

Absence of Hole Confinement in Transition-Metal Oxides with Orbital Degeneracy

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We investigate the spectral properties of a hole moving in a two-dimensional Hubbard model for strongly correlated t_{2g} electrons. Although superexchange interactions are Ising-like, a quasi-one-dimensional coherent hole motion arises due to effective three-site terms. This mechanism is fundamentally different from the hole motion via quantum fluctuations in the conventional spin model with SU(2) symmetry. The orbital model describes also propagation of a hole in some e_g compounds, and we argue that orbital degeneracy alone does not lead to hole self-localization.

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One of the fundamental problems in solid state physics consists in understanding the motion of an electron or hole coupled to the other degrees of freedom in a material. In many cases, the other degrees of freedom (spin, orbital, or phonon excitations) can increase the mass of the carrier and possibly localize it. For example, the undoped parent compound of high- T_c cuprates is an antiferromagnetic (AFM) Mott insulator due to electron-electron repulsion. A hole doped into it was at first thought to be localized because its movement would disturb the AFM background and thus cost energy [1]. Only two decades later it was found that quantum spin fluctuations heal the background and lead to a coherent hole motion [2,3]; see Figs. 1(a) and 1(b). This shows how important it is to critically assess any approximation used and to identify possible mechanisms of the coherent hole motion.

Novel aspects of localization occur in *orbital* models. The superexchange (SE) is then no longer SU(2) symmetric, and the lower symmetry [4] leads to anisotropy and often to frustrated interactions [5]. The resulting variety of possible scenarios [6] make compounds with orbital degree of freedom at once very interesting for material science and challenging to theory. The most relevant systems of strongly correlated orbitals are (nearly) degenerate e_g or t_{2g} orbitals. While e_g orbitals are of interest in colossal magnetoresistance manganites, t_{2g} bands are relevant to, e.g., cubic titanates [7] or vanadates [8].

Two e_g orbitals describe the two-dimensional (2D) ferromagnetic (FM) planes of LaMnO₃ [9], K₂CuF₄ [10], or Cs₂AgF₄ [11]. The ground state of the orbital SE model has Ising-like alternating orbital (AO) order, where quantum fluctuations are largely suppressed [12]. Nevertheless, a hole doped into the AO background finds a way to move in manganites; see Figs. 1(c) and 1(d). Here, we investigate hole motion in a FM plane with two active t_{2g} orbitals (similar to manganites, spin excitations could contribute for the AFM coupling between the FM planes [13]), where AO order is also Ising-like and interorbital hopping is excluded. This situation arises when a crystal field splits

t_{2g} orbitals in d^1 or d^2 systems. If one of the three t_{2g} orbitals is either empty (d^1) or fully occupied (d^2), the remaining two can form the AO order, as, e.g., in the planes of Sr₂VO₄ [14] with possible weak FM order [15]. In addition to t_{2g} compounds, this “ t_{2g} model” also describes e_g orbitals in the above-mentioned fluorides, where a crystal field induces AO order of $d_{z^2-x^2}$ or $d_{z^2-y^2}$ orbitals [10,11], so both quantum fluctuations and interorbital hopping are quenched and cannot generate coherent quasiparticle (QP) propagation [9], shown in Fig. 1.

In this Letter, we show that a hole doped in a state with alternating t_{2g} orbitals is not confined but finds a way to move *coherently* via three-site effective hopping terms arising from SE, i.e., even in a model with strictly nearest-neighbor hopping. For the present orbital model, long-range hopping is not expected to be important, because it is straightforward to verify that: (i) the second neighbor hoppings flip the orbital flavor [16], so they do not contribute to QP dispersion, while (ii) the third neighbor hoppings, which conserve the orbital flavor, are considerably smaller than the three-site terms for realistic parameters. These latter SE terms are often neglected [17], but here they play a central role and determine QP propagation. This finding contradicts naive expectations of absence of coherent hole motion for the Ising-like SE in the present t_{2g} orbital model. We investigate the spectral function by a combination of analytic and numerical methods to arrive at unbiased conclusions.

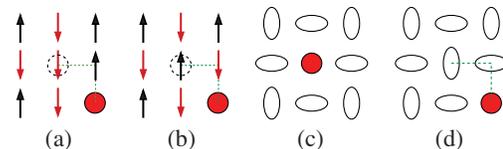


FIG. 1 (color). Hole motion in the spin model with SU(2) symmetry (a),(b), and in the e_g orbital model (c),(d). Defects in the AFM background (a) caused by hole hopping are healed (b) by spin fluctuations, while for a hole in the AO state (c), interorbital hopping does not generate defects at all (d).

A FM plane with t_{2g} AO order is described by interacting spinless fermions that undergo one-dimensional (1D) hopping with conserved orbital flavor:

$$\mathcal{H} = -t \sum_{\langle ij \rangle \parallel b} a_i^\dagger a_j - t \sum_{\langle ij \rangle \parallel a} b_i^\dagger b_j + U \sum_i n_{ia} n_{ib}. \quad (1)$$

Here a_i^\dagger (b_i^\dagger) creates an electron with flavor a (b) that is allowed to move by hopping t along the b (a) axis and cannot hop along the a (b) axis in a cubic system [4]; U gives the energy of a doubly occupied site. Apart from its applicability to t_{2g} and certain e_g orbital systems, the t_{2g} Hamiltonian (1) is of high theoretical interest, because it presents—to our knowledge—the only possibility to derive Ising SE from a Hubbard-like model [18]. In the regime of large on-site Coulomb repulsion $U \gg t$, where the undoped system is a Mott insulator, it reduces to an orbital t - J^z model in a similar way as the Hubbard model does reduce to the spin t - J model [19]. The model Hamiltonian, $\mathcal{H}_{t-J^z} = \mathcal{H}_t + \mathcal{H}_{J^z}$, consists of:

$$\mathcal{H}_t = -t \sum_{\langle ij \rangle \parallel b} \tilde{a}_i^\dagger \tilde{a}_j - t \sum_{\langle ij \rangle \parallel a} \tilde{b}_i^\dagger \tilde{b}_j, \quad (2)$$

$$\mathcal{H}_{J^z} = \frac{1}{2} J \sum_{\langle ij \rangle} \left(T_i^z T_j^z - \frac{1}{4} \tilde{n}_i \tilde{n}_j \right). \quad (3)$$

Here $\tilde{a}_i^\dagger = a_i^\dagger (1 - n_{ib})$ and $\tilde{b}_i^\dagger = b_i^\dagger (1 - n_{ia})$ are the creation operators in the restricted space without double occupancies, as in the spin t - J model [19], and the sum in Eq. (3) includes each bond $\langle ij \rangle$ only once. The corresponding density operators are \tilde{n}_{ia} and \tilde{n}_{ib} , with the total on-site density $\tilde{n}_i = \tilde{n}_{ia} + \tilde{n}_{ib}$. $T_i^z = \frac{1}{2}(\tilde{n}_{ia} - \tilde{n}_{ib})$ stands for the z th component of the pseudospin operator and $J = 4t^2/U$ is the SE energy. A hole inserted into the b orbital of the AO ground state can move by hopping t only along the bonds $\langle ij \rangle \parallel a$, i.e., only in one direction; see Fig. 2. This first step costs excitation energy $E_1 = \frac{3}{4}J$, and further steps build a string of orbital excitations with ever-increasing energy [Fig. 2(a)]. In contrast to the spin Hubbard model with isotropic hopping, the severe restrictions on hole hopping remove all mechanisms of healing the defects in the AO state—not only the quantum fluctuations but even the Trugman loop processes [20], which would lead to a coherent propagation, are here excluded. Consequently, the spectral function $A(\mathbf{k}, \omega)$ for the t - J^z model obtained from the self-consistent Born approximation (SCBA) is independent of momentum \mathbf{k} and consists of a ladder spectrum (not shown), with well separated peaks similar to Refs. [2,3]. Surprisingly, the spectral function for the full orbital model (1) obtained within the variational cluster approach (VCA) [21] exhibits a distinct coherent low-energy mode [22] [shown by a solid line in Fig. 3(a)], with a 1D dispersion that depends on the orbital.

The VCA is a variational method based on exact diagonalization (ED) combined with the self-energy functional approach [23], and has its roots in perturbative cluster approaches for the Hubbard model [24]. The method is

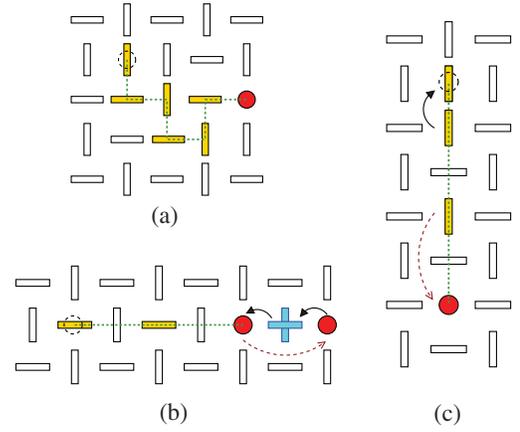


FIG. 2 (color). Artist's view of possible hole hopping processes in the orbital model (1) with AO order ($t \ll U$). Dashed (red) circles indicate initial (final) positions of the hole. The dotted lines connecting them give the hole paths, while the shaded (yellow) rectangles mark electrons that changed orbital due to hole motion. When the hole moves by hopping t (a), it has to turn by 90° in each step and thus creates a zigzag string of frustrated bonds. In contrast, in (b) the hole moves along a direction via effective three-site terms (4) generated by intermediate states with two holes separated by a double occupancy (blue cross). (c) The hole moving in the other direction after frustrating three bonds.

ideally suited to the present problem, because it combines an unbiased solution of the full Hubbard-like Hamiltonian (1) on a small cluster (here 10 sites) with access to the thermodynamic limit. Spontaneous symmetry breaking in ordered phases is incorporated by optimizing an appropriate “fictitious” field, in our case a staggered orbital field. Consequently, the resulting optimal state has almost perfect (Ising-like) AO order. To control our results, we also performed ED on (4×4) -site clusters and likewise found a 1D dispersion with similar bandwidth.

The above results suggest that the t - J^z model with its dispersionless ladder spectrum cannot reproduce the spectral density of the full model (1). The puzzle is resolved by noticing that the complete low-energy model in second order includes also three-site terms

$$\mathcal{H}_{3s}^{(0)} = -\frac{1}{4} J \sum_{\{imj\} \parallel a} \tilde{b}_i^\dagger \tilde{n}_{ma} \tilde{b}_j - \frac{1}{4} J \sum_{\{imj\} \parallel b} \tilde{a}_i^\dagger \tilde{n}_{mb} \tilde{a}_j, \quad (4)$$

where $\{imj\}$ denotes three adjacent sites in a row $\parallel a$ (or a column $\parallel b$) with m in the middle. This effective hopping term is also obtained perturbatively from Eq. (1) by allowing one double occupancy next to the inserted hole, and is thus again of the order $\propto t^2/U$ as the SE term (3). Figure 2(b) illustrates how a b electron moves over an occupied a orbital and interchanges with the hole without affecting the AO order. This leads to 1D free propagation on two sublattices: $\varepsilon_{\mathbf{k}}^a = \frac{1}{2} J \cos(2k_x)$ for the one with occupied b orbitals, and $\varepsilon_{\mathbf{k}}^b = \frac{1}{2} J \cos(2k_y)$ for the other. If we include $\mathcal{H}_{3s}^{(0)}$ and treat the effective strong-coupling

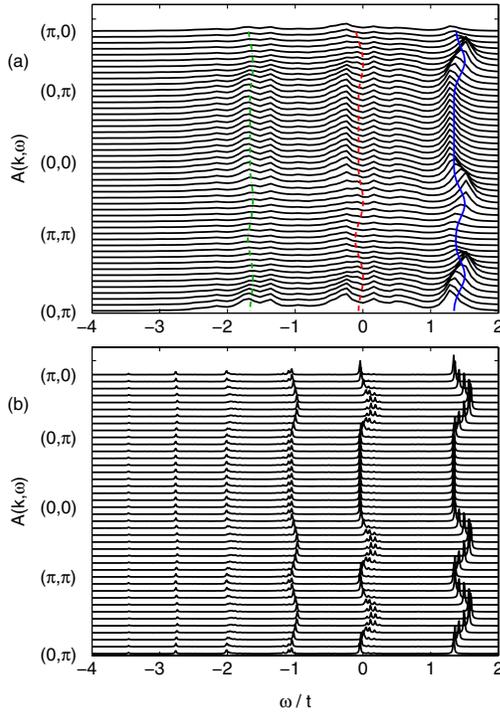


FIG. 3 (color online). Spectral function $A(\mathbf{k}, \omega)$ for b orbitals at $U = 8t$ along the main directions of the 2D Brillouin zone obtained for (a) the t_{2g} orbital Hubbard model (1) in the VCA with a 10-site cluster; (b) the effective model \mathcal{H}_{eff} (5) in the SCBA. First moments of the VCA spectra calculated using the energy intervals $[0.9t, 2t]$, $[-t, 0.9t]$, and $[-2.5t, -t]$ are shown by solid and dashed lines in (a).

model

$$\mathcal{H}_{\text{eff}} = \mathcal{H}_t + \mathcal{H}_{J^z} + \mathcal{H}_{3s}^{(0)}, \quad (5)$$

the low-energy QP state indeed becomes dispersive, similar to the VCA, both in ED for a 4×4 cluster (not shown) and in the SCBA; see Fig. 3(b). However, the free dispersion $\propto J$ is strongly renormalized; see below. For a single hole, the QP weight is almost \mathbf{k} independent in the SCBA. In contrast, in the VCA for the orbital Hubbard model (1) the spectral weight of the QP state decreases strongly between $\mathbf{k} = (0, 0)$ and $\mathbf{k} = (\pi, \pi)$, similar to the decrease of the spectral weight in the spin Hubbard model [25]. This reflects the fundamental difference between the Hilbert spaces of the full (1) and the effective strong-coupling model (5).

One finds that not only the first but also all subsequent peaks have distinct 1D dispersions in the SCBA [Fig. 3(b)]. We attribute this to a 1D propagation analogous to the QP state, but started after an even number of t hopping steps have generated string defects in the AO state. The positions of all peaks evolve from the excitations of the t - J^z ladder spectrum where the hole is confined, and the pseudogap between the QP and the second peak scales as $(J/t)^{2/3}$, similar to the spin t - J^z model [2]. Such processes cannot be properly included within the VCA using a 10-site cluster,

where too few momenta \mathbf{k} are available. The dispersive features of Fig. 3(b) are thus replaced by several maxima and the spectral weight is transferred between them when \mathbf{k} changes. Remarkably, the first moments calculated for these structures (within the relevant energy regimes) show again similar \mathbf{k} dependence [Fig. 3(a)] as that found in the SCBA.

Figure 4 compares QP features of the present t_{2g} model, obtained within the VCA and the SCBA, with those for the spin Hubbard/ t - J model. In the latter case, the bandwidth W at small J (large U) is, as expected, approximately linear in J [26], both in the VCA (Hubbard model), as well as in the SCBA (t - J model), with and without three-site terms. The t - J model with three-site terms gives the complete second-order perturbation result for the Hubbard model, and consequently its bandwidth agrees with the VCA data for somewhat larger J (smaller U) than the t - J model without three-site terms; see Fig. 4(a). In the t_{2g} case, where there is no bandwidth $\sim J$ coming from the quantum fluctuations, the propagation via three-site terms leads to a bandwidth that is nearly quadratic in J . The additional power arises from the structure of the wave function [27], which renormalizes the bare three-site bandwidth $\sim J/4$ by an additional factor $\propto J$ related to the QP weight a_{QP} . It is instructive to take a closer look at the contributions of the three-site terms to the bandwidth of the usual SU(2) symmetric t - J model: The difference between the SCBA results with [dotted line in Fig. 4(a)] and without (dashed line) three-site terms is almost exactly twice the t_{2g} bandwidth. This factor of 2 is due to the reduction by $1/2$ of the t_{2g} SE (3) when compared to the spin model.

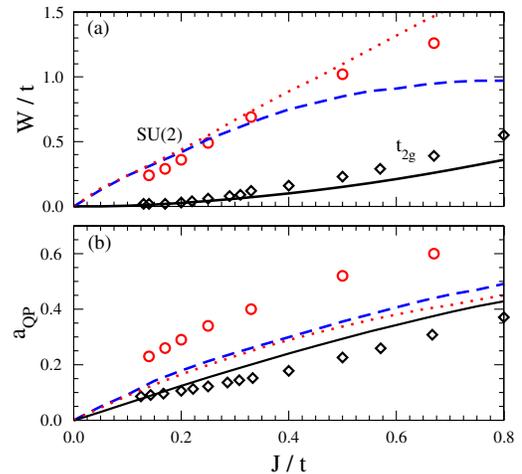


FIG. 4 (color online). QP properties for the t_{2g} orbital (t_{2g}) and for the conventional SU(2)-symmetric spin model [SU(2)] for increasing J/t : (a) bandwidth W/t , and (b) weight a_{QP} averaged over the 2D Brillouin zone. Diamonds (orbital) and circles (spin) denote the VCA data for the Hubbard-like models, lines give the SCBA results for the orbital t - J^z model with three-site terms (5) (solid line), as well as the usual spin t - J model with (dotted line) and without (dashed line) three-site terms.

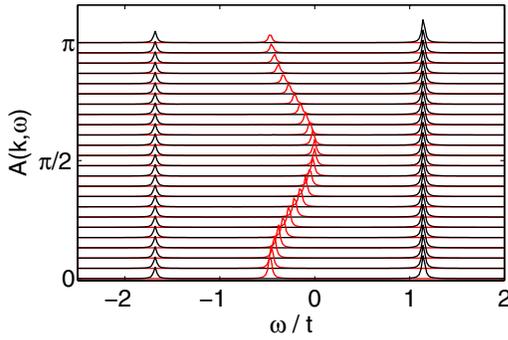


FIG. 5 (color online). Spectral function for the 1D t_{2g} orbital model obtained in the VCA for $U = 8t$. Excitations at $\omega \approx 1.2t$ and $\omega \approx -1.7t$ correspond to a hole doped into a localized a orbital, while a hole doped into a mobile b orbital gives a free propagation with dispersion $\omega_k = \frac{1}{4}J \cos k + c$.

In case of the QP weight a_{QP} [Fig. 4(b)], the three-site terms have almost no impact on the Hubbard model and the SCBA results for both models do not differ strongly. While the VCA gives a similar weight for t_{2g} orbitals as the SCBA, the values for the spin Hubbard model are considerably larger. Since the SCBA has been shown to give the same results as quantum Monte Carlo data for the t - J model extrapolated to the thermodynamic limit [28], we believe that the weight given by the VCA is affected by finite-size effects, which appear to be weaker in the more classical t_{2g} model.

Finally, we discuss the 1D chain along the a axis [Eq. (1) without the first term], because it gives even clearer insight into the role played by the three-site terms. In one dimension, electrons of only one (b) flavor can hop, similar to e_g electrons in a 1D FM chain [29]. Without three-site terms, one finds just three dispersionless peaks, one arising from a hole in the mobile orbital and two from the localized orbital [29]. With three-site terms, the hole doped into a mobile b orbital propagates; see Fig. 5. Its bandwidth is exactly J ; i.e., we observe the full three-site dispersion because hole motion is here not renormalized by string excitations. For a hole doped into a localized a orbital, t - J^z and Hubbard-like model give two identical dispersionless features, which arise from hopping to the sites next to the initial site of the hole. One might expect the three-site terms to have some effect because they allow the hole to move over the entire chain instead of being confined to just three sites. However, the energy gain due to this delocalization is small, and the corresponding spectral feature is invisible in Fig. 5. For the same reason, processes displayed in Fig. 2(c) are not seen in the 2D spectra of Fig. 3 either, where they might be expected to show up as higher-energy excitations with a dispersion complementary to that of the first peak.

To summarize, we have investigated hole motion in a background with t_{2g} —and in some cases e_g —AO order, and have found a coherent hole motion via three-site terms. This mechanism is fundamentally different from the ones

established so far in spin systems (quantum fluctuations), or for e_g electrons in manganites (interorbital hopping). It can also be distinguished from hole motion via direct longer-range hopping terms because it behaves differently under particle-hole transformation. Furthermore, the present model provides a realistic case with purely classical Ising SE interactions. As one still finds coherent hole motion, we argue that the hole confinement and dispersionless ladder excitation spectrum of the t - J^z model—while being attractive mathematical ideas—are *never* realized in transition-metal oxides.

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