

Spinon-orbiton repulsion and attraction mediated by Hund's rule

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We study the impact of Hund's-rule coupling on orbital excitations, as, e.g., measured in inelastic resonant x-ray scattering. We find that the interpretation in terms of spin-orbit separation, which has been derived for one-dimensional systems without Hund's rule, remains robust in its presence. Depending on whether or not the orbital excitation includes a spin excitation, Hund's rule leads to an attractive or repulsive interaction between spinon and orbiton. Attraction (repulsion) leave clear signatures through a transfer of spectral weight to the lower (upper) edge of the spectrum.

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I. INTRODUCTION

Low-dimensional quantum systems have long been of special interest due to the intriguing and often counterintuitive properties they can host when quantum fluctuations and interactions come together. Arguably the strangest concept at play is fractionalization, where the electron, an elementary particle, behaves as if it were split into parts with fractional charges (e.g., in two-dimensional fractional quantum-Hall states) or into a charge separated from its spin [1]. This last concept, spin-charge separation, applies to an electron or hole propagating in a one-dimensional Mott insulator. Spin and charge can then be considered as propagating as “spinon” and “holon” with different velocities, as has been studied theoretically [2,3] and verified experimentally using angle-resolved photoemission spectroscopy [4,5].

More recently, spin-charge separation has been complemented by the idea of spin-orbit separation [6]. This also occurs in one-dimensional Mott insulators, but it involves an electron being excited into some unoccupied higher-energy orbital instead of being removed from the system. As has been pointed out theoretically, the orbital excitation can then be considered in a manner analogous to a hole and similarly separates into “spinon” and “orbiton.” Experimentally, this has been verified using resonant inelastic x-ray scattering (RIXS), which can address orbital excitations, in a cuprate chain compound [7].

The theory behind the analogy of spin-orbit and spin-charge separation rests on a mapping [6] of the orbital excitation onto a hole-removal excitation that, strictly speaking, breaks down in the presence of Hund's-rule coupling [8]. However, it must be assumed to be present in any realistic material description and can—depending on the material at hand—be strong. We thus want to assess how far the mapping and the scenario of spin-orbit separation can be trusted and to identify the impact of Hund's rule on orbital excitations.

We show in this paper that an interpretation in terms of spinon and orbiton survives to a very large degree and that the main effect of Hund's-rule coupling is an interaction between

spinon and orbiton. The issue of spinon-holon interaction in the t - J model has been discussed analytically in the supersymmetric limit [9], where some exact results can be obtained, and it turned out to be rather subtle [10,11]. Numerically, spinon-holon attraction has been followed from the t - J^z model, where it leads to a bound state, to the isotropic t - J model, where it was concluded to be present but too weak for a bound state [12]. The present work indicates that orbital excitations provide an intriguing window into the interactions between fractionalized excitations: they can address the repulsive as well as the attractive regime, and for strong Hund's-rule coupling, signatures of spinon-orbiton interaction become quite pronounced.

II. ORBITAL EXCITATIONS IN ANTIFERROMAGNETS

We consider here two orbitals per site, denoted by 1 for the low-energy and 2 for the high-energy state, and the limit of strong on-site Coulomb repulsion U , i.e., we neglect charge fluctuations. Second-order perturbation theory with intersite hopping t as a small parameter t/U then gives a Kugel-Khomskii-type model [13] with the general form

$$H = 2 \sum_{\langle i,j \rangle} \left(\vec{S}_i \cdot \vec{S}_j + \frac{1}{4} \right) A_{ij} + \sum_{\langle i,j \rangle} K_{ij} + \Delta \sum_i T_i^z, \quad (1)$$

where \vec{S}_i describes a spin $S = \frac{1}{2}$ at site i and $T_i^z = \frac{1}{2}(n_2 - n_1)$ is the z component of the orbital pseudospin. Operators A_{ij} and K_{ij} depend on the orbital degrees of freedom; see below. Bonds $\langle i, j \rangle$ run over nearest neighbors (NN), but they can be chosen to include longer-range interactions. Strong crystal field $\Delta \gg t$ ensures that only lower-energy orbital 1 is occupied in the ground state.

Since states with two electrons on one site enter the perturbation theory as (virtual) intermediate states, A_{ij} and K_{ij} depend on the on-site interactions. It is helpful to first consider any doubly occupied site to have energy U , regardless of its spin and orbital occupation. This neglects dominant processes such as Hund's-rule coupling but brings out the dominant terms

$$A_{ij}^{(0)} = \frac{4t^2}{U} \left[\left(T_i^z T_j^z + \frac{1}{4} \right) + \frac{1}{2} (T_i^+ T_j^- + \text{H.c.}) \right], \quad K_{ij}^{(0)} = 0. \quad (2)$$

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(We assume identical hopping t in both orbitals.) When strong crystal-field splitting Δ enforces orbital polarization $T_i^z \equiv -\frac{1}{2}$, the second term in $A_{ij}^{(0)}$ is inactive in the ground state, and the first term leads to antiferromagnetic (AFM) Heisenberg coupling of the spins in (1).

An orbital excitation is then induced into the AFM state, e.g., by RIXS, which allows for this excitation to come with or without a spin flip. The two excitations can be distinguished in experiment [14,15] and are described by operators

$$T^+(k) = \frac{1}{\sqrt{L}} \sum_{j,\sigma} e^{ikj} c_{j,2,\sigma}^\dagger c_{j,1,\sigma}, \quad (3)$$

$$(S^x T^+)(k) = \frac{1}{\sqrt{L}} \sum_{j,\sigma} e^{ikj} c_{j,2,-\sigma}^\dagger c_{j,1,\sigma}, \quad (4)$$

where $c_{j,2,\sigma}^\dagger$ ($c_{j,1,\sigma}$) creates (annihilates) an electron with spin $\sigma = \pm 1 = \uparrow, \downarrow$ in the empty orbital 2 (occupied orbital 1) on site j . k denotes crystal momentum and runs over the first Brillouin zone of the L -site chain.

The excitations move via the second term in $A_{ij}^{(0)}$ [see Eq. (2)], and it turns out that the spin in the upper orbital 2 is conserved and has no impact on either kinetic or potential energy [6,16]. Excitations with and without spin flip are thus equivalent and can be mapped onto a spinless hole moving in an AFM background. This mapping between orbital excitations and hole dynamics has been used extensively to analyze RIXS in one-dimensional cuprates [7,17,18] and two-dimensional iridates [16,19,20].

III. RICHER STRUCTURE OF THE DOUBLY OCCUPIED SITE

However, the above considerations can only be applied to materials in which Hund's-rule coupling J_H is much smaller than on-site charge repulsion U . Since U is expected to depend more strongly on screening effects than J_H [24,25], the important ratio $\frac{J_H}{U}$ can differ significantly even for closely related compounds [26]. To extend the analysis of orbital excitations to materials with larger $\frac{J_H}{U}$, the description of two electrons on one site has to take into account processes beyond pure charge interactions.

Let U denote Coulomb repulsion felt by two electrons in the same orbital on the same site i . Due to reduced overlap of the wave functions, their interaction $U' = U - \Delta U < U$ is weaker if they occupy different orbitals. In that case, Hund's-rule coupling $-2J_H \hat{S}_{i,1} \hat{S}_{i,2}$ moreover favors their ferromagnetic (FM) alignment. Finally, a "pair hopping" J_P involves a doubly occupied high-energy orbital 2 and is suppressed here by the large crystal field. The full Kugel-Khomskii Hamiltonian is given by

$$\begin{aligned} A_{ij} = & \frac{4Ut^2}{U^2 - J_P^2} \left(T_i^z T_j^z + \frac{1}{4} \right) + \frac{4J_H t^2}{U^2 - J_H^2} \left(T_i^z T_j^z - \frac{1}{4} \right) \\ & + \frac{2U't^2}{U^2 - J_H^2} (T_i^+ T_j^- + \text{H.c.}) \\ & - \frac{2J_P t^2}{U^2 - J_P^2} (T_i^- T_j^- + \text{H.c.}) \end{aligned} \quad (5)$$

and

$$\begin{aligned} K_{ij} = & -\frac{4Ut^2}{U^2 - J_P^2} \left(T_i^z T_j^z + \frac{1}{4} \right) + \frac{4U't^2}{U^2 - J_H^2} \left(T_i^z T_j^z - \frac{1}{4} \right) \\ & + \frac{2J_H t^2}{U^2 - J_H^2} (T_i^+ T_j^- + \text{H.c.}) \\ & + \frac{2J_P t^2}{U^2 - J_P^2} (T_i^- T_j^- + \text{H.c.}). \end{aligned} \quad (6)$$

We use here relations $J_H = J_P$ and $\Delta U = 2J_H$, which arise naturally for symmetry-related orbitals [27,28], but we have checked that deviations do not significantly alter our results.

We apply (Lanczos) exact diagonalization to Hamiltonian (1) with orbital operators (5) and (6). To reach longer chains, only states with at most one electron in the higher-energy orbital 2 are kept, which does not affect results in our limit of large crystal-field splitting. Exact diagonalization is complemented with spin-cluster-perturbation theory [29], which gives limited access to momentum points not directly available on the directly solved cluster and which has been previously applied to orbital excitations [8].

IV. NUMERICAL RESULTS AND SPINON-ORBITON INTERACTION

Figure 1 shows spectra for excitations (3) and (4) without and with a spin flip, for increasing deviation from the high-symmetry case (2). At small $J_H/U = 1/20$, excitations with and without spin flip look nearly identical; see Figs. 1(a) and 1(b). They also strongly resemble the $J_H = 0$ result [6], which in turn corresponds to the one-particle spectral density of the supersymmetric t - J model [30] with $t = J/2$. The spectrum can then be described in terms of a spinon and a holon interacting via a phase string [23], where the role of the holon is taken here by the "orbiton." The lens-shaped dominant feature can thus be identified with the one-spinon-one-holon (orbiton) part of the spectrum, while the additional small weight at higher energy toward $k = \pi$ comes from states with three spinons [21,22].

At larger $J_H/U = 3/20$, the lens can still be recognized, albeit with a broadened energy range. However, spectral weight has clearly shifted to its high-energy (low-energy) edge for the pure orbital (combined spin-orbital) excitation; see Figs. 1(c) and 1(d). Finally at $J_H/U = 5/20 = 1/4$, energy range has further increased and spectral weight is almost completely located on the upper (lower) side without (with) a spin flip. For the combined spin-orbital excitation, Fig. 1(f) shows features such as the "spinon" and "holon" branches familiar from the t - J model, however the "holon" is broadened.

To interpret the features and understand their origin, corrections to (2) in first-order of $\frac{J_H}{U}$ can be analyzed. The part K_{ij} decoupled from spins no longer vanishes,

$$\begin{aligned} K_{ij}^{(1)} = & \frac{4t^2}{U} \left[\frac{\Delta U}{U} \left(T_i^z T_j^z - \frac{1}{4} \right) + \frac{J_H}{2U} (T_i^+ T_j^- + \text{H.c.}) \right. \\ & \left. + \frac{J_P}{2U} (T_i^- T_j^- + \text{H.c.}) \right], \end{aligned} \quad (7)$$

where the last term $\propto J_P$ is suppressed by crystal-field splitting Δ and the first term $\propto \Delta U$ gives a small overall energy shift.

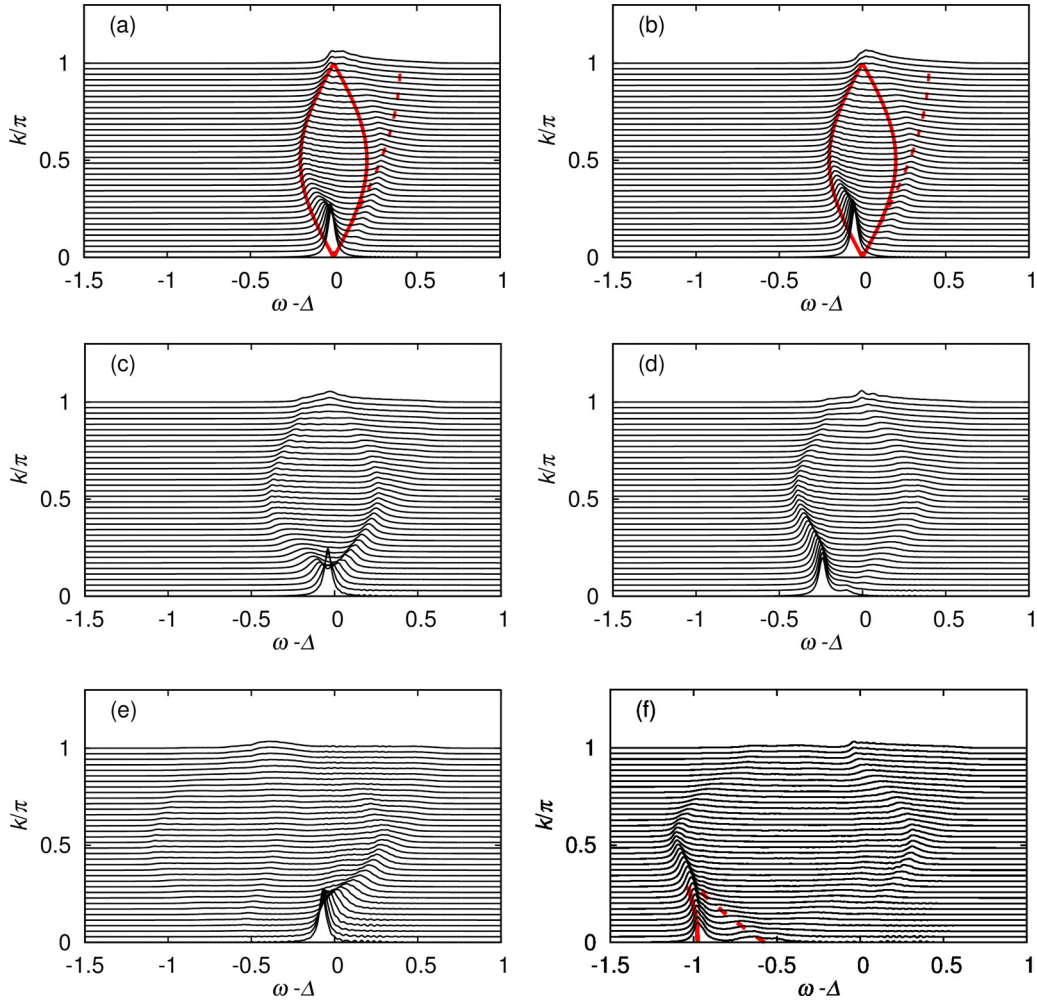


FIG. 1. Orbital excitations with increasing $J_H = J_P = \Delta U/2$. Spectra for the pure orbital excitation Eq. (3) without a spin flip are shown in the left column for (a) $J_H = t = U/20$, (c) $J_H = 3t = 3U/20$, and (e) $J_H = 5t = U/4$. The right column gives spectra for the spin-orbital excitation Eq. (4), which includes a spin flip, for (b) $J_H = t$, (d) $J_H = 3t$, and (f) $J_H = 5t$. The broad solid (dashed) lines in (a) and (b) give the approximate support of the one-spinon–one-holon (three-spinon–one-holon) part of the one-particle spectrum of the t - J model [21,22]. This support is consistent with a phenomenological analysis in terms of a spinon and holon interacting via a phase string [23], and it corresponds to the orbital excitation spectrum for $J_H = 0$. In (f), the broad solid and dashed lines are guides to the eye following the peaks of the branches identified as “spinon” and “orbiton” branches. Results obtained with spin CPT based on $L = 24$ sites, Coulomb repulsion $U = 20t$, crystal field $\Delta = 10t$; spectra were broadened by a Lorentzian with width $\eta = 0.05t$.

The second term $\propto J_H$ allows the excited orbital to move without an *on-site* spin flip, so that the spin of the *excitation* can flip. In the 1D chain, this can induce additional spinons and is a likely reason for, e.g., the broadening of the “holon” branch in Fig. 1(f). Despite its rather minor role here, we expect this term to have a more decisive effect in higher dimensions, where it would allow the orbital excitation to travel “freely” through an AFM ordered state without creating a string potential.

Corrections to A_{ij} are

$$A_{ij}^{(1)} = \frac{4t^2}{U} \left[\frac{J_H}{U} \left(T_i^z T_j^z - \frac{1}{4} \right) + \frac{\Delta U}{2U} (T_i^+ T_j^- + \text{H.c.}) - \frac{J_P}{2U} (T_i^- T_j^- + \text{H.c.}) \right], \quad (8)$$

where the last term is again suppressed. The second terms here and in Eq. (2) have exactly the same form and the same sign, so that the main effect of $U' < U$ is to increase orbiton hopping

relative to spin superexchange [16]. This in turn increases bandwidth and makes the orbiton faster than the spinon, so that we recover the “usual” spinon-holon scenario in Fig. 1(f).

Finally, the first term of (8) becomes negative between two sites with different orbital occupation because $T_i^z T_j^z = -\frac{1}{4}$ in that case, while the term vanishes for identical orbitals and $T_i^z T_j^z = +\frac{1}{4}$. Negative A_{ij} implies FM spin-spin coupling in the Kugel-Khomskii Hamiltonian (1). This is opposite to the AFM coupling between identical orbitals that comes from the first term of (2), which in turn vanishes between sites with different orbital occupation. The sign change of the magnetic interaction driven by different orbital occupation, known as Goodenough-Kanamori rules [31,32], often contributes to complex magnetic orderings in the presence of orbital degrees of freedom. We argue here that this effect also mediates an interaction between the excited orbital and the spinon (see Fig. 2) that plays out differently for the excitations with and without a spin flip.

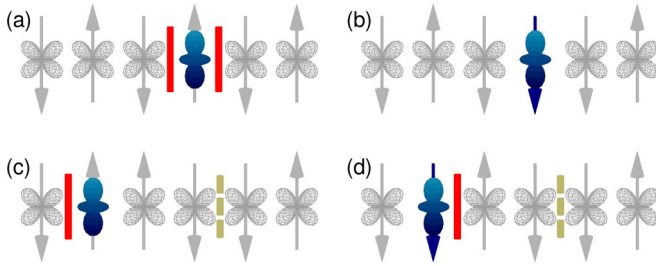


FIG. 2. Illustration of the effective spinon-orbital repulsion/attraction. In (a) and (b), orbital excitations without/with spin flip are created; the spinon and orbitor are located on the same spot. In (c) and (d), the orbitor has moved two sites, leaving behind the spinon. Broken vertical bars between sites indicate spinons; solid bars denote violations of Goodenough-Kanamori rules on bonds with alternating orbitals.

In (short-range) AFM order, the excited electron after a process [Eq. (4)] with spin flip has the same spin as its two neighbors in lower-energy orbitals [see Fig. 2(b)], exactly the situation energetically favored by the first term of (8). If the excited orbital (i.e., the orbitor) moves away, leaving behind a domain wall (i.e., the spinon), it is generally found between spins of opposite sign, so that only one bond can profit from FM exchange. The other bond, indicated by a red vertical bar in Fig. 2(d), is AFM and raises the energy for separating the spinon and orbitor; the orbitor and spinon thus see an attractive potential. In contrast, an orbital excited *without* a spin flip in process [Eq. (3)] has spin opposite to its two neighbors [see Fig. 2(a)], and both bonds pay energy. Separated from the spinon and sitting between an up and a down spin, it has spin parallel to one of its neighbors [see Fig. 2(d)], which reduces energy cost. Accordingly, the spinon and orbitor repel each other here.

However, the illustration shown in Fig. 2, with its perfectly ordered Ising spins, overemphasizes spinon-orbitor attraction because it suggests that the domain wall costs energy $\propto J = \frac{4t^2}{U}$ anywhere except at the site of the orbitor. This interaction, indicated by broken vertical bars in Fig. 2, would be independent of J_H and indeed binds the spinon and holon together in the t - J^z model [33]. While the effect has been found too weak to induce a bound state in the spin-isotropic t - J model with its half-filled spinon-sea ground state, it is sizable on finite chains [12].

Fortunately, Hund's-rule-driven spinon-orbitor interaction can be distinguished from this "baseline" interaction even on

small systems by use of open boundary conditions (OBC). To do so, we set the crystal-field splitting to a negative value at one site $i_2 = \frac{L}{2}$ near the center of an OBC chain. In the ground state, orbital 2 thus has one electron at site i_2 , and the AFM state in orbital 1 has at least one domain wall, which can sit either around site i_2 or at the open ends. At $J_H = 0$, both positions have equal weight. In the presence of $J_H > 0$, the preferred position depends on total $S^z = \frac{1}{2}(N_\uparrow - N_\downarrow)$: For $S^z = 1$ (describing the case with a spin flip), the domain wall is found predominantly around i_2 while it prefers the open chain ends for $S^z = 0$, indicating attraction and repulsion, respectively. Numerical spectra in Fig. 1 do not show bound states, but the buildup of spectral weight at lower (higher) excitation energies can be explained by including such a spinon-holon interaction into a phenomenological description [23] in terms of a spinon and a holon.

V. CONCLUSIONS

Fractionalization of the electron into spin and charge has long been realized as an intriguing property of one-dimensional systems. The question of interactions between the fractionalized parts then naturally arises, even if they are not strong enough to glue the electron back together. We find here that orbital excitations can offer insights into this aspect of spin-charge separation that are not easily accessible from one-particle excitations.

Orbital excitations had been shown to exhibit spin-orbit separation in analogy with spin-charge separation, with the orbitor taking the role of the holon. We have seen here that Hund's rule leads to an attraction or repulsion between spinon and orbitor, depending on whether the excitation includes a spin flip or not. Their microscopic origin can be understood as a dynamic signature of the Goodenough-Kanamori rules that favor FM (AFM) spins on bonds with different (identical) orbitals. Hund's-rule-induced interactions are not strong enough to induce (anti)bound states, but they lead to clear signatures by shifting spectral weight to the upper (lower) edge of the one-spinon-one-orbitor part of the spectrum.

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