A frustrated three-dimensional antiferromagnet: stacked $J_1$–$J_2$ layers

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Abstract

We study a frustrated 3D antiferromagnet of stacked $J_1$–$J_2$ layers. The intermediate ‘quantum spin liquid’ phase, present in the 2D case, narrows with increasing interlayer coupling and vanishes at a triple point. Beyond this, there is a direct first-order transition from Néel to columnar order. Possible applications to real materials are discussed.

1. Introduction

The study of frustrated quantum antiferromagnets remains an active field, characterised by strong interplay between theory and experiment. A model which has been studied extensively (see [1–15] and references therein) is the so-called ‘$J_1$–$J_2$ model’, a spin-1/2 system on the two-dimensional square lattice with nearest- and next-nearest-neighbour interactions of strength $J_1$ and $J_2$, respectively, both being antiferromagnetic. The Néel order at $T = 0$, which pertains for $J_2 = 0$, is destabilised by the frustrating $J_2$ interaction and vanishes at around $J_2/J_1 \simeq 0.4$. In the opposite limit, of large $J_2$, a columnar (often ambiguously termed ‘collinear’) ordered phase occurs, in which successive columns (or rows) of spins alternate in direction. The columnar phase becomes unstable at $J_2/J_1 \simeq 0.6$. A magnetically disordered region thus exists in the region $0.4 \lesssim J_2/J_1 \lesssim 0.6$, the nature of which remains not fully resolved.

It is of interest to ask what happens to this intermediate phase, and indeed to the entire phase diagram, when these $J_1$–$J_2$ layers are coupled, perhaps weakly, in the third direction, forming a three-dimensional structure. This is the main topic of this paper. It has already been studied by Schmalfuß et al [16] using the coupled-cluster and rotation-invariant Green’s function methods. They found that the disordered region becomes narrower as a function of $J_2$ when $J_3$ is increased and vanishes entirely at $J_2/J_1 \simeq 0.2$–0.3. A more recent paper [17] has used a different approach and obtained a much higher value, $J_3/J_1 \simeq 0.67$. We will discuss these results further in the conclusions. However, it is clear that there is a need to resolve this large discrepancy. We will address the question using series expansion methods [18] in both Néel and columnar phases, as well as first-order spin-wave theory.

It has been argued recently that the layered materials Li$_2$VOSiO$_4$ and Li$_2$VOGeO$_4$ are well represented by the spin-1/2 $J_1$–$J_2$ model with $J_2 \gg J_1$, i.e. in the columnar phase. In [19] high-temperature expansions for the specific heat and magnetic susceptibility for the purely 2d model were used to try to constrain the values of the exchange parameters by fitting to the experimental data. At the same time an ab initio local density approximation (LDA) calculation of the exchange parameters $J_1$, $J_2$ and $J_3$ yielded (0.75, 8.8 and 0.25 K) for the Si material and (1.7, 8.1 and 0.19 K) for the Ge system [19]. This suggests that the coupling in the third direction is by no means negligible and ought to be included in a fitting procedure.

We mention also that a popular, though not universally accepted, scenario to understand the magnetic properties of the recently discovered superconducting iron pnictides is via a spin-1 layered $J_1$–$J_2$ model [20, 21]. While we do not consider these systems explicitly here, our results may have some relevance.

Our model is a three-dimensional spin-1/2 antiferromagnet on a tetragonal lattice, as shown in figure 1(a). Frustrating $J_2$ interactions occur in the $x$–$y$ plane but not in the third direction. All interactions are antiferromagnetic.

The Hamiltonian, in standard notation, is

$$H = J_1 \sum_{i<j} S_i \cdot S_j + J_2 \sum_{i<k} S_i \cdot S_k + J_3 \sum_{i<l} S_i \cdot S_l$$

(1)
where the summations are over the three classes of coupling, respectively.

In section 2, we present our zero-temperature calculations and results, for the ground state energy and sublattice magnetisation (order parameter), for both the Néel and columnar phases. Our results support a phase diagram of the form shown in figure 1(b) and we estimate the location of the triple point at $J_2/J_1 = 0.54(3)$ and $J_3/J_1 = 0.16(3)$ Our series results are compared with the results of linear spin-wave theory. In section 3 we use series methods to compute the 1-magnon dispersion curves in both ordered phases, and again compare our results with the spin-wave predictions. The spin-wave theory follows standard lines and, for completeness, is outlined in the appendix. In section 4 we present our conclusions and discussion.

2. Ground state bulk properties

The series expansion method is based on perturbative calculations for a sequence of finite connected clusters, which are then combined to obtain a series for the bulk system. In practice it is possible to treat of the order of $10^6$ different clusters and to obtain series of the order of 10–20 terms, respectively. The Hamiltonian is written in the standard form

$$H = H_0 + \lambda V,$$

where $H_0$ has a simple known ground state. In the present work we use an Ising expansion in which $H_0$ consists of the diagonal $S_i^z S_i^z$ terms and the quantum fluctuations are included in the perturbation $V$. Series are obtained in powers of $\lambda$ and extrapolated to $\lambda = 1$ via standard Padé or differential approximant methods. The interested reader is referred to the book [18] for more detail.

2.1. Néel phase

It is well known that the unfrustrated square lattice ($J_2 = J_3 = 0$) has reduced Néel order in the ground state. This is a two-sublattice structure with spins pointing in opposite directions on the two sublattices. For technical reasons it is useful to perform a spin rotation on the B sublattice, making the unperturbed ground state ferromagnetic. The Hamiltonian for a lattice of $N$ sites is then written as

$$H = -\frac{1}{4} (2J_1 - 2J_2 + J_3) N + H_0 + \lambda V$$

with

$$H_0 = J_1 \sum_{\langle ij \rangle} \left( \frac{1}{4} - S_i^z S_j^z \right) + J_2 \sum_{\langle ik \rangle} \left( S_i^z S_k^z - \frac{1}{4} \right)$$

$$+ J_3 \sum_{\langle ij \rangle} \left( \frac{1}{4} - S_i^z S_j^z \right)$$

and

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

$$+ \frac{1}{2} J_3 \sum_{\langle ij \rangle} \left( S_i^+ S_j^- + S_i^- S_j^+ \right).$$

We have added and subtracted constant terms to make the unperturbed energy zero.

We have obtained series for the ground state energy $E_0$ and the magnetisation $M$ to order $\lambda^9$. The rapid proliferation of clusters with three bond types (there are 320 274 with nine or fewer sites) limits the length of the series obtainable. The data are far too extensive to present, but can be provided on request. The values of $E_0$ and $M$ can then be estimated, with some uncertainty, for any values of $J_2$ and $J_3$.

2.2. Columnar phase

In the columnar phase the spins on alternating columns in a particular $x$–$y$ plane will point in opposite directions. This satisfies all of the strong $J_2$ bonds but leaves half of the $J_1$ bonds frustrated. In adjacent planes these are shifted by one lattice spacing, leaving all of the $J_3$ bonds satisfied. This structure has an additional twofold degeneracy as columns can equally well be ‘rows’. Again it is convenient to carry out a spin rotation on ‘down’ sites, yielding

$$H = -\frac{1}{4} (2J_2 + J_3) N + H_0 + \lambda V$$

with

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

$$+ J_2 \sum_{\langle ik \rangle} \left( \frac{1}{4} - S_i^z S_k^z \right) + J_3 \sum_{\langle ij \rangle} \left( \frac{1}{4} - S_i^z S_j^z \right)$$

and

$$V = \frac{1}{2} J_2 \sum_{\langle ij \rangle} \left( S_i^+ S_j^- + S_i^- S_j^+ \right) + \frac{1}{2} J_3 \sum_{\langle ik \rangle} \left( S_i^+ S_k^- + S_i^- S_k^+ \right)$$

$$+ \frac{1}{2} J_3 \sum_{\langle ij \rangle} \left( S_i^+ S_j^- + S_i^- S_j^+ \right).$$
2.3. Results

Series were computed and analysed for the cases $J_3 = 0, 0.05, 0.1, 0.2, 0.5$ and $1.0$ and a range of values of $J_2$ in both the Néel and columnar phases. We set $J_1 = 1$ throughout.

Figure 2 shows estimates of the ground state energy per site versus $J_2$ for two cases, $J_3 = 0$ (figure 2(a)) and $J_3 = 1.0$ (figure 2(b)). The energy is evaluated by forming Padé approximants to the series and evaluating these at $\lambda = 1$. Error bars, where shown, represent confidence limits, based on the degree of consistency between high-order approximants.

As is apparent, the behaviour of the energy estimates is quite different in the two cases. For $J_3 = 0$, i.e. the $J_1$–$J_2$ square lattice model, the energy estimates in the intermediate range $J_2 = (0.4, 0.6)$ become erratic and the two curves, from the Néel and columnar series, respectively, do not appear to join. However, for $J_3 = 1$, the estimates are quite precise and the two curves cross near $J_2 \simeq 0.55$. The clear difference in slope of the two branches at the crossing point indicates a direct first-order transition between the two phases.

To analyse the magnetisation series we first performed a Huse transformation \cite{22} to a new variable $x = 1 - \sqrt{1 - \lambda}$, to remove the square-root singularity at $\lambda = 1$, expected from spin-wave theory. This procedure has been used in earlier work on the 2D antiferromagnet \cite{22, 23}. Padé approximants to the new series were then evaluated at $x = 1$.

The results are shown in figure 3 and again display a striking difference between the two cases $J_3 = 0$ and 1. For $J_1 = 0$ we see clear evidence of an intermediate non-magnetic phase, with Néel and columnar magnetisations vanishing near $J_2 \simeq 0.4$ and $J_2 \simeq 0.6$, respectively. The error bars are large near the transition points and it is not possible to say whether
the transitions are first or second order. It is generally believed that the columnar-disordered transition is first order while the Néel-disordered transition appears second order but may, in fact, be weakly first order [15]. On the other hand, for $J_3 = 1$ (figure 3(b)) it is clear that both magnetisations remain finite throughout their phase. This is consistent with a direct first-order transition.

Proceeding in this way with other values of $J_3$, we construct a phase diagram as shown in figure 4, which confirms the schematic phase diagram in figure 1(b). As is apparent, the disordered phase narrows as the interplane coupling $J_3$ is increased and vanishes at the estimated point $J_2/J_1 = 0.54 \pm 0.03$, $J_3/J_1 = 0.16 \pm 0.03$. This value of $J_3$ is a little lower than found previously [16].

3. Excitations

It is also of interest to investigate the spectrum of single-magnon excitations in the ordered phases and to see how these change on increasing the interlayer coupling $J_3$. Standard series methods exist for calculating the excitation energy throughout the Brillouin zone [18], and we utilise these in the following.

3.1. Néel phase

Series were obtained to order $\lambda^7$, based on 255 196 distinct clusters of up to eight sites. In figure 5 we show dispersion curves for two values of the frustrating interaction $J_2 = 0.0, 0.4$ for $J_3 = 0.5$, along various symmetry lines in the Brillouin zone, as shown. For comparison we also show the results of linear spin-wave theory.

Several features are worth noting:

(1) A very pronounced dip develops at wavevector $(\pi, 0, 0)$ (and, by symmetry, also at $(0, \pi, 0)$) with increasing $J_2$. Linear spin-wave theory has the gap vanishing at $J_2 = 0.5$ (see the appendix).

(2) There is a distinct ‘shoulder’ at $(0, 0, \pi/2)$, which is not seen in linear spin-wave theory. Indeed, along the whole line $(0, 0, k_z)$ the energy depends only weakly on $J_2$.

(3) Linear spin-wave theory reproduces the overall dispersion curves reasonably well, but systematically underestimates the excitation energies.

We note also that the series become very irregular near the Goldstone point $k = 0$ and the Padé approximants do not accurately show the vanishing of the gap at this point. This has been noted in previous work [24] and an indirect way found to overcome this problem, which also allows calculation of the spin-wave velocity. However, we do not address this further in the present work.

3.2. Columnar phase

Series were obtained to order $\lambda^7$, based on 490 487 distinct clusters with up to eight sites, and four possible bond types. In figure 6 we show dispersion curves for $J_3 = 0.5$ and $J_2 = 1.0, 0.6$ along various symmetry lines in the Brillouin zone. The predictions of linear spin-wave theory are shown as dashed lines.

The most notable feature is the depressed excitation energy along the line $(\pi/2, \pi, 0)$ to $(0, \pi, 0)$. The dispersion curve in this region flattens as $J_2$ decreases from 1.0 to 0.6 in both cases (a) and (b). According to linear spin-wave theory this whole branch becomes soft at $J_2/J_1 = 0.5$, the classical transition point. Again linear spin-wave theory tends to underestimate the excitation energy, but not as badly as for the Néel phase. The same difficulty with the Goldstone point $k = 0$, discussed above for the Néel phase, occurs here.

3.3. The energy gap at $(0, \pi, 0)$

As the phase transition is approached, from either the Néel or columnar side, we expect increasing quantum fluctuations at wavevector $k = (0, \pi, 0)$, which should be manifest through a decreasing energy gap at this point. As mentioned above, linear spin-wave theory predicts a vanishing excitation energy at this $k$ point at the classical transition point $J_2/J_1 = 0.5$. Hence the variation of the gap is of some interest, and our results are shown in figure 7, as a function of $J_2$, for two values of the coupling ratio $J_3/J_1 = 0.5$.
Figure 6. Series estimates of the single-magnon dispersion relations in the columnar phase at
(a) $J_2/J_1 = 1.0$, $J_3/J_1 = 0.5$, $k_z = 0$ (upper curve) and
$J_2/J_1 = 0.6$, $J_3/J_1 = 0.5$, $k_z = 0$ (lower curve), and
(b) $J_2/J_1 = 1.0$, $J_3/J_1 = 0.5$, $k_z = \pi/2$ (upper curve) and
$J_2/J_1 = 0.6$, $J_3/J_1 = 0.5$, $k_z = \pi/2$ (lower curve). The dashed lines
are linear spin-wave predictions. Values of $q = k/\pi$ along the path
are shown at the bottom.

Figure 7. Series estimates of the energy gap $\epsilon(k)$ at $k = (0, \pi, 0)$ as a function of $J_2$
(a) $J_3 = 0$ and (b) $J_3 = 0.5$. Filled circles—Néel phase; filled triangles—columnar phase.
The dashed lines are linear spin-wave predictions in each phase.

4. Conclusions

We have used series expansion methods to study a system of stacked $J_1$–$J_2$ layers with antiferromagnetic coupling $J_3$
between the layers. This model is applicable to the layered materials Li$_2$VOSiO$_4$ and Li$_2$VOGeO$_4$, as well as being of
intrinsic theoretical interest. In agreement with earlier work
of Schmalfuß et al [16], we find that the disordered region of the phase diagram (the ‘intermediate phase’) becomes
narrower as the interlayer coupling $J_3$ is increased, and
vanishes completely at a triple point, beyond which there is
a direct first-order transition between the Néel and columnar
phases. We estimate the location of the triple point as $J_2/J_1 = 0.54 \pm 0.03$, $J_3/J_1 = 0.16 \pm 0.03$. Reference [16] uses,
in fact, two methods: a rotation-invariant Green’s function
method and a coupled-cluster approach (CCM). The results
are rather different. The CCM, presumably more accurate,
gives $J_3/J_1 \approx 0.23$, a value comparable with, but somewhat
higher than, our estimate. The CCM method requires an
extrapolation of a sequence of approximations and this
may be problematic (see, e.g., the application of the CCM
between the two ordered phases is first order at this value of $J_3$.

Figure 7(a) shows the energy gap at $J_3 = 0$. Note that,
whereas linear spin-wave theory has the gap vanishing at all
 couplings in the columnar phase, it is in fact finite at larger
$J_2$, as given by a modified spin-wave theory and earlier series
results [13]. Note also that the energy gaps in both phases
extrapolate to zero before reaching the classical transition
point $J_2/J_1 \approx 0.5$, providing further evidence of a disordered
intermediate phase when $J_3 = 0$. Taken at face value, the
results indicate that the gap remains finite at the estimated
transition points $J_2 \simeq 0.4$ and $J_2 \simeq 0.6$, which would indicate
first-order transitions. Naive extrapolations are not entirely
reliable near a critical point, however, and a more precise
check of this point would be of interest. A Dlog Padé analysis
of the series in $\lambda$ gives no consistent estimates of the critical
point in this case.

In the case $J_3/J_1 = 0.5$ (figure 7(b)) the gaps remain of
substantial size right up to the direct first-order transition and
the gaps appear to be of roughly equal magnitude in both
phases at the transition. This is a further sign that the transition
to the ‘Union-Jack’ antiferromagnet [26], in particular the results in Table II). Thus we believe our series results to be more accurate. A more recent calculation [17], using an effective field theory approach, concludes that the triple point (they refer to this as a critical end point, assuming the Néel to disordered transition is second order) gives $J_2/J_1 \approx 0.56$, $J_3/J_1 \approx 0.67$, which is much higher than both [16] and our result. In yet another recent calculation, Majumdar [25] has carried out a spin-wave calculation to second order ($1/S^2$) and concludes that the intermediate phase remains present even at $J_3 = 1$, which seems surprising, in view of the other works referred to above.

It has been suggested by a referee that the series approach, starting as it does from a broken symmetry state, may overestimate the extent of the ordered phases and thus explain the discrepancy between our result and CCM. However, previous studies [27, 10] have shown that rotation symmetry is restored to correlators calculated by series methods in the rotationally invariant limit $\lambda = 1$. We do not believe that our approach overestimates the widths of the ordered phases to any significant degree. It is also significant that a ‘self-consistent’ spin-wave theory [21] gives (in our units) $J_3/J_1 \approx 0.13$ at the tricritical point.

As well as mapping out the ground state phase diagram, we have computed magnon dispersion curves along symmetry lines in the Brillouin zone, in both the Néel and columnar phases, for various parameter values. This would allow a more critical evaluation of the validity of the model for the materials mentioned above, when experimental inelastic neutron scattering results for magnon energies become available.

The present work represents, to our knowledge, the first study of this model by series expansion methods. We emphasise the importance of studying such frustrated models by a variety of methods. The large spread of results obtained by different authors using different methods only confirms this. We believe that series methods are, at present, the most reliable method for studying frustrated spin models, where a quantum Monte Carlo approach is not possible.

Finally we mention another recent study [28] of a different generalisation of the two-dimensional $J_1$–$J_2$ model, in which the frustrating $J_2$ interactions are included in all spatial directions. That model appears also to have an interesting phase diagram, and could be studied by series methods.

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Appendix A. Linear spin-wave theory

In the appendix we present the results of linear spin-wave theory (LSWT) for the model, for both the Néel and columnar phases. The results have been used in the main text to compare with the more accurate series results.

The basic procedures of LSWT are well known so we will not include all details.

A.1. Néel phase

There are two equivalent sublattices A, B and two sets of bosons describing spin deviations from the classical Néel state. After Fourier transformation the Hamiltonian is, to quadratic order

$$H = -NS^2(2J_1 - 2J_2 + J_3) + 2S(2J_1 - 2J_2 + J_3)$$

$$+ 4SJ_1 \sum_k \gamma_k (a_k^\dagger a_k^\dagger + a_k b_k)$$

$$+ 2SJ_2 \sum_k \mu_k (a_k^\dagger a_k^\dagger + b_k b_k)$$

$$+ 2SJ_3 \sum_k \nu_k (a_k^\dagger b_k^\dagger + a_k b_k)$$

(A.1)

with

$$\gamma_k = \frac{1}{2}(\cos k_x + \cos k_y), \quad \mu_k = \cos k_x \cos k_y,$$

$$\nu_k = \cos k_z.$$  

This is then diagonalised by a standard Bogoliubov transformation, yielding

$$H = E_0 + \sum_k \omega_k (A_k^\dagger A_k + B_k^\dagger B_k)$$

(A.3)

with

$$\omega_k = 2S\sqrt{P_k^2 - Q_k^2}$$

$$P_k = 2J_1 + J_3 - 2J_2(1 - \mu_k)$$

$$Q_k = 2J_1 \gamma_k + J_3 \nu_k.$$ 

The ground state energy is

$$E_0 = -NS(S + 1)(2J_1 - 2J_2 + J_3) + \sum_k \omega_k$$

(A.5)

and the sublattice magnetisation is

$$M = S + \frac{1}{2} - \frac{2S}{N} \sum_k P_k \omega_k.$$ 

We note that for small $k$

$$P_k + Q_k = 2J_1 + J_3 - \frac{1}{2} (J_1 + J_2)(k_x^2 + k_y^2)$$

$$+ \frac{1}{2} J_3 k_z^2 + \cdots$$

(A.7)

$$P_k - Q_k = \frac{1}{2} (J_1 - J_2)(k_x^2 + k_y^2) + \frac{1}{2} J_3 k_z^2 + \cdots$$

so there is a linear Goldstone mode at $k = 0$, provided $J_2 < J_1/2$. The phase becomes unstable for $J_2 > J_1/2$.

Another special case occurs at $k = (\pi, 0, 0)$. Let $k' = k - (\pi, 0, 0)$ be small, then

$$P_k + Q_k = 2J_1 - 4J_2 + \frac{3}{2} k_z^2 + J_2 (k_x^2 + k_y^2) + \cdots$$

$$P_k - Q_k = 2J_1 - 4J_2 + \frac{3}{2} J_3 + J_2 (k_x^2 + k_y^2)$$

(A.8)

$$- \frac{1}{2} J_3 k_z^2 + \cdots.$$ 

Hence the gap at $k' = 0$ vanishes at $J_2 = J_1/2$, regardless of $J_3$, corresponding to a phase transition.
A.2. Columnar phase

There are again two sublattices, with the A and B sites forming successive columns. The Fourier-transformed Hamiltonian is now

\[
H = -NS^2(2J_2 + J_3) + 2S \sum_k (J_1 \cos k_x + 2J_2 + J_3)(a_k^\dagger a_k + b_k^\dagger b_k) \\
+ 2S \sum_k (J_1 \cos k_x + 2J_2 \mu_k + J_3 \nu_k)(a_k^\dagger b_k^\dagger + a_k b_k).
\]

(A.9)

A Bogoliubov transformation then yields

\[
H = E_0 + \sum_k \omega_k (A_k^\dagger A_k + B_k^\dagger B_k)
\]

(A.10)

with

\[
E_0 = -NS(S + 1)(2J_2 + J_3) + \sum_k \omega_k
\]

(A.11)

and

\[
\omega_k = 2S \sqrt{p_k^2 - Q_k^2}, \\
P_k = J_1 \cos k_x + 2J_2 + J_3
\]

(A.12)

The sublattice magnetisation is again given by the formula ((A.6)).

For small \(k\)

\[
P_k - Q_k = \frac{1}{2}(J_1 + 2J_2)k_x^2 + \frac{1}{2}(2J_2 - J_1)k_y^2 \\
+ \frac{1}{2}J_3k_z^2 + \cdots
\]

(A.13)

which shows that again there is a Goldstone mode at \(k = 0\), which is stable for \(J_2 > J_1/2\), but becomes unstable at \(J_2 = J_1/2\).

Another special case is \(k = (k_x, \pi, 0)\), when

\[
P_k - Q_k = (2J_2 - J_1)(1 + \cos k_x) + \cdots
\]

(A.14)

Hence \(\omega_k\) vanishes along this line at \(J_2 = J_1/2\), corresponding again to the transition point.

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