

# High-temperature expansion for frustrated magnets: Application to the $J_1$ - $J_2$ model on the BCC lattice

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## Abstract

We present the high-temperature expansion up to 11th order for the specific heat  $C$  and the uniform susceptibility  $\chi_0$  and up to 9th order for the structure factor  $S_{\mathbf{Q}}$  of the frustrated spin-half  $J_1$ - $J_2$  Heisenberg model on the BCC lattice. We consider ferromagnetic as well as antiferromagnetic nearest-neighbor exchange  $J_1$  and frustrating antiferromagnetic next-nearest-neighbor exchange  $J_2$ . We discuss the influence of frustration on the temperature dependence of these quantities. Furthermore, we use the HTE series to determine the critical temperature  $T_c$  as a function of the frustration parameter  $J_2$ .

*Keywords:* quantum magnetism, frustration, structure factor, high-temperature expansion

## 1 Introduction

Magnetic systems with strong frustration are currently in the focus of active theoretical and experimental research [1, 2].  $J_1$ - $J_2$  Heisenberg models, i.e., models with competing nearest-neighbor (NN) exchange coupling  $J_1$  and next nearest-neighbor (NNN) exchange coupling  $J_2$  can serve as canonical systems to study the interplay of quantum effects, thermal fluctuations and frustration. On bipartite lattices, such as the square or the BCC lattices, the strength of frustration  $J_2/J_1$  can be continuously tuned (where the limits  $J_2 = 0$  and  $J_2 \rightarrow \infty$  represent frustration-free systems), thus allowing to study frustration driven effects in some detail. The corresponding  $J_1$ - $J_2$  Heisenberg Hamiltonian is given by

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{[i,j]} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where  $(\mathbf{S}_i)^2 = s(s+1)$ , and  $\langle i,j \rangle$  denotes NN and  $[i,j]$  denotes NNN bonds. For antiferromagnetic (AFM) NNN bonds,  $J_2 > 0$ , the spin system is frustrated irrespective of the sign of  $J_1$ . The frustration present in the model makes the theoretical treatment of the model challenging. There are several investigations on the ground state properties of two-dimensional (2D)

[3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16] and three-dimensional (3D) [17, 18, 19, 20, 21]  $J_1$ - $J_2$  model. From these studies it became evident that the 2D and 3D models behave differently. From numerous studies of the 2D model with AFM  $J_1$  it is now clear that there is an intermediate non-magnetic quantum phase separating the two semiclassical magnetically ordered ground-state phases, see e.g. Refs. [5, 6, 7, 8, 9, 10, 11, 12]. However for ferromagnetic (FM)  $J_1$  there is a controversial discussion on the existence of such a non-classical intermediate phase [13, 14, 15, 16]. On the other hand, for the corresponding 3D BCC model, there are strong arguments that there is a direct first-order transition at  $J_2 = J_2^c$  between the two magnetically ordered phases present for small (i.e.,  $J_2 < J_2^c$ ) and large (i.e.,  $J_2 > J_2^c$ ) values of  $J_2$  [17, 18, 19, 21]. Much less studied are the finite-temperature properties of these models. In case of strong frustration the quantum Monte Carlo approach is not applicable.[22] Therefore, reliable theoretical data for strongly frustrated quantum spin systems are notoriously rare. Bearing in mind the very active experimental research in the field of frustrated quantum magnetism [1] theoretical methods to calculate thermodynamic properties of frustrated magnets are highly desirable.

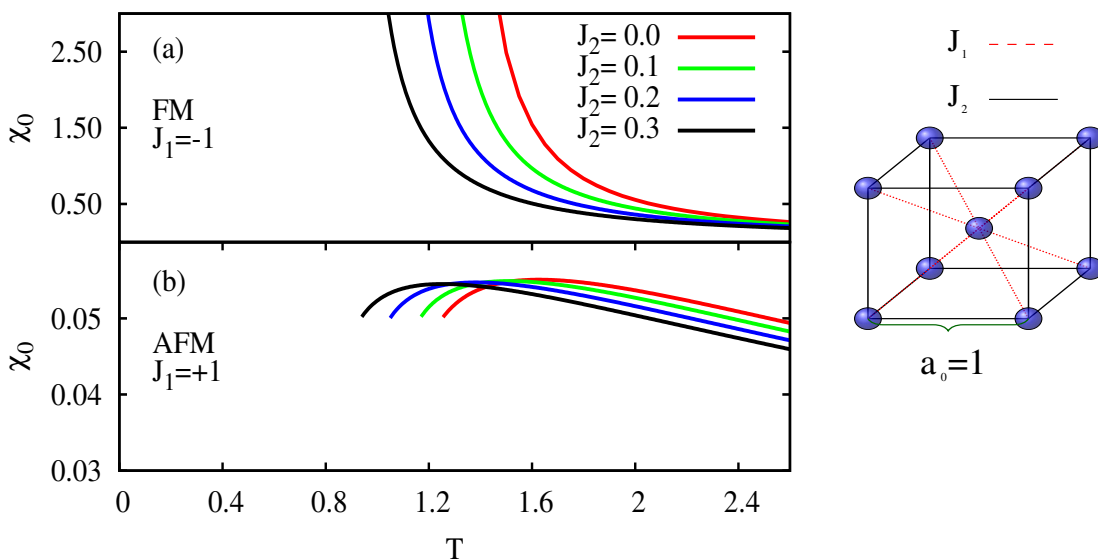


Figure 1: Left: Uniform susceptibility  $\chi_0$  as a function of the temperature  $T$  in the FM (a) and the AFM (b) regime for  $|J_1| = 1$  and different values of the frustrating interaction  $J_2$ . Right: Illustration of the  $J_1$ - $J_2$  model on the BCC lattice.

A universally applicable method to determine thermodynamic quantities for magnetic systems (including frustrated ones) is the high-temperature expansion (HTE). In 1950ies and 1960ies this method was developed and then widely applied to various Heisenberg magnets, see Ref. [23] and references therein. An enormous progress could be achieved in the application of the HTE over the last 20 years by using computer algebraic tools, see e.g. [24, 25, 26, 27, 28, 29]. Very recently the present authors have published a HTE algorithm (encoded as a C++ program available at URL <http://www.uni-magdeburg.de/jschulen/HTE/>) to calculate the HTE series of the uniform susceptibility and the specific heat for general spin- $s$  Heisenberg models up to 10th order. In [28] we have demonstrated that the susceptibility and the specific-heat data obtained from the 10th order HTE can be used to discuss thermodynamic properties of

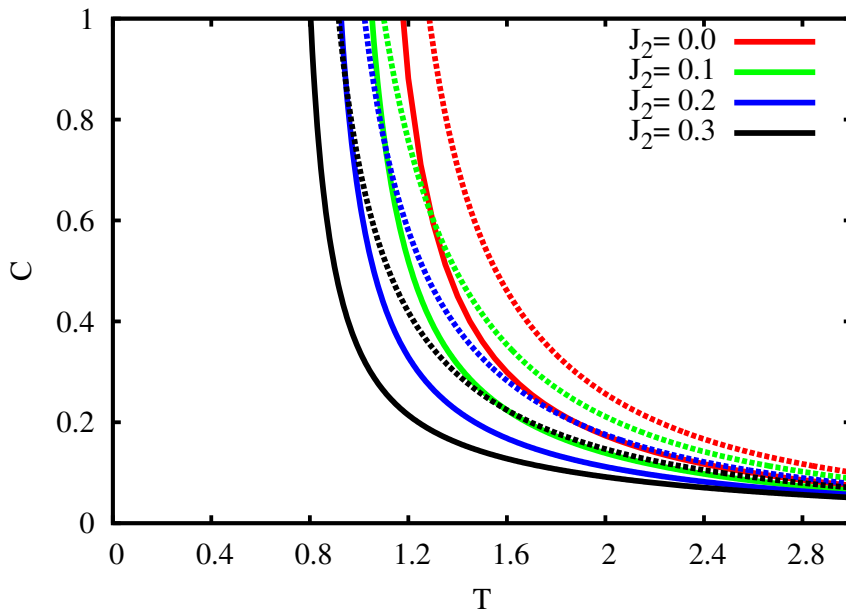


Figure 2: Specific heat  $C$  as a function of temperature  $T$  in the FM ( $J_1 = -1$ , solid lines) and the AFM ( $J_1 = +1$ , dashed lines) regime for different values of  $J_2$ .

Heisenberg systems down to moderate temperatures of about  $T/s(s+1) \sim 0.4 \dots 0.5J$ .

In the present paper we apply our HTE algorithm to investigate the thermodynamics of the  $J_1$ - $J_2$  Heisenberg model, see Eq. (1), on the BCC lattice and we compare the models with AFM NNN and FM NN  $J_1$ . A sketch of the model is given in the right panel of Fig. 1. We focus on spin quantum number  $s = 1/2$ . In 3D models there is typically a conventional phase transition from the high-temperature paramagnetic phase with magnetic short-range order to the low-temperature phase with magnetic long-range order. We focus here on the paramagnetic regime, i.e.  $T \geq T_C$ , which is accessible by HTE, and we discuss the influence of the frustration on the main thermodynamic quantities, such as the uniform susceptibility  $\chi_0(T)$ , the specific heat  $C(T)$ , the structure factor  $S_{\mathbf{Q}}(T)$  and (last but not least) the critical temperature  $T_c$ , where the phase transition takes place.

## 2 Uniform susceptibility and specific heat

We start with the discussion of the uniform susceptibility  $\chi_0$  and the specific heat  $C$ . For that we can use the HTE algorithm presented in [28]. For the specific lattice at hand we have extended the HTE to order eleven. It is well-known that Padé approximants of the HTE series extend the region of validity of the HTE series down to lower temperatures  $T$  [23]. The Padé approximants in form of ratios of two polynomials  $[m, n] = P_m(x)/R_n(x)$  of degree  $m$  and  $n$  provide an analytic continuation of a function  $f(x)$  given by a power series. Since typically approximants with  $m \sim n$  provide best results, we use here either the  $[5, 6]$  or the  $[6, 5]$  approximants, where we choose that approximant that does not have unphysical poles for temperatures in the region of interest. The Padé approximant  $[6, 5]$  for the uniform susceptibility

$\chi_0$  is shown in Fig. 1. For FM  $J_1 = -1$ , we find the characteristic divergence of  $\chi_0$  at a critical temperature (Curie temperature)  $T_c$ . We will use the critical behavior in Sec. 4 to determine  $T_c$  as a function of  $J_2$ . From Fig. 1(a) it is already obvious that the transition is shifted to lower values of  $T$ . For AFM  $J_1 = +1$  the susceptibility  $\chi_0$  exhibits a maximum. This maximum is related to the critical (Néel) temperature. However, the HTE is not able to reproduce the expected kinklike shape of the maximum in  $\chi_0$ . Hence the position  $T_{\max}$  may yield only a crude estimate of the Néel temperature, see Sec. 4. Nevertheless, the effect of the frustrating bond  $J_2$  is clearly seen. To get a more reliable determination of the Néel temperature by a HTE series only the divergence of the staggered susceptibility, or, alternatively, of the structure factor is appropriate, see Ref. [18, 30] and also the next section.

Next we discuss the specific heat  $C$ . It is known that  $C$  exhibits a cusplike singularity at  $T_c$ , but no divergence, see, e.g., [31, 32, 33]. The Padé approximant [5, 6] for  $C$  is shown in Fig. 2. For both regimes,  $J_1 = -1$  and  $J_1 = +1$ , specific heat behaves quite similar. The drastic upturn corresponds to the expected cusplike singularity. This upturn takes place at higher temperatures for AFM  $J_1$  than for FM  $J_1$ , thus yielding a first indication that the Néel temperature is larger than the Curie temperature [30, 34].

### 3 Structure factor

As already mentioned above a reliable determination of the Néel temperature requires a HTE series of the staggered susceptibility or the structure factor at the magnetic wave vector of the Néel state  $\mathbf{Q}_N$ . Both quantities, the staggered susceptibility,  $\chi_{\mathbf{Q}_N}$ , and the structure factor,  $S_{\mathbf{Q}_N}$ , behave quite similarly. Approaching a critical point from above, i.e.  $T \rightarrow T_c^+$ , one has  $\chi_{\mathbf{Q}_N} \sim \beta S_{\mathbf{Q}_N}$  [36]. Since the staggered susceptibility is not directly accessible in experiments we prefer here to use the structure factor defined by

$$S_{\mathbf{Q}} = \frac{1}{N} \sum_{n,m} \cos(\mathbf{Q}(\mathbf{r}_n - \mathbf{r}_m)) \langle S_n^z S_m^z \rangle \quad (2)$$

which can be measured by neutron-scattering experiments. We use here as basis vectors of the direct lattice  $\mathbf{a}_i = \frac{1}{2}(-1, 1, 1); \frac{1}{2}(1, -1, 1); \frac{1}{2}(1, 1, -1)$ , cf. the right panel of Fig. 1, and of the reciprocal lattice  $\mathbf{b}_i = 2\pi(0, 1, 1); 2\pi(1, 0, 1); 2\pi(1, 1, 0)$ . Then the magnetic wave vector  $\mathbf{Q}_M$  of FM state is  $\mathbf{Q}_M = \mathbf{Q}_F = (0, 0, 0) = (2\pi, 2\pi, 0)$  and of the AFM Néel state is  $\mathbf{Q}_M = \mathbf{Q}_N = (2\pi, 2\pi, 2\pi)$ , cf. Ref. [20]. To get  $S_{\mathbf{Q}}$  we have extended our HTE code to calculate the spin-spin correlation functions  $\langle S_n^z S_m^z \rangle$  entering  $S_{\mathbf{Q}}$  up to 9th order (that corresponds to 10th order for the susceptibility).

We show our 9th-order HTE data for  $S_{\mathbf{Q}}$  at  $T = 2$  in Fig. 3. The well-pronounced maximum at the corresponding magnetic wave vector  $\mathbf{Q}_M$  is clearly seen. An increase of  $J_2$  leads to a suppression of the maximum, thus indicating the weakening of magnetic short-range order by frustration. The divergence of  $S_{\mathbf{Q}_M}$  as approaching the critical temperature is shown in the insets of Figs. 3(a) and (b). Another interesting feature of  $S_{\mathbf{Q}}$  is also visible in Figs. 3(a) and (b): Although the frustration leads to a decrease of  $S_{\mathbf{Q}_M}$ , we find an increase of  $S_{\mathbf{Q}}$  at  $\mathbf{Q} = \mathbf{Q}_C = (\pi, \pi, \pi)$ . This increase of  $S_{\mathbf{Q}_C}$  with growing  $J_2$  is a precursor of the so-called collinear AFM phase present for  $J_2 > J_2^c$ . From previous studies [17, 18, 19, 21] it is known that for  $T = 0$  the critical  $J_2^c$  for  $s = 1/2$  is close its classical value  $J_2^{c,\text{clas}} = 2/3$ , i.e. the maximum frustration  $J_2 = 0.5$  used in Fig. 3 is still noticeably below  $J_2^c$ .

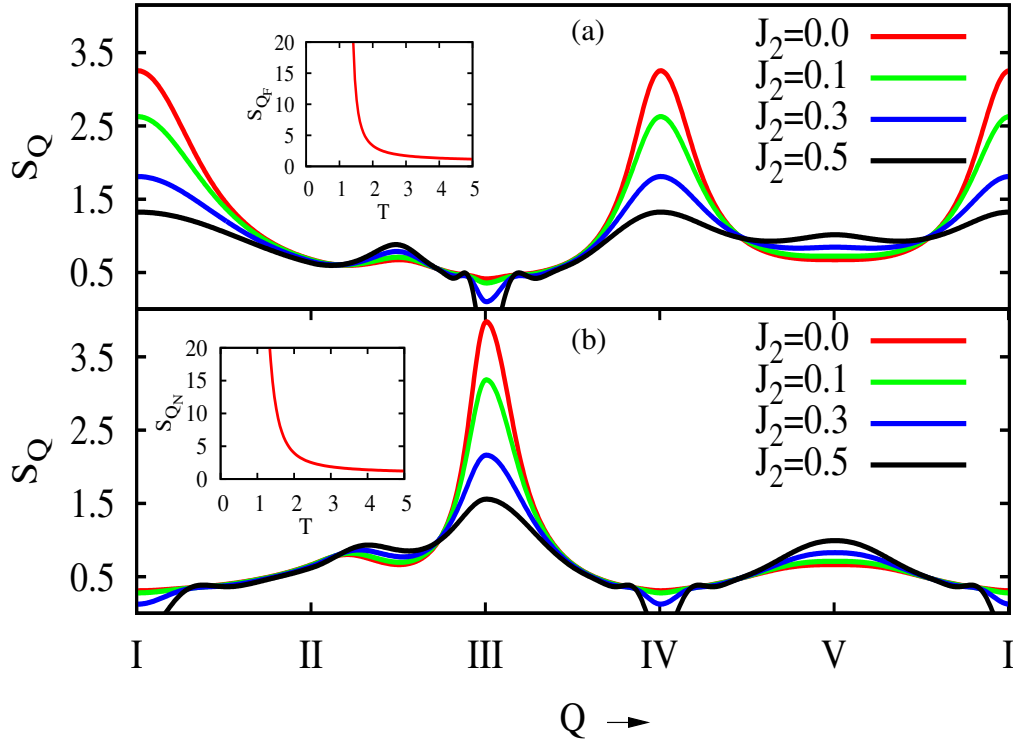


Figure 3: Structure factor  $S_{\mathbf{Q}}$  as a function of the wave vector  $\mathbf{Q} = (Q_x, Q_y, Q_z)$  for different values of  $J_2$  along several paths between characteristic points (I,II, III, IV,V) in the Brillouin zone (I=  $(0, 0, 0)$ ; II=  $(0, 0, \pi)$ ; III=  $(2\pi, 2\pi, 2\pi)$ ; IV=  $(2\pi, 2\pi, 0)$ ; V=  $(\pi, \pi, \pi)$ ). The temperature is set to  $T = 2$ . Inset: Height of the the maximum in  $S_{\mathbf{Q}}$  at the corresponding magnetic wave vector  $\mathbf{Q} = \mathbf{Q}_M$  in dependence on  $T$  for  $J_2 = 0$ . (a): FM regime ( $J_1 = -1$ ),  $\mathbf{Q}_M = \mathbf{Q}_F = (0, 0, 0)$ . (b): AFM regime ( $J_1 = +1$ ),  $\mathbf{Q}_M = \mathbf{Q}_N = (2\pi, 2\pi, 2\pi)$ .

## 4 The critical temperature $T_c$

We use the HTE series for the structure factor  $S_{\mathbf{Q}}$  at  $\mathbf{Q} = \mathbf{Q}_M$  to calculate the critical temperature  $T_c$ . Assuming critical behavior of the form  $S_{\mathbf{Q}_M} \propto (T - T_c)^{-\epsilon}$ , we can use the well-elaborated technique of the so-called differential approximants (DA) to extract  $T_c$  from the HTE series of  $S_{\mathbf{Q}_M}$ , for details see [21, 37, 38]. In Fig. 4 we compare our results with data obtained by a second-order Green's function approach (GFA) [21, 35] as well as data obtained by analyzing the HTE series of the staggered susceptibility [18]. We notice that all approaches yield similar results. There is a significant reduction of the critical temperature  $T_c$  by frustration. Likely, and as indicated by the GFA data,  $T_c(J_2)$  goes to zero as  $J_2$  approaches  $J_2^c$ . On the other hand, there is a noticeable difference between the GFA and HTE data, whereas our HTE data obtained from the structure factor agree reasonably well with those of [18] obtained from

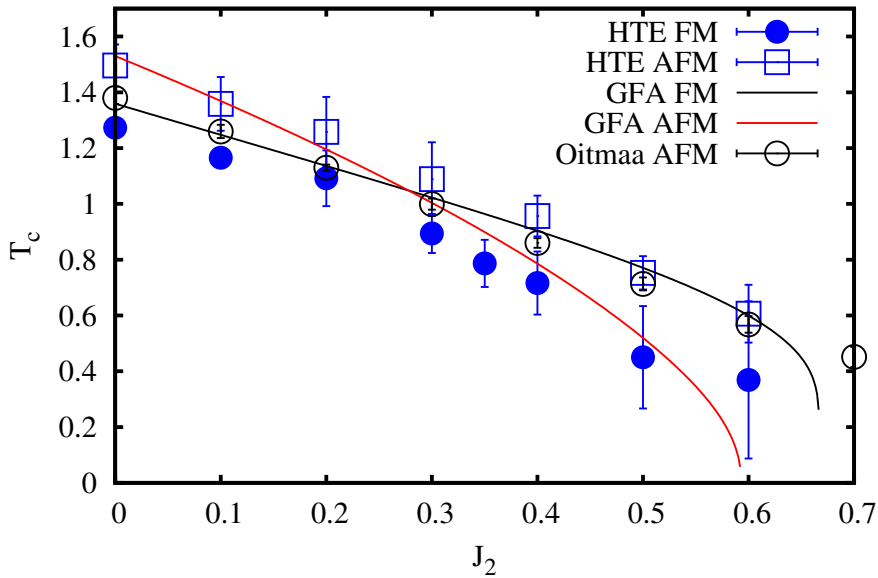


Figure 4: Critical temperatures  $T_c$  as a function of the frustrating NN coupling  $J_2$ . Our data are labeled by 'HTE AFM' and 'HTE FM', the other labels correspond to data taken from [18] ('Oitmaa AFM'), [21] ('GFA FM') and [35] ('GFA AFM'). The labels 'FM and 'AFM' correspond to  $J_1 = -1$  and  $J_1 = +1$ , respectively.

the staggered susceptibility. From a previous comparison between HTE and Monte-Carlo data for unfrustrated systems [28] we may expect that in the regime of weak frustration the HTE data for  $T_c$  are more accurate than the GFA data. Another point concerns the comparison of the Curie temperature  $T_C$  with the Néel temperature  $T_N$ . For unfrustrated Heisenberg magnets one finds the relation  $T_N > T_C$  [30, 34]. By contrast to the GFA results, we find that our HTE data obey this relation in the entire region up to  $J_2 \sim 0.6$  accessible by the HTE approach. We mention that the position  $T_{\max}$  of the susceptibility maximum for AFM  $J_1 = +1$ , cf. Fig. 1(a) in Sect.2, is indeed only about 10% larger as our  $T_N$  values presented in Fig. 4.

## 5 Summary

We have applied the high-temperature expansion (HTE) in high orders to investigate the thermodynamic quantities of frustrated spin-half  $J_1$ - $J_2$  Heisenberg model on the BCC lattice in the short-range ordered phase at  $T \geq T_c$ . We consider FM as well as AFM NN exchange  $J_1$ . The main focus is on the influence of the frustrating AFM NNN coupling  $J_2$  on the specific heat, the uniform susceptibility, the structure factor as well as the critical temperature  $T_c$ . The presented data can be used to get information on the ratio  $J_2/J_1$ , e.g., from susceptibility measurements. Moreover, our data for the critical temperature as a function of  $J_2$  also provide an information on this ratio. Interestingly, the wave-vector dependence of the structure factor, although calculated for  $T > T_c$ , shows some indications of the zero-temperature quantum phase transition at  $J_2 = J_2^c$ , if  $J_2$  becomes sufficiently close to  $J_2^c$ . The present investigations are focused on

theoretical aspects, however, there might be some relevance for FM compounds [39, 40].

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