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Rapid Note

Plaquette ground state in the two-dimensional SU(4) spin-orbital model

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Abstract. In order to understand the properties of Mott insulators with strong ground state orbital fluctuations, we study the zero temperature properties of the $SU(4)$ spin-orbital model on a square lattice. Exact diagonalizations of finite clusters suggest that the ground state is disordered with a singlet-multiplet gap and possibly low-lying $SU(4)$ singlets in the gap. An interpretation in terms of plaquette $SU(4)$ singlets is proposed. The implications for $LiNiO_2$ are discussed.

PACS. 75.10.Jm Quantized spin models – 11.30.-j Symmetry and conservation laws – 75.40.Mg Numerical simulation studies

Orbital degeneracy is a very common feature of Mott insulators. In most cases it is lifted by a cooperative Jahn-Teller distortion at relatively high temperature, and the low energy physics can be described by a pure spin model, with an effective Hamiltonian, hence a magnetic order, that depends on the orbital ordering [1]. In the past few years, this picture has been challenged in a number of systems, and the possibility to get a spin liquid is now well established [2–4]. But it seems that there are even more exotic systems that do not undergo a cooperative Jahn-Teller distortion in spite of the orbital degeneracy. The best example is probably $LiNiO_2$, in which no orbital or magnetic order has been detected down to very low temperature [5]. The minimal model to describe this system is the $SU(4)$ spin-orbital model defined by the Hamiltonian

$$H = J \sum_{\langle i,j \rangle} \left(2\mathbf{s}_i \cdot \mathbf{s}_j + \frac{1}{2} \right) \left(2\boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j + \frac{1}{2} \right) \qquad (1)$$

on the triangular lattice. In this model, $\mathbf{s}_i$ are spin-1/2 operators that describe the spin degrees of freedom of Ni^{3+}, while $\boldsymbol{\tau}_i$ are pseudo-spin-1/2 operators that describe the orbital degeneracy associated to the two e_g orbitals. While additional terms arising from the anisotropy in hopping integrals and the Hund's rule coupling will destroy the symmetry between spin and pseudo-spin and favour parallel alignment of the spins of a pair of neighbouring sites, the absence of ordering may be traced back to the properties of the Hamiltonian (1). As suggested by Li *et al.* [6], the ground state is a liquid of resonant plaquette $SU(4)$ singlets. Note that this model is not equivalent to the model of reference [4]. Although both models possess $SU(4)$ symmetry, the low-energy physics is completely different. Hamiltonian (1) in 1-dimension (1D) has been solved by Bethe Ansatz [7,8], by numerical methods [9–12] and field theory methods [13], and the ground state is a spin-orbital liquid. In 2D, the proposed plaquette ground state of model (1) was mainly based on variational wavefunction or mean field theory [6], and more work is clearly needed to put these ideas on a firm ground.

In this article, we present a detailed analysis of the low-energy properties of the $SU(4)$ model on the square lattice using symmetry analysis and exact diagonalizations of finite clusters. While the most relevant compound $LiNiO_2$ is a quasi-two-dimensional system made out of triangular planes, we shall start with the square lattice for simplicity. We will discuss possible differences between the triangular and the square lattices at the end. The choice of exact diagonalization as a numerical method was motivated by the fact that other methods that have been successfully used in the 1D case cannot be applied here. In particular, the Quantum Monte Carlo algorithm used by Frischmuth *et al.* [11] suffers from a severe minus sign problem in 2D lattices.

Let us start with some symmetry considerations that will be very useful throughout the paper. First of all, the $SU(4)$ symmetry implies that $s^z = \sum_i s_i^z$, $\tau^z = \sum_i \tau_i^z$ and $s\tau^z = \sum_i s_i^z \tau_i^z$ are good quantum numbers, and all numerical results have been obtained by diagonalizing

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the Hamiltonian in sectors defined by a given set $(s^z, \tau^z, s\tau^z)$. Besides, the first Casimir operator, the equivalent of the square of the total spin in $SU(2)$, is given for N sites by $C_{1\ldots N} = (1/32)(\mathbf{A}_{\rm tot})^2$, where the components of $\mathbf{A}_{\rm tot}$ are the fifteen generators of the $SU(4)$ algebra and are given by $2\sum_{i=1}^{N} s_i^\alpha$, $2\sum_{i=1}^{N} \tau_i^\alpha$ and $4\sum_{i=1}^{N} s_i^\alpha \tau_i^\beta$, $\alpha, \beta = x, y, z$. Since the Casimir operator of any irreducible representation (IR) can be easily calculated with the tools of group theory [14], this operator is useful to find out to which IR a given state belongs. The values of C for various IR's, classified according to their dimensionality d, are listed in Table 1. The Hamiltonian can also be written in terms of on site fifteen-component vectors as

$$H = \frac{J}{4} \sum_{\langle i,j \rangle} (\mathbf{A}_i \cdot \mathbf{A}_j + 1). \tag{2}$$

This allows to rewrite the Hamiltonian of several small systems in terms of the Casimir operators of sub-systems using identities such as $\mathbf{A}_i \cdot \mathbf{A}_j = (1/2)(32 C_{ij} - \mathbf{A}_i^2 - \mathbf{A}_j^2)$. In this case, all eigenvalues and degeneracies can be deduced from the possible IR's for each sub-system. As we shall see below, this allows a full diagonalization for systems with 2 and 4 sites, as well as for 8 sites with periodic boundary conditions where the the dimension of the Hilbert space is already 65 536.

Table 1. Dimension (d) and Casimir operator eigenvalue (C) of some irreducible representations of $SU(4)$.

d	1	4	6	10	15	20	20	20	35	45
C	0	15/32	5/8	9/8	1	3/2	39/32	63/32	3	2

Let us now present the results we have obtained for several systems. Since the interpretation we will give at the end of the paper heavily relies on the properties of the 2 and 4 sites clusters, we include them here for convenience.

(i) *Two sites (pair):* in terms of spins and orbitals, the ground state is 6-fold degenerate (spin singlet × orbital triplet and *vice versa*) with energy $-J$. The other 10 states are degenerate with energy $+J$. In $SU(4)$ language, this means that the only accessible IR's have dimension 6 and 10. So it is impossible to build an $SU(4)$ singlet with only two sites, as already emphasized by Li *et al.* [6].

(ii) *Four sites (plaquette):* the Hamiltonian can be rewritten as

$$H = 4J(C_{1234} - C_{13} - C_{24} + 1/4). \tag{3}$$

The ground state is an $SU(4)$ singlet with the pairs (13) and (24) in the IR of dimension 6, and its energy is $-4J$. It minimizes the energy per bond and is thus a very stable object. The first excited state is 50-fold degenerate with energy $-2J$. This corresponds to twice the adjoint IR of dimension 15 with the pairs (13) and (24) in the IR's of dimension 6 and 10 (resp. 10 and 6) and to one the IR's of dimension 20 with both (13) and (24) in the IR of dimension 10. Several pictures of the ground state, all useful for some purpose, can be given. The first one is the fermionic representation of reference [6] and corresponds to the linear combination of all possible configurations with all 4 sites different, the relative coefficients being the sign of the permutation. One can also write this wavefunction as an antisymmetric combination of spin $SU(2)$ singlets along the horizontal bonds times orbital $SU(2)$ singlets along the vertical bonds minus the bond exchanged state (see Fig. 1).

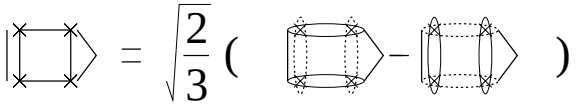

Fig. 1. The $SU(4)$ singlet on a four site cluster in terms of spin (solid) and orbital (dotted) $SU(2)$ singlets.

Finally, if one considers the 4-site cluster as two coupled pairs, the ground state can be written in terms of pair ground state only. Since the ground state minimizes the energy of each bond, this means that the energy of the bonds that couple the pairs is completely recovered by lifting the degeneracy of the ground state manifold of two independent pairs. This is another way to understand why the plaquettes play such a special role.

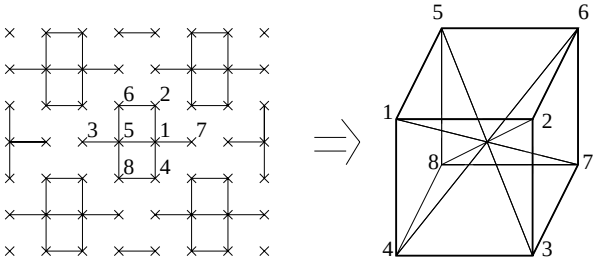

Fig. 2. Embedding of the 8-site cluster in the square lattice, and equivalent connectivity to a cube with diagonals.

(iii) *Eight sites:* the eight-site cluster with periodic boundary conditions has the topology of a cube with diagonals (see Fig. 2). This allows one to write the Hamiltonian as

$$H = 4J(C_{1\ldots8} - C_{1368} - C_{2457} + 1). \tag{4}$$

The ground state is a four-fold degenerate $SU(4)$ singlet of energy $-8J$ with the sets (1368) and (2457) in the IR of dimension 20 which is realized twice for 4 sites [6] and has a Casimir equal to 3/2. The first excited singlet has energy $-4J$ and is highly degenerate. It is above the first multiplet $(-6J)$, so that there are exactly 4 singlets below the first multiplet.

(iv) *Sixteen sites:* for that cluster, the only way to get the spectrum is to perform exact diagonalizations. With 4 degrees of freedom per-site, the numerical task is roughly equivalent to 32 sites for spin 1/2, and using the current facilities, this is the largest cluster we could do. To reduce the size of the Hilbert space, we used the 3 $SU(4)$

quantum numbers as well as spatial symmetries. The results are given in Figures 3b and 4. The ground state is a non-degenerate singlet of energy $-17.351J$. The first excitations are singlets. There are three singlet excitations below the first multiplet. The dispersion of the singlets shows a clear minimum at the X-point, *i.e.* $\mathbf{k} = (0, \pi)$ and $(\pi, 0)$.

Let us now discuss these results. The first striking feature is that some basic quantities have a very small size dependence between 8 and 16 sites [15]. For instance, the ground-state energy per site is $-J$ for 8 sites and $-1.084425J$ for 16 sites. More interestingly, the singlet-multiplet gap is equal to $2J$ for 8 sites and $1.999809J$ for 16 sites (see Fig. 3). This is a clear evidence that there is a very short correlation length in the system. Besides, although we have results for only two different-size systems, the fact that the singlet-multiplet gap is almost a constant within 10^{-4} strongly suggests that it will remain finite in the thermodynamic limit. This suggests that the ground state does not have long-range order, and we must be dealing with some kind of spin-orbital liquid.

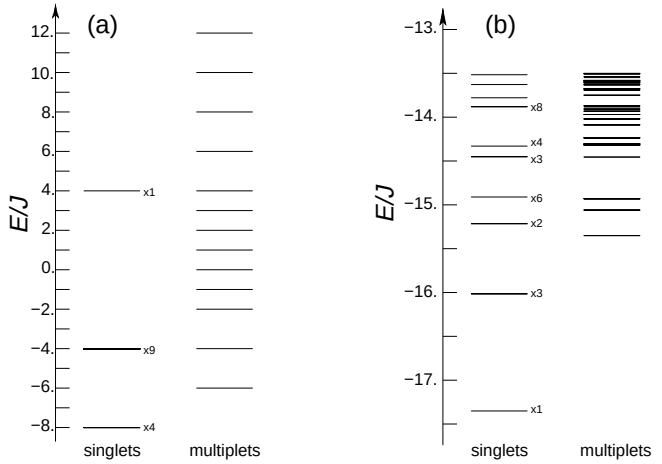

Fig. 3. Distribution of the low-lying states in the singlet *vs.* multiplet representations of $SU(4)$ on the 8-site cluster (a) and on the 16-site cluster (b). The degeneracy is indicated for the low-lying singlets.

To characterize this spin-orbital liquid, we have analyzed in more details the singlet sector. Let us start with the 8-site cluster. The ground state energy is twice the energy of a 4-site plaquette. This suggests that the plaquette picture of Li *et al.* [6] should be a very good starting point. In an 8-site cluster, there are 18 plaquette coverings, which generate a Hilbert space of dimension 14. This is identical to the dimension of the singlet subspace as we checked by diagonalizing the total Casimir operator. So the ground-state must be a linear combination of the plaquette coverings. Note that a single plaquette covering is not a ground state of the Hamiltonian: using the fermionic representation of the plaquette ground state, one can easily check that the mean value of the Hamiltonian in a plaquette covering is only $-6J$, quite far from the

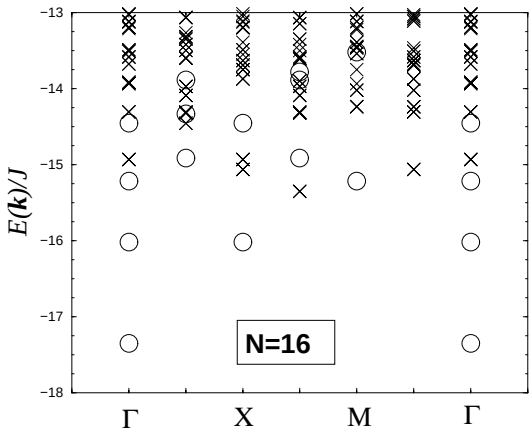

Fig. 4. Dispersion of low-lying states of the $N = 16$ $SU(4)$ spin-orbital model on a square lattice, through the 6 non-equivalent points of the first Brillouin zone. Circles stand for $SU(4)$ singlet states, and crosses for $SU(4)$ multiplets.

ground-state energy $-8J$. So the energy gained through the resonance between plaquette coverings is crucial to get $-J$ per site in the ground state. In other words, the exact ground state of the eight-site cluster is a resonant plaquette state, a generalization of the resonant-valence bond studied in the context of high temperature superconductivity. For the 16-site cluster, this picture turns out to remain very accurate. In that case, there are 30 plaquette coverings, which is much smaller than the dimension of the total singlet subspace (24 024). To check whether this subspace provides a good variationnal basis for the ground state, we have numerically calculated the projection of the ground state on an orthonormalized basis of this resonant plaquette subspace, and we find that the weight of the ground-state in that subspace is 93.5% of the total weight. In view of the small relative size of this resonant plaquette subspace, this number is quite impressive.

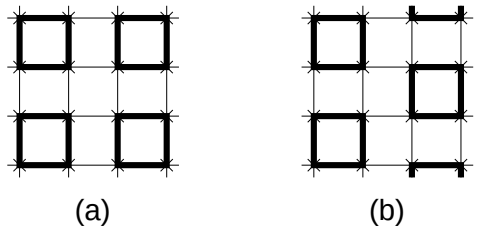

Fig. 5. Two different kinds of plaquette coverings on the square lattice: (a) a symmetric square lattice of plaquettes; (b) a column (shifted) covering.

Another argument in favour of this resonant plaquette description is provided by the dispersion of the 16-site cluster. First of all, we note that 30% of the weight is actually carried by the 4 symmetric plaquette coverings. They correspond to the plaquette covering of Figure 5a, and the three coverings deduced by translation. Now the natural singlet excitations from such a state consist of shifting either rows or columns of plaquettes (see Fig. 5b). Note

however that it is impossible to do both simultaneously. So we expect to find quasi-1D low-lying singlet excitations along $(0, \pi)$ and $(\pi, 0)$. This is precisely the case for the 16-site cluster shown in our numerical calculations, with a clear local minimum at the X-point for singlet excitations.

Since we have good evidence that a singlet-multiplet gap will remain in the thermodynamic limit, it is natural to ask whether singlet excitations will remain within this gap in the thermodynamic limit, as for instance in the case of the $SU(2)$ antiferromagnet on the Kagomé lattice [16]. While we cannot decide whether some singlets will remain within the gap, it is quite unlikely that they will form a continuum, like for the Kagomé lattice: the number of low-lying singlets does not increase from 8 to 16 sites – it is equal to 4 in both cases – and they are shifted to higher energy on going form 8 to 16 sites. This result might be explained by a simple counting argument. Because of the impossibility to shift simultaneously rows and columns, the number of plaquette coverings of the square lattice does not increase exponentially with the size of the system, but with the square root of the size. So we do not expect to be able to build a continuum with a number of states that remains significant in the thermodynamic limit. This should be contrasted to the Kagomé case, where a simple counting argument could reproduce the exponential increase of the number of low-lying singlets [17]. In that respect, the $SU(4)$ model on the triangular lattice might be different. In that case, it is possible to generate new plaquette coverings by local modifications (see Fig. 6), and the number of plaquette coverings increases exponentially with the number of sites. So, if plaquette coverings provide the lowest singlet states for the triangular lattice, a continuum of low-lying $SU(4)$ singlets might be present in the singlet-multiplet gap. This would be consistent with the specific heat data of $LiNiO_2$, where no gap was observed [5].

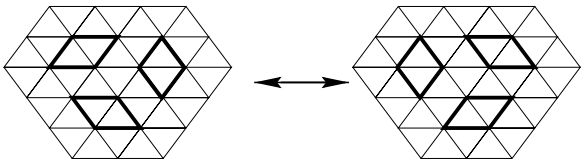

Fig. 6. Two plaquette coverings of the triangular lattice obtained by local permutation of three plaquettes.

Finally, let us comment on the physical properties of Mott insulators that can be described by a 2D spin-orbital model in the vicinity of the $SU(4)$ symmetry. The correlation length appears to be so short, and the singlet-multiplet gap so large, that small perturbations will not close the gap. So in the absence of coupling to other degrees of freedom the system is expected to remain disordered with a gap to magnetic excitations. Besides, the cooperative Jahn-Teller mechanism that lifts the orbital degeneracy will also have to overcome this singlet-multiplet gap since the $SU(4)$ singlet ground-state of a plaquette explicitly requires two orbital degrees of freedom (see Fig. 1). As a consequence, the orbitals will not order unless the electron-phonon coupling is strong enough. This picture is consistent with the properties reported so far for $LiNiO_2$ [5]. As we noticed above, the structure of the singlet sector might be different for the triangular lattice, however. The analysis of this model is under progress.

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