Topological Phases emerging from Spin-Orbital Physics

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Abstract We study the evolution of spin-orbital correlations in an inhomogeneous quantum system with an impurity replacing a doublon by a holon orbital degree of freedom. Spin-orbital entanglement is large when spin correlations are antiferromagnetic, while for a ferromagnetic host we obtain a pure orbital description. In this regime the orbital model can be mapped on spinless fermions and we uncover topological phases with zero energy modes at the edge or at the domain between magnetically inequivalent regions.

Keywords Spin-orbital order · Charge dilution · Doped Mott insulator · Majorana modes

1 Introduction

Transition metal oxides are fascinating materials where several degrees of freedom (i.e., spin, orbital, charge, etc.) couple and, from a theoretical point of view, need to be treated on equal footing in order to provide reliable predictions. In undoped 3d Mott insulators large on-site Coulomb interactions localize electrons and the coupling between transition metal ions is controlled by a low-energy spin-orbital superexchange introduced first by Kugel and Khomskii [1]. As for spins, orbital degrees of freedom have a quantum character and can drive strong fluctuations which end in destroying long range order [2] or lead to exotic novel types of magnetic order [3]. However, such cases are rare in $e_g$ systems and, typically, long range order in both spin and orbital sector develops in perovskite lattices [4], with the corresponding correlations following the Goodenough-Kanamori rules [5]. A well known example is the spin-orbital order in LaMnO$_3$ [6], with different energy scales for spin and orbital order [7]. However, there are numerous deviations from these rules caused either by superexchange on non-linear bonds [8], or by lattice frustration such as for instance in LiNiO$_2$ [9], or by spin-orbital entanglement [10], or, finally, by the presence of next nearest neighbor hopping [11]. In $t_{2g}$ systems, orbital superexchange has leading contributions with SU(2) symmetry along a given cubic direction thus orbital fluctuations are much stronger [12] than in $e_g$ and a spin-orbital liquid emerging from intrinsic frustration is more likely to occur [13][14][15]. On the other hand, ordered states may be even stabilized by orbital fluctuations [16] as for instance in LaVO$_3$ [17] and Ca$_2$RuO$_4$ [18] — in this latter case spin-orbit coupling also plays a role [19]. Quantum fluctuations and spin-orbital entanglement [20] are of great importance in this class of materials and may lead to novel phenomena as superconductivity in the pnictides driven by competing symmetries at orbital degeneracy [21], or spectacular topological structure of the excited states in the one-dimensional (1D) SU(2)$\otimes$XY model [22], or, finally, dimerised phases [23].

Doping of Mott insulators adds another charge degree of freedom in spin-orbital systems and leads to several remarkable phenomena. Recently short range charge-density wave called stripe phase was reported in doped cuprates [24]. It has been suggested that the critical charge, orbital, and spin fluctuations near the quantum critical point provide the pairing interaction
In doped cuprates, the holes doped in $t_{2g}$ orbitals may be mobile due to three-site terms \([26]\) or self-organization in stripe phases \([27]\). However, the formation of orbital molecules makes 1D insulating zigzag states kinetically more favorable than metallic stripes \([28]\). Insulating state is also found \([29]\) when holes are confined near charge defects in $Y_{1-x}Ca_xVO_3$ \([30]\).

In contrast, neutral defects in spin-orbital systems lead to orbital dilution (with a local increase of spin to $S = \frac{3}{2}$) \([31]\) and the changes in spin-orbital order \([32]\), or charge dilution \([33]\) (with invariant spin $S = 1$ states). These phenomena are distinct from the orbital dilution in cuprates where holes remove simultaneously spin and orbital degree of freedom \([34]\). The $t_{2g}$ systems with charge dilution are unexplored yet — they will likely play a major role in future functional materials and, possibly, in novel electronic devices. The purpose of the paper is to investigate the consequences of charge dilution in a $t_{2g}$ system due to the substitution of a $d^0$ by a $d^2$ transition metal ion. Such type of doping allows to uniquely design a spin-orbital correlated environment with an orbital degree of freedom having an inequivalent charge character. Indeed, for $d^2$ and $d^4$ valence configurations, the empty orbital (i.e., holon) and the doubly occupied state (i.e., doublon) set the orbital degree of freedom, respectively. As an experimental motivation we mention, among the various emergent phenomena and the many possible hybrid oxides that we consider, the formation of orbital molecules makes 1D insulating zigzag states \([35]\), and \((\text{ii})\) Mn-substituted single crystals of $Sr_2Ru_2-xMnxO_7$ rapidly drive an unusual metal-insulator transition and E-type antiferromagnetic (AF) order at low doping \([36]\). The theoretical search for the consequences of holon-doublon substitution is performed for a 1D ring and we analyze both spin and orbital correlations around the charge defect. We give reasons why the FM regime is well designed to search for topological aspects of the present model.

### 2 Spin-orbital physics and charge dilution

We consider a 1D ring made of $d^4$ transition metal ions in the insulating regime, with one $d^2$ charge defect, see Fig. 1. The physics of the undoped system is governed by a spin-orbital superexchange model which is equivalent, through an electron-hole transformation, to that introduced for vanadates \([37]\). It depends on two Kanamori’s parameters: the intraorbital Coulomb element $U_2$ and Hund’s exchange $J_2$ for $t_{2g}$ electrons \([3]\), responsible for the high spin states with spin $S = 1$ at the host $d^4$ ions. In the regime of strong electron interactions we obtain a spin-orbital model with spin $S = 1$ at every site and an orbital degree of freedom described by a pseudospin $T = \frac{1}{2}$. Since we work in one dimension and with $t_{2g}$ orbitals, we select the cubic axis $c$ with the active orbitals \([16]\): $|a\rangle \equiv |yz\rangle$ and $|b\rangle \equiv |xz\rangle$.

However, the situation becomes less familiar when some of the $d^4$ ions are substituted by the $d^2$ ones. In the regime of low doping, all the bonds will be either between two $d^4$ ions, called host bonds, or between $d^2$ and $d^4$ ions around an impurity site — these we call hybrid bonds. The superexchange Hamiltonian for both kinds of bonds has a generic form (all the bonds are along the cubic axis $c$ in the 1D chain) \([32]\).

$$H_b = J_b \sum_{\langle i,j \rangle} \left\{ K^{(b)}_{i,i+1}, S_i \cdot S_{i+1} + Q_{i,i+1}^{(b)} \right\},$$

(1)

where the label $b = \{0, h\}$ stands for the type of bond and the operators \(\{K^{(0)}_{i,i+1}\}\) and \(\{Q_{i,i+1}^{(0)}\}\) act in the orbital space at two sites, \(\{i, i+1\}\). These operators differ fundamentally for the hybrid and host bonds, i.e., for the host they take the U(1) symmetric form of,

$$K^{(0)}_{i,i+1} = A_K \tau^x_i \tau^x_{i+1} + B_K \left( \tau^x_i \tau^x_{i+1} + \tau^y_i \tau^y_{i+1} \right) + C_K,$$

(2)

$$Q_{i,i+1}^{(0)} = A_Q \tau^x_i \tau^x_{i+1} + B_Q \left( \tau^x_i \tau^x_{i+1} + \tau^y_i \tau^y_{i+1} \right) + C_Q,$$

(3)

whereas for the hybrid bonds the symmetry is lowered,

$$K^{(h)}_{i,i+1} = D_K \tau^x_i \tau^x_{i+1} + E_K \tau^y_i \tau^y_{i+1} + F_K \tau^z_i \tau^z_{i+1} + G_K,$$

(4)

$$Q_{i,i+1}^{(h)} = D_Q \tau^x_i \tau^x_{i+1} + E_Q \tau^y_i \tau^y_{i+1} + F_Q \tau^z_i \tau^z_{i+1} + G_Q.$$  

(5)

Here, $\tau^x$, $\tau^y$, and $\tau^z$ are the Pauli operators describing doublon/holon fluctuating between $|a\rangle$ and $|b\rangle$ orbitals. They are defined by the Pauli matrices $\sigma$ as

$$\tau_i = \left( a_i^T b_i^T \right) \cdot \sigma \cdot \left( a_i b_i \right)^T,$$

(6)

where hardcore boson operators $a_i$ and $b_i$ create holon or doublon in the orbitals $|a\rangle$ and $|b\rangle$, respectively.

Coefficient $J_b$ in the spin-orbital model \([14]\) is a superexchange constant and is given by $J_b = 4U_2/\Delta$ and $J_b = 2T^2/\Delta$ (note that the excitations which provide the main contribution on the hybrid bond go in one
direction only), where \( t_0 = (b = 0, h) \) is a hopping amplitude along host-host or impurity-host bond, and \( U_2 \) is the Hubbard interaction for the host \((d^3)\) atoms. \( \Delta \) is a typical excitation energy scale in the virtual process \( d^2d^1 \rightarrow d^3d^3 \) (or charge transfer energy) given by,

\[
\Delta \equiv L_c + 2U_1 - 3U_2 - 6(J_1 + J_2). 
\]  

(7)

Here \( U_1 \) and \( J_1 \) are Hubbard and Hund’s interactions at the \( d^2 \) impurity site, \( L_c \) is the energy mismatch of the electronic levels at \( b \); since \( \Delta \) must be positive and relatively large, this implies that \( L_c > U_1 \) must be the largest energy scale in the system. The coefficients \( A_{K(Q)}, \ldots, C_{K(Q)} \) and \( D_{K(Q)}, \ldots, G_{K(Q)} \) in Eqs. 2–5 are numerical constants depending on microscopic parameters of the ions: \( A_{K(Q)}, \ldots, C_{K(Q)} \) depend only on host’s parameter \( \eta_h \) and \( D_{K(Q)}, \ldots, G_{K(Q)} \) both on host’s and impurity’s Hund’s exchange \( \eta_1 \) and \( \eta_2 \), where

\[
\eta \equiv \frac{J_2}{U_2}, \quad \eta_1 \equiv \frac{J_1}{\Delta}, \quad \eta_2 \equiv \frac{J_2}{\Delta}.
\]  

(8)

All these \( \eta \)'s measure the relative strength of Hund’s exchange with respect to typical excitation energy — in case of host bonds it is \( U_2 \) whereas for hybrid bonds it is \( \Delta \). The exact functional forms of these coefficients are complicated and will be reported elsewhere.

The properties of the host and hybrid bonds are the following: a single host’s bond is always FM in spin and AF in orbital sector because of an orbital singlet which is formed on a bond [10]. This however is not stable when there are more than one bond — for a longer 1D system as for \( L = 8 \) chain considered here we find AF spin correlations for low \( \eta \) turning FM in a high \( \eta \) limit. The case of a hybrid bond is much simpler: despite the complicated form of the Hamiltonian [1] it always gives AF spin correlations accompanied by FM \( \langle \tau_i^+ \tau_{i+1}^- \rangle \) orbital correlations. Because of these intrinsic difference between host and hybrid bonds it is essential to check the ground state properties of a finite system with single impurity, see Fig. 1.

In Fig. 2 we show the ground state spin and orbital correlations obtained for a closed chain of \( L = 8 \) sites with a single \( d^2 \) impurity, see Fig. 1. The results are shown as functions of \( \eta \) for fixed values of \( J_0, J_h, \eta_1, \) and \( \eta_2 \) which weakly influence the overall behavior. Due to translational invariance one finds four inequivalent bonds, see Fig. 1. There are two regimes: (i) AF with total spin \( S = 2 \) \((\sum_i S_i^z = 2)\) for \( \eta < 0.09 \), (ii) FM with \( S = 6 \) \((\sum_i S_i^z = 6)\) for \( \eta > 0.09 \) (however the hybrid impurity bonds are always AF). In the AF regime at \( \eta = 0 \) all the spin correlations are AF, but a level crossing occurs at \( \eta = 0.033 \) where the magnetic moment delocalizes from the impurity to its two neighbors, remaining nearly constant within these three sites.

Surprisingly, for increasing \( \eta < 0.09 \) the spin correlations between second and third neighbors of impurity become weak FM due to spin-orbital entanglement, but the remaining spin correlations are AF. In the FM regime all the host bonds have almost saturated FM spin correlations, \( \simeq +1 \), while they tend to the classical value of \( -1 \) for increasing \( \eta \) on hybrid bonds, see Fig. 2(b). The orbital \( \langle \tau_i^+ \tau_{i+1}^- \rangle \) correlations behave more regularly; they are AF for host bonds and FM for hybrid bonds in both regimes of \( \eta \), see Fig. 2(b). For the off-diagonal orbital correlations we define the conventional \( \tau_i^\pm \) operators as \( \tau_i^\pm \equiv \frac{1}{2}(\tau_i^x \pm i\tau_i^y) \) (here \( \tau_i^{x,y} \) are normalized to \( \pm 1 \)). It turns out that \( \langle \tau_i^+ \tau_{i+1}^- \rangle \) correlations are significant only for the host bonds and \( \langle \tau_i^+ \tau_{i+1}^- \rangle \) only for hybrid bonds and they are always AF, see Fig. 2(c).

To investigate the spin-orbital entanglement we introduce covariances for the various correlators,

\[
C_{\sigma}^{\tau_\tau_{i+1}} = \langle S_iS_{i+1}\tau_i^\sigma \tau_{i+1}^\sigma \rangle - \langle S_iS_{i+1} \rangle \langle \tau_i^\sigma \tau_{i+1}^\sigma \rangle, \quad \sigma = \pm
\]

(9)

\[
C_{\sigma}^{\tau_\tau_{i+1}} = \langle S_iS_{i+1}\tau_i^\sigma \tau_{i+1}^\sigma \rangle - \langle S_iS_{i+1} \rangle \langle \tau_i^\sigma \tau_{i+1}^\sigma \rangle + H.c.
\]

(10)

with \( \sigma = \pm \). In Fig. 2 we show the spin-orbital covariances in the AF and FM regime. One finds that both
Fig. 3 Ground state spin-orbital covariances \( C_{i,i+1}^{\alpha,\beta} \) on the bonds for a closed chain system of length \( L = 8 \) (Fig. 1), obtained via exact diagonalization as functions of \( \eta \) for orbital correlations: (a) \( \langle \tau_i^x \tau_{i+1}^x \rangle \); (b) \( \langle \tau_i^x \tau_{i+1}^y \rangle \) for the host (hybrid) bonds. Left (right) column — AF (FM) host with small (large) \( \eta \). Parameters: \( J_0 = 1, J_h = 2, \eta_1 = \eta_2 = 1 \).

Fig. 4 Orbital model results obtained in Hartree-Fock (Eq. 11) in the regime of FM host as a function of \( \eta_2 = J_2/\Delta \): (a) bond correlations \( \langle \tau_i^x \tau_{i+1}^x \rangle \) and \( \pm \langle \tau_i^y \tau_{i+1}^y \rangle \) (solid and dashed lines), with color convention as in Fig. 1; (b) magnified view of (a) for \( \eta_2 \to 0 \); (c) orbital couplings \( A_{xx} \) and sum \( A_{xx} + A_{yy} \) (solid and dashed) for hybrid bonds, and (d) topologically relevant quantities, \( \Im D_{x/2} \) and \( D_{0} D_{x} \) (solid and dashed). Parameters: \( J_0 = 1, J_h = 2, \eta_1 = 4, \eta = 0.2 \).

3 Topological states in the orbital model

Factorization of spin and orbital degrees of freedom is allowed in the FM regime and leads to an effective orbital-only Hamiltonian,

\[
H_{i,j}^0 = \frac{1}{4} J_0 \frac{1}{1 - 3\eta} \tau_i \tau_j, \\
H_{i,j}^h = J_h (A_{xx} \tau_i^x \tau_j^x + A_{yy} \tau_i^y \tau_j^y + A_{zz} \tau_i^z \tau_j^z),
\]

(11)

for the host and hybrid bonds, respectively. This purely orbital Hamiltonian can be mapped on spinless fermions by the Jordan-Wigner transformations.

For symmetry reasons we find an exact relation \( A_{zx} \equiv -A_{xz} \), and we also get that \( A_{xx} \) and \( A_{yy} \) almost compensate each other so their sum \( A_{xx} + A_{yy} = \delta \) has a relatively small amplitude. It is however important to point out that \( \delta \neq 0 \) because in the representation of Jordan-Wigner fermions \( \delta \) is proportional to the pairing amplitude and its finite value can induce a topological non-trivial state. All the \( \{ A_{\alpha,\alpha} \} \) coefficients are functions of \( \eta_1 \) and \( \eta_2 \), while we find that the dependence on \( \eta_1 \) is very weak. Thus, we fix \( \eta_1 = 4 \) (\( \eta \) is already fixed as \( \eta = 0.2 \)) and we show the behavior of the \( A_{\alpha,\alpha} \) coupling in Fig. 3(c). We note that \( \eta_2 = 0 \) is a high symmetry point where \( A_{\alpha,\alpha} \equiv 0.5 \) for any \( \eta_1 \).

Hence, by means of the Hartree-Fock decoupling we deal with fermion-interaction term \( \langle \tau_i^x \tau_{i+1}^x \rangle \) in a self-consistent manner and we obtain the bond \( \langle \tau_i^x \tau_{i+1}^x \rangle \) orbital correlations for a periodic (and infinite) system with one \( d^2 \) impurity per every \( L = 8 \) sites. We find that in the present parameter regime the \( \langle \tau_i^x \tau_{j+1}^x \rangle \) vanish and one gets only the kinetic terms \( \langle \tau_i^x \tau_{i+1}^x \rangle \) and \( \langle \tau_i^y \tau_{i+1}^y \rangle \). For the host bonds they are all AF while for the hybrid ones the \( xx \) correlations are AF and \( yy \) ones are FM, see Fig. 3(a). Interestingly, we obtain a discontinuous transition at \( \eta_2 = 0^+ \) between anisotropic and isotropic phases — the difference between \( xx \) and \( yy \) correlations is triggered by any finite \( \eta_2 \), see Fig. 3(b). Finally, at finite \( \eta_2 \) one always gets a regime with a non-trivial topological phase with respect to the Jordan-Wigner fermionic representation.

Indeed, the fermionic Hamiltonian in the momentum space is given by a matrix \( H_k \) that belongs to the BDI Altland-Zirnbauer class [35]. Thus, it can have a non-trivial \( Z \) topological number. The topological invariant can be determined by looking at \( H_k \) in the
eigen-basis of the chiral symmetry where it consists of two anti-diagonal blocks $u_k$ and $u_k^\dagger$. The determinant of $u_k$, $D_k \equiv \det u_k$, is a complex number which yields a non-trivial topological number if it winds around the $(0, 0)$ point in the complex plane as $k$ changes from 0 to $2\pi$. In present case this happens if: (i) the imaginary part of $u_{k/2}$ is non-vanishing and (ii) the determinants $D_0$ and $D_{\pi}$ have opposite signs. In Fig. 5(d) we observe indeed these conditions hold as long as $\eta_2 > 0$.

Recently, the topological phase diagram of a 1D tight-binding model of spinless electrons with an inhomogeneous distribution of pairing centers has been investigated [39]. The Hamiltonian includes inhomogeneities generated by diluted pairing centers with a given distribution profile in the unit cell of length $L$. For a periodic configuration with momentum $k$ we get,

$$\mathcal{H} = \sum_{p=1}^{L} \left\{ t_p c_{k,p}^\dagger c_{k,p+1} + \Delta_p c_{k,p}^\dagger c_{-k,p+1}^\dagger + \mu_p c_{k,p}^\dagger c_{k,p} + \text{H.c.} \right\},$$

(12)

with $c_{L+1,k} \equiv e^{ik} c_{1,k}$ and $\{\Delta_p\}$ being the nearest neighbor hopping and on-bond pairing amplitudes. There, we have found the topological invariant that can be generally expressed in terms of the physical parameters for any pairing center configuration [39].

Here, we emphasize the occurrence of edge states and present the spectra around zero energy for a closed and open system, see Figs. 5(a) and 5(b). We note that for an open system there are two zero-energy states appearing in the gap. These are Majorana end modes that arise as a consequence of the bulk-boundary correspondence in a topologically non-trivial configuration. In Figs. 5(c) and 5(d) the spatial occupation probabilities $\rho_i$ for the two zero energy states are explicitly shown in order to confirm their degree of localization on the right/left edges of the 1D chain. We also point out that the modification of the kinetic term with the inclusion of long-range hopping is expected to lead to multiple Majorana end modes both in spinless [40] and spinfull $p$-wave superconducting chains [11].

4 Discussion and Summary

In conclusion, we have studied a one-dimensional hybrid $d^2$-$d^4$ system with a single $d^2$ spin-orbital correlated host. Remarkably, the exact diagonalization analysis allows to single out regimes for which the orbitals and spins can be factorized if the host configuration is FM. By this decoupling one finds interacting orbital pseudospins exhibiting fully isotropic exchange for the host bonds and fully anisotropic for the hybrid ones. A Jordan-Wigner transformation and Hartree-Fock decoupling allow, then, to map the system on non-interacting fermions and to find topological non-trivial states. Unexpectedly, a topological non-trivial state occurs for any finite value of $J_2$, i.e., the amplitude of the Hund’s coupling at the host’s ions. For a long chain we explicitly demonstrate that Majorana-like modes occur at the edge of the system. We argue that inhomogeneous topological patterns [12] can be achieved in the present spin-orbital scenario with Majorana modes occurring, for instance, at the boundary of the FM region if the impurities drive a magnetic configuration that has alternating FM with AF domains.

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