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Ordered phases in the frustrated fcc lattice antiferromagnet

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We study the ordered phases of the $S = 1/2$ fcc Heisenberg antiferromagnet, with first- and second-neighbor interactions, using perturbative series expansions at zero temperature. From calculations of ground-state energies we determine the regions of stability of semiclassical AF1, AF2, and AF3 phases. For the pure nearest-neighbor case our results suggest that AF1 is favored over AF3, a result in disagreement with linear spin-wave theory, but in agreement with other more recent work. We also investigate the two possible AF2 phases and locate the crossover between AF3 and AF2 at $J_2/J_1 \sim 0.535$.

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I. INTRODUCTION

Studies of antiferromagnetism in the face-centered-cubic (fcc) Heisenberg model go back to the 1960s or even earlier [1–3]. The geometric frustration inherent in this structure allows the possibility of different ordered states and renders the theoretical analysis difficult. The inclusion of additional second-neighbor exchange interactions, which are expected to play a role in real materials, can add further frustration through competing interactions.

There is a large body of theoretical and experimental work in this field [4–9], including measurement of excitations in the well known materials NiS_2 [10] and MnS_2 [11], as well as the discovery of new materials [12] (and references therein).

Early theoretical studies, mostly based on "effective-field" methods, have led to the identification of a number of "classical" phases, known as AF1, AF2, and AF3 (also termed type-1, type-2, and type-3 in the literature). We show these schematically in Fig. 1. The interaction strengths, which we denote J_1 and J_2, will determine the regions of stability of each of these phases and lead to a phase diagram with possible crossovers between different phases, as shown, for example, in Ref. [7].

In both AF1 and AF3 phases each spin has four like and eight unlike nearest neighbors. Thus, with nearest-neighbor interactions only and in the absence of quantum fluctuations, these classical phases will be degenerate. The lifting of this degeneracy by quantum fluctuations has been studied recently [13], using linear spin-wave theory, with the conclusion that AF3 is favored, although the energy difference per spin is only a few parts in 10^4. More recent calculations [14] have, however, come to the opposite conclusion, which is in accord with our calculations below. In the AF1 phase the second-neighbor pairs are all alike, whereas in AF3 four are alike and two opposite. Thus ferromagnetic J_2 will favor AF1 and antiferromagnetic J_2 will favor AF3. For larger antiferromagnetic J_2 a new phase AF2 or, rather, two phases AF2a and AF2b will be possible. These phases have six like and six unlike nearest neighbors, while all six second neighbors are unlike. Thus, classically, these are degenerate for all J_2. However, it has been argued [8] that quantum fluctuations favor AF2a. The crossover point between AF3 and AF2, classically at $J_2/J_1 = 0.5$, does not appear to have been accurately determined in the quantum case.

The present paper reports studies of the frustrated $J_1 - J_2$ fcc model for the extreme quantum case $S = 1/2$, using series expansion methods [15,16]. Details of the calculation are given in Sec. II and the results presented and discussed in Sec. III. Section IV gives an overall summary.

II. SERIES EXPANSIONS AT ZERO TEMPERATURE

We consider the isotropic Heisenberg model, defined by the usual Hamiltonian

$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle ik \rangle} \mathbf{S}_i \cdot \mathbf{S}_k, \tag{1}$$

where the sums are over first- and second-neighbor bonds, respectively, the $\mathbf{S}$ are quantum spin-$1/2$ operators, and we assume antiferromagnetic $J_1 > 0$. J_2 can be of either sign, although we focus mainly on the case of antiferromagnetic J_2 as well.

The series expansion method at $T = 0$ is based on separating the Hamiltonian in the usual perturbative form $H = H_0 + \lambda V$, where H_0 has a simple known ground state, usually without quantum fluctuations, and V represents the remaining terms. In the present work we use "Ising expansions," in which H_0 consists of the diagonal (zz) terms in the Hamiltonian and V consists of the transverse terms.

We use the "linked cluster" approach [15,16], in which perturbation series are computed for a sequence of connected clusters of increasing size, and these are then combined appropriately to yield series for any extensive quantity, such as the ground-state energy, magnetization, etc. for the bulk lattice. The coefficients of the resulting series, up to an order determined by the clusters with the largest number of sites, are numerically exact.

In carrying out these calculations, we choose H_0 to exhibit a particular type of magnetic order and thus a separate calculation is needed for each candidate phase. We need to distinguish between four bond types, which we label by κ, as

TABLE I. Bond numbers and classical energies of AF1, AF2, and AF3 ground states.

Type κ	AF1	AF2	AF3
1	2N	3N	2N
2	4N	3N	4N
3	3N	0	2N
4	0	3N	N
E_{cl}	$-\frac{1}{2}J_1 + \frac{3}{4}J_2$	$-\frac{3}{4}J_2$	$-\frac{1}{2}J_1 + \frac{1}{4}J_2$

follows:

$$\kappa = 1 \quad \text{1st nhbr like spins} \quad (++), (--),$$
$$\kappa = 2 \quad \text{1st nhbr unlike spins} \quad (+-),$$
$$\kappa = 3 \quad \text{2nd nhbr like spins} \quad (++), (--),$$
$$\kappa = 4 \quad \text{2nd nhbr unlike spins} \quad (+-).$$

Table I gives the number of each bond type in the three ground states AF1, AF2, and AF3 and the classical energy of each.

It is also convenient to transform the spin operators on the "up" sites $(S^x, S^y, S^z) \rightarrow (S^x, -S^y, -S^z)$ to give a uniform unperturbed ground state and to separate out the classical energy for each cluster. The Hamiltonians for each bond type then take the form

$$\kappa = 1, \quad H_0 = -J_1 \sum_{\langle ij \rangle}^{(1)} \left(\frac{1}{4} - S_i^z S_j^z \right),$$
$$V = \frac{1}{2} J_1 \sum_{\langle ij \rangle}^{(1)} (S_i^+ S_j^- + S_i^- S_j^+),$$
$$\kappa = 2, \quad H_0 = J_1 \sum_{\langle ij \rangle}^{(2)} \left(\frac{1}{4} - S_i^z S_j^z \right),$$
$$V = \frac{1}{2} J_1 \sum_{\langle ij \rangle}^{(2)} (S_i^+ S_j^+ + S_i^- S_j^-),$$

TABLE III. Padé approximant values of energies of AF1 and AF3 phases at $T = 0$. The superscript l denotes results obtained from the logarithmic derivative series.

	AF1		AF3	
	$t = 0$	$t = 0.5$	$t = 0$	$t = 0.5$
[4,6]	−0.7253	−0.7240	−0.7231	−0.7221
[5,5]	−0.7250	−0.7252	−0.7229	−0.7228
[6,4]	−0.7250	−0.7244	−0.7229	−0.7224
[4,5]	−0.7247	−0.7237	−0.7227	−0.7218
[5,4]	−0.7249	−0.7239	−0.7228	−0.7220
[4,4]	−0.7252	−0.7225	−0.7231	−0.7206
$[4,5]^l$	−0.7250	−0.7251	−0.7229	−0.7229
$[5,4]^l$	−0.7252	−0.7237	−0.7230	−0.7218
$[4,4]^l$	−0.7249	−0.7235	−0.7227	−0.7216

$$\kappa = 3, \quad H_0 = -J_2 \sum_{\langle ij \rangle}^{(3)} \left(\frac{1}{4} - S_i^z S_j^z \right),$$
$$V = \frac{1}{2} J_2 \sum_{\langle ij \rangle}^{(3)} (S_i^+ S_j^- + S_i^- S_j^+),$$
$$\kappa = 4, \quad H_0 = J_2 \sum_{\langle ij \rangle}^{(4)} \left(\frac{1}{4} - S_i^z S_j^z \right),$$
$$V = \frac{1}{2} J_2 \sum_{\langle ij \rangle}^{(4)} (S_i^+ S_j^+ + S_i^- S_j^-),$$

where the superscripts on the summations designate the bond type κ.

Another modification which is commonly used to improve convergence of the series is to add and subtract a diagonal term $H_0 \rightarrow H_0 + t\sum_i S_i^z$, $V \rightarrow V - t\sum_i S_i^z$, where t is an adjustable parameter. This is particularly useful in the J_1, J_2 case, where small energy gaps in the spectrum of H_0 could lead to small energy denominators in the perturbation series. We have used this technique, with $t = 0.5$, 1.0. It is of course

TABLE II. Coefficients of AF1 and AF3 energy series for $t = 0.0, 0.5$.

AF1		AF3	
$t = 0.0$	$t = 0.5$	$t = 0.0$	$t = 0.5$
−0.500000000000D+00	−0.500000000000D+00	−0.500000000000D+00	−0.500000000000D+00
0.000000000000D+00	0.000000000000D+00	0.000000000000D+00	0.000000000000D+00
−0.333333333333D+00	−0.250000000000D+00	−0.333333333333D+00	−0.250000000000D+00
0.222222222222D+00	0.625000000000D−01	0.222222222222D+00	0.625000000000D−01
−0.282804232804D+00	−0.600322420635D−01	−0.274933862434D+00	−0.577628968254D−01
0.481946334089D+00	0.572992941390D−01	0.464719702696D+00	0.563404470585D−01
−0.980165915126D+00	−0.777241341673D−01	−0.943220784740D+00	−0.766621778647D−01
0.222522844917D+01	0.111189395904D+00	0.213374470566D+01	0.109697507750D+00
−0.543376663230D+01	−0.173819812516D+00	−0.518390582543D+01	−0.170885483881D+00
0.139765332577D+02	0.282379109908D+00	0.132639837657D+02	0.277344869098D+00
−0.374005417279D+02	−0.478771665557D+00	−0.353155798676D+02	−0.469641935449D+00

TABLE IV. Estimated ground-state energies of AF1 and AF3 phases from different methods.

	AF1	AF3
LWST [13]	−0.744028	−0.745553
SCSWT [14]	−0.72425	−0.72160
CC [14]	−0.7267(3)	−0.7244(3)
Series ($t = 0$)	−0.7250(3)	−0.7230(3)

necessary to check that this does not introduce systematic errors.

III. RESULTS

The calculations are limited by the rapid increase in the number of clusters of increasing size, made larger by the need to distinguish bond types. For the nearest-neighbor case, where there are two bond types, there are a total of 955 116 distinct clusters with 10 or fewer sites for AF1 and 1 674 334 for AF3, from which we have computed series to 10th order. In Table II we give the coefficients of the energy series for the pure J_1 model, for both $t = 0$ and $t = 0.5$, where t is the convergence parameter discussed above.

We note that the AF1 and AF3 series are identical up to the terms in λ^4, but differ from fourth order onwards. This is consistent with the results of Ref. [8].

The series have been analyzed using standard Padé approximant methods, using both the direct series and the logarithmic derivative. Table III shows values of the energies of AF1 and AF3 phases from a set of high-order approximants.

As is apparent, the energies of the two phases are very close, but those of AF1 lie consistently below those of AF3. The inclusion of the t term, as discussed above, gives some variation in the third digit, but does not, in the present case, result in better consistency among the different approximants.

Table IV compares our estimates of the ground-state energies of both phases with previous results, obtained from linear spin-wave theory (LSWT) [13], a self-consistent spin-wave theory (SCSWT) [14], and a numerical coupled-cluster (CC) calculation [14].

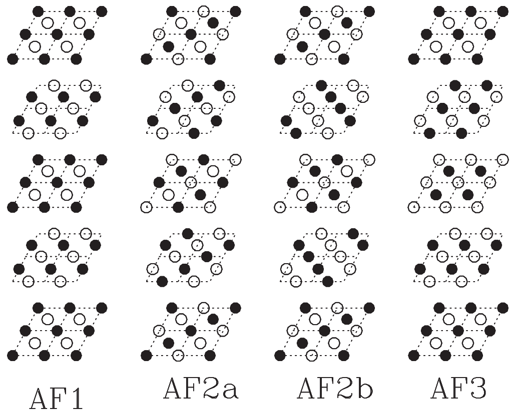

FIG. 1. Ordered phases of the $J_1 - J_2$ fcc antiferromagnet. Solid and open circles denote up and down spins with respect to a quantization axis. The z axis, in the vertical direction, is expanded for clarity.

As can be seen our series results are very consistent with other most recent estimates, falling between the SCSWT and CC values. They all indicate that, for $J_2 = 0$, the AF1 phase is selected over AF3 by quantum fluctuations. The earlier LSWT result therefore does not give a correct prediction.

We now turn to the more general case with both interactions J_1 and J_2. Here the number of clusters grows even more rapidly with cluster size and we have computed series to eighth order in general.

First, we consider the AF2a and AF2b phases which, classically, are degenerate for all values of J_2. In Table V we give the coefficients of the energy series for the particular case $J_2 = 0.7$, for parameter values $t = 0, 1$.

Again, we note that the series are identical through third order and differences begin at the λ^4 term. Analysis of these series gives the energy estimates shown in Table VI.

The estimates are less regular than those in Table II, for the pure J_1 case. This is to be expected, due to both the competing interactions and the shorter series. However, we note that, for every different approximant, the AF2a phase has

TABLE V. Coefficients of the AF2a and AF2b energy series for $J_2 = 0.7$ for parameter values $t = 0, 1$.

AF2a		AF2b	
$t = 0.0$	$t = 1.0$	$t = 0.0$	$t = 1.0$
−0.525000000000D+00	−0.525000000000D+00	−0.525000000000D+00	−0.525000000000D+00
0.000000000000D+00	0.000000000000D+00	0.000000000000D+00	0.000000000000D+00
−0.339375000000D+00	−0.211048951049D+00	−0.339375000000D+00	−0.211048951049D+00
0.333984375000D+00	0.491290527654D−01	0.333984375000D+00	0.491290527654D−01
−0.580362830237D+00	−0.640871825598D−01	−0.563534631188D+00	−0.607531973356D−01
0.126254314588D+01	0.594980686405D−01	0.122809301350D+01	0.603421771454D−01
−0.318910716158D+01	−0.807625851448D−01	−0.310415495749D+01	−0.803667425705D−01
0.894628857272D+01	0.113129149300D+00	0.867461095606D+01	0.112551278350D+00
−0.270084841561D+02	−0.175616037870D+00	−0.260614406845D+02	−0.174251584932D+00

TABLE VI. Estimates of the ground-state energies of AF2a and AF2b phases, from Padé approximant analysis of series for $J_2 = 0.7$ and $t = 1.0$. The superscript l denotes approximants to the logarithmic derivative series.

	AF2a	AF2b
[3,5]	−0.7280	−0.7235
[4,4]	−0.7262	−0.7218
[5,3]	−0.7279	−0.7232
[3,4]	−0.7260	−0.7213
[4,3]	−0.7260	−0.7214
[3,3]	−0.7263	−0.7219
$[3, 4]^l$	−0.7238	−0.7207
$[4, 3]^l$	−0.7284	−0.7237
$3, 3^l$	−0.7255	−0.7211

lower energy. The same result is found for other J_2 values. We thus conclude that, for the AF2 phases, the AF2a case is more stable and is selected by quantum fluctuations, although the energy difference is small.

Finally, we compare the energies of AF1, AF2, and AF3 phases for a range of J_2 values and construct a phase diagram. In Fig. 2 we show the estimated ground-state energies and magnetizations versus J_2/J_1 for all three phases. Uncertainties are, unless shown by error bars, within the size of the data points. Where error bars are shown, they represent "confidence limits," based on consistency between different approximants and not true statistical errors. The magnetization series are, as usual, more erratic, but not excessively so.

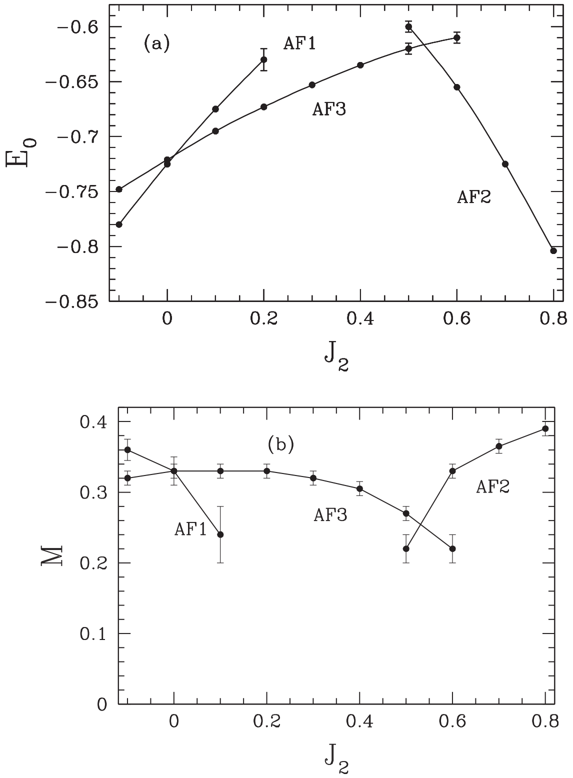

FIG. 2. (a) Ground-state energies of different phases versus J_2 and (b) magnetizations. Unless shown by error bars, uncertainties are believed to be no larger than the data points. The lines are guides for the eye.

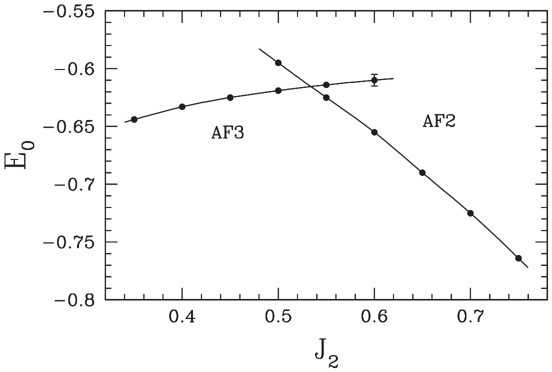

FIG. 3. Energies of AF3 and AF2 phases in the crossover region. The lines are guides for the eye.

It is evident that for $J_2 < 0$ the AF1 phase is stable. For $0 \leqslant J_2 \leqslant 0.5$ the AF3 phase is the stable ground state, while for $J_2 \geqslant 0.5$ the phase AF2 has lowest energy. The transitions are seen to be strongly first order. Quantum fluctuations lead to a reduction of the ground-state magnetization below the classical value 0.5, with the strongest reduction at the AF1/AF3 boundary, where the magnetization is reduced by some 50%.

Figure 3 shows the energies of AF3 and AF2 phase on a finer scale in the crossover region. We estimate the location of the transition at $J_2/J_1 = 0.535 \pm 0.005$.

IV. DISCUSSION

We have used series expansions to compute the ground-state energies and magnetizations of various ordered phases of the spin-1/2 Heisenberg antiferromagnet on the fcc lattice, with both first- and second-neighbor exchange interactions. This allows us to properly include quantum fluctuations at $T = 0$ and to determine their effect in selecting among degenerate classical states.

In particular, we present evidence that in the pure nearest-neighbor case the AF1 phase is favored over AF3, in agreement with the most recent other work [14]. We also find that for large antiferromagnetic J_2 the AF2a phase is selected over AF2b, which also agrees with previous work [8].

The crossover between AF3 and AF2 phases, which classically occurs at $J_2 = 0.5$, is estimated to be shifted, by quantum fluctuations, to $J_2 \sim 0.535$. We are not aware of any previous calculation of this shift.

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