Spiral versus modulated collinear phases in the quantum axial next-nearest-neighbor Heisenberg model

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Motivated by the discovery of spiral and modulated collinear phases in several magnetic materials, we investigate the magnetic properties of Heisenberg spin $S = 1/2$ antiferromagnets in two and three dimensions, with frustration arising from second-neighbor couplings in one axial direction [the axial next-nearest-neighbor Heisenberg (ANNNH) model]. Our results clearly demonstrate the presence of an incommensurate spiral phase at $T = 0$ in two dimensions, extending to finite temperatures in three dimensions. The crossover between Néel and spiral order occurs at a value of the frustration parameter considerably above the classical value 0.25, a sign of substantial quantum fluctuations. We also investigate a possible modulated collinear phase with a wavelength of four lattice spacings and find that it has substantially higher energy and hence is not realized in the model.

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I. INTRODUCTION

Frustrated magnetic materials continue to provide a fruitful interaction between experiment and theory [1]. In particular, large quantum fluctuations in systems with low spin and low dimensionality, coupled with frustration, can lead to novel states [2], quite different from the usual Néel state of classical antiferromagnetism.

Magnetic frustration can arise from the lattice structure itself, as in triangular and kagome systems, or from the presence of additional further neighbor interactions which favor a different type of order from that which would arise from nearest-neighbor interactions alone. One such scenario is the inclusion of second-neighbor interactions along one axial direction, in square or cubic lattices, referred to as the ANNNH (axial next-nearest-neighbor Heisenberg) model [3–6].

The ANNNH model is the obvious quantum extension of the Ising version, the axial next-nearest-neighbor Ising (ANNNI) model, which was much studied primarily in connection with modulated phases in both magnetic and alloy systems [7,8]. The ANNNI model was found to have an extremely rich finite-temperature phase diagram, in both two and three dimensions, with modulated phases having both constant and continuously varying wave vectors.

Our motivation for studying the quantum ANNNH model is twofold. First, there are now a number of materials where commensurate-incommensurate transitions and modulated spiral and collinear phases have been recently observed to arise [9–12]. For example, in the materials $Lu_{1-x}Sr_xMnSi$, cycloidal antiferromagnetic order is argued to arise from an axial next-neighbor interaction [10]. In the material $BiMn_2PO_6$, also a number of commensurate and incommensurate phases are observed, driven by the spatial anisotropy of the interactions in a three-dimensional spin system [11]. On the other hand, the material FeSe shows a "pair-checkerboard" collinear magnetic order [12]. It would be interesting to establish if such phases also arise in ANNNH models, like in their Ising counterpart.

Second, on general grounds one expects that the presence of further neighbor interactions will favor spiral phases, in which the average moment varies sinusoidally with a wave vector along the frustration axis. It is well known that quantum fluctuations can stabilize collinear phases [13]. Thus, it is interesting to ask if additional modulated collinear phases are stabilized in these systems due to quantum fluctuations.

We find that such spiral phases do indeed arise in the quantum models [6]. In two dimensions, long-range order only arises at zero temperature, but in three-dimensional (3D) systems, such phases extend to finite temperatures, and there is a Lifshitz point where Néel, spiral, and paramagnetic phases meet [14]. We find that, despite strong quantum fluctuations, the ANNNH model does not support modulated collinear phases. Instead, the parameter region for the stability of the collinear Néel phase is substantially enhanced by quantum fluctuations.

We consider a Heisenberg model with Hamiltonian

$$H = J_0 \sum_{\langle ij\rangle}^{(0)} \mathbf{S_i}\cdot\mathbf{S_j} + J_1 \sum_{<ik>}^{(1)} \mathbf{S_i}\cdot\mathbf{S_k} + J_2 \sum_{<il>}^{(2)} \mathbf{S_i}\cdot\mathbf{S_l}, \tag{1}$$

where the sums are over nearest-neighbor bonds perpendicular to the modulation axis, nearest-neighbor bonds along the modulation axis, and next-nearest pairs along the modulation axis, with coupling constants J_0, J_1, J_2, respectively. This is shown in Fig. 1 for the two-dimensional (2D) case. The $\mathbf{S_i}$ are quantum spin $S = 1/2$ operators. In the present work we consider all interactions to be antiferromagnetic ($J_i > 0$), although other cases could be treated in a similar way.

The phase diagram for classical spins is well known, but we repeat the argument here for completeness. The energy of a classical spiral ground state is

$$\frac{E}{NS^2} = -nJ_0 + J_1 \cos q + J_2 \cos 2q, \tag{2}$$

where q is the angle between neighboring spins in the modulation direction and $n = 1(2)$ for the square (simple-cubic) lattice. Minimization gives

$$\begin{aligned} q &= \pi \quad J_2/J_1 < 1/4 \\ &= \pi - \cos^{-1}\left(\frac{J_1}{4J_2}\right) \quad J_2/J_1 > 1/4. \end{aligned} \tag{3}$$

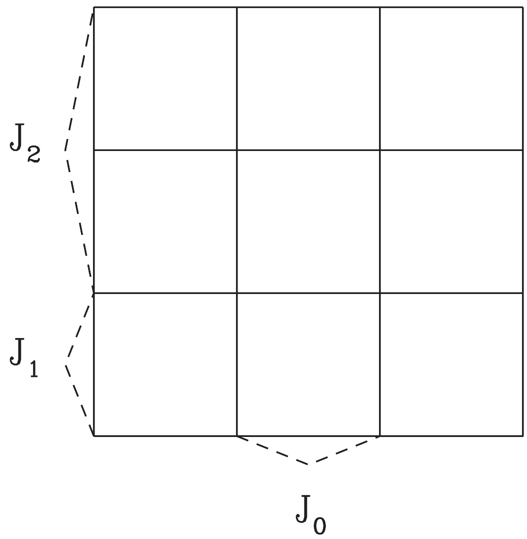

FIG. 1. Coupling constants of the ANNNH model in two dimensions.

Thus the small J_2 Néel phase becomes an incommensurate spiral with wave vector q at the transition point $J_2 = J_1/4$. It can also be seen that in the large J_2 limit, where $q \to \pi/2$, a collinear phase in which each column has two spins "up" followed by two spins "down," with neighboring columns ordered antiferromagnetically, will become degenerate with the spiral. Such a phase has been termed [10] "pair-checkerboard," but we will refer to it as a "2+2 phase" (see Fig. 2). Such a phase occurs in the ANNNI model for large frustration and, while in the classical vector case [Eq. (3)] it only occurs as a limiting case, its stability in the quantum case has not been investigated previously, to our knowledge.

A number of studies of the quantum ANNNH model were reported in the 1980s [3–5] using bosonic Hamiltonians obtained via standard Holstein-Primakoff or Dyson-Maleev transformations. These studies, which focused only on the case of ferromagnetic J_0, J_1, encountered difficulties in treating quantum corrections about the classical states in a consistent way. In any case, these analytic approaches are essentially large S theories, and their reliability for $S = 1/2$ is uncertain. The quantum antiferromagnetic model was studied in two dimensions by Zinke *et al.* [6] using a coupled-cluster method focusing on the spiral order and its pitch angle. We present various comparisons with their study for the two-dimensional case.

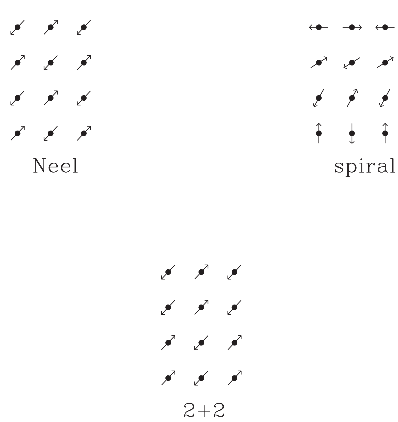

FIG. 2. The Néel, spiral, and 2+2 states.

Our aim in the present work is to explore the physics of this model for spin 1/2, using series expansion methods [15,16]. This approach has been amply demonstrated to give reliable results for quantum spin models and is a method of choice for models with strong frustration, where quantum Monte Carlo methods are defeated by the infamous "minus sign" problem. In the following sections we derive and analyze series for the ground-state energy and magnetization for both the 2D and 3D models. In Sec. IV we compute series at high T for spin-spin correlations and for the structure factor $S(\mathbf{q})$. This analysis clearly shows that the large J_2 phase is an incommensurate spiral, in agreement with the coupled-cluster work [6]. Following the 2D work, in Sec. IV we treat the 3D model and present results at both $T = 0$ and high T. Clear differences from the 2D case are demonstrated. Finally, in Sec. V we summarize our results and suggest possible extensions of this work.

II. GROUND STATE OF THE 2D ANNNH MODEL

We use the linked-cluster method [15,16] to obtain series for the ground-state energy and magnetization. In this approach, series are computed for a sequence of finite connected clusters, and these are combined to obtain series in the thermodynamic limit of a bulk lattice. For each finite cluster, the Hamiltonian is decomposed in the usual perturbative form $H = H_0 + \lambda V$, where H_0 describes a simple system with known ground state and V is treated perturbatively to high order. In the present work we use "Ising expansions" in which H_0 consists of the diagonal $S_i^z S_j^z$ terms, and V consists of the transverse quantum fluctuations. Thus the SU(2) symmetry is broken by the choice of H_0, which reflects the order in the chosen phase but is restored in the limit $\lambda = 1$. Provided there is no singularity for $0 < \lambda < 1$ the true ground state will be reached in this limit. We refer the readers to more detailed expositions [15,16] for further details of the method.

A. Néel phase

To derive series to eighth order we need to consider clusters of up to eight sites, having three bond types. There are a total of 10 644 distinct such clusters for the 2D case. The ground-state energy and magnetization are expressed in the form

$$E_0/N = \sum_{n=0}^{\infty} a_n \lambda^n, \tag{4}$$

$$M = \sum_{n=0}^{\infty} b_n \lambda^n, \tag{5}$$

with the coefficients (a_n, b_n) computed to 12-figure accuracy. The series are analyzed by standard Padé approximant methods to yield estimates of E_0 and M for any values of the exchange constants (J_0, J_1, J_2). In the present work we choose $J_0 = J_1 = 1$ and plot quantities versus the frustration parameter J_2/J_1.

B. 2+2 phase

The 2+2 phase has a four-sublattice structure, and it is necessary to distinguish two types of J_1 bond, between like and unlike spins. This results in a total of 22 613 clusters with four bond types, to eighth order. The derivation and analysis of the series then proceeds in the same way as above.

C. Spiral phase

To carry out an Ising expansion for a noncollinear ordered phase, we transform to a local basis in which each spin is directed along its local z axis. This results in a Hamiltonian of the form

$$H = -\tfrac{1}{4}(J_0 + J_1\cos\theta + J_2\cos 2\theta)N + H_0 + \lambda V, \qquad (6)$$

with

$$H_0 = J_0 \sum_{\langle ij\rangle}^{(0)} \left(-S_i^z S_j^z + \frac{1}{4}\right) + J_1\cos\theta \sum_{\langle ij\rangle}^{(1)} \left(-S_i^z S_j^z + \frac{1}{4}\right)$$
$$+ J_2\cos 2\theta \sum_{\langle ij\rangle}^{(2)} \left(S_i^z S_j^z - \frac{1}{4}\right), \qquad (7)$$

and

$$V = -\frac{1}{2}J_0 \sum_{\langle ij\rangle}^{(0)} (S_i^+ S_j^+ + S_i^- S_j^-)$$
$$-\frac{1}{4}J_1 \sum_{\langle ij\rangle}^{(1)} \big[(1+\cos\theta)(S_i^+ S_j^+ + S_i^- S_j^-)$$
$$-(1-\cos\theta)(S_i^+ S_j^- + S_i^- S_j^+)$$
$$+2\,\sin\theta\big(S_i^+ S_j^z + S_i^- S_j^z - S_i^z S_j^+ - S_i^z S_j^-\big)\big]$$
$$-\frac{1}{4}J_2 \sum_{\langle ij\rangle}^{(2)} \big[(1-\cos 2\theta)(S_i^+ S_j^+ + S_i^- S_j^-)$$
$$-(1+\cos 2\theta)(S_i^+ S_j^- + S_i^- S_j^+)$$
$$-2\,\sin 2\theta\big(S_i^+ S_j^z + S_i^- S_j^z - S_i^z S_j^+ - S_i^z S_j^-\big)\big], \qquad (8)$$

where the superscripts 0, 1, 2 refer to the three bond types, and θ is the angle between successive spins in columns. (Actually the angle is $\pi - \theta$ in the original picture, before a rotation of axes.) This Hamiltonian contains the angle θ as a parameter, and this is not known *a priori*. Thus we choose a range of values, plot the energy as a function of θ, and choose the correct θ from the minimum.

In practice, the minimum is quite shallow and it is difficult to choose θ with high precision. However, this does not seriously affect the energy estimates. We calculated series for various θ values at intervals of 5 deg and estimated the θ values where the energy are minimum. The values of θ at the minima were in rough agreement with the coupled-cluster calculation of Ref. [6]. We got estimates for θ values for $J_2 =$ 1.0, 0.8, 0.7, 0.6, and 0.5 of 80, 75, 70, 60, and 45 deg, respectively. Thus the pitch angle $q = \pi - \theta$ was somewhat below the classical value for $J_2 > 0.7$ but became sharply larger for smaller J_2 values.

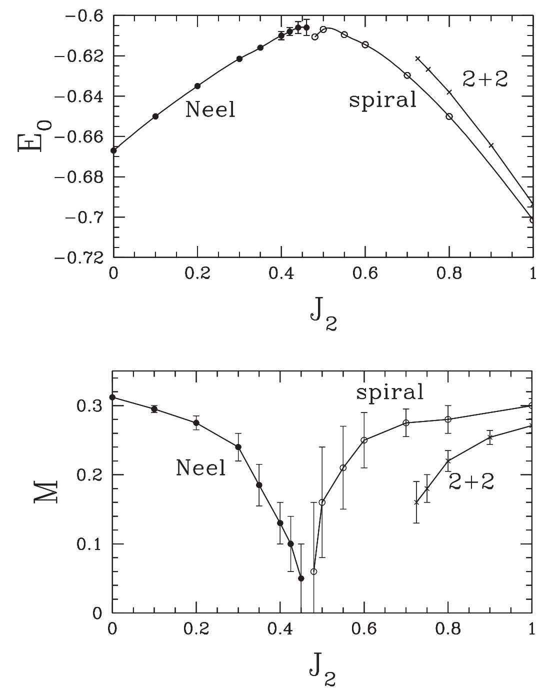

FIG. 3. Ground-state energy (upper panel) and magnetization (lower panel) as a function of J_2 for the 2D ANNNH model.

D. Results

Figure 3 shows the ground-state energy and magnetization versus J_2/J_1 for the 2D ANNNH model, for the Néel, spiral, and 2+2 phases obtained from our series. The series have been analyzed by standard Padé approximant techniques, using both the direct series and the logarithmic derivative. The latter are found to give slightly more stable results, but the two approaches are broadly consistent. Where error bars are shown in the figures, they represent "confidence limits" based on the spread of different approximants.

We first comment on the ground-state energy. These series are very regular, and any uncertainty is estimated to be no larger than the plotted points, except very near the transition point. The Néel and spiral series appear to meet smoothly at a point near $J_2 = 0.47 \pm 0.02$, well above the classical transition point 0.25. We note that in the coupled-cluster study the Néel order was found to continue up to a J_2 value of approximately 0.4. The transition is consistent with a continuous transition, although the match of the energy of spiral and Néel phases is not perfect. This is probably caused by the uncertainty in determining the pitch angle and the spiral-state ground-state energy near the transition. In Fig. 3(a) we also plot the energy of the 2+2 phase. This clearly lies at higher energy and is thus not a stable phase. It seems that as $J_2 \to \infty$, the energies of the spiral and 2+2 phases become asymptotically equal, as for the classical case.

The magnetization series are less regular, and the error bars become quite large near the transition point. The most interesting feature is that both the Néel and spiral phase magnetizations appear to be dropping to zero at the transition, between 0.45 and 0.5. Thus quantum fluctuations in this 2D model are large enough to destroy the long-range order at this point. Indeed, we cannot exclude the possibility of a (very) narrow nonmagnetic phase. We also show the magnetization for the 2+2 phase, but, since this phase has higher energy, it is of little significance.

III. HIGH-T SERIES FOR $S(\mathbf{q})$

High-temperature series [15] provide a complementary approach for studying the nature of magnetic orders. Although the Mermin-Wagner theorem precludes any finite-temperature ordered phase in the 2D model, it is expected that, as the temperature is lowered, the correlations that build up will reflect the nature of the order which occurs at $T = 0$. High-T expansions for a correlator $\langle S_i^z S_j^z \rangle$ can be developed as

$$C(\mathbf{r}) = \frac{1}{Z} \sum_{n=0}^{\infty} \frac{(-1)^n}{n} Tr\{S_0^z S_{\mathbf{r}}^z H^n\} \beta^n, \tag{9}$$

where $\beta = 1/k_B T$, and Z is the partition function, which is itself expanded as a series in β. We note that since we are in a paramagnetic phase, the correlations have full rotational symmetry and it suffices to compute the (zz) correlators. From these we compute a high-T series, in powers of β, for the static structure factor

$$S(\mathbf{q}) = \sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}} C(\mathbf{r}), \tag{10}$$

which should develop a peak at whatever $\mathbf{q}$ value reflects the $T = 0$ order.

To compute the structure factor series to eighth order, for general $\mathbf{q}$, would require a correlator series for all cluster space types with eight or fewer bonds, a total of over 600 000 distinct clusters. However, for $\mathbf{q}$ in the modulation direction, this number can be reduced considerably by effectively calculating correlator series between horizontal rows of spins. This requires only 76 712 clusters.

The $S(\mathbf{q})$ series converges rapidly at high T (small β) and can be evaluated using Padé approximants down to about $t = k_B T/J_1 \approx 0.5$. We have carried out such an analysis for $\mathbf{q} = (\pi, q_z)$ for various J_2 and results are shown in Fig. 4 for the temperature $t = 0.5$. Below this t the series becomes too erratic. We see that for $J_2 = 1.0$ there is a clear peak at $q_z = 0.58\pi$, corresponding to an angle of 76 deg. As J_2 is decreased, the peak broadens and moves to larger q_z (smaller angles). Note that $q_z = \pi/2$ would correspond to a modulation wavelength of a lattice spacing, as for the 2+2 structure, whereas $q = \pi$ corresponds to the Néel phase. The peak positions do not change significantly with temperature.

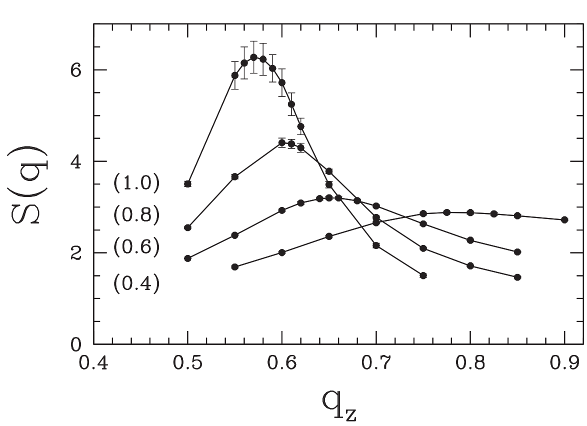

FIG. 4. Structure factor $S(q)$ for the 2D ANNNH model,with q in units of π/a, for different values of J_2 as calculated from high-T series expansions. The values of J_2 for the different plots are indicated within brackets.

IV. THE 3D ANNNH MODEL

We have used the same approach to study the ANNNH model on the simple-cubic lattice. The ground-state energy and magnetization are shown in Fig. 5.

The following points can be noted: (1) The energy series are very regular, and the curves meet smoothly at $J_2 = 0.34 \pm 0.01$. The Néel series can be accurately continued well beyond this point, as shown. We also note the maximum in the spiral phase energy near 0.4, which then drops again to meet the Néel curve smoothly. This feature occurs in the classical case and is also apparent, though less clearly, in the 2D case (Fig. 3). The Néel-to-spiral crossover point, at $J_2 \approx 0.34$, is again well above the classical value 0.25, but the difference is less than in the 2D case, reflecting the smaller quantum fluctuations in higher dimension. (2) The magnetization series are less regular, and this is reflected in the error bars, although the size of the uncertainty is exaggerated by the scale chosen for the figure. We note that the magnetization decreases on approaching the crossover point from either side, but only by ~10%. Unlike the 2D case, the magnetization does not drop to zero, again showing that quantum fluctuations are less dominant.

As in the 2D case, we have also derived high-T series for the structure factor $S(q)$. There is, however, one important

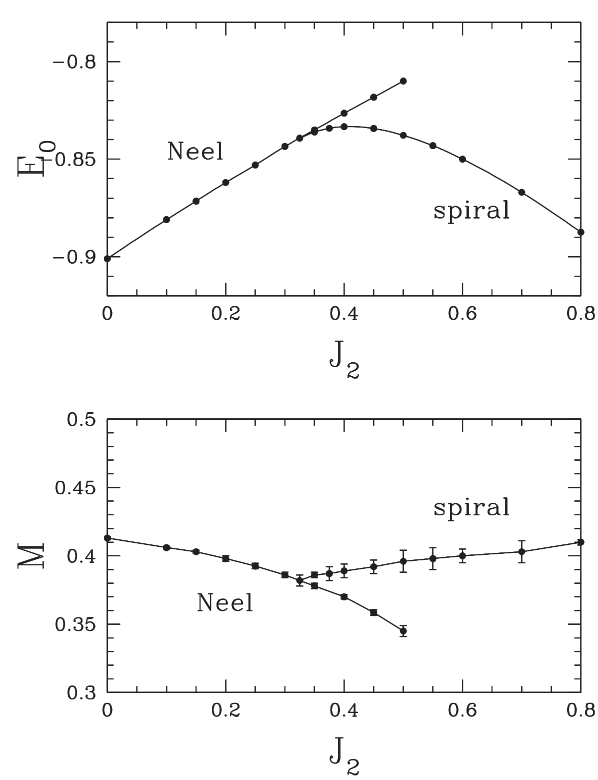

FIG. 5. Ground-state energy (upper panel) and magnetization (lower panel) for the 3D ANNNH model from series expansions.

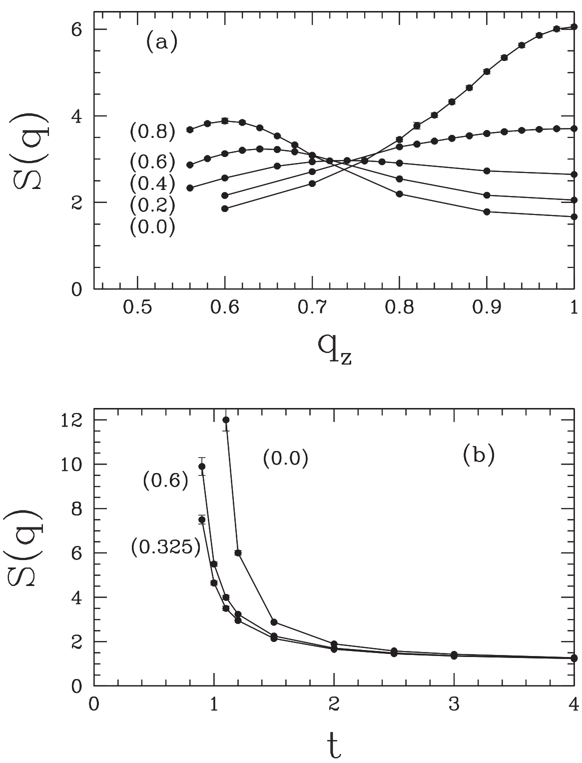

FIG. 6. (a) Structure factor $S(q)$ versus q_z at a temperature $t = 1.2$ for several values of J_2 for the 3D ANNNH model. (b) Structure factor at the critical wave vector versus temperature for several values of J_2. The values of J_2 for the different plots are indicated within brackets.

difference. In 3D the system will retain long-range magnetic order at finite temperature, up to some critical temperature $T_c(J_2)$. On approaching $T_c(J_2)$ from above, the structure factor $S(q)$ at the appropriate wave vector is expected to diverge in the thermodynamic limit, reflecting the development of long-range correlations at the critical temperature. Thus we may expect to be able to estimate the locus of this critical line from our series. Some results are shown in Fig. 6.

Figure 6(a) shows $S(q)$ versus q_z for various J_2. For $J_2 = 0.0,0.2$ the maximum is at $q_z = 1.0$, corresponding to Néel order. For larger J_2 the peak moves continuously to smaller q_z: $q_z \approx 0.73$ for $J_2 = 0.4$, $q_z \approx 0.64$ for $J_2 = 0.6$, $q_z \approx 0.6$ for $J_2 = 0.8$. This is indicative of an incommensurate spiral phase. The q_z values are consistent with those found to give the lowest ground-state energy in the $T = 0$ spiral phase. The point where the peak begins to move away from $q_z = 1$ is close to $J_2 = 0.325$.

We have also analyzed the $S(q)$ series to estimate the values of the critical temperature as a function of J_2. While the eighth-order series are too short to provide estimates of high precision, Padé approximants to the logarithmic derivative series do show a fairly consistent pole on the positive real axis, corresponding to a line of critical points.

In Fig. 6(b) we plot the value of $S(q_c)$, at the critical wave vector q_c, versus reduced temperature $t = k_B T / J_0$. A strong divergence is clearly seen. Our best estimates for the critical temperatures are 1.09 ($J_2 = 0.0$), 0.82 ($J_2 = 0.325$), 0.89 ($J_2 = 0.6$). For $J_2 = 0$, the isotropic simple-cubic nearest-neighbor model, a more precise estimate is available from longer series [17]. There are, as far as we know, no previous estimates of the critical line for the 3D ANNNH model. The critical temperature is lowest near $J_2 = 0.325$, which is also where the peak in $S(q)$ moves away from $q_z = 1$. This is a Lifshitz point [14], where paramagnetic, Néel ordered, and spiral phases meet and coexist.

V. SUMMARY AND DISCUSSION

We have used a combination of perturbation series at $T = 0$ and high-T expansions to investigate the nature of magnetic order, and the magnetic phase diagram in the quantum spin $S = 1/2$ ANNNH model, in both two and three dimensions. While it is easy to show, for classical vector spins, that an incommensurate spiral phase exists for large frustration J_2, previous analytic studies for quantum spins have encountered difficulties. Our study confirms that the classical picture remains qualitatively correct. However, quantum fluctuations shift the crossover point between Néel and spiral phases substantially. In the two-dimensional case, we also looked for possible modulated collinear phases. However, we found that even the most robust of those, the 2+2 phase, has a rather high energy and hence is not stabilized. Hence, we conclude that such modulated collinear phases are unlikely to arise in the model.

For the 2D model we find that the magnetizations in both Néel and spiral ground states appear to tend continuously to zero at the crossover point. This was not expected and is reminiscent of the behavior in the $J_1 - J_2$ model, where there is an intermediate nonmagnetic phase. We see no evidence for such a phase here, although we cannot exclude the possibility of a very narrow phase of this kind. In the 3D model, the magnetizations clearly cross over at a finite value.

In the 3D model, the magnetic phases extend to finite temperature, and we have estimated the position of the critical line, and of the Lifshitz point, where paramagnetic, Néel, and spiral phases coexist.

In the large J_2 limit a collinear phase, the "2+2 phase," becomes asymptotically degenerate with the $q_z = \pi/2$ spiral, both having a modulation wavelength of four lattice spacings. Such a phase, termed "pair-checkerboard," was found to exist in the FeSe monolayer system [12]. We find that in the ANNNH model, such a phase always has higher energy than the spiral. Thus, if it exists as a stable phase, a more complex Hamiltonian would be indicated.

ACKNOWLEDGMENTS

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