac{kT_c}{J}$ by $\lim_{n\to\infty} q_n = \frac{kT_c}{J}$.

We made a linear fit of our HTE data for q_n including data points for $n = 5, \ldots, 10$ to get an approximate value for T_c. Our results for T_c are shown in Fig. 2. For comparison we also show T_c data for the simple-cubic ferromagnet, where precise Monte Carlo data are available for $s = 1/2$ (Ref. 46) and for $s \to \infty$ (Ref. 47), which yield an impression of the accuracy of the HTE estimate of T_c. Except for the $s = 1/2$-pyrochlore case the q_n data follow reasonably well a straight line (see also the error bars in Fig. 2). For the $s = 1/2$-pyrochlore ferromagnet the linear fit of the q_n data due to extremely large fluctuations of the data fails.[48] The comparison with the Monte Carlo data for the simple-cubic ferromagnet demonstrates that indeed the HTE series up to order 10 for the susceptibility may yield accurate values for T_c. Already the poles in the Padé approximants provide reasonable results (we have about 14% deviation from Monte Carlo data for $s = 1/2$ and about 9% for $s \to \infty$). The linear fit of q_n is even very close to the Monte Carlo results.

Comparing the pyrochlore and simple cubic lattices we find that T_c is significantly lower for the pyrochlore lattice. (Note that a simple molecular field approximation would lead to identical values of T_c, since both lattices have the same coordination number.) A similar finding was reported in Ref. 49, where the Curie temperatures of stacked square and a stacked kagome ferromagnets were compared. In analogy to the discussion in Ref. 49 we may attribute the lower T_c values of the pyrochlore lattice to geometric frustration. For the ferromagnetic ground state frustration is irrelevant, i.e. the ground-state energies are identical for the pyrochlore and simple-cubic ferromagnets. However, due to frustration the upper bound of the eigenenergies (related to the absolute value of ground-state energy of the corresponding antiferromagnet) is much lower for the pyrochlore ferromagnet than that for the simple-cubic lattice. Hence, one can expect that excited states with antiferromagnetic spin correlations have lower energy for the pyrochlore ferromagnet resulting in a larger contribution to the partition function at a certain finite temperature T in comparison with the simple-cubic ferromagnet.

B. The kagome Heisenberg antiferromagnet

The two-dimensional (2D) kagome antiferromagnet is one of the most interesting and challenging spin models. There are numerous papers investigating the ground state of the $s = 1/2$ case, see, e.g., Refs. 5–17, 50, 51 and references therein, but so far no conclusive answer on the nature of the ground state and the existence of a spin gap has been found. The finite-temperature properties are also widely discussed for the spin-1/2 model, including the analysis of HTE series.[32, 52–58] On the other hand, there are several kagome compounds with spin quantum number $s > 1/2$. We mention, the jarosite compounds with $s = 5/2$ (see, e.g., Refs. 59, 60), the magnetic compounds $KCr_3(OD)_6(SO_4)_2$ (Ref. 61) and $SrCr_{9p}Ga_{12-9p}O_{19}$ (Ref. 62) with $s = 3/2$, and the recently synthesized $BaNi3(OH)_2(VO4)_2$ (Ref. 63) compound with $s = 1$. For the $s = 5/2$ one may expect that a classical Monte Carlo approach[18–22] might be reasonable, but for $s = 1$ and for $s = 3/2$ certainly quantum effects are important. However, we will see that at least for the specific heat the classical Monte Carlo data significantly deviate from the data for $s = 5/2$, see below.

We present the HTE series for χ and C up to order 10 and for arbitrary s in Appendix B. Remember that in Ref. 32 the HTE series for $s = 1/2$ for χ (C) was given up to order 15 (16).[64] As a benchmark test we first compare in Figs. 3 and 4 our HTE-Padé data with available data for $s = 1/2$ model[32] and for the classical model.[20] This comparison leads to the conclusion that (i) the [4,6] and [5,5] Padé approximants are favorable

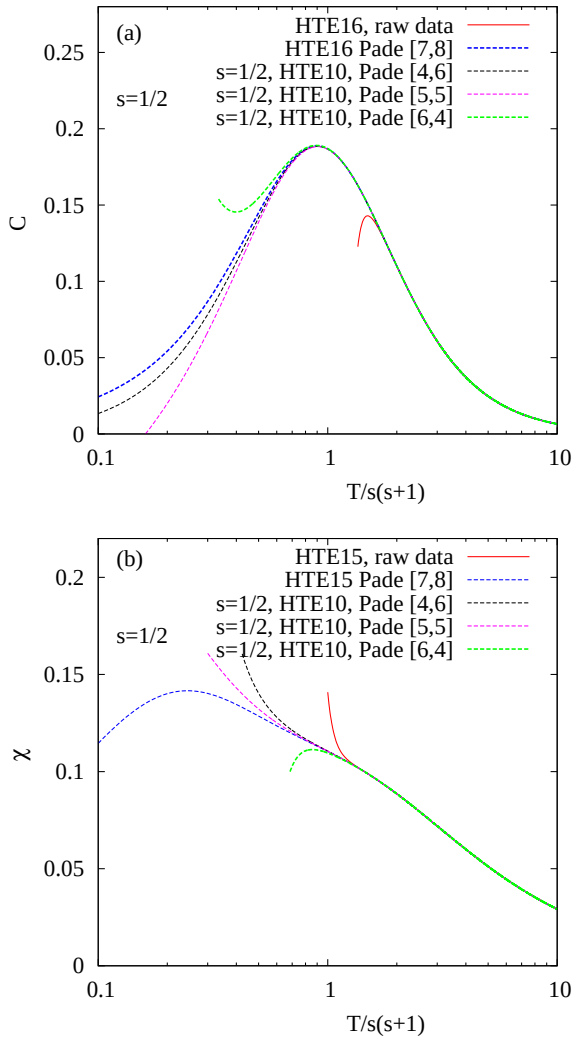

FIG. 3: (Color online) Specific heat C (a) and susceptibility χ (b) of the $s = 1/2$ kagome Heisenberg antiferromagnet. For comparison we show the raw data of the 15th/16th order HTE and the corresponding Padé [7,8] approximant taken from Ref. 32.

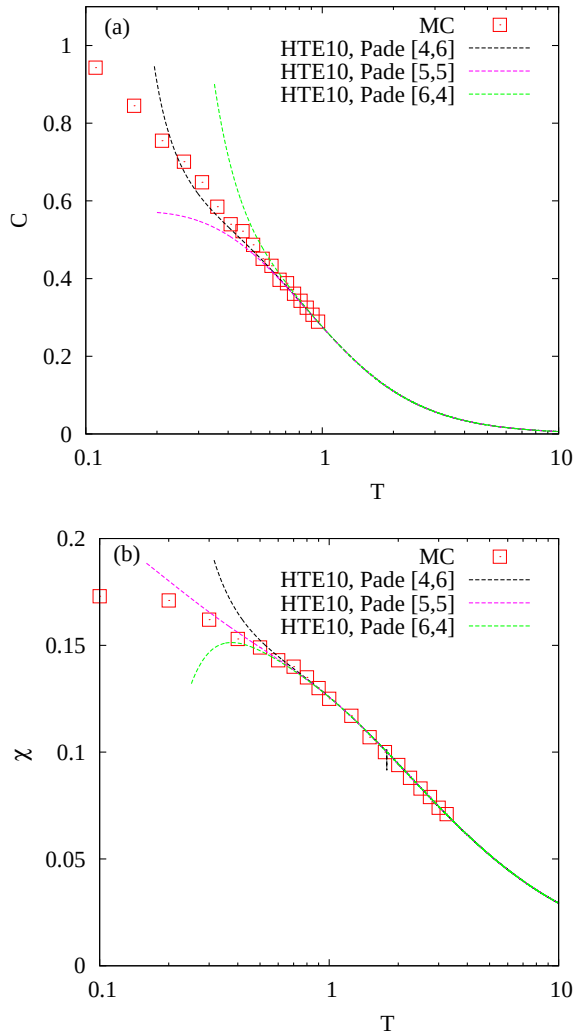

FIG. 4: (Color online) Specific heat C (a) and susceptibility χ (b) of the classical kagome Heisenberg antiferromagnet. For comparison we show the Monte Carlo data (MC) taken from Ref. 20.

and that (ii) our HTE-Padé data are quite accurate down to temperatures $T/s(s+1) \sim 0.5$ ($T \sim 0.4$) for $s = 1/2$ ($s \to \infty$). In particular, the maximum in C present for the $s = 1/2$ model at $T/s(s+1) \sim 0.9$, cf. Refs. 32, 52–57, is correctly described by our Padé approximants. Note, however, that for $s = 1/2$ there are indications for a second low-temperature maximum in $C(T)$ below $T/s(s+1) = 0.1$, see Refs. 32, 53, 54, 56, which is not covered by our HTE approach. Another characteristic features is the shoulder present in $\chi(T)$ for $s = 1/2$ at about $T/s(s+1) = 1$, which is also well described by our Padé approximants.

In Figs. 5 and 6 we compare the $\chi(T)$ (Fig. 5) and $C(T)$ data (Fig. 6) for spin quantum numbers $s = 1/2, 1, \ldots, 7/2, \infty$. The susceptibility data clearly show that all curves for $s > 1/2$ form a narrow bundle in the temperature range accessible by our approach. Only for $s = 1/2$ the $\chi(T)$ curve is out of this bundle. Hence, one can argue that for $s > 1/2$ quantum effects in χ are almost negligible at normalized temperatures $T/s(s+1) \gtrsim 0.4$. The situation is quite different for the specific heat C, cf. Fig. 6. The maximum in $C(T)$, already mentioned above for $s = 1/2$, is evidently dependent on the spin quantum number s: Its position $T_{max}/s(s+1)$ is shifted to lower normalized temperatures and its height C_{max} increases with growing s. Hence, the quantum effects seem to be important also for quite large values of s. The basic difference in the influence of s on $\chi(T)$ and $C(T)$ can be attributed to an exceptional density of low-lying singlet excitations, see e.g. Ref. 6. These nonmagnetic excitations are irrelevant for χ but important for C. Hence,

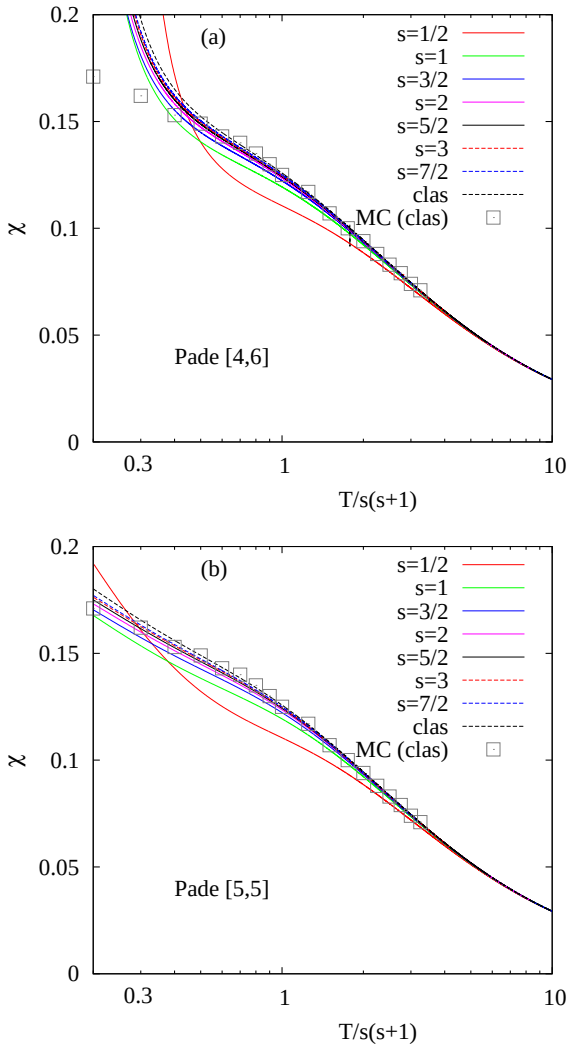

FIG. 5: (Color online) HTE data for the susceptibility χ of the spin-s kagome Heisenberg antiferromagnet, (a) Padé [4,6], (b) Padé [5,5]. For comparison we show the Monte Carlo data (MC) taken from Ref. 20.

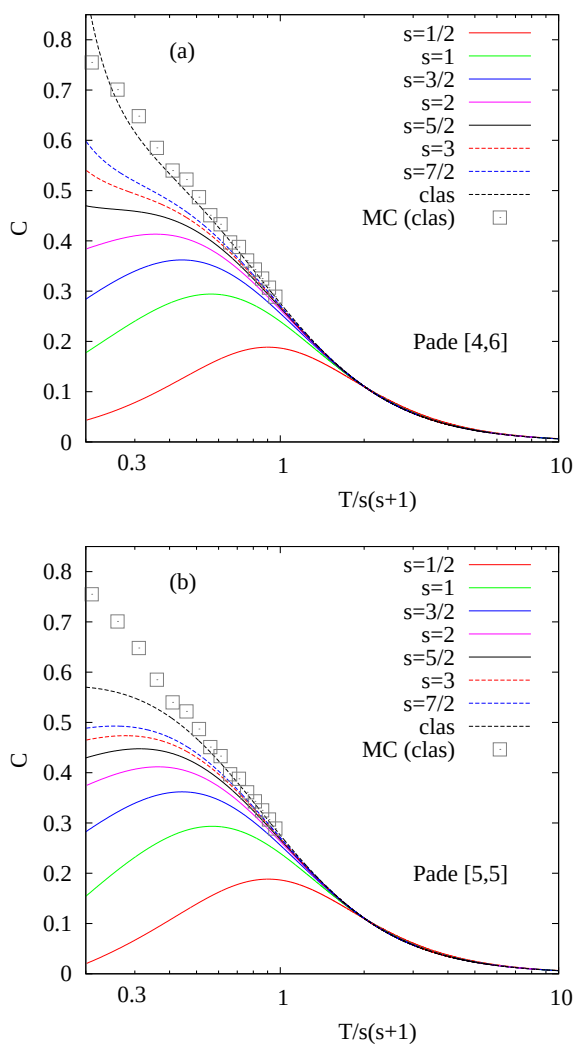

FIG. 6: (Color online) Specific heat C of the spin-s kagome Heisenberg antiferromagnet, (a) Padé [4,6], (b) Padé [5,5]. For comparison we show the Monte Carlo data (MC) taken from Ref. 20.

our HTE-Padé data for $C(T)$ can be understood as an indirect indication for the existence of a unusual large density of low-lying singlet excitation also for $s > 1/2$. We show the position $T_{max}/s(s+1)$ and the height C_{max} as a function of $1/s$ in Fig. 7. From Fig. 6a it is obvious that for the [4,6]-Padé approximant a maximum is exists only for $s < 5/2$. The tendency of how the classical limit is approached is clearly visible from our HTE data. There is indeed a remarkably strong dependence on the spin quantum number. The slope of the corresponding curves shown in Fig. 7 is even increasing when approaching the classical limit $1/s = 0$. Let us mention that for the above discussion of the maximum of the specific heat the 8th order HTE presented in our previous paper[39] is not appropriate, since the corresponding [4,4] Padé approximant exhibits an unphysical pole in the vicinity of the maximum for $s > 1/2$.

IV. CONCLUSIONS

In this paper we present the HTE series up to 10th order of the specific heat C and the uniform susceptibility χ for Heisenberg models with arbitrary exchange patterns $J_{\mu\nu}$ and spin quantum number s. Our HTE scheme is encoded in a C++ program using as input the exchange matrix $J_{\mu\nu}$ and spin quantum number s. Using Padé approximants for the HTE series the scheme can be used to discuss thermodynamic properties of general Heisenberg systems down to moderate temperatures of about $T/s(s+1) \sim 0.4 \ldots 0.5$ and thus for the interpretation of experimental data in rather wide temperature

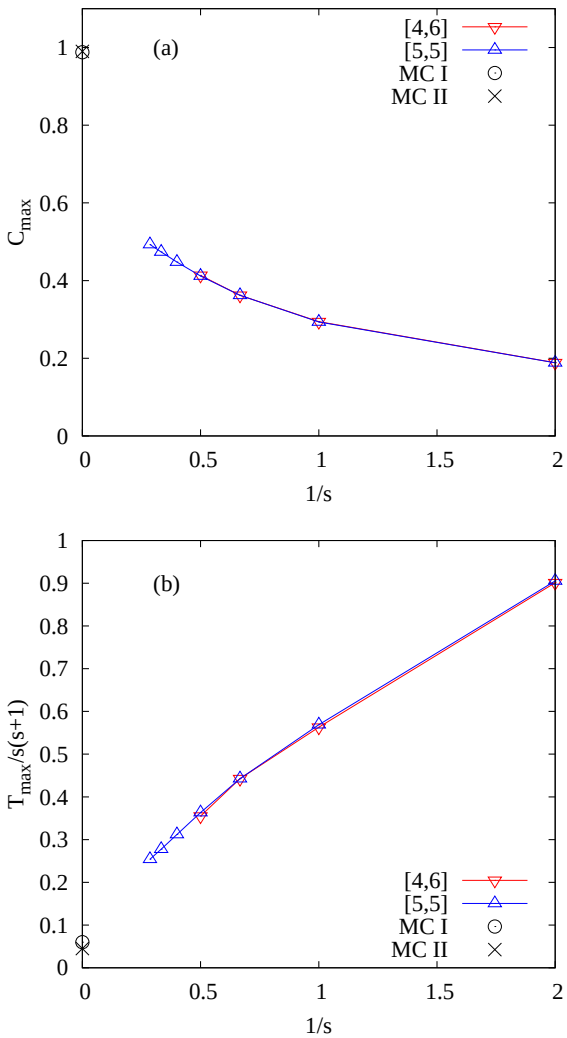

FIG. 7: (Color online) Height C_{max} (a) and position T_{max} (b) of the maximum in the specific heat C in dependence on the inverse spin quantum number $1/s$ of the kagome Heisenberg antiferromagnet. The Monte Carlo results for $s \to \infty$ are taken from Ref. 20 (MC I) and Ref. 21 (MC II). Note, however, that the maximum in $C(T)$ for the classical model is not well-pronounced, rather there is a fairly broad region of high values of C.[21]

range, especially if other precise methods such as the quantum Monte Carlo method or the finite-temperature density matrix renormalization group approach are not applicable. We apply our scheme to the 3D pyrochlore ferromagnet to calculate the Curie temperature T_c in dependence on the spin quantum number s. Comparing T_c of the pyrochlore ferromagnet with corresponding values for the simple-cubic ferromagnet we find that the triangular configuration of bonds present in the pyrochlore lattices leads to a noticeable lowering of T_c. Using our HTE scheme for the kagome antiferromagnet we discuss the influence of s on the temperature dependence of C and χ. While the effect of s on χ in the accessible temperature range is rather weak, there is a well-pronounced shift of the maximum in the temperature dependence of specific heat to lower renormalized temperatures $T/s(s+1)$ when increasing s.

Acknowledgment

JR thanks A. Hauser and M. Maksymenko for fruitful discussions.

Appendix A: The high-temperature expansion for the susceptibility and the specific heat for the Heisenberg model on the pyrochlore lattice

The general formulas for the susceptibility and the specific heat for the Heisenberg model on the pyrochlore lattice with NN exchange constant J up to 8th order can be found in Ref. 39. For the sake of consistency with this reference we have set $\beta = \frac{1}{k_B T}$ in appendix A and B which is slightly different from the definition in section II. The formulas for the 9th and 10h order read for the susceptibility

$$\chi(\beta) = \frac{N}{J}\sum_{n=1}^{\infty} c_n (J\beta)^n \tag{A1}$$

$$c_9 = \frac{1}{5143824000} r^2(-2710665 + 142840908r - 2195288001r^2 + 14497581366r^3 - 45972407664r^4 + 77794619872r^5 - 82650432896r^6 + 46730617088r^7)$$

$$c_{10} = -\frac{1}{169746192000} r^2(51519240 - 2994073848r + 51386055291r^2 - 396940170060r^3 + 1579391570694r^4 - 3442568263344r^5 + 4692701814464r^6 - 4374573206272r^7 + 2124654831616r^8).$$

and for the specific heat

$$C(\beta) = Nk \sum_{n=2}^{\infty} d_n (J\beta)^n \tag{A2}$$

$$d_9 = -\frac{1}{285768000} r^2 (-1807110 + 91861560r - 1255862151r^2 + 6268644864r^3 - 8882615472r^4 - 1691186688r^5 - 21317760r^6 + 1042017280r^7)$$

$$d_{10} = -\frac{1}{6286896000} r^2 (-25759620 + 1451298330r - 22610800701r^2 + 142189820847r^3 - 349296723134r^4 + 154955752848r^5 + 102919717624r^6 + 82927576960r^7 - 11100907520r^8),$$

where $r = s(s+1)$.

Appendix B: The high-temperature expansion for the susceptibility and the specific heat for the Heisenberg model on the kagome lattice

The general formulas for the Heisenberg model on the kagome lattice with NN exchange constant J read for the susceptibility

$$\chi(\beta) = \frac{N}{J} \sum_{n=1}^{\infty} c_n (J\beta)^n \tag{B1}$$

$$c_1 = \frac{1}{3} r$$

$$c_2 = -\frac{4}{9} r^2$$

$$c_3 = \frac{1}{9} r^2 (-1 + 4r)$$

$$c_4 = -\frac{4}{405} r^2 (3 - 28r + 37r^2)$$

$$c_5 = \frac{1}{4860} r^2 (-45 + 702r - 1892r^2 + 1328r^3)$$

$$c_6 = -\frac{1}{510300} r^2 (1728 - 35946r + 164289r^2 - 207896r^3 + 102576r^4)$$

$$c_7 = \frac{1}{6123600} r^2 (-8694 + 218916r - 1401381r^2 + 2888772r^3 - 2251248r^4 + 909184r^5)$$

$$c_8 = -\frac{1}{22963500} r^2 (15390 - 446256r + 3538764r^2 - 10535337r^3 + 12202552r^4 - 7318640r^5 + 2416640r^6)$$

$$c_9 = \frac{1}{7715736000} r^2 (-2710665 + 87954822r - 807482331r^2 + 3091042674r^3 - 5118502560r^4 + 4009481184r^5 - 2113197952r^6 + 518354176r^7)$$

$$c_{10} = -\frac{1}{1273096440000} r^2 (257596200 - 9180862110r + 93799827171r^2 - 426255134022r^3 + 931126345494r^4 - 977085756168r^5 + 621427831616r^6 - 280517703040r^7 + 48779713280r^8)$$

and for the specific heat

$$C(\beta) = Nk \sum_{n=2}^{\infty} d_n (J\beta)^n \tag{B2}$$

$$d_2 = \frac{2}{3}r^2$$
$$d_3 = -\frac{1}{9}r^2(-3+4r)$$
$$d_4 = -\frac{2}{45}r^2(-3+23r+3r^2)$$
$$d_5 = \frac{1}{162}r^2(9-126r+116r^2+48r^3)$$
$$d_6 = -\frac{1}{68040}r^2(-1728+33426r-102969r^2-19464r^3+2144r^4)$$
$$d_7 = -\frac{1}{97200}r^2(-1242+29556r-150039r^2+96676r^3+64544r^4+20992r^5)$$
$$d_8 = \frac{1}{1093500}r^2(7695-213084r+1435806r^2-2537523r^3-539132r^4+58400r^5+186680r^6)$$
$$d_9 = \frac{1}{214326000}r^2(903555-28196370r+227949579r^2-634514526r^3+285950568r^4+230120832r^5$$
$$+135526080r^6+14890240r^7)$$
$$d_{10} = -\frac{1}{18860688000}r^2(-51519240+1775187630r-16326321219r^2+59250202038r^3-69170925596r^4$$
$$-15707506528r^5-728311984r^6+9196378240r^7+3884989440r^8).$$

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