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Orbital differentiation in Hund metals

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Orbital differentiation is a common theme in multi-orbital systems, yet a complete understanding of it is still missing. Here, we consider a minimal model for orbital differentiation in Hund metals with a highly accurate method: We use the numerical renormalization group as real-frequency impurity solver for a dynamical mean-field study of three-orbital Hubbard models, where a crystal field shifts one orbital in energy. The individual phases are characterized with dynamic correlation functions and their relation to diverse Kondo temperatures. Upon approaching the orbital-selective Mott transition, we find a strongly suppressed spin coherence scale and uncover the emergence of a singular Fermi liquid and interband doublon-holon excitations. Our theory describes the diverse polarization-driven phenomena in the $t_{2g}$ bands of materials such as ruthenates and iron-based superconductors, and our methodological advances pave the way towards real-frequency analyses of strongly correlated materials.

I. INTRODUCTION

The discovery of superconductivity in the iron pnictides and chalcogenides [1, 2] (FeSCs) has led to renewed interest in multi-orbital systems. Both theoretical and experimental studies of these systems have uncovered the remarkable phenomenon of orbital differentiation: in an almost degenerate manifold of $d$ states, some orbitals are markedly more correlated than others. For instance, in FeSe$_{1-x}$Te$_{x}$ [3], LiFeAs [4], and K$_{0.76}$Fe$_{1.72}$Se$_2$ [5], among the $t_{2g}$ states, only the $xy$ orbital disappears from photoemission spectra as temperature is raised. Orbital differentiation is also seen in tunneling experiments [6] and is a key ingredient in theoretical frameworks to describe FeSCs [7–9]. It is not unique to the FeSCs; it has further been documented in the ruthenates [10] and likely takes place in all Hund metals [11, 12].

An extreme form of orbital differentiation is the orbital-selective Mott transition (OSMT) [13], where some orbitals become insulating, while others remain metallic. Despite its importance, the OSMT in three-band systems has not yet been systematically investigated with a controlled method enabling access to low temperatures, where Fermi liquids form. Controversial questions include: For a given sign of crystal-field splitting, which orbitals localize? Is the OSMT of first or second order? Do correlations enhance or reduce orbital polarization as one approaches the OSMT? Is it true that quenching of orbital fluctuations makes the orbitals behave independently? Do the itinerant electrons in the OSM phase (OSMP) form a Fermi liquid? Finally, how are the precursors of the OSMT related to the physics of Hund metals?

In this paper, we use a minimal model (see motivation below) for orbital differentiation in Hund metals to answer these questions in a unified picture. Our conceptual arguments are supported by a numerical method of unprecedented accuracy: we use the numerical renormalization group (NRG) [14] as real-frequency impurity solver for dynamical mean-field theory (DMFT) [15], extending the tools of Ref. 16 from full SU(3) to reduced orbital symmetry. Whereas different bandwidths directly lead to different effective interaction strengths among the orbitals (as extensively studied for two-orbital models; see [17] for a list of references), we focus here on the more intricate case where a crystal field shifts one orbital in energy w.r.t. two degenerate orbitals [18–22]. Thereby, we can isolate polarization effects and drive the system through band+Mott insulating, metallic, and OSM phases, reminiscent of Ca$_2$RuO$_4$ [13], Sr$_2$RuO$_4$ [23], and FeSCs, respectively.

Theoretically, the OSMP has been under debate both w.r.t. the precise form of the (conducting) self-energy [18, 21, 24–26] and w.r.t. subpeaks in the insulating spectral function [20, 26–28]. Whereas previous studies were limited by finite-size effects of exact diagonalization or finite temperature in Monte Carlo data (requiring analytic continuation), our NRG results yield conclusive numerical evidence down to the lowest energy scales. We give a detailed phase diagram including coexistence regimes (lacking hitherto) and characterize the individual phases with real-frequency properties and their relation to Kondo temperatures spanning several orders of magnitude. Upon approaching the OSMT, we find a strongly suppressed spin coherence scale and uncover the emergence of a singular Fermi liquid [24, 29–32] and interband doublon-holon excitations [33–36] (both of which were previously realized only separately and in two-orbital models).

II. MODEL AND METHOD

The Hamiltonian of our three-orbital Hubbard model is given by

$$
\hat{H} = -t \sum_{\langle ij \rangle m \sigma} \hat{d}_{i m \sigma}^\dagger \hat{d}_{j m \sigma} + \sum_i \hat{H}_{\text{int}}[\hat{d}_{i m \sigma}] + \sum_{i m} \epsilon_m \hat{n}_{i m},
$$

where $\epsilon_m$ is the chemical potential of the $m$th orbital, $\hat{d}_{i m \sigma}^\dagger$ is the creation operator for a hole in orbital $m$, spin $\sigma$, and site $i$, and $\hat{n}_{i m}$ is the corresponding number operator.
where \( \hat{d}_{m\sigma} \) creates an electron on lattice site \( i \) in orbital \( m \in \{1, 2, 3\} \) with spin \( \sigma \in \{\uparrow, \downarrow\} \). The first term describes nearest-neighbor hopping within each orbital on the lattice of uniform amplitude \( t = 1 \), which thus sets the unit of energy. As local interaction, we use the following “minimal rotationally invariant” form \([12, 16, 37, 38]\),

\[
H_{\text{int}}[\hat{d}_{m\sigma}] = \frac{1}{2} J \hat{n} + \left(U - \frac{3}{2} J\right) \hat{n}(\hat{n} - 1) - J \hat{S}^2.
\]

Here, \( \hat{S} = \sum_m \hat{S}_m \) is the total spin operator; \( \hat{n} = \sum_m \hat{n}_m \), \( \hat{n}_m = \sum_{\sigma} \hat{n}_{m\sigma} \), and \( \hat{n}_{m\sigma} = \hat{d}^\dagger_{m\sigma} \hat{d}_{m\sigma} \) are number operators with expectation values \( n, n_m \), and \( n_{m\sigma} \), respectively. This interaction yields an intraorbital Coulomb interaction with expectation values \( n \), \( n_m \), and \( n_{m\sigma} \). Starting from the symmetric case \((\Delta = 0, p = 0)\), we also obtain a sufficiently accurate resolution at high energies via arbitarily small energy scales very accurately, we also observe a coexistence region when approaching \( \Delta \) from below or above, giving rise to the definitions \( \Delta_{\text{p} \uparrow} \approx 0.3 \), \( \Delta_{\text{p} \downarrow} \approx 0.6 \). If we decrease \( \Delta \) starting from \( \Delta = 0, p \) increases until it saturates for \( \Delta \leq \Delta_{\text{p} \uparrow} \approx -0.85 \) at \( p = 0.5 \). This regime constitutes the

\[
\Delta > 0 \quad \epsilon_1 \quad \Delta = 0 \quad \Delta < 0 \quad \epsilon_3
\]

FIG. 1. Illustration of the onsite energies \( \epsilon_1 - \Delta = \epsilon_2 = \epsilon_3 \) and impurity occupations. Due to Hund’s coupling, spins are aligned; shaded arrows symbolize a symmetric distribution among the degenerate orbitals. The different phases portrayed are (a) a band+Mott insulator for large, positive \( \Delta \), (b) an orbitally symmetric metal for vanishing \( \Delta \), and (c) the OSMP for large, negative \( \Delta \), yet \( |\Delta| \lesssim 2 J \). After a particle-hole transformation and the identification \( 1 \leftrightarrow xy, 23 \leftrightarrow xz/yz \), (a) and (b) mimic properties of the \( t_{2g} \) orbitals of CaRuO\(_4\) and Sr\(_2\)RuO\(_4\), respectively; with a half-filled \( xy \)-orbital and further metallic ones, (c) resembles the situation in FeSCs.

### III. CRYSTAL-FIELD SPLITTING

As we tune \( \Delta \), the system undergoes (for suitable interaction strength) several phase transitions. The nature of the different phases can be easily understood by looking at the occupations in the atomic limit (Fig. 1) \([19, 21]\): For large \( \Delta > 0 \), the 1-orbital has highest energy; both electrons reside in the half-filled 23-doublet and are likely to form a Mott insulator \([46]\). For the symmetric model at \( \Delta = 0 \), the two electrons are equally distributed among the three degenerate orbitals with occupation \( n_m = 2/3 \) each, giving rise to metallic behavior (for not too strong interaction). Finally, for large \( \Delta < 0 \), the filling of the lowest orbital is eventually increased up to half filling, \( n_1 = 1 \), and the remaining electron occupies the quarter-filled 23-doublet. For intermediate interaction strengths \([47]\), the half-filled 1-orbital is Mott-insulating while the quarter-filled 23-doublet remains metallic, thereby realizing an OSMP. By decreasing \( \Delta \) even further, one reenters a metallic \((1 < n_1 < 2)\) and ultimately a band-insulating phase \((n_1 = 2)\).

These considerations anticipate the mechanism driving the phase transitions \([18–22]\): \( \Delta \) primarily induces orbital polarization, i.e., it changes the relative filling of the orbitals. Starting from the orbitally symmetric, metallic phase, the different orbitals can become band-insulating or undergo a filling-driven Mott transition. If there are partially filled orbitals of different occupations and/or degeneracies, as in Fig. 1(c), this leads to different critical interaction strengths for the Mott transition, and an OSMP can be realized.

We now investigate the precise nature of these phase transitions as function of \( \Delta \) for fixed \( U, J, n \). Figure 2(a) shows the orbital polarization, \( p = n_1 - n_{23} \). Starting from the symmetric case \((\Delta = 0, p = 0)\) and increasing \( \Delta, p \) decreases to its minimum \( p = -1 \) \( \Delta \). For large \( \Delta > 0 \), we observe a coexistence region when approaching \( \Delta \) from below or above, giving rise to the definitions \( \Delta_{\text{p} \uparrow} \approx 0.3 \), \( \Delta_{\text{p} \downarrow} \approx 0.6 \). If we decrease \( \Delta \) starting from \( \Delta = 0, p \) increases until it saturates for \( \Delta \leq \Delta_{\text{p} \uparrow} \approx -0.85 \) at \( p = 0.5 \). This regime constitutes the
OSMP, for which we find no hysteresis w.r.t. $\Delta$. Clearly, the $\Delta$-driven OSMT is much more second-order-like than the ordinary Mott transition at $\Delta > 0$. We also note that, while $p$ appears differentiable at the OSMT, $\text{Var}(p)$ exhibits a kink [cf. Fig. 8(a)]. The OSMP is stable from $\Delta_{\text{reg}}$ down to $\Delta \simeq -1.5$, where one enters a strongly polarized ($p > 0.5$) metallic phase (not shown).

To address the effect of correlations on orbital differentiation, we examine the difference in the real part of the self-energies, $\delta \Sigma = \text{Re}\Sigma_1 - \text{Re}\Sigma_{23}$, which adds to a renormalized $\Sigma$. Whereas $\delta \Sigma$ increases with increasing $\Delta > 0$ at both $\omega \in (\infty, 0)$, the $\delta \Sigma(0)$ curve (only shown for metallic solutions) bends upward for $\Delta < -0.3$, thereby counteracting the splitting. (c) The full width at half maximum of the quasiparticle peak, $\delta \omega_{\text{qp}}$, confirms the metallic vs. insulating character. In the coexistence regime, either $\delta \omega_{\text{qp}} = 0$ or $\delta \omega_{\text{qp}} > 0$ for all orbitals alike. (d) The orbital and spin Kondo temperatures are clearly separated ($T_{K,\text{orb}} \simeq 0.5$, $T_{K,\text{sp}} \simeq 0.05$ at $\Delta = 0$). Strikingly, $T_{K,\text{sp}}$ strongly decreases with increasing $|\Delta|$ and vanishes altogether in the OSMP (out of range on the log scale).

The difference, $\delta \Sigma = -(U-3J)p/2$, increases monotonically with $\Delta$ (via $p$) for $U-3J > 0$, such that interactions overall enhance orbital differentiation [12]. However, the renormalization of $\Delta$ at low energies must be determined numerically. Figure 2(b) displays $\delta \Sigma$ at $\omega \in (0, \infty)$: $\delta \Sigma(0)$ is smaller in magnitude than $\delta \Sigma|_{\omega=\infty}$ and increases monotonically with $\Delta$ only for $\Delta > -0.3$. For $\Delta < -0.3$, $\delta \Sigma(0)$ bends upward and eventually increases with decreasing $\Delta$, thereby counteracting the splitting.

Next, Fig. 2(c) shows the width of the quasiparticle peak, $\delta \omega_{\text{qp}}$, of the spectral function (cf. Fig. 4) to confirm the conducting vs. insulating character of the different phases. For positive and negative $\Delta$, we indeed find that the 23- and 1-orbital(s), respectively, undergo a Mott transition, with gradually decreasing $\delta \omega_{\text{qp}}$. The sharp decline in $\delta \omega_{\text{qp}}$ around $|\Delta| \sim 0.3$ corresponds to the formation of a subpeak (see below). For $\Delta > 0$, the 1-orbital shows a slight increase of $\delta \omega_{\text{qp}}$ and eventually becomes band-insulating, while, for $\Delta < 0$, $\delta \omega_{\text{qp}}$ of the 23-orbitals decreases until it saturates in the OSMP. Note that the quasiparticle weight, $Z_{\text{qp}} = (1 - \partial_\omega \text{Re}\Sigma_m(0))^{-1}$, often used to describe the single-orbital Mott transition, is not ideal to characterize the full range of orbital differentiation: For $\Delta > 0$, when the 1-orbital gets emptied out, $Z_1$ increases although the whole quasiparticle peak gradually disappears; for large $\Delta < 0$, $Z_1$ of the insulating 1-orbital does not vanish throughout the OSMP, yet $Z_{23} = 0$ in the metallic 23-orbitals, as further explained below.

We complete our phase diagram by showing in Fig. 2(d) the $\Delta$-dependence of Kondo temperatures, defined as the energy scale at which the corresponding susceptibility, $\chi''$, is maximal [cf. Fig. 4(c)]. As typical for Hund metals [12, 16], we observe spin–orbital separation in terms of Kondo scales: orbital fluctuations are screened at much higher energies than spin fluctuations ($T_{K,\text{orb}}^p \gg T_{K,\text{sp}}^p$). While $T_{K,23}$ characterizes orbital fluctuations within the 23-doublet, $T_{K,1}^p$ describes those between the (separated) 1-orbital and the 23-doublet [cf. Eq. (D2)] and reduces to the bare energy scale $\sim |\Delta|$ for large splitting. At sizable $J$, both orbitals have the same $T_{K,1}^p$ [50], and, strikingly, $T_{K,23}^p$ strongly decreases with increasing $|\Delta|$.

This can be understood as follows: It is well-known that finite $J$ decreases $T_{K,1}^p$ [12, 51, 52], as it splits the impurity groundstate manifold. Intuitively, a smaller groundstate degeneracy implies a reduced effective hybridization and thus a reduced Kondo temperature. For $J > 0$ and finite $\Delta$, the groundstate degeneracy is reduced even further, particularly for $\Delta < 0$, see Fig. 3. Moreover, the DMFT self-consistency suppresses the low-energy hybridization strength of the orbital approaching the Mott transition. In the OSMP, $A_1(0)$ and $T_{K,1}^p$ eventually vanish altogether.

Let us now examine in detail how the spectral functions
change with $\Delta$ in the metallic phase. Figures 4(a,b) show that, for both positive and negative $\Delta$, the most important change with stronger correlations occurs in the orbital(s) approaching a Mott transition (main panels). The other orbitals (insets) mostly adjust the spectral weight. At $\Delta = 0$ [gray lines in Figs. 4(a–c)], the spectral functions exhibit the typical shoulder in the quasiparticle (qp) peak [16, 52] (below half filling at $\omega < 0$). In Ref. [52], this has been explained as the combination of a sharp SU(2) spin Kondo resonance ("needle" with width $\propto T_{K}^{\text{orb}}$) and a wider SU(3) orbital Kondo resonance ("base" with width $\propto T_{K}^{\text{sp}}$). If we first stay with the orbitally symmetric case [Fig. 4(c)] and use $J$ and $E_{\text{at}} = U - 2J$ as tuning parameters [52], we can reduce $T_{K}^{\text{sp}}$ by increasing $J$ while only mildly affecting $T_{K}^{\text{orb}}$. As a consequence, the needle sharpens while the wide base remains, revealing a subpeak.

Similarly, increasing $|\Delta|$ drastically decreases $T_{K}^{\text{sp}}$ [Figs. 2(d), 4(d)] and causes a thin qp needle. Additionally, finite $\Delta$, which acts in orbital space similar to a magnetic field in spin space, splits the qp base. For $|\Delta| \gtrsim T_{K}^{\text{orb}}$, the orbital Kondo resonance is destroyed and subpeaks on both sides of $\omega = 0$ remain. In fact, the orbital-rideshoulder is remarkably accurately centered at $-T_{K}^{\text{orb}}(\Delta = 0)/2$ [Fig. 4(c)], and crosses over to an interband double-holon excitation at $\Delta < 0$ (see below) for $|\Delta| \gtrsim T_{K}^{\text{orb}}(\Delta = 0)/2$. Note that the authors of Ref. [38] similarly marked strong influence of $J$ by $J \gtrsim T_{K}^{\text{orb}}(J = 0)$.

Generally, finite $\Delta$ amplifies Hund-metal features in some orbitals while suppressing them in others. This is apparent in spectral functions (Fig. 4) as well as self-energies, see Fig. 5. For $\Delta = 0$, we find the typical [23, 53] inverted slope in Re$\Sigma$ for small $\omega < 0$ and kink in Re$\Sigma$ for small $\omega > 0$ (with Im$\Sigma$ related by Kramers–Kronig transform). These features are enhanced as the orbital becomes more correlated, and suppressed as it becomes less correlated. The degree of correlation follows from proximity to half filling: $n_{1}$ approaches 1 as $\Delta$ increases; $n_{23}$ approaches 1 as $\Delta$ decreases.

IV. OSMP

For $\Delta \leq -0.85$, $T_{K}^{\text{sp}}$ and the qp needle vanish altogether; the 1-orbital becomes a Mott insulator while the 23-doublet retains spectral weight at $\omega = 0$ [Fig. 6(a)]. In the metallic orbitals, Luttinger pinning [54] via the semicircular lattice density of states $\rho$, with $\mathcal{A}_{23}(0) = \rho(x_{n})$ and $\int_{-\infty}^{\infty} \rho(x)dx = n_{23,\sigma}$, is fulfilled throughout [leading to $\pi\mathcal{A}_{23}(0) \approx 0.85 - 0.91$ at quarter filling $n_{23,\sigma} = 1/4$]. Yet, the spectral function of the half-filled 1-orbital strongly differs from a single-orbital Mott insulator. Next to the standard Hubbard bands, charge fluctuations in the 23-doublet enable interband double-holon excitations (previously identified in a two-band DMFT+DMRG study [35]; cf. [33, 34] for experimental signatures) in the insulating spectral function. Here, they occur at energies $\Delta$ and $\Delta + 2J$, as derived in Appendix B. These gap-filling states give $A_{1}$ its soft form. They are shifted with $\Delta$, leading to a "tilt" of $A_{1}$ around $\omega = 0$. A hard gap is revealed when pushing the subpeaks apart (via $J$) and decreasing their weight (via $E_{\text{at}} = U - 2J$) by suppressing 23-charge fluctuations [Fig. 6(b)]. The subpeaks’ distinct nature [43, 55] is further underlined in plots of the momentum-
The self-energy of the insulating 1-orbital diverges. In Fig. 7(a), we see that the singularity of $\Sigma_1$ is not bound to $\omega = 0$; instead, its position shifts with $\Delta$. This implies that $Z_1 = 1/(1 - \frac{\partial}{\partial \omega} \text{Re} \Sigma_1(0))$ does not vanish throughout the OSMP and is thus not suited to mark the insulating character of the 1-orbital in the OSMP. A low-energy zoom of the self-energy in the metallic 23-orbitals [Fig. 7(b)] reveals strong deviations from the standard zero-temperature FL form, $\text{Re} \Sigma_{\text{SFL}} = a + \omega$ and $\text{Im} \Sigma_{\text{SFL}} = -|c|\omega^2$. Instead, it exhibits logarithmic singularities that can be well fitted [dashed lines in Fig. 7(b)] to the SFL relations [24, 32, 56]

$$\text{Re} \Sigma_{\text{SFL}} = \tilde{a} + \tilde{b} \text{sgn}(\omega) \ln^{-3}|\omega/T^*|,$$

$$\text{Im} \Sigma_{\text{SFL}} = -|c| \ln^{-2}|\omega/T^*|.$$ 

The logarithmic singularity in $\Sigma_{23}$ implies that $Z_{23} = 0$ despite the conducting character of the 23-orbitals with finite spectral weight at the Fermi level [Fig. 6(a)]. To further scrutinize the singularity, we consider the logarithmic derivative of the imaginary part of $\Sigma_{23}$,

$$\mathcal{L}(z) = \frac{d \ln[-\text{Im} \Sigma_{23}(z)]}{d \ln z},$$

both for real frequencies, $z = \omega + i0^+$ with $\omega \in \mathbb{R}$, and for imaginary frequencies, $z = i\omega \in (2\pi + 1)i\pi T$. This quantity is well suited to discriminate between singularities of logarithmic or fractional power-law type:

$$-\text{Im}\Sigma(z) = |c| z^\alpha \quad \Rightarrow \quad \mathcal{L}(z) = \alpha,$$

$$-\text{Im}\Sigma(z) = |c| \ln^{-2}(z/T^*) \quad \Rightarrow \quad \mathcal{L}(z) = -2 \ln^{-1}(z/T^*) \quad \xrightarrow{z \to 0} 0.$$ 

In Fig. 7(c), we clearly see that $\mathcal{L}(0) = 0$, confirming the logarithmic nature of the singularity. Note that a smoothening postprocessing was used to suppress minor

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FIG. 6. Characterization of the OSMP. (a) Spectral functions showing the insulating and metallic character of the 1- and 23-orbital(s), respectively. Interband doublon-holon excitations are seen as subpeaks in $A_1$, whose position shifts with $\Delta$, leading to a tilt of $A_1$ around $\omega = 0$; the $A_{23}$ curves all lie on top of each other. (b) Close up of the insulating spectral function at variable $U$ (only in this panel), with $J/U = 1/6$ fixed. Increasing $J$ shifts the right subpeak towards larger $\omega$, and increasing $E_{\text{at}} = U - 2J$ decreases the weight of the subpeaks by suppressing charge fluctuations in the 23-doublet. Both effects help to reveal a hard spectral gap. (c) Diverging spin (solid lines) and regular orbital (dashed) susceptibilities. (We again plot $4A_{23}$) (d) NRG flow diagram of the rescaled, lowest-lying energy levels at characteristic level spacing $\sim |\omega|$. The legend provides charge $Q_m$, total spin $S$, and SU(2) orbital $T_{23}$ quantum numbers. The groundstate carries a residual spin $1/2$ since the contribution to the impurity spin from the insulating 1-orbital cannot be screened. The SFL nature entails that the flow approaches the Fermi-liquid fixed point (where the first and second as well as third and fourth excitations become degenerate) only asymptotically.

Resolved spectral function, shown in Appendix C, where one can also see how the widths of the 23-qp peak and 1-orbital subpeaks narrow together with increasing $E_{\text{at}}$.

As the insulating 1-orbital does not contribute to spin screening, the OSMP inherits properties of an under-screened (spin) Kondo effect [32], as manifested in a divergent spin susceptibility [Fig. 6(c)]. Within our DMFT description of the OSMP, the impurity electron in the 1-orbital and that in the 23-doublet form a combined spin 1, due to Hund’s coupling. However, the 1-orbital hybridization ($\propto A_1$) has zero weight at low enough energies. Hence, given the diagonal hybridization, only the 23-contribution to the impurity spin can be screened, while its 1-orbital contribution remains unscreened. The underscreened Kondo effect in turn leads to the singular Fermi-liquid (FL) fixed point only asymptotically [30].
oscillations in very small values of $\text{Im}\Sigma$. The imaginary-
frequency data $\mathcal{L}(i\omega)$, available for $|i\omega| \geq \pi T$, perfectly
matches the low-frequency behavior but does not suffi-
tice to follow the decay up to $\mathcal{L}(0) = 0$. In fact, if the
imaginary-frequency data were available only in a limited
temperature range, as is the case in Monte Carlo studies,
say, $T \gtrsim 10^{-3}$ and $|i\omega| \gtrsim \pi \cdot 10^{-3}$, one might easily be
tempted to conclude that $\mathcal{L}(i\omega)$ saturates at $\alpha \approx 0.5$.

V. CONCLUSION

We have shown that DMFT+NRG can be used to study three-orbital Hubbard models with reduced or-
bital symmetry, used this method to accurately describe
polarization-driven phase transitions induced by a crystal
field $\Delta$, and uncovered the rich real-frequency structure
inherent in the interplay of Hund-metal physics and or-
bital differentiation. Our analysis leads to a conclusion
of major conceptual significance: The popular notion
that orbital screening, facilitated by $J$, makes the orbitals
behave almost independently [8–10, 12, 18, 26, 46, 57]
(as seen, e.g., in static correlations [18, 26, 57], cf. also
Fig. 8(a)) misses the importance of spin fluctuations.
It must be revised when looking at dynamic correlation
functions, as (i) a suppressed hybridization in one orbital
suppresses the spin Kondo temperature of all orbitals (at
sizable $J$), (ii) charge fluctuations in some orbitals enable
interband doublon-holon excitations [35] in the spectrum
of other orbitals, and (iii) the presence of localized spins
implies singular Fermi-liquid behavior of the remaining
itinerant electrons [32].

With our methodological advances, NRG is ready to be
used as real-frequency impurity solver in a DFT+DMFT
description of three-orbital materials with reduced orbital
symmetry. Future studies should further investigate the
stability of the OSMP against interorbital hopping [58].

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Appendix A: Additions to the phase diagram

In the discussion of the phase diagram in Fig. 2, we
mentioned that the polarization $p = \langle \hat{p} \rangle$, with
$\hat{p} = \hat{n}_1 - \hat{n}_{23}$ and $\hat{n}_{23} = (\hat{n}_2 + \hat{n}_3)/2$, varies with $\Delta$ in a differ-
entiable way throughout the OSMT. Regarding the nature
of the phase transition, it is then interesting to note that
$\text{Var}(\hat{p}) = \langle \hat{p}^2 \rangle - \langle \hat{p} \rangle^2$ and $\text{Cov}(\hat{n}_1, \hat{n}_{23}) = |\langle \hat{n}_1 \hat{n}_{23} \rangle - \langle \hat{n}_1 \rangle \langle \hat{n}_{23} \rangle|$ exhibit a kink at the OSMT. The latter shows that static, interband
correlations are rather weak (plot shows 5 Cov). (b) Two
different versions of an effective crystal field (shown only for
metallic solutions), $\Delta + \Sigma(0)$ as relevant for electronic degrees of
freedom and $\Delta = \Delta_1 \cdot \Sigma(1) - \Delta_2 \cdot \Sigma(23)$) for
quasiparticle excitations. Both show similar behavior: They
depend monotonically on $\Delta$ in a region around $\Delta = 0$ but
bend upward for large, negative $\Delta$, counteracting the splitting.

FIG. 8. Additions to the $\Delta$ phase diagram. (a) $\text{Var}(\hat{p}) = \langle \hat{p}^2 \rangle - \langle \hat{p} \rangle^2$ and $\text{Cov}(\hat{n}_1, \hat{n}_{23}) = |\langle \hat{n}_1 \hat{n}_{23} \rangle - \langle \hat{n}_1 \rangle \langle \hat{n}_{23} \rangle|$ exhibit a kink at the OSMT. The latter shows that static, interband
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quasiparticle excitations. Both show similar behavior: They
depend monotonically on $\Delta$ in a region around $\Delta = 0$ but
bend upward for large, negative $\Delta$, counteracting the splitting.

Appendix B: Doublon-holon excitations

The spectrum of the insulating 1-orbital in the OSMP
can be qualitatively explained from the atomic level struc-
ture. In the atomic limit, the groundstate consists of
eigenstates of the impurity Hamiltonian with one electron
in the 1- and 23-orbital(s) each (the first contribution
to $|G\rangle$ in Fig. 9(b), marked in red, is a representative).
However, the metallic character of the 23-orbitals implies charge fluctuations, such that the actual groundstate also contains admixtures from states where the 23-levels of the impurity are empty or doubly occupied [second and third contribution to \( |G\) in Fig. 9(b)]. At fixed filling, the residual charge is carried by the bath [second “ket” in the tensor-product notation of Fig. 9(b)].

At large interaction, the first term of \( |G\) with impurity occupation 2 is dominant. Single-particle and -hole excitations in the 1-orbital on top of this state mark the Hubbard bands [first “column” in Fig. 9(b)]. Single-particle and -hole excitations to the other contributions make states accessible which are inaccessible in the atomic limit [second and third “column” in Fig. 9(b)]. If we relate these states to the dominant part of the groundstate, we can identify them as interband doublon-holon excitations [35]: the charge on the impurity remains the same while an electron is removed in the 23-orbital and added in the 1-orbital (dashed blue line), or added in the 23-orbital and removed in the 1-orbital (dashed green). The location of the excitations in the 1-orbital spectral function (right, vertical axis) can be deduced from the atomic energy levels [see Eqs. (B7) and (B9)].

**Charge fluctuations in the 23-orbitals**

Charge fluctuations in the 23-doublet on top of the dominant groundstate contribution connect the states shown in Fig. 9(a) with atomic energies

\[
\begin{align*}
E_{\text{HB,23}}^+ &= \epsilon_1 + 2\epsilon_{23} + 3(U - 2J), \\
E_{\text{HB,23}}^- &= \epsilon_1.
\end{align*}
\]

The energy cost for the respective transitions, giving the position of Hubbard bands in the 23-doublet, is

\[
\begin{align*}
\delta E_{\text{HB,23}}^+ &= E_{\text{HB,23}}^+ - E_G = \epsilon_{23} + 2(U - 2J), \\
\delta E_{\text{HB,23}}^- &= E_{\text{HB,23}}^- - E_G = -\epsilon_{23} - (U - 2J).
\end{align*}
\]

Equilibrium at filling 2 is thus obtained when

\[
\delta E_{\text{HB,23}}^+ = \delta E_{\text{HB,23}}^- \quad \Rightarrow \quad \epsilon_{23} = -\frac{3}{2}(U - 2J).
\]

Inserting the values \( U = 6 \) and \( J = 1 \) mostly used, this means \( \epsilon_{23} = -6 \) and \( \delta E_{\text{HB,23}}^+ = 2 \), corresponding to the bumps in \( \Delta_{23} \) at \( \omega = \pm 2 \) [Fig. 6(a)].
Hubbard bands in the 1-orbital

Single-particle and -hole excitations in the 1-orbital on top of the dominant groundstate contribution lead to the states shown in the first “column” of Fig. 9(b) with energies

\[ E_{\text{HB},1}^+ = 2\epsilon_1 + \epsilon_{23} + U + (U - J) + (U - 2J), \]
\[ E_{\text{HB},1}^- = \epsilon_{23}. \]

Excitations to these states mark the 1-orbital Hubbard bands, which are found in the spectral function at

\[ \delta E_{\text{HB},1}^+ = E_{\text{HB},1}^+ - E_G = \epsilon_1 + 2U - J, \]
\[ -\delta E_{\text{HB},1}^- = E_G - E_{\text{HB},1}^- = \epsilon_1 + U - 2J. \]

Inserting the value for \( \epsilon_1 = \Delta + \epsilon_{23} \) from Eq. (B4) yields

\[ \delta E_{\text{HB},1}^+ = \Delta + \frac{3}{2}U + 2J, \]
\[ -\delta E_{\text{HB},1}^- = \Delta - \frac{1}{2}U + J. \]

If we further insert the values \( \Delta = -1, U = 6, \) and \( J = 1 \) of Fig. 6(a), we get the peak positions \( -3 \) and \( 4 \). Increasing \( U \) up to \( 8 \), with \( J = U/6 \) as in Fig. 6(b), increases their magnitude up to \( -3\frac{2}{3} \) and \( 5\frac{2}{3} \), respectively. These numbers match the curves in Fig. 6(a,b) very well.

Doubloon-holon subpeaks

The doubloon-holon excitation energies are found from single-particle or -hole excitations on top of the subleading contributions to the groundstate with an empty or doubly occupied 23-doublet [second and third “column” of Fig. 9(b)]. The atomic energies of the excited states are

\[ E_{d_{1},d_{23}}^+ = 2\epsilon_1 + U, \]
\[ E_{d_{1},d_{23}}^- = 2\epsilon_{23} + (U - 2J). \]

The energy difference to the dominant groundstate contribution [dashed lines in Fig. 9(b)] gives the position of the subpeaks in the insulating spectral function. Using \( \epsilon_1 - \epsilon_{23} = \Delta \), we have

\[ \delta E_{d_{1},d_{23}}^+ = E_{d_{1},d_{23}}^+ - E_G = \Delta + 2J, \]
\[ -\delta E_{d_{1},d_{23}}^- = E_G - E_{d_{1},d_{23}}^- = \Delta. \]

Interestingly, these peak positions only depend on the difference of the energy levels, \( \Delta \), and on Hund’s coupling, \( J \). Inserting the values for Fig. 6(a) gives \( -1 \) and \( +1 \), and those for Fig. 6(b) yield \( -1 \) and \( 1 + U/3 \), in perfect agreement with the plots.

Both the charge fluctuations in the 23-doublet and the interband doubloon-holon excitations are determined by the same subleading contributions to the groundstate (such as the terms with coefficients \( |\beta|^2 \) and \( |\gamma|^2 \) in Fig. 9).

Hence, the widths of the quasiparticle peak in the 23-doublet and the subpeaks in the 1-orbital are closely tied together. By increasing \( E_{at} = U - 2J \), one can then decrease both the widths of the 23-quasiparticle peak and the 1-subpeaks. On the other hand, by tuning \( \Delta \) and \( J \) at constant \( E_{at} \), one can shift the positions of the 1-subpeaks, while the weights of the 23-quasiparticle peak and the 1-subpeaks remain roughly the same.

Appendix C: Momentum-resolved spectral function

In Fig. 10, we plot the local spectral function, \( A(\omega) \), together with the momentum-resolved one, \( A(\omega, \epsilon_k) \). As explained in the caption, strong particle-hole asymmetry, decreasing quasiparticle weight, and localization of the 1-electrons can be nicely seen. Moreover, it is interesting to observe that the crossover between the \( \omega < 0 \) shoulder and the interband doubloon-holon subpeak at \( \Delta \) is accompanied by a transfer of spectral weight from \( \epsilon_k < 0 \) to \( \epsilon_k > 0 \). In the OSMP, the doubloon-holon subpeak at \( \omega < 0, \epsilon_k > 0 \) can be very well distinguished from the Hubbard band at \( \omega < 0, \epsilon_k < 0 \). Especially in the momentum-resolved plot, these interband doubloon-holon subpeaks resemble the intraband doubloon-holon subpeaks known from the single-orbital strongly correlated metallic phase [43, 55].

Appendix D: Susceptibilities

Here, we give the definitions for the various susceptibilities computed. The total spin operator is given by \( \hat{S} = \sum_m \hat{S}_m \) with \( \hat{S}_m = \frac{1}{2} \sum_{\sigma\sigma'} \hat{d}^\dagger_m \tau_{\sigma\sigma'} \hat{d}^\dagger_m \) and Pauli matrices \( \tau \). We further define \( \hat{S}_{23} = (\hat{S}_2 + \hat{S}_3)/2 \), and mainly compute the spin susceptibilities

\[ \chi_{1}^{\text{sp}} = \frac{1}{3} \sum_{\alpha=1}^3 \langle \hat{S}_1^\alpha \rangle \hat{S}_1^\alpha \omega, \quad \chi_{23}^{\text{sp}} = \frac{1}{3} \sum_{\alpha=1}^3 \langle \hat{S}_{23}^\alpha \rangle \hat{S}_{23}^\alpha / \omega. \]

Further, we use the angular-momentum operator \( \hat{L} \) with \( \hat{L}_m = i \sum_{m'} \epsilon_{mm'm'} \hat{d}_{m'\sigma}^\dagger \hat{d}_{m'\sigma} \) and compute orbital susceptibilities according to \( \hat{L}_{23} = (\hat{L}_2 + \hat{L}_3)/2 \) and

\[ \chi_{1}^{\text{orb}} = \langle \hat{L}_{23} \rangle / \langle \hat{L}_{23} \rangle, \quad \chi_{23}^{\text{orb}} = \langle \hat{L}_1 \rangle / \langle \hat{L}_1 \rangle. \]

In fact, as the system exhibits full SU(2) orbital symmetry in the 23-doublet, we can also use the fully symmetrized \( \hat{T}_{23} = \frac{1}{2} \sum_{m, m' \in \{2,3\}} \hat{d}_{m\sigma}^\dagger \tau_{mm'} \hat{d}_{m'\sigma} \) and

\[ \chi_{23}^{\text{orb}} = \frac{1}{3} \sum_{\alpha=1}^3 \langle \hat{T}_2^\alpha \rangle \hat{T}_2^\alpha / \omega. \]

In the literature, orbital susceptibilities are sometimes computed from charge fluctuations in the individual orbitals. In this language, with \( \hat{n}_{23} = (\hat{n}_2 + \hat{n}_3)/2 \), one has

\[ \chi_{1}^{\text{ch}} = \langle \hat{n}_1 \rangle / \langle \hat{n}_1 \rangle, \quad \chi_{23}^{\text{ch}} = \langle \hat{n}_{23} \rangle / \langle \hat{n}_{23} \rangle. \]
FIG. 10. Local \(A(\omega)\) and momentum-resolved \(A(\omega, \epsilon_k)\) spectral functions for varying \(\Delta\) in the metallic phase (left panel) and for varying \(U\) (\(J = U/6\) fixed) in the OSMP (right). Note that, within DMFT, the \(k\) dependence enters only via \(\epsilon_k\), and we set the half-bandwidth to 2. (a) Already at \(\Delta = 0\), \(A(\omega)\) and \(A(\omega, \epsilon_k)\) reveal a strong particle-hole asymmetry. (b,c) As we decrease \(\Delta\), the 1-orbital is pushed towards half filling, the quasiparticle weight decreases, and \(A(\omega, \epsilon_k)\) reveals an almost flat dispersion. Interestingly, the spectral weight from the \(\omega < 0\) shoulder is continuously transferred from negative to positive \(\epsilon_k\). (d) In the OSMP, the quasiparticle weight in the 1-orbital has vanished; the Hubbard band in \(A(\omega, \epsilon_k)\) at \(\omega < 0\) is found at \(\epsilon_k < 0\) while the subpeak is distinctively centered at \(\epsilon_k > 0\) (note the altered color scale). The logarithmic singularities in the 23-orbitals are contained in the very sharp structure around \(\omega = 0\). (e,f) With increasing \(E_{2d} = U - 2J\), the widths of the 23-quasiparticle peak and, consequently, the widths of the 1-subpeaks decrease. With increasing \(J\), the position of the right subpeak shifts to higher energies [cf. Eq. (B9)]. The distinct nature of the interband doublon-holon excitations and the Hubbard bands becomes clearly visible; they resemble the intraband doublon-holon subpeaks in the single-orbital strongly correlated metallic phase [43, 55].
Using orbital SU(2) symmetry with $\chi^{orb}_2$ from the position of the maximum of the orbital susceptibility, $\langle n_1|n_1\rangle_\omega$ and $\langle n_1|n_{23}\rangle_\omega$ (dashed lines).

In the fully symmetric case at $\Delta = 0$, we can also extract the spin and orbital Kondo temperatures from

$$\chi^s = \frac{1}{Z} \sum_{m=1}^{3} \langle \hat{n}_m | \hat{n}_m \rangle_\omega \quad \text{and} \quad \chi^{orb} = \frac{1}{Z} \sum_{m=1}^{3} \langle \hat{n}_23 | \hat{n}_23 \rangle_\omega + \chi^{orb}_2.$$  

(D5)

where $T = \frac{1}{Z} \sum_{m} \sum_{m',\sigma} \hat{d}_{m\sigma} g_{mm'} \hat{d}_{m'\sigma}$ with SU(3) Gell-Mann matrices normalized as $\text{Tr}[g^a, g^b] = 2\delta_{a,b}$.

For illustration, we finally show in Fig. 11 intra- and inter-orbital susceptibilities of spin and number operators. As the inter-orbital ones, $\langle \hat{n}_1 | \hat{n}_{23} \rangle_\omega = \frac{1}{N} \sum_{n=1}^{N} \langle \hat{S}_n | \hat{S}_{23} \rangle_\omega$ and $\langle \hat{n}_23 | \hat{n}_{23} \rangle_\omega$, change sign within $0 < \omega < \infty$, they are shown in absolute value. We see that the orbital Kondo scale, read off from the position of the maximum in $\chi^{orb}$ (dashed-line), can also be determined from orbital-resolved charge susceptibilities (dashed lines), corresponding to their explicit relation given in Eq. (D5). It is interesting to note that spins align, meaning $\langle \hat{n}_1 | \hat{n}_{23} \rangle_\omega > 0$, for $|\omega| \lesssim J = 1$ due to Hund's coupling, and the individual charges antagonize, meaning $\langle \hat{n}_23 | \hat{n}_{23} \rangle_\omega < 0$, for $|\omega| \lesssim U/2 = 3$ to minimize the Coulomb repulsion.

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For $\Delta < 0$, the Mott transition of the half-filled 1-orbital depends mainly on $U$, with a critical $U_c \approx 6$ similar to the single-orbital case [15]. The Mott transition in the quarter-filled, 23-doublet requires much stronger interaction [48, 49]. Hence, our choice $U = 6$ and $J = 1$ is close to the minimal interaction strength required for the OSMP.

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