Phase Diagram of the BCC $S = \frac{1}{2}$ Heisenberg Antiferromagnet with First and Second Neighbor Exchange

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We use linked-cluster series expansions, both at $T = 0$ and high temperature, to analyse the phase structure of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet with competing first and second-neighbor interactions on the 3-dimensional body-centred-cubic lattice. At zero temperature we find a first-order quantum phase transition at $J_2/J_1 \simeq 0.705 \pm 0.005$ between AF$_1$ (Néel) and AF$_2$ ordered phases. The high temperature series yield quite accurate estimates of the bounding critical line for the AF$_1$ phase, and an apparent critical line for the AF$_2$ phase, with a bicritical point at $J_1/J_2 \simeq 0.71$, $kT/J_1 \simeq 0.34$. The possibility that this latter transition is first-order cannot be excluded.

I. INTRODUCTION

The occurrence of competing exchange interactions in magnetic materials can give rise to a rich variety of magnetic ordered states, and of phase transitions between them. Studies of such phenomena, within the “molecular” or “mean-field” approximation go back half a century or more. It is perhaps surprising that open questions remain, but, at least for quantum models, this is the case.

We study in this paper, the spin-$\frac{1}{2}$ Heisenberg antiferromagnet on the body-centered-cubic (bcc) lattice, with first- and second-neighbor interactions. The Hamiltonian is

$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$  \hspace{1cm} (1)$$

where the summations are over first and second-neighbor pairs respectively and the exchange constants $J_1, J_2 > 0$. For our purposes we need to divide the structure into four interpenetrating sublattices, which we denote $A_1, A_2, B_1, B_2$ and illustrate in Fig. 1(a). Each of these sublattices has a face-centered-cubic (fcc) structure. The $J_1$ interactions couple the $A$ and $B$ sublattices, while the interactions $J_2$ couple $A_1$ to $A_2$, and $B_1$ to $B_2$ only.

The first question to ask concerns the ground state, at $T = 0$. For classical vector spins it is easy to show that this will be the Néel or AF$_1$ phase for $J_2/J_1 < 2/3$, and the AF$_2$ phase for $J_2/J_1 > 2/3$. In the AF$_1$ phase all $A$ spin point in the direction of an arbitrary unit vector $\mathbf{n}$ while $B$ spins point in the opposite direction $-\mathbf{n}$. In the AF$_2$ phase each of the $A$ and $B$ sublattice is itself Néel ordered. Thus we may choose $A_1$ spins in direction $\mathbf{n}$, $A_2$ spins in direction $-\mathbf{n}$, $B_1$ spins in direction $\mathbf{n}'$ and $B_2$ spins in direction $-\mathbf{n}'$, with $\mathbf{n}$ and $\mathbf{n}'$ completely independent. In the quantum case these will not be exact eigenstates but, as usual in quantum antiferromagnetism, will be modified by quantum fluctuations. Furthermore in the AF$_2$ phase the direction $\mathbf{n}'$ will be locked to either $\pm \mathbf{n}$, leading to a discrete 2-fold Ising symmetry added to the O(3) spin symmetry.

This model has similarities to the so-called “$J_1 - J_2$ model” on the square lattice, which has been much studied in recent years. In that case there is now strong evidence for an intermediate “spin-liquid” phase between the Néel and AF$_2$ (the “collinear phase”), with the possibility of an even richer structure. Motivated by this similarity, Schmidt et al. have studied the bcc model at $T = 0$, using exact diagonalizations and linear spin-wave theory. They
find a direct first-order quantum phase transition between AF$_1$ and AF$_2$ phase at $J_2/J_1 \simeq 0.7$, with no intermediate phase. This can be understood in terms of the diminishing effect of quantum fluctuations in the 3-dimensional system.

In Section 2 we use linked-cluster series expansions at $T = 0$ to study ground state properties of the model. We obtain rather precise results for both the ground state energy and order parameters, confirming the scenario of Ref. [5] and obtaining a somewhat more precise estimate of the quantum phase transition point.

A second interesting aspect of the model is the phase structure at finite temperatures. There will be transition lines in the $(T, J_2/J_1)$ plane where the low-temperature ordered phases meet the high-temperature disordered phase, and the locations and nature of these transition lines are not well known. The earliest attempt to address these questions, beyond mean-field theory, was the 6th-order high-temperature series work of Pirnie et al.$^6$. They obtained series for the appropriate staggered susceptibilities for both AF$_1$ and AF$_2$ ordering for the $J_1 - J_2$ bcc lattice (for general spin), and estimated critical temperatures via Dlog Padé approximants, in the usual way. They were unable to obtain precise values for the critical exponent $\gamma$. More recently Pan$^7$ has computed an 8th-order staggered susceptibility series for the Néel phase for the case $J_2 = 0$, and obtained the critical temperature as $kT_c/J_1 = 1.384 \pm 0.005$. (This is actually a thermodynamic perturbation expansion, which can generate a high-temperature series but can also used to study properties in the ordered phase).

In Section 3 we report on the derivation and analysis of new extended high temperature series for this system. In particular we have computed staggered susceptibilities for the full $J_1 - J_2$ model to 10th order in the AF$_1$ phase and 9th order in the AF$_2$ phase, adding 4 and 3 terms respectively to the work of Ref. [6]. These longer series allow reasonably precise estimates to be made of both critical temperature and exponents. There is one caveat. There are arguments that the AF$_2$ to paramagnetic transition is first-order. If that is indeed the case then our series are presumably seeing a spinodal line within the ordered phase, and the actual transition temperature is higher.

The arguments for a first-order transition come from two sources. Firstly for the Ising version of this system both Monte Carlo simulations$^8$ and a combination of high and low-temperature series$^9$ support, fairly conclusively, the existence of a “fluctuation-induced” first-order transition. Secondly, a renormalization group treatment$^8,10$ of the appropriate $n = 6$ component Landau-Ginzburg-Wilson Hamiltonian for the AF$_2$ transition has no stable fixed point, suggesting a first-order transition. However this analysis is based on an $\epsilon$-expansion about $d = 4$ and, in our view, while persuasive, is not conclusive.
Fig. 2: Ground state energy versus $J_2/J_1$ from Ising expansions for the AF$_1$ and AF$_2$ phases. The crossing point at $0.705\pm0.005$ identifies the first-order quantum phase transition. Uncertainties in the series extrapolation are no larger than the symbols. Lines joining symbols are guides to the eye.

II. EXPANSIONS AT $T = 0$

Our approach is to use high-order linked-cluster expansions, where the Hamiltonian is written as

$$ H = H_0 + xV $$

where $H_0$ is the Ising Hamiltonian

$$ H_0 = J_1 \sum_{\langle ij \rangle} \langle 1 \rangle S_i^z S_j^z + J_2 \sum_{\langle ij \rangle} \langle 2 \rangle S_i^z S_j^z $$

and

$$ V = \frac{1}{2} J_1 \sum_{\langle ij \rangle} \langle 1 \rangle (S_i^+ S_j^- + S_i^- S_j^+) + \frac{1}{2} J_2 \sum_{\langle ij \rangle} \langle 2 \rangle (S_i^+ S_j^- + S_i^- S_j^+) $$

is treated as a perturbation. Quantities are expanded as power series in $x$, and then evaluated at the isotropic limit $x = 1$, by Padé and differential approximants. We have used this technique successfully in many previous studies of quantum antiferromagnets. In the present work we are able to compute series through $x^8$ in both Néel and AF$_2$ region.

Figure 2 shows the ground state energy versus exchange ratio $J_2/J_1$, the two branches corresponding to the two phases which can occur. The results are very similar to those of Ref. obtained from extrapolation of exact diagonalization results. The two branches clearly cross, confirming that the ground state transition between AF$_1$ and AF$_2$ phases is first-order. Our estimated transition point is $J_2/J_1 = 0.705 \pm 0.005$. 
FIG. 3: Staggered magnetizations (order-parameters) and correlator discriminants (Eq. 5) versus $J_2/J_1$, in both the AF$_1$ and AF$_2$ phases. $\Delta C_1$, $\Delta C_2$ refer to first and second neighbour correlators respectively. In the AF$_2$ phase the superscripts $++$ and $+-$ refer to first neighbour correlators for sites with like spins and unlike spins. The thin vertical line denotes the transition. The lines joining symbols are guides to the eye.

We have also computed series for two other physical quantities: staggered magnetization in each phase $M$, and the correlator discriminant

$$\Delta C = |3 \langle S^z_i S^z_j \rangle - \langle S_i \cdot S_j \rangle|$$

(5)

which is a measure of the breaking of spin rotation symmetry. These results are shown in Figure 3. We look first at the magnetization. In the Néel phase this starts from $\sim 0.435$ and decreases monotonically to approx. 0.34 at the transition point. In the AF$_2$ phase the curve start from 0.424, the result for the simple cubic lattice, and remains quite flat, with a downward curvature near the transition point. Our results show a small upward curve near $J_2/J_1 \approx 1$, but this is most likely an artifact. Our results are similar to, but considerably more precise than, those of ref. [5].

Results for the correlators are new. Rather than show all correlators, we show in Figure 3 only the quantities $\Delta C$ in Eq. 5 for first and second-neighbor pairs. A nonzero value indicates spontaneous breaking of spin rotation symmetry. Of course our expansion starts from a symmetry-broken state, and is thus biased. However an identical calculation for the $J_1 - J_2$ square lattice clearly shows $\Delta C \to 0$ at the boundary between the Néel and disordered phase. The fact that this does not happen here is clear and strong evidence that the Néel phase does not vanish continuously, but undergoes a first-order transition to the AF$_2$ phase. The second-neighbor discriminant $\Delta C_2$ in the AF$_2$ phase is particularly interesting, showing a small but clearly discernable increase on approaching the transition. This is unexpected.

In summary our $T = 0$ series confirm the occurrence of two phase, AF$_1$ and AF$_2$ with a first-order quantum phase
FIG. 4: Phase diagram of the BCC $J_1 - J_2$ antiferromagnet. The solid line is the AF$_1$-paramagnetic critical line, with universal $n = 3$ exponents. The dashed line is the AF$_2$-paramagnetic transition line, as determined from our series. The short vertical dotted line is the first-order AF$_1$-AF$_2$ transition line, extending from the bicritical point (or critical end-point) to the quantum phase transition point on the $T = 0$ axis.

transition at $J_2/J_1 = 0.705 \pm 0.005$. We now turn to the finite temperature phase structure.

III. HIGH TEMPERATURE EXPANSIONS

The standard approach to determining finite temperature critical points and exponents for quantum (and classical) models is via high-temperature expansions. For the bcc(1,2) antiferromagnet this was first done in Ref. [6]. One includes an appropriate staggered field $F$ in the Hamiltonian

$$ H' = H - F \sum_i \eta_i S_i^z $$

(6)

where $\eta_i = \pm 1$ on the different sublattices, reflecting the type of order expected at low temperatures, expands the partition function

$$ Z = \text{Tr}[e^{-\beta H'}] = \sum_{n=0}^{\infty} \frac{(-\beta)^n}{n!} \text{Tr}((H - FM)^n) $$

(7)

and compute the appropriate staggered susceptibility

$$ \chi = -\frac{\partial^2}{\partial F^2} \left( \frac{1}{N} \ln Z \right) $$

(8)

in the form

$$ \chi = 1 + \sum_{n=1}^{\infty} a_n(\alpha) K^n $$

(9)
where $K = \beta J_1$, $\alpha = J_2/J_1$ and a constant has been factored out. Ref. [6] gives the $a_n(\alpha)$ for both AF$_1$ and AF$_2$ phases through $n = 6$. We have extended these series by 4 terms in the AF$_1$ phase and 3 terms in the AF$_2$ phase. The coefficient are given in Table I. Our results agree fully with previous shorter series for the AF$_1$ phase, but there is a discrepancy at 6th order for the AF$_2$ series. We believe that in the second last row of the last table in Appendix II of Ref. [6] the entry 994944 should be replaced by -2230656. With this substitution we find agreement for both $S = \frac{1}{2}$ and $S = 1$ (for which we have also derived short series).

The series have been analysed via standard Padé approximant methods. Table II shows the critical temperature and exponent estimates obtained from a direct analysis of $\frac{d}{dK} \ln \chi(K)$. As is apparent, for the smaller value of $\alpha = J_2/J_1$, there is a consistent pole and an exponent around 1.4, which is the value expected for the Heisenberg ($n = 3$) universality class. As $\alpha$ increases the series become less regular. This is due to a pole on the negative real axis lying closer to the origin (“ferromagnetic singularity”). Nevertheless the direct analysis is consistent with $\gamma \simeq 1.4$ along the entire line. Assuming this, it is possible to increase the precision in $K_c$. The resulting critical line is shown in Fig. 3. In the AF$_2$ phase the series are less regular and consequently the analysis is less precise. In Table III we show some of the raw analysis results. As is evident the higher-order Padé’s show a consistent physical singularity down to at least $J_2/J_1 = 0.8$, although there is again an interfering singularity on the negative real axis. The exponent estimates show a clear decreasing trend from $J_1/J_2 = 0$ (while corresponds to the Néel phase of the simple cubic lattice, for which we expect a second-order transition with the universal exponent 1.40) for increasing $J_1/J_2$. For example, for $J_1/J_2 = 1.25$ ($J_2/J_1 = 0.8$), the estimated $\gamma < 1$. Now, if the AF$_2$-paramagnetic phase boundary is second-order, we would expect critical exponents appropriate to an $n = 6$ component order parameter. Thus the series will be affected by a crossover from $n = 3$ to $n = 6$, presumably followed by a second crossover to the bicritical point. In a relatively short series this can give rise to varying critical exponents, as observed here. On the other hand there are arguments, mentioned previously, for this transition to be first-order. If that is the case then the transition will lie above our curve in Fig. 4, and the apparent divergence locates a spinodal line within the order phase. The “bicritical point” is then, in fact, a “critical-end point”.

### IV. CONCLUSIONS

We have used a combination of series expansions at $T = 0$ and conventional high-temperature series to elucidate the phase diagram of the quantum $S = \frac{1}{2}$ Heisenberg antiferromagnet on the BCC lattice with nearest and next-nearest-neighbor interactions. The ground state properties of this model have not, to our knowledge, been previously studied by series methods. We identify a first-order quantum phase transition at $J_2/J_1 \simeq 0.705 \pm 0.005$ between the two possible types of antiferromagnetic order. We have extended previous high temperature series by 4 and 3 terms, respectively in the AF$_1$ and AF$_2$ phases. These longer series allow a fairly precise estimate of the AF$_1$-paramagnetic critical line, and confirm $n = 3$ universality along this line. The nature of the AF$_2$-paramagnetic line remains enigmatic. We find a consistent line of poles, which may represent a true critical line. However we cannot exclude the possibility of a first-order transition at somewhat higher temperatures.

In the corresponding Ising case the AF$_2$-paramagnetic transition was shown to be first-order by a careful analysis using both high and low temperature series. The former approach cannot be used here since low temperature series cannot be obtained. It would be of some interest to study this system using quantum Monte Carlo methods, although a “sign problem” would be expected. A thorough Monte Carlo study for the classical Heisenberg system would also appear very worthwhile. To our knowledge no such studies have been attempted.
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TABLE I: Susceptibility polynomials $a_n(\alpha)$ (Eq. 9) for the spin-$\frac{1}{2}$ bcc(1,2) Heisenberg antiferromagnet. To avoid fractions, the values of $2^{n+1}(n+1)!a_n(\alpha)$ are given.

| n=1  | 16, -12 |
| n=2  | 160, -288, 72 |
| n=3  | 2048, -5424, 4032, -528 |
| n=4  | 31096, -110640, 132168, -55680, 4950 |
| n=5  | 557456, -2470992, 4070976, -2947488, 825120, -56196 |
| n=6  | 11495460, -61270752, 127951356, -131155632, 65324256, -13458816, 693273 |
| n=7  | 269007424, -1670678976, 2421885472, -5589739344, 403451024, -1489565472, 238760256, -9630816 |
| n=8  | 7026835032, -4991146560, 148837552176, -240765473760, 226149075384, -123139222944, 35480202336, -4568142528, 156881934 |
| n=9  | 202835216096, -1620119828784, 5556372919872, -10631250963456, 12358650036192, -8857806164208, 380668562496, -888255818640, 9455942080, -2810097960 |
| n=10 | 6406925312668, -56883245890656, 220319073129360, -48724824539392, 674224356164040, -60229832497512, 344178723975168, -120654960988368, 23412924414240, -21163553822240, 52557775149 |

TABLE II: Estimates of the critical temperature $K_c$ and exponent $\gamma$ (in brackets) from Dlog Padé approximants to the Néel phase staggered susceptibility series.

| $J_2/J_1$ | 0.0     | 0.2     | 0.4     | 0.6     |
|-----------|---------|---------|---------|---------|
| [3/4]     | 0.7233(1.391) | 0.8881(1.427) | 1.1614(1.418) | 1.6710(1.117) |
| [4/4]     | 0.7248(1.410) | 0.8839(1.391) | 1.1634(1.429) | 1.7446(1.314) |
| [3/5]     | 0.7251(1.414) | 0.8844(1.396) | 1.1635(1.430) | 1.7706(1.416) |
| [5/4]     | 0.7281(1.459) | 0.8896(1.454) | 1.1704(1.478) | 1.6534(1.087) |
| [4/5]     | 0.7199(1.371) | 0.8859(1.410) | 1.1606(1.415) | 1.6534(1.087) |
| negative pole | -1.0     | -0.8     | -0.68   | -0.6    |
TABLE III: Estimates of the critical temperature $K_c = J_2/kT_c$ and exponent $\gamma$ (in brackets) from Dlog Padé approximants to the AF$_2$ phase staggered susceptibility series.

| $J_1/J_2$ | 0.0   | 0.5   | 1.0   | 1.25  |
|-----------|-------|-------|-------|-------|
| [3/3]     | 1.0218(1.232) | 1.0698(1.299) | –     | –     |
| [2/4]     | 1.0318(1.287) | 1.0709(1.304) | –     | –     |
| [3/4]     | 1.0540(1.404) | 1.0847(1.367) | 1.221(1.232) | 1.387(0.966) |
| [4/4]     | 1.0627(1.461) | 1.0827(1.357) | 1.233(1.282) | 1.369(0.920) |
| [3/5]     | 1.0639(1.472) | 1.0828(1.358) | 1.234(1.288) | 1.370(0.924) |
| negative pole | -1.5 | -0.7 | -0.5 | -0.4 |