

Intrinsic Coupling of Orbital Excitations to Spin Fluctuations in Mott Insulators

Krzysztof Wohlfeld,¹ Maria Daghofer,¹ Satoshi Nishimoto,¹ Giniyat Khaliullin,² and Jeroen van den Brink¹

¹*IFW Dresden, P.O. Box 27 01 16, D-01171 Dresden, Germany*

²*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*

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We show how the general and basic asymmetry between two fundamental degrees of freedom present in strongly correlated oxides, spin and orbital, has very profound repercussions on the elementary spin and orbital excitations. Whereas the magnons remain largely unaffected, orbitons become inherently coupled with spin fluctuations in spin-orbital models with antiferromagnetic and ferro-orbital ordered ground states. The composite orbiton-magnon modes that emerge fractionalize again in one dimension, giving rise to spin-orbital separation in the peculiar regime where spinons are faster than orbitons.

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In transition metal oxides, different $3d$ orbitals near the Fermi level can have similar energy and thereby contribute to low-energy physics. In the presence of strong correlations charge fluctuations become suppressed and a Mott insulator is realized when there is a commensurate number of electrons per unit cell. The effective Hamiltonian that emerges, often referred to as the Kugel-Khomskii (KK) Hamiltonian, can be expressed in terms of spin and orbital operators [1]. From a formal viewpoint the spin and orbital operators are very similar because they form identical algebras, but the way in which these operators enter into realistic KK Hamiltonians is very different.

While the spin wave function is in essence rotationally invariant and the coupling between spin operators therefore $SU(2)$ symmetric, the symmetry in orbital space is much lower due to the ubiquitous crystal field acting on the orbital wave functions in crystals. This reduced symmetry causes the orbital-orbital interaction to be very anisotropic in space and often inherently frustrated, which causes exotic effects such as macroscopic degeneracy of the ground state or the emergence of non-Abelian topological excitations in the case of the compass or Kitaev models, respectively [2], which may be relevant for quantum computation [3,4]. We will show that this asymmetry between spin and orbital degrees of freedom has fundamental repercussions on the coupling of the elementary spin and orbital excitations—the magnons and orbitons, respectively: whereas the magnons remain largely unaffected, orbitons become inherently coupled with spin fluctuations in spin-orbital models with antiferromagnetic (AFM) and ferro-orbital (FO) ordered ground states. This is relevant in the experimental context as substantial progress is being made in measuring orbitons [5,6] and their dispersion, in particular, in resonant inelastic x-ray scattering (RIXS) [7,8], where we predict the coupling of the orbiton to magnetic fluctuations to be clearly discernible.

In the standard approach the complex problem of intertwined spin-orbital excitations is solved using a mean-field decoupling, i.e., considering magnons in a fixed orbital

background or orbitons in a fixed spin background. While such an approach can work well to obtain the correct spin and orbital orderings consistent with the Goodenough-Kanamori rules [1] and in some cases with ferromagnetic (FM) order [9], we show here that it fails to describe orbital excitations even qualitatively correctly for a number of spin-orbital models. We avoid this decoupling by mapping the coupled orbiton-magnon dynamics onto the well-controlled problem of a hole propagating in a magnetic background: the extensively studied single hole t - J model. In particular, we find that in 1D the orbital excitation fractionalizes into freely propagating spinon and orbiton, giving rise to spin-orbital separation in the peculiar regime where spinons are faster than orbitons.

Model and problem statement.—The generic form of the KK Hamiltonian [1] in the Mott-insulating limit is

$$\mathcal{H} = 2J \left(\sum_{\langle i,j \rangle} H_{i,j}^S H_{i,j}^T + \sum_i H_i^T \right), \quad (1)$$

where i and j are lattice sites and on each bond $\langle i, j \rangle$ the spin-spin interaction is $H_{i,j}^S$ whereas the orbital-orbital one is $H_{i,j}^T$. To capture the generic differences between orbitons and magnons, it is enough to break the $SU(2)$ rotation symmetry in orbital space, which is here achieved by the large local crystal field breaking the degeneracy between the orbitals, expressed as $H_i^T = \frac{E_z}{2J} T_i^z$. We keep for simplicity the rotational symmetry in the interactions so that for the spins $H_{i,j}^S = \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4}$ and for the orbitals $H_{i,j}^T = \mathbf{T}_i \cdot \mathbf{T}_j + \frac{1}{4}$, where \mathbf{S} (\mathbf{T}) are the spin (orbital) operators that fulfill the $SU(2)$ algebra for $S = 1/2$ ($T = 1/2$) spins (pseudospins). The constant $J > 0$ gives the energy scale of the spin-orbital superexchange, and the symmetry breaking field for the orbitals is E_z . Note that for the case of $E_z = 0$ (not considered here), this model has an $SU(4)$ symmetry even higher than the combined $SU(2) \times SU(2)$ symmetries, which results in the ground state given by the Bethe ansatz and composite spin-orbital gapless excitations in addition to the separate spin and orbital ones (see, e.g.,

Ref. [10]). This model describes the low-energy physics, determined by singly occupied sites, of a two-orbital Hubbard model in the limit of a large on-site Coulomb repulsion U and vanishing Hund's exchange J_H , cf. Ref. [9].

Here we are interested in the orbital excitations of the model Eq. (1) when $E_z \gg J$; i.e., the orbital splittings are larger than the magnetic coupling energy. This is a realistic regime for many strongly correlated compounds such as 1D or 2D cuprates [11]. As one degree of freedom is completely polarized, the ground state is easily found and given by all electrons occupying a single orbital in an AFM state. But while decoupling spin and orbital degrees of freedom works here for the ground state, it is not at all appropriate for orbital excitations—this is the problem investigated below.

Decoupling of spin and orbital sector.—The ground state $|\psi\rangle = |\psi_S\rangle \otimes |\psi_O\rangle$ of Eq. (1) is described by the ground state $|\psi_S\rangle = |\text{AFM}\rangle$ of an AFM Heisenberg system formed by spins in the lower-energy orbital, i.e., a FO ordered state $|\psi_O\rangle = |\text{FO}\rangle$. The orbital excitations are reached by flipping an orbital, i.e., by promoting an electron at site j from the occupied lower orbital to the empty higher band at the same site, expressed by the orbital raising operator T_j^+ . The momentum-dependent orbital excitation is given by $T_k^+ = \sum_j e^{ikj} T_j^+$ [$T_k^- = (T_k^+)^\dagger$], and the spectral function describing its dynamics is

$$O(k, \omega) = \frac{1}{\pi} \lim_{\eta \rightarrow 0} \text{Im} \langle \psi | T_k^- \frac{1}{\omega + E_\psi - \mathcal{H} - i\eta} T_k^+ | \psi \rangle. \quad (2)$$

First we discuss the orbiton spectral function by taking the orbital excitation to be independent of the magnetic excitation. One can then rewrite the orbital operators by

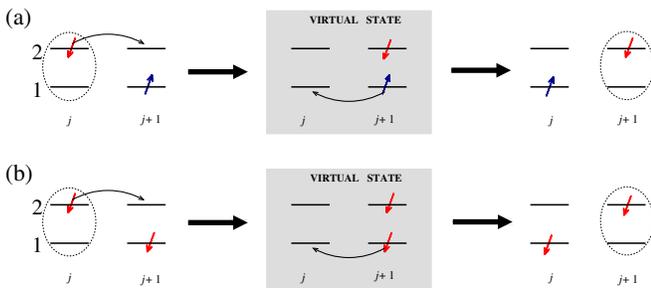


FIG. 1 (color online). Two superexchange processes moving an electron in an excited orbital (indicated by oval) from site j to its neighbor $j+1$. (a),(b) Orbiton motion when spins along the bond are antiparallel or parallel, respectively; see text. The states in the gray middle panels are not part of the low-energy Hilbert space corresponding to (1). These virtual excitations within the full two-orbital Hubbard model illustrate the origin of those superexchange interactions of Hamiltonian (1) that propagate the orbiton; note that the spin of the excited electron is conserved.

use of Holstein-Primakoff bosons (see, e.g., Ref. [12]), keep only quadratic terms in the expansion (orbital-wave theory), and, noticing that the ground state $|\psi_O\rangle$ does not contain bosons, obtain

$$O(k, \omega) = \delta[\omega - \omega_{\text{OW}}(k)], \quad (3)$$

with a mean-field orbital-wave dispersion $\omega_{\text{OW}}(k) = E_z - \frac{1}{2}zJ_{\text{OW}}(1 - \gamma_k)$. Here, z is the coordination number, γ_k is the lattice structure factor, and the effective orbital exchange constant $J_{\text{OW}} = 2J\langle \psi_S | \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} | \psi_S \rangle$. The orbital excitation on the mean-field level is thus a quasiparticle with a cosinlike dispersion with period 2π : for example, in 1D we obtain an effective reduced $J_{\text{OW}} \simeq -0.4J \ll J$, cf. the thick line in Fig. 2. An analogous procedure for magnons in FM planes with alternating orbitals (AO) has been applied to LaMnO₃ or KCuF₃, and similarly yields magnons with a reduced bandwidth, but without any other trace of the AO order, in agreement with experiment [13]. We will see, however, that for orbitons this framework of mean-field decoupling is greatly oversimplified.

Mapping onto an effective t - J model.—The orbital-wave approximation puts all the impact of the AFM order into J_{OW} , which is a mean-field average of the sum of two distinct superexchange processes in which the orbital excitation may propagate through the lattice. The first one, which corresponds to $(T_j^+ T_{j+1}^- + \text{H.c.})(S_j^+ S_{j+1}^- + \text{H.c.})$ processes in Hamiltonian (1), allows for orbiton propagation when the spins on the bond are antiparallel; see Fig. 1(a). The second one, which corresponds to $(T_j^+ T_{j+1}^- + \text{H.c.})(S_j^z S_{j+1}^z + 1/4)$ processes in Hamiltonian (1), allows for orbiton propagation when the spins on the bond are parallel; see Fig. 1(b). Crucially (see next paragraph), this figure illustrates that in both cases the spin of the electron in the upper orbital 2 is conserved during the orbiton propagation. This is because spin-orbital

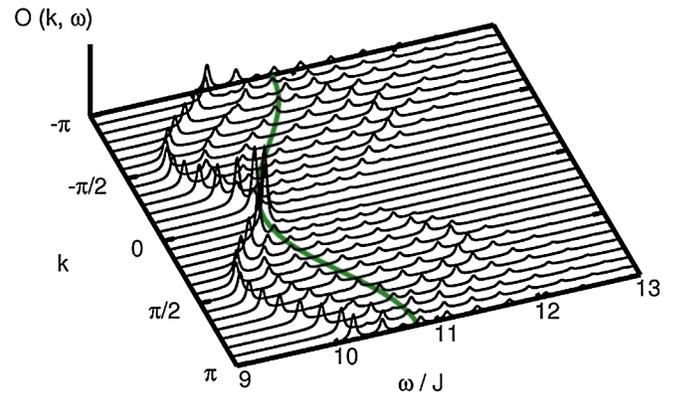


FIG. 2 (color online). Spectral function $O(k, \omega)$ of orbital excitation obtained via the mapping onto the t - J model, Eq. (6), evaluated using Lanczos exact diagonalization on a 28 site chain. A broadening $\eta = 0.03J$ and $E_z = 10J$. The thick line shows orbital excitation in a mean-field (orbital-wave) approach, Eq. (3).

Hamiltonian (1) is, as mentioned above, a low-energy limit of the two-orbital Hubbard model with the Hund's exchange $J_H = 0$, and the spins of individual electrons in the superexchange process cannot be flipped (see middle panels of Fig. 1). In more realistic spin-orbital models the Hund's exchange is finite [1], but the processes which would not conserve the electron's spin in the excited orbital are small ($\propto J_H/U$) and thus could be neglected.

To be explicit, we now focus on the 1D case and employ a Jordan-Wigner transformation [14]. 2D and 3D cases are discussed later. We thus introduce $S_j^+ = \beta_j e^{i\pi Q}$, $S_j^- = e^{-i\pi Q} \beta_j^\dagger$, $S_j^z = \frac{1}{2} - n_{j\beta}$, where $Q = \sum_{l=1}^{j-1} n_{l\beta}$ and β_j^\dagger create spinons while $T_j^+ = e^{-i\pi Q} \alpha_j^\dagger$, $T_j^- = \alpha_j e^{i\pi Q}$, $T_j^z = n_{j\alpha} - \frac{1}{2}$, where $\bar{Q} = \sum_{l=1}^{j-1} n_{l\alpha}$ and α_j^\dagger creates a pseudospinon. Since the spin of the propagating electron in the upper orbital 2 is conserved (see above), one may calculate the spectral function $O(k, \omega)$ for, e.g., spin-up in the upper orbital. We are then allowed to replace $T_k^\dagger \rightarrow \sum_j e^{ikj} T_j^\dagger (\frac{1}{2} + S_j^z) = e^{-i\pi \bar{Q}} \alpha_j^\dagger (1 - n_{j\beta})$ in Eq. (2), and thus terms in the Hamiltonian that create or annihilate both a spinon and a pseudospinon at the same site lead to a vanishing contribution to the spectral function. Phase factors cancel in one dimension and we obtain

$$O(k, \omega) = \frac{1}{\pi} \lim_{\eta \rightarrow 0} \text{Im} \langle \bar{\psi} | \alpha_k \frac{1}{\omega + E_{\bar{\psi}} - \tilde{\mathcal{H}} - E_z - i\eta} \alpha_k^\dagger | \bar{\psi} \rangle, \quad (4)$$

with the effective fermionic Hamiltonian

$$\begin{aligned} \tilde{\mathcal{H}} = & -\frac{1}{2} J \sum_{\langle i,j \rangle} (\beta_i^\dagger \alpha_i \alpha_j^\dagger \beta_j + \alpha_i \alpha_j^\dagger + \text{H.c.}) \\ & + J \sum_{\langle i,j \rangle} \left[\frac{1}{2} (\beta_i^\dagger \beta_j + \text{H.c.}) - \frac{1}{2} (n_{i\beta} + n_{j\beta}) + n_{i\beta} n_{j\beta} \right], \end{aligned} \quad (5)$$

and an implicit constraint $\forall_j \beta_j^\dagger \beta_j + \alpha_j^\dagger \alpha_j \leq 1$. Here $|\bar{\psi}\rangle$ is a tensor product of the magnetic ground state $|\psi_S\rangle = |\text{AFM}\rangle$ expressed in terms of spinons and a vacuum state for pseudospinons [recall that we consider here a single orbitor only; this also allowed us to skip quartic terms in pseudospinons in Eq. (5)].

At this point, we observe that the resulting effective Hamiltonian is in fact a Hamiltonian for the t - J model written in terms of the Jordan-Wigner fermions with the above constraint [15]. By introducing the electron operators $p_{j\uparrow} = \alpha_j^\dagger$, $p_{j\downarrow} = \alpha_j^\dagger \beta_j e^{i\pi Q}$ acting in the restricted Hilbert space without double occupancies, we obtain

$$O(k, \omega) = \frac{1}{\pi} \lim_{\eta \rightarrow 0} \text{Im} \langle \bar{\psi} | p_{k\uparrow}^\dagger \frac{1}{\omega + E_{\bar{\psi}} - \tilde{\mathcal{H}} - E_z - i\eta} p_{k\uparrow} | \bar{\psi} \rangle, \quad (6)$$

with the t - J Hamiltonian

$$\tilde{\mathcal{H}} = -t \sum_{\langle i,j \rangle, \sigma} (p_{i\sigma}^\dagger p_{j\sigma} + \text{H.c.}) + J \sum_{\langle i,j \rangle} \left(\mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} n_i n_j \right), \quad (7)$$

where $n_j = \sum_\sigma n_{j\sigma}$ and the hopping parameter t is defined as $t = J/2$ [16]. The ground state $|\bar{\psi}\rangle$ is now the tensor product of a vacuum state for holes and the $|\psi_S\rangle$ state. We have thus mapped the single orbitor in the FO and AFM chain, with dynamics governed by Hamiltonian (1), onto a single hole doped into the undoped AFM chain with its dynamics governed by Hamiltonian (7).

Numerical results for the t - J model.—To flesh out the resulting coupling between orbitons and spin fluctuations we use Lanczos exact diagonalization to evaluate Eq. (6) on a finite chain (28 sites). The spectral function is shown in Fig. 2: the spectrum differs qualitatively from the orbital-wave result shown as a thick line in Fig. 2. It now consists of multiple peaks (expected to merge into incoherent spectrum in the thermodynamic limit) instead of one single excitation. There is a dominant feature at the lower edge of the spectrum, but its periodicity is π reflecting the doubled unit cell of the AFM order.

The t - J model with $J > t$ [16] is not easily accessible in the Hubbard-type models, as it would formally correspond to small on-site interaction U , where the t - J model is no longer valid. In this regime, the spinon moves faster than the holon, and the entire lower edge of the spectrum is thus given by ‘‘holon’’ states [17]. If the orbitor takes the place of the holon as argued here, this exotic behavior should be observable in RIXS experiments and one would expect a dominant excitation with orbitor character with dispersion $\omega \approx E_z - 2t \sin|k|$ at the bottom of the spectrum. The latter would be expected to extend up to $\omega \approx E_z + \sqrt{J^2 + 4t^2} - 4tJ \cos k$ and to contain an intermediate feature still well visible within the continuum with the dispersion of the purely orbitor character scaling as $\omega \approx E_z + 2t \sin|k|$.

Figure 3 illustrates how electron exchange processes can let an orbital excitation propagate through the system after creating a spinon in the first step. The spinon itself moves via spin flips $\propto J > t$, faster than the orbitor, and the two get well separated. The orbital-wave picture, on the other hand, would require the orbital excitation to move without creating the spinon in the first step. As can be inferred from Fig. 3, this is only possible for imperfect Néel AFM spin order so that the averages of processes shown in Figs. 1(a) and 1(b) are finite.

2D and 3D.—Remarkably, the standard OW picture becomes even worse in higher dimensions: in 2D (3D) cubic lattices, the mean-field orbital coupling J_{OW} almost vanishes due to $\langle \psi_S | \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} | \psi_S \rangle \approx -0.08$ (-0.05); the orbital dynamics is then entirely governed by coupling to the spin fluctuations [18]. The t - J model description of a single orbitor [Eqs. (6) and (7)] is general and valid for any dimension: the mapping rests entirely on the fact that spin

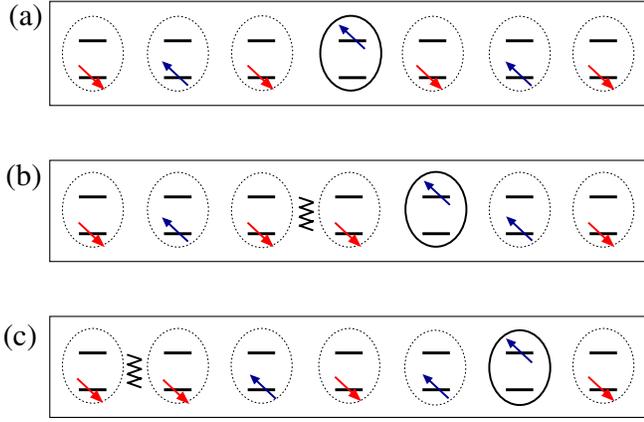


FIG. 3 (color online). Schematic representation of the orbital motion and the induced spin fluctuations giving rise to spin-orbital separation in 1D. The first hop of the excited state [(a) \rightarrow (b)] creates a spinon (wavy line) that moves via spin exchange $\propto J$. The next hop [(b) \rightarrow (c)] does not produce any extra spinons: an “orbiton” freely propagating as a “holon” with an effective hopping $t \sim J/2$ is created.

on the excited orbital is conserved. In particular, the Jordan-Wigner fermionization applied to the 2D case gives the t - J model expressed in terms of Jordan-Wigner fermions [15] and consequently the 2D version of Eqs. (6) and (7); see [19] for details.

A very good approximate solution for the higher-dimensional t - J model with $J > t$ can be obtained by perturbation theory, because $J > t$ corresponds to weak coupling; see Eq. (7) of Ref. [20]. The solution shows that in 2D or 3D a single orbiton in the undoped AFM system described by the t - J model cannot fractionalize due to the magnetic string effect. Still, the orbiton dressed with spin fluctuations is mobile on a renormalized scale [20]. For example, the 2D case directly corresponds to the result in Ref. [21] which gives $\omega \approx E_z - x_1 + x_2(\cos k_x + \cos k_y)^2 + x_3(\cos 2k_x + \cos 2k_y)$ for the orbiton dispersion (x_i are positive parameters $\propto t^2/J$ when $J > t$, cf. Ref. [21] for exact values). We thus expect to observe, e.g., in high resolution RIXS experiment on 2D cuprates, a spectrum similar to the one-particle Green’s function of the higher-dimensional t - J model, with a low-energy quasiparticle orbiton peak $\propto \omega$ and additional incoherent part [21]. Crucially, while the parameters x_i depend on J/t and would vary in more realistic spin-orbital models [16], the general shape of the orbiton dispersion ω —the minimum at $(\pi/2, \pi/2)$, saddle point at $(\pi, 0)$, maxima at $(0, 0)$ and (π, π) —is robust reflecting the fact that the coherent motion of orbiton is possible only within a given spin sublattice.

No analogue in spin sector.—Let us now revisit the analogue where the spin is polarized (FM) and the orbital sector shows AO order due to, e.g., a large Jahn-Teller effect. An example would be the FM and AO planes in LaMnO_3 or KCuF_3 [13]. Many studies have shown that the

orbital degrees of freedom merely renormalize the spin excitation in this case and do not change the periodicity of magnons (see, e.g., Ref. [22]). This qualitative difference is caused by the fact that in realistic cases the Jahn-Teller stabilized AO order is much more classical and robust, thus suppressing the creation of orbital excitations by the excited spin and giving larger weight to the pure spin excitation. In other words, spin waves are typically below the orbital gap and well protected by the underlying $SU(2)$ symmetry and Goldstone theorem.

In conclusion, we have shown that orbitons in realistic spin-orbital models with AFM spin and ferro-orbital ground state are so strongly coupled to the spin excitations that the usual mean-field decoupling of two sectors breaks down. In fact, we have presented an exact mapping of the problem onto an effective t - J model and have shown that the orbiton in such models behaves like a single hole in an undoped AFM Mott insulator. However, since the typical superexchange parameters for KK spin-orbital models lead to $J > t$, the study of the orbiton problem provides access to a regime of the t - J model which has been thought to be “unphysical” in terms of single-band Hubbard models. Finally, in 1D signatures of spin-orbital separation are expected again in the peculiar regime where spinons are faster than orbitons.

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