Topic area: **CMMSP**

Phase Diagram for a Triangular Lattice t-J-V Model for the Novel Superconductor Na_xCoO_2

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The recent discovery [1] of superconductivity in the layered material $Na_xCoO_2 \cdot yH_2O$, in the region $x \simeq 0.35$, $y \simeq 1.3$, following earlier observations of unusually large thermopower [2] and strong magnetic correlations [3], are indicative of strong electronic correlations. It is apparent that the physics of this material is, in many respects, similar to that of the cuprate superconductors, for which no complete theory yet exists. The important structure elements in the material are the weakly coupled CoO_2 planes. In the parent material ($x = 0$) cobalt exists as Co^{4+} with spin $S = \frac{1}{2}$. Intercalation of sodium electron dopes the CoO_2 layers, producing Co^{3+} with $S = 0$. Hydration is believed to weaken the coupling between planes, leading to an enhanced 'two-dimensionality', and presumably enhancing quantum fluctuations.

An important difference between the CoO_2 planes in this material and the CuO_2 planes in the cuprate superconductors is that the Co ions form a triangular lattice, rather than the square lattice in the cuprates. Some authors [3, 4] have argued that this will lead to new physics, in particular to the 'resonating valence bond' (RVB) picture originally suggested by Anderson as the foundation of high T_c superconductivity [5]. On the other hand there is, by now, strong evidence that the ground state of the triangular $S = \frac{1}{2}$ antiferromagnet is not a spin liquid but has long-range order [6]. Nevertheless it is possible that RVB physics might be important in the doped system.

It is clear that the phase diagram of $Na_xCoO_2 \cdot yH_2O$, which has not yet been fully explored, will be very complex. In addition to the superconducting phase (in the approximate range $0.25 < x < 0.35$), there will also be a charge-ordered insulating phase (near $x = \frac{1}{2}$) and paramagnetic and 'Curie-Weiss' metallic phases [7].

The natural theoretical model for strongly correlated electron systems is the large-U Hubbard model, or its derivative $t - J$ model. A number of such model calculations for Na_xCoO_2 have been reported [4, 8–11]. However these have all been at the mean-field level and their validity is uncertain.

We have recently used series expansion methods at zero temperature to study a modified $t - J$ model for the triangular lattice of Co ions [12]. This approach properly includes quantum fluctuations and has been used successfully in related studies. The Hamiltonian is taken to be

$$H = -t \sum_{\langle ij \rangle} P(c^\dagger_{i\sigma} c_{j\sigma} + \text{h.c.})P + J \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4}) + V \sum_{\langle ij \rangle} (1 - n_i)(1 - n_j) \qquad (1)$$

The first term represents electron hopping, with P a projection operator which excludes double occupancy of any site; the second term is the usual $t - J$ exchange term; the final term is a nearest neighbour hole-hole repulsion, as introduced in Ref. [9]. The constants t, J, V are parameters of the model. Another parameter of the model is the electron density $n =$ average

no. of electron/site. In our previous work [12], and in the present paper we choose $n = 2/3$, which corresponds to $x = 1/3$ in Na_xCoO_2, near the boundary of the superconducting phase.

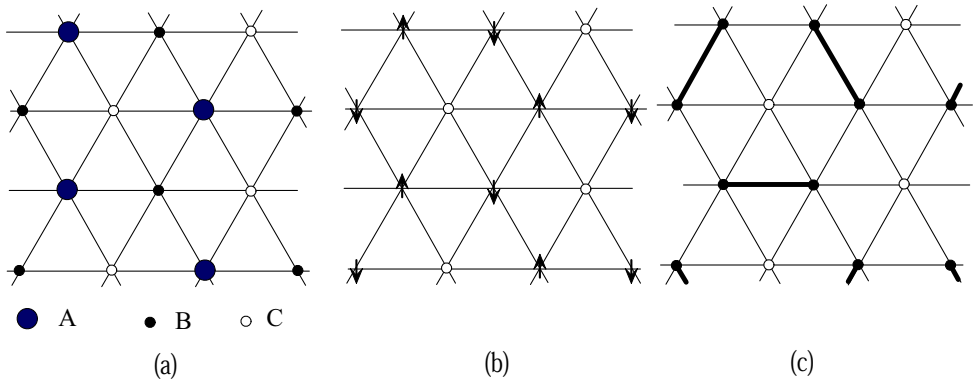

Figure 1: (a) The three sublattice structure of the triangular lattice; (b) The Néel antiferromagnetic phase with sublattice C unoccupied; (c) The dimer phase with sublattice C unoccupied.

The triangular lattice can be subdivided into three sublattices (A,B,C) as shown in Fig. 1(a). For $n = 2/3$ and large V the electrons will charge order, perfectly, occupying two of the three sublattices (chosen to be A,B). These occupied sites form a honeycomb lattice, and it is known that the fully occupied honeycomb lattice shows antiferromagnetic Néel order [13]. An interesting question is whether the ability of the electrons to hop to the third sublattice can destabilize the antiferromagnetic order or promote ground states with different symmetries. Our results show that for small V a number of different ground state phases, including charge ordered, Néel ordered, dimerized, ferromagnetic, phase separated and short-range antiferromagnetic phases can arise.

To derive long perturbation series the Hamiltonian must be decomposed in the usual way $H = H_0 + \lambda H_1$, where the unperturbed Hamiltonian H_0 is exactly solvable, and H_1 represents the remaining terms. We compute series for the ground state energy E_0, the sublattice magnetization M, the difference between sublattice electron densities and the difference between electron hopping probabilities between different sublattices. The series are extrapolated to $\lambda = 1$, the full Hamiltonian, by Padé approximant methods. From these results we are able to infer a (partial) phase diagram.

The decomposition of the Hamiltonian $H = H_0 + \lambda H_1$, has been done in two different ways. In the first approach, known as an *Ising expansion*, we take

$$H_0 = \sum_{\langle ij \rangle} [J(S_i^z S_j^z - \frac{n_i n_j}{4}) + V(1-n_i)(1-n_j)] \tag{2}$$

and the remaining terms as H_1. This is particularly appropriate for large J, where we expect antiferromagnet order. Fig. 1(b) shows the unperturbed Néel ordered ground state for this case. The Ising expansions for various quantities have been computed to order λ^{11}. The other approach, appropriate for a spin gapped phase, is to start with a dimerized ground state, in which the electrons form singlet dimers. Several dimerization patterns are possible for the

honeycomb lattice [14]. We choose the so-called *columnar* pattern, illustrated in Fig. 1(c). The *dimer expansions* have been computed to order λ^8 for various ground state properties, and for the triplet excitation spectrum.

We present here a selection of results, supplementing those in Ref. [12]. Figure 2(a) shows the ground state energy as a function of parameters V, J as obtained from Ising expansions. The dashed line shows the energy of the ferromagnetic state. The crossover indicates a transition from ferro- to antiferromagnetic or dimer order. Fig. 2(b) gives the sublattice magnetization as a function of V, J. We note the robustness of the antiferromagnetic phase for $t < 0$, whereas for $t > 0$ a transition from spin order to a metallic phase occurs. Further results are given in Ref. [12].

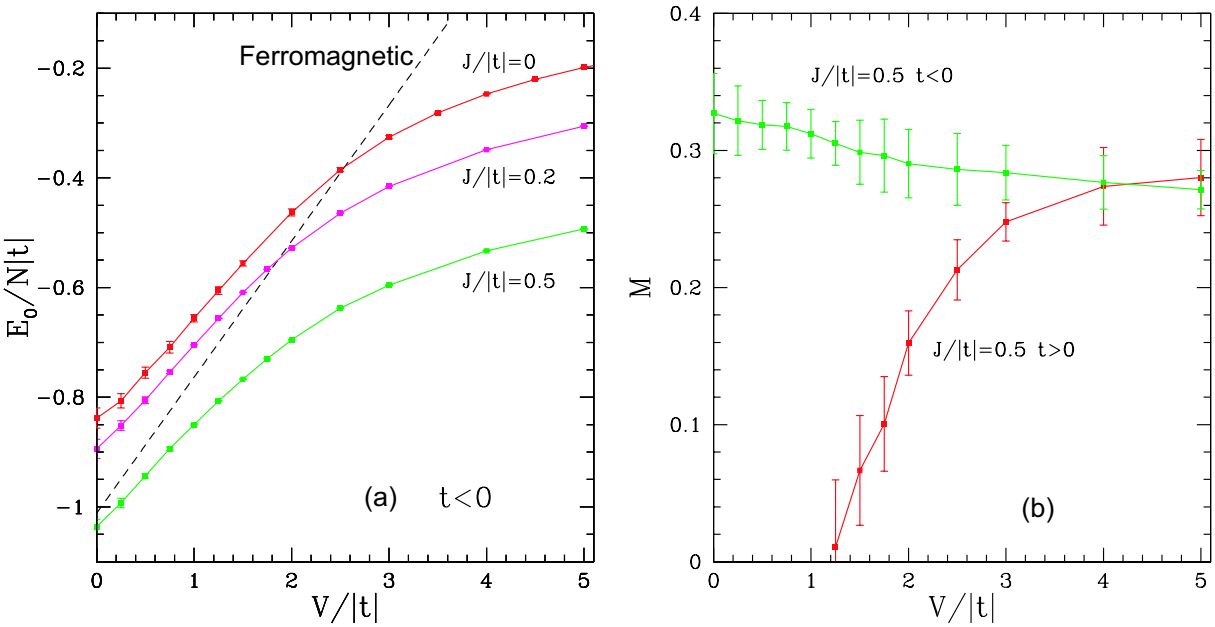

Figure 2: Results from Ising expansions: ground state energy (a) and sublattice magnetization (b) versus V for various values of J. The results are discussed in the text.

The results are summarized in a schematic phase diagram (Fig. 3). For small t the antiferromagnetic charge ordered charge ordered phase is favoured, except near $V = 0$ when phase separation (PS) occurs. The later phase is identified by comparing respective ground state energies.

We find that the sign of the hopping matrix element t plays a significant role in the properties of the system. First consider negative t. In this case, charge order is very robust, and for a substantial range of $|t|/J$ values, extends even to $V = 0$. Also increasing $|t|$ causes the magnetization on the honeycomb sites to increase well beyond the value in the pure Heisenberg model. For $V = J = 0$, the Nagaoka ferromagnetic state is the ground state. However, singlet, possibly dimerized phases compete with the Nagaoka state even at very small V and J values. For positive t, hopping reduces Néel and charge order. Below a critical V, spontaneous charge and Néel order vanish over most of $t/(J+t)$, except a small region near $t/(J+t) = 0.3$, where a weak Néel and charge order may exist down to $V = 0$. From the series expansions one cannot conclusively show that the Néel and charge order vanish simultaneously, but the numerics is consistent with a single transition. At this transition, the symmetry between the three sublattices is restored both with respect to occupancy and hopping. Also, the dimerized

and singlet states are never favoured for positive t. Thus, for small V and large t, there appears to be a phase transition to an itinerant state with short-range antiferromagnetic correlations.

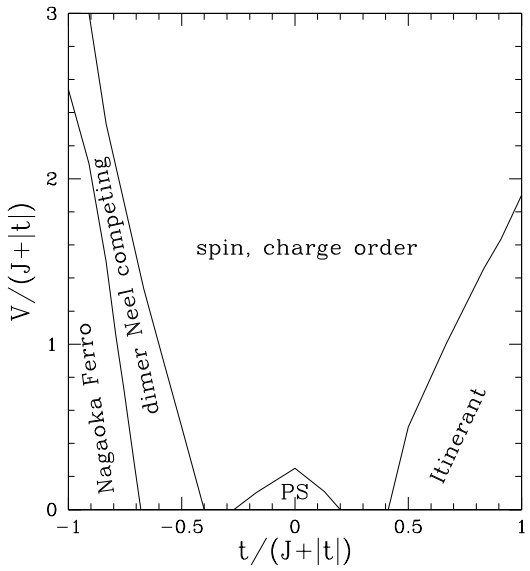

Figure 3: Schematic phase diagram for the triangular lattice $t-J-V$ model with $n = 2/3$. The various phases are discussed in the text.

The triangular lattice $t-J-V$ model with 2/3 electron density clearly has a very rich phase diagram. Variation of the electron density will certainly lead to even richer behaviour. The series method used here is not able to allow for variable doping. It is difficult to relate these calculations directly to the $Na_xCoO_2 \cdot yH_2O$ material, but our model calculations clearly exhibit some features to be expected. An important issue is the sign of t in the real material. Recent ARPES measurements [15] suggest $t < 0$.

This work is supported by a grant from the Australian Research Council and by US National Science Foundation grant number DMR-0240918.

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