A Frustrated Three-Dimensional Antiferromagnet: Stacked $J_1 - J_2$ Layers

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We study a frustrated three-dimensional antiferromagnet of stacked $J_1 - J_2$ layers. The intermediate 'quantum spin liquid' phase, present in the two-dimensional 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, characterized by a strong interplay between theory and experiment. An archetypal model, which has been extensively studied, is the '$J_1 - J_2$ model', where $S = ½$ spins are located on the sites of a square lattice, with 1st and 2nd neighbour antiferromagnetic interactions J_1, J_2 [1]. This model shows two kinds of magnetic order at zero temperature, conventional Néel order for small J_2 and 'columnar' order for large J_2. In the intermediate region $0.4 < J_2/J_1 < 0.6$, strong frustration destroys long-range magnetic order and leads to a disordered 'quantum spin liquid' phase.

It has been argued [2] that the layered materials Li_2VOSiO_4 and Li_2VOGeO_4 are well represented by this model with $J_2 >> J_1$, i.e. in the columnar phase. However electronic structure (LDA + U) calculations suggest that the coupling between layers, J_3, is by no means negligible. This means that J_3 ought to be included in any fitting to experimental data. It also raises the question of the nature of the overall phase diagram of a model of coupled $J_1 - J_2$ layers. The present work is motivated by this question. We employ the well-established technique of linked-cluster series expansions at temperature $T = 0$ [3].

In Figure 1 we show the structure of the model, and a schematic phase diagram. Our calculations confirm the form of this phase diagram.

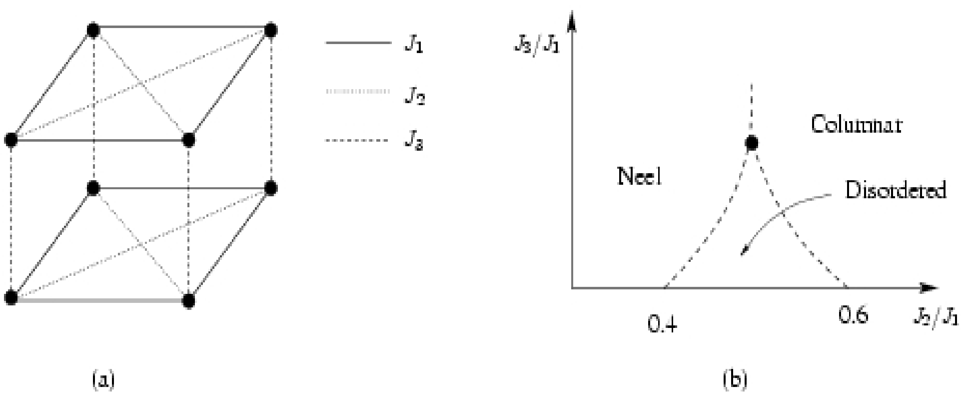

Fig. 1. (a) Tetragonal unit cell of the model; (b) Conjectured phase diagram at $T = 0$. The transition lines are (probably) first-order and the solid circle is thus a triple point.

2. Method and Results

2.1 Ground State Properties

Expansions about Néel and columnar ordered Ising ground states yield estimates of the ground state energy and magnetization for any choice of the parameters J_1, J_2, J_3. Figures 2 and 3 show these quantities versus J_2/J_1 for two values of the interplane coupling J_3.

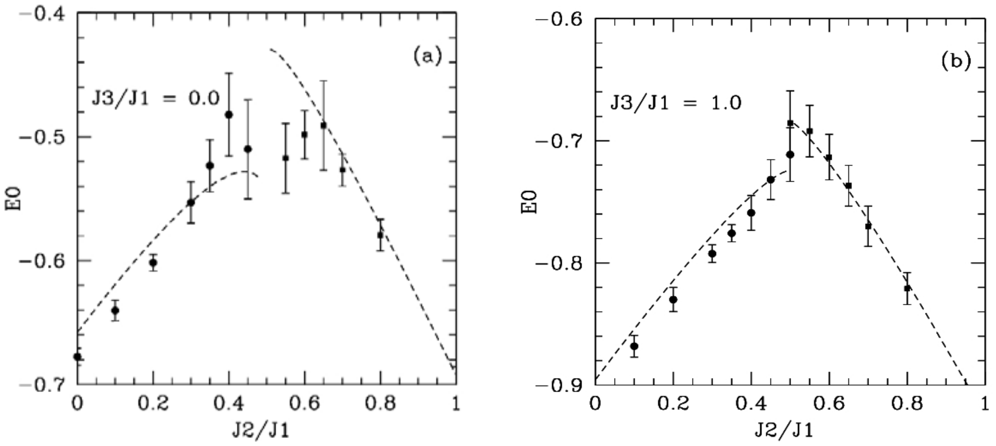

Fig. 2. Series estimates of the ground state energy per site for (a) $J_3/J_1 = 0.0$, and (b) $J_3/J_1 = 1.0$. Filled and open circles are from Néel and columnar phase expansions, respectively. The dashed lines are linear spin-wave theory predictions.

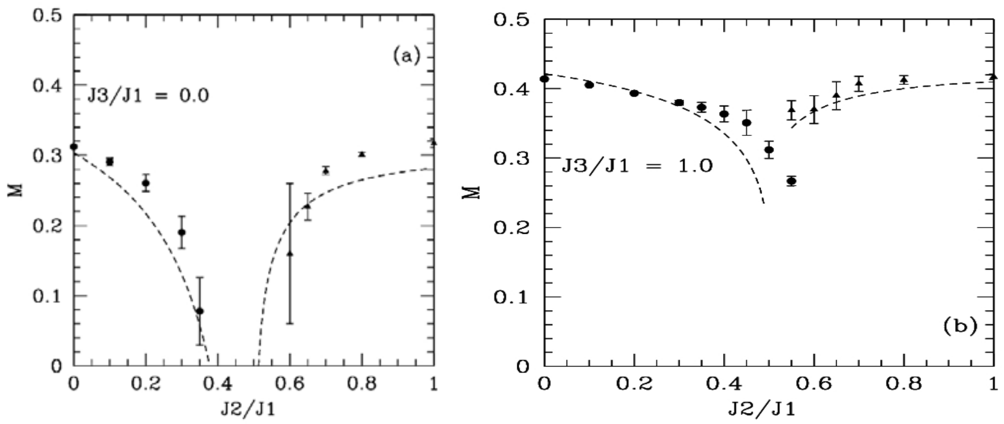

Fig. 3. Series estimates of the magnetization for (a) $J_3/J_1 = 0.0$, and (b) $J_3/J_1 = 1.0$. Filled and open circles are from Néel and columnar phase expansions, respectively. The dashed lines are linear spin-wave theory predictions.

We note a striking difference between the two cases. For $J_3/J_1 = 0$ the existence of an intermediate phase is clearly seen, particularly from the magnetization curves. On the other hand, for $J_3/J_1 = 1.0$, the energy branches clearly meet, albeit with different slopes, indicative of a direct first-order transition. The magnetization curves (Fig. 3b) show that the magnetization in each phase remains finite at the transition.

Such calculations, for a range of J_3 values, leads us to conclude that the intermediate phase shrinks to zero at a point $J_2/J_1 = 0.54 \pm 0.03$, $J_3/J_1 = 0.16 \pm 0.03$, consistent with the schematic phase diagram in Fig. 1.

2.2 Magnon Excitations

Our series technique is also able to determine the energies of magnon excitations of the model. Fig. 4 shows a set of typical results, for the Néel phase, for $J_3 = 0.5$, and two values of J_2.

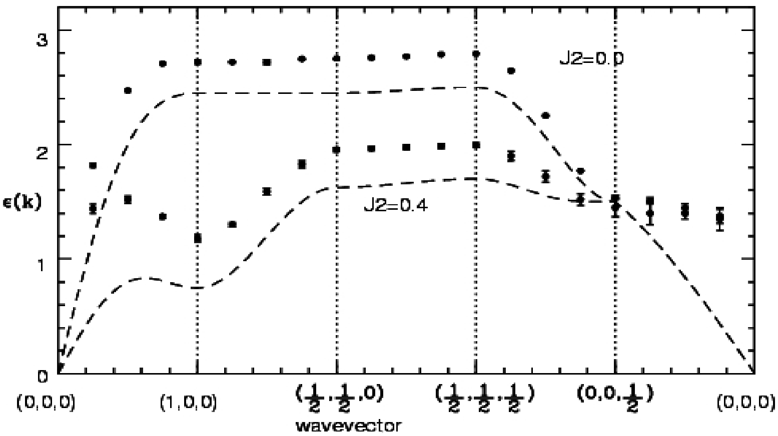

Fig. 4. Magnon dispersion curves along symmetry directions in the Brillouin zone, for $J_3 = 0.5$ and two values of J_2. The dashed lines are linear spin-wave predictions.

As can be seen in Fig. 4, we are able to identify fine details of the dispersion curves. In particular, we see a pronounced dip forming at wavevector (π,0,0) as the phase boundary is approached, and a pronounced shoulder near (0,0,π/2). Linear spin-wave theory is in qualitative agreement with the presumably more accurate series results, but generally underestimates the excitation energies.

We have also calculated dispersion curves in the columnar ordered phase. Details and examples will be given in a future paper [5].

3. Conclusions

A frustrated spin ½ model antiferromagnet, consisting of frustrated $J_1 - J_2$ antiferromagnetic layers coupled by an antiferromagnetic J_3, which is believed to provide a good description of the layered materials Li_2VOSiO_4 and Li_2VOGeO_4 has been studied by series expansion methods.

We find that the magnetically disordered spin-liquid phase, which has been identified as a prominent feature in the single layer $J_1 - J_2$ model, becomes narrower with increasing interlayer coupling J_3, and vanishes at a triple point, beyond which there is a direct first-order transition between Néel and columnar magnetic order. The location of the triple point is estimated as $J_2/J_1 = 0.54 \pm 0.03$, $J_3/J_1 = 0.16 \pm 0.03$. This is a little lower than found in an earlier calculation [4]. We also compute dispersion curves for magnon excitations, which will provide a stronger test of the applicability of the model, when and if inelastic neutron scattering data become available for these materials.

Details of these calculations will be given in a forthcoming paper [5]. In the three-dimensional model the magnetically ordered phases will persist to finite temperatures, up to some critical surface $T_c(J)$. This will be the subject of future work.

The model studied here is also of possible relevance for understanding the magnetic properties of the recently discovered iron pnictide superconductors, but this remains controversial [6].

Acknowledgments

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References

[1] Sirker J *et al.* 2006 *Phys. Rev. B* **73** 1184420; and references therein.
[2] Rosner H *et al.* 2002 *Phys. Rev. Lett.* **88** 186405; 2003 *Phys. Rev. B* **67** 014416
[3] Oitmaa J, Hamer C J and Zheng W-H 2006 *Series Expansion Methods for Strongly Interacting Lattice Models* (Cambridge Univ. Press).
[4] Schmalfuss D *et al.* 2006 *Phys. Rev. Lett.* **97** 157201
[5] Hamer C J, Rojas O and Oitmaa J, *to be submitted to Phys. Rev. B.*
[6] Holt M *et al.* 2011 Preprint arXiv:1010.5551v1