Renormalized dispersing multiplets in the spectrum of nearly Mott localized systems

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The spectrum of the strongly correlated systems usually shows resonant peaks at finite energy, with examples in the 115 Ce family which are reproduced by the dynamical mean-field theory. A similar structure has been seen recently in the orbitally selective Mott (OSM) phase of two-band model, known as doublon-holon bound state, with implications on the fate of such phase in the zero Hund’s coupling limit. We show that these features can be captured with the slave-particle methods once their Hilbert space is taken into account. We use slave-spin calculations, justifiable in the limit of large dimensions, to explicitly demonstrate this and compare the results with dynamical mean-field theory.

I. INTRODUCTION

The study of the properties of quantum materials, including high-temperature superconductors, requires understanding strongly correlated systems in two or three dimensions, a task which is theoretically very challenging. Dynamical mean-field theory (DMFT), one of the very few tools at our disposal, provides a systematic interpolation between the atomic limit where the correlations are important and the non-interacting limit of band theory, and is exact in the limit of infinite dimensions. However, analytical insight into the result is often formidable as a result of the self-consistency loop.

There have been lots of efforts to produce analytically tractable understanding using slave-particle mean-field theories. While when treated exactly all these methods in principle agree, the approximation schemes used for analytical/numerical tractability causes discrepancies, for example on the fate of the so-called orbitally selective Mott (OSM) phase in absence of Hund’s coupling, that has been a source of confusion. The simplest version of the phenomena appears in a two-band Hubbard model with orbital-dependent tunnelling and local interaction. Slave spin methods predict that when the ratio of the bandwidths of the two bands \( r = t_2/t_1 \) is close to one, the two bands undergo a transition between Mott-phase and the metallic phase at the same value of Hubbard \( U \), the so-called locking effect, whereas when the bandwidth ratio is smaller than a threshold of about \( r_c \approx 0.2 \), there is a region of OSM phase in the phase diagram, in which one band is metallic and the other band is itinerant. This agrees with some and disagrees with other DMFT calculations. Moreover, the general consensus is that Hund’s coupling \( J_H \) favours OSM phase and decreases \( r_c \).

In a recent study, some of us used a density-matrix renormalization group (DMRG) impurity solver to obtain very accurate DMFT results on the two-orbital problem. The existence of an OSM phase was obtained by previous work. In the absence of a Hund’s interaction, Núñez Fernández et al found locking, irrespective of the ratio of the bandwidths. Moreover, when including an inter-orbital Coulomb interaction \( U_{12} \), they identified a resonant feature in the spectral function of the localized orbital in the OSM phase, dubbed holon-doublon excitonic peak corresponding to a virtual bound state at energy scales of about \( \Delta = U - U_{12} \).

There have been previous studies of holon-doublon peaks in the literature of the single-band Hubbard model where, due to the limited size of the local Hilbert space, the holon-doublon boundstate necessarily forms between nearby sites. In contrast, the two-site, two-orbital model provided in with on-site interaction shows that their holon-doublon boundstate forms in the two orbitals of the same site and in this case they form well-defined quasiparticle peaks.

The problem of understanding the origin of finite-energy multiplets in the spectrum of the strongly correlated quantum materials is general and not limited to the two-orbital case mentioned above. Here, we show that slave-particle mean-field methods are fully capable of capturing these finite-energy spectral features, and in particular, the holon-doublon peak. Similar methods has been applied in the past to analyze the spectrum of mixed valence compounds, including Pu pnictides and chalcogenides and the 115 Ce family: CeIrIn\(_5\), CeCoIn\(_5\), and CeRhIn\(_5\).

The structure of the paper is the following: In section II, we describe the general formalism of the method, as well as various approximation schemes. Section III applies the general method of section II to study the spectral functions of single and two-band lattices, including the holon-doublon bound state, using \( Z_2 \) slave-spin method. In section IV we compare numerical results from slave-spin to DMFT. Appendix A contains a comparison between exact diagonalization and slave-spin method applied to the two-orbital two-site problem. Appendix B and C contain diagonalization of the slave-spin Hamiltonian, and the spectral representation of slave-particle Green’s function, respectively.

II. GENERAL FORMALISM

In slave-particle methods we introduce a parton construction for the fermionic operator, \( d_\alpha = \hat{z}_\alpha f_\alpha \), where \( \hat{z} \) has its own Hilbert space (without loss of generality we restrict our discussion to the simpler cases in which \( z \) and \( f \) share the same index). As a result the Hilbert space is enlarged to the tensor product of that of the \( f \) and \( z \) particles. A constraint, usually imposed on averages via few Lagrange multipliers, ensures that the averaged physical parameters are computed in the originally restricted part of the extended Hilbert space.
As a next step
\[ H = \sum_{\langle ij \rangle \alpha \beta} t_{ij}^\alpha \hat{d}^\dagger_{i\alpha} \hat{d}_{j\beta} + \sum_i H_{\text{int}}[i] \] (1)
is mean-field decoupled into \( H_{MF} = H_f + H_z \) where
\[ H_z = \sum_{\langle ij \rangle \alpha \beta} J_{ij}^\alpha \hat{z}_{i\alpha}^\dagger \hat{z}_{j\beta} + \sum_i H_{\text{int}}[i] \] (2)
and
\[ H_f = \sum_{\langle ij \rangle \alpha \beta} \tilde{t}_{ij}^\alpha f^\dagger_{i\alpha} f_{j\beta} \] (3)
with the parameters given by
\[ \tilde{t}_{ij}^\alpha = t_{ij}^\alpha \left\langle \hat{z}_{i\alpha}^\dagger \hat{z}_{j\beta} \right\rangle, \quad J_{ij}^\alpha = t_{ij}^\alpha \left\langle f_{i\alpha} \hat{f}_{j\beta} \right\rangle \] (4)

Here, \( H_f \) describes the renormalized fermionic bands, whereas \( H_z \) describes renormalized atomic structure. The real advantage is that the constraints can be used to absorb \( H_{\text{int}}[i] \) entirely in \( H_z \). The ordered (disordered) phases of the \( z \)-lattice, are usually associated with itinerant (Mott-localized) phases of the original electrons.

The mean-field decoupling mentioned above neglects fluctuations in time that couple between Mott-localization in one band and itineracy in the other. Calculating the renormalized non-interacting band \( G_f(k) \) is narrow enough (at, or close to, a partial Mott transition), the atomic features of the \( G_d(\tau) \) can be resolved, whereas in the metallic regime usually \( G_f \) is broad and those features are washed out after convolution. We use this method to show that various (renormalized) atomic multiplets can be identified in the complex many-body spectrum of the multi-channel Hubbard model. In addition to the bare atomic orbitals, this contains additional multiplets arising due to an interplay between Mott-localization in one band and itineracy in the other.\(^9\) We demonstrate this explicitly, by comparing slave-particle methods to the solution of a two site problem considered before\(^9\) and then generalizing to the lattice.

Inserting (7) in (6) and going to momentum space
\[ G_{d,\alpha\beta}(k,\tau) = Z_{\alpha\beta} G_{f,\alpha\beta}(k,\tau) + \mathcal{P}(\tau) - Z_{\alpha\beta} \sum_q G_{f,\alpha\beta}(q,\tau), \] (9)
i.e. \( G_d \) contains a \( k \)-dependent part coming from renormalized non-interacting band \( G_f \) plus some \( k \)-independent atomic structure in the last term. In particular in the Mott phase \( Z = 0 \) no \( k \)-dependence exist.

A second point of this paper is that the two limits of mean-field decoupling and single-site approximation generally do not commute with each other. This is schematically represented in Fig. (1). To see this, consider doing a single-site approximation, first. This is achieved by the single-site DMFT, according to which the lattice problem is mapped to an effective impurity problem embedded within a conduction bath
\[ H_{\text{imp}} = \sum_{k\alpha\beta} (V_{k\alpha\beta} d_{k\alpha}^\dagger \hat{c}_{k\beta} + h.c.) + H_{\text{int}} + H_c, \] (10)

without lack of generality, in the following we restrict the discussion to the diagonal \( \alpha = \beta \) elements. The function \( \Pi(\tau) \) “knows” about the renormalized atomic physics as seen from its spectral representation
\[ \Pi_{\alpha\alpha}(\nu_n) = e^{-\Omega/T} \sum_{nm} \frac{e^{-E_n/T} - e^{-E_m/T}}{\nu_n + E_n - E_m} |\langle n| \hat{z}_{\alpha} |m \rangle|^2 \] (8)
The impurity problem is solved first. Extracting \( \Sigma_I(z) \) from the \( G_D^{-1}(z) = z - \Delta(z) - \Sigma_I(z) \), and assuming \( \Sigma_{int}(k, z) = \Sigma_I(z) \) is local, the lattice Green’s function \( G(k, z) = [z - \epsilon_k - \Sigma_I(z)]^{-1} \) is computed. From this and Eq. (11), a new hybridization function \( \Delta(z) \) is extracted and the loops is repeated until convergence is reached.

The slave-particle methods can be used as an impurity solver for this DMFT loop. The resulting Hamiltonian

\[
H_{imp} = \sum_{k\alpha}\langle V_{k\alpha}^{\dagger}\hat{a}_{k\alpha}^\dagger \hat{c}_{k\beta} + h.c. \rangle + H_{int}\{\hat{z}_\alpha\} + H_c
\]

is still interacting. Various approximate schemes exist to solve this impurity problem, including non-crossing approximation (NCA)\(^{18,20}\) and one-crossing-approximation (OCA).\(^{20}\) For the purpose of studying multiplets and the comparison to the mean-field solution above, it suffices to settle on a mean-field decoupling. As a result, Eq. (12) gives \( H_{imp} = H_{imp,f} + H_{z,ss} \) where \( H_{z,ss} \) is the same as in Eq. (5), but \( H_{imp,f} \) is given by

\[
H_{imp,f} = \sum_{k\alpha}\langle \hat{V}_{k\alpha'}\hat{a}_{l\alpha}^\dagger \hat{c}_{k\beta} + h.c. \rangle + \sum_{k\alpha} \epsilon_k \hat{c}_{k\alpha}^\dagger \hat{c}_{k\alpha}
\]

where \( \hat{V}_{k\alpha'} = V_{k\alpha} z_{\alpha'} \). The impurity Green’s function is then

\[
G_{d,\alpha\beta}(\tau) = \Pi_{\alpha\beta}(\tau) G_{f,\alpha\beta}(\tau)
\]

The mean-field parameters are obtained from minimization of \( F = F_f + F_z(a) - 2a z \) and are given by\(^8\)

\[
a_\alpha = -\frac{2}{z_\alpha} \int \frac{d\omega}{\pi} f(\omega) \text{Im} \left[ G^{\alpha\alpha}(\omega + i\eta) \right], \quad z_\alpha = \frac{dF_z}{da_\alpha}
\]

It is clear that even though \( G_{d}(\tau) \) is factorizable, in this scheme \( G_{int}(k, \tau) \) does not factorize, as opposed to (6), and consequently the multiplets are generally dispersing.

A. Single-band

In the single-band case, the Hamiltonian has the general impurity form of Eq. (10) with \( \alpha = \uparrow, \downarrow \) and the interaction \( H_{int} = U\hat{n}_\uparrow\hat{n}_\downarrow \) where \( \hat{n}_\sigma = d^\dagger d - 1/2 \). Using slave-spin we identify \( \hat{n}_\sigma = \tau_\sigma^z/2 \), so that

\[
H_{z,ss} = a_\uparrow \tau_\uparrow^z + a_\downarrow \tau_\downarrow^z + U \tau_\uparrow^z \tau_\downarrow^z
\]

where \( a_\sigma = 2f z_\sigma \) and \( f = -0.212D \) is the average kinetic energy for a Bethe lattice of bandwidth \( D = 2t \). A better choice of basis is

\[
|\psi_1^+\rangle = \frac{|\uparrow\uparrow\rangle \pm \downarrow\uparrow\rangle}{\sqrt{2}}, \quad |\psi_2^\pm\rangle = \frac{|\uparrow\downarrow\rangle \pm \downarrow\uparrow\rangle}{\sqrt{2}}
\]

Here, \( |\uparrow\rangle \) or \( |\downarrow\rangle \) refers to the eigen-states of \( \tau_\sigma^z \) operator, where \( \sigma = \uparrow, \downarrow \). In the paramagnetic regime \( a_\uparrow = a_\downarrow = a \) and with p-h symmetry, \( |\psi_1,2\rangle \) decouples and in the basis of \( |\psi_1,2\rangle \) the Hamiltonian reduces to \( H_{z,ss} = a \tau_+^z + (U/4)\tau_-^z \) with \( a = 2a \). This two-state problem can be diagonalized with an \( SO(2) \) rotation

\[
\begin{pmatrix}
\psi_1 \\
\psi_2
\end{pmatrix} =
\begin{pmatrix}
\cos \theta & \sin \theta \\
-\sin \theta & \cos \theta
\end{pmatrix}
\begin{pmatrix}
\psi_1^+ \\
\psi_2^+
\end{pmatrix}
\]

where tan \( 2\theta = 2a/(E_2 - E_1) \) and the eigen-energies are

\[
E_{g/e} = U/4 \mp \sqrt{U^2/4 + a^2}
\]

III. \( Z_2 \) SLAVE SPIN MEAN-FIELD

The above discussion was general. In this section, we focus on the \( Z_2 \) slave-spin method,\(^4\) where at half-filling \( z_\alpha = \tau_\alpha^z \) and the constraint \( 2f_\alpha f_\alpha = \tau_\alpha^z + 1 \) is applied on average, via a Lagrange multiplier \( \lambda_\alpha \). Here \( \tau_\alpha^z \) with \( a = x, y, z \) are Pauli matrices that square to 1. Due to particle-hole symmetry, these Lagrange multipliers vanish \( \lambda_\alpha = 0 \) at the saddle point.\(^8\)

The fact that the U(1) charge is carried by the \( f_\alpha \) in this method, indicates that as long as \( t_{ij} = t_{ij} \tau_i^z \tau_j^z \) is non-zero (even if \( \tau_\alpha^z = 0 \) the bulk is conducting and the system is not in a Mott phase.\(^{21}\) Therefore, we only consider the limit of large dimensions where single-site approximation is valid and the relation \( \langle \tau_\alpha^z \tau_\beta^z \rangle = \langle \tau_\alpha^z \rangle^2 \) is satisfied and \( \langle \tau_\alpha^z \rangle = 0 \) is equivalent to Mott localization.\(^8,22\)

Note that \( E_g(\alpha \to 0) \approx -2a^2/U \). The mean-field study of this problem has been discussed in the past.\(^3,8,23\) In the case of the Hubbard model, \( z_\alpha = \langle \psi_g | \tau_\alpha^z | \psi_g \rangle = \sin 2\theta \). By minimizing the free energy, it can be shown that \( Z = |z_\sigma|^2 = \sin^2 2\theta \) follows the Brinkman-Rice theory\(^24\) \( Z = [1 - u^2] \theta(1 - u) \) where \( \theta(x) \) is the Heaviside step function, \( u = U/U_c \) and \( U_c = 16/J' = 3.36 \times 2t \). In the case of an Anderson impurity, \( Z = |z_\sigma|^2 \) plays the role of “order parameter” for the Kondo physics.\(^8\) In both cases, the function \( \Pi(\tau) \) at low temperature is equal to

\[
\Pi(\tau) = \langle T \tau_\tau^z \tau_\tau^z \rangle = Z + (1 - Z)e^{-\tau_\tau^z \Delta E}
\]

\[\text{(20)}\]
where $\Delta E = E_{c} - E_{g}$ and $Z = \sin^{2}(2\theta_{g})$. Multiplying by $G_{f}(\tau)$ and Fourier transforming

$$G_{ff}^{\alpha}(\omega) = ZG_{f}^{\alpha}(\omega) + (1 - Z)\left[G_{f}^{\alpha}(\omega + \Delta E)\theta(\omega < -\Delta E) + G_{f}^{\alpha}(\omega - \Delta E)\theta(\omega > \Delta E)\right]$$

(21)

Here, $\theta(\omega)$ appears as a low-temperature limit of $f(\omega + \Delta E) + n_{f}(\pm \Delta E)$ (Appendix C). The real part $G_{ff}^{\alpha}(\omega)$ follows from Eq. (21) using Hilbert transform.

On a Bethe lattice of bandwidth $2t$ with density of states $\rho(\epsilon) = (\pi t)^{-1}\sqrt{1 - (\epsilon/2t)^{2}}$, we can plot the spectrum as a function of frequency $\omega$ and the Bethe lattice energy $\epsilon$. Fig. (2) shows a comparison between the ($\epsilon$-resolved and integrated) spectral function, as computed from Eq. (21) with a Brinkman-Rice $Z$ and a lattice $G_{f}$, vs. a DMFT calculation with a slave-spin impurity solver where $G_{f}$ is the Greens' function of the impurity. Note that the Hubbard bands are featureless in (a) but disperse in (c) and there are slight differences in $Z$. However, close to the Mott transition, slave-spins behave poorly as the impurity solver for DMFT due to the fact that $\Delta(z) = z - \Delta_{1} - G_{1oc}^{\alpha}$ is non-analytical, the so-called non-causality of the impurity solver.

### B. Two-band model

In the two-band model, the impurity problem is given by Eq. (10) with $\alpha = 1 \uparrow, 1 \downarrow, 2 \uparrow, 2 \downarrow$. To make a connection to

\[ E = 2U - \Delta \]

\[ \sqrt{2a_{2}} \]

\[ \sqrt{2a_{1}} \]

\[ |\psi_{12}\rangle \]

\[ |\psi_{21}\rangle \]

\[ C_{1} \]

\[ C_{2} \]

\[ \text{holon-doublon state} \]

FIG. 3. The relevant sector of the slave-spin Hamiltonian (see Appendix B), with states given in Eqs. (24). At OSM phase $\alpha_{2} = 0$, and the Hamiltonian breaks into $C_{1}$ and $C_{2}$ sectors, with the ground state in sector $C_{1}$ due to larger level-repulsion and lower energies. The transitions caused by $\tau_{1\alpha}^{c}$ and $\tau_{1\alpha}^{v}$ are indicated by blue and red arrows, respectively. The bright-red arrow is to the holon-doublon bound-state $|\psi_{21}\rangle$.

Ref. 9 we choose the same form of simplified interaction

\[ H_{\text{int}} = U \sum_{\alpha} \sum_{m=1}^{2} \tilde{n}_{\alpha m\uparrow} \tilde{n}_{\alpha m\downarrow} + U_{12} \sum_{\sigma\sigma'} \sum_{m=1}^{2} \tilde{n}_{1\sigma\uparrow} \tilde{n}_{2\sigma'} \]

\[ = \sum_{\alpha} \left\{ \frac{U}{2} \left( \tilde{n}_{1\uparrow} + \tilde{n}_{1\downarrow} + \tilde{n}_{2\uparrow} + \tilde{n}_{2\downarrow} \right)^{2} - \frac{U}{2} \right\} \]

\[ - \Delta(\tilde{n}_{1\uparrow} + \tilde{n}_{1\downarrow})(\tilde{n}_{2\uparrow} + \tilde{n}_{2\downarrow}) \] (22)

where $\tilde{n}_{\alpha} \equiv n_{\alpha} - 1/2$ and $\Delta = U - U_{12}$ is the difference between the intra and inter-orbital Coulomb couplings. We will drop the $-U/2$ constant term in the following. While this form of the interaction is simpler to follow, we have obtained qualitatively similar results with a more general Kanamori Hamiltonian. Within slave-spin method this becomes

\[ H_{z,ss} = \sum_{m\sigma} a_{m\sigma} \tau_{m\sigma}^{c} + H_{\text{int}}[\tilde{n}_{\alpha} \rightarrow \tau_{\alpha}^{c}] \] (23)

As before, $a_{m\sigma} = 2J_{m} z_{m\sigma}$ and $J_{m} \propto D_{m}$ and in the paramagnetic regime, $a_{m\sigma} = a_{m}$ and $z_{m\sigma} = z_{m}$. The Hamiltonian is a $16 \times 16$ matrix, and the full calculation of the wavefunctions, eigen-energies and the correlation functions $\Pi_{11}(\tau)$ and $\Pi_{12}(\tau)$ are done numerically.

Here in order to get an analytical insight, we rather make a simplifying assumptions that the system is already in an OSM regime, $\alpha_{2} = 2J_{22} z_{2}$ = 0. As a result, the relevant sectors of the Hamiltonian (Appendix B), shown diagrammatically in Fig. (3) breaks into two $2 \times 2$ matrix blocks with level repulsion $\alpha_{1} = 2a_{1}$ and $\alpha_{2} = \sqrt{2a_{2}}$. The vertical axis is the energy. The diagonal elements of the matrix are represented by black/gray dots and the off-diagonal matrix elements are represented by the black/gray lines connecting the dots. The basis and the diagonal energies are
\[ |\psi_{11}\rangle = \frac{|\uparrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\rangle}{\sqrt{2}} \frac{|\uparrow\uparrow\uparrow\rangle + |\uparrow\downarrow\uparrow\rangle}{\sqrt{2}} \], \qquad E_{11} = 0
\]
\[ |\psi_{21}\rangle = \frac{|\uparrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\rangle}{\sqrt{2}} \frac{|\uparrow\uparrow\uparrow\rangle + |\uparrow\downarrow\uparrow\rangle}{\sqrt{2}} \], \quad E_{21} = \Delta
\]
\[ |\psi_{12}\rangle = \frac{|\uparrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\rangle}{\sqrt{2}} \frac{|\uparrow\uparrow\uparrow\rangle + |\uparrow\downarrow\uparrow\rangle}{\sqrt{2}} \], \quad E_{12} = \frac{U}{2}
\]
\[ |\psi_{22}\rangle = \frac{|\uparrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\rangle}{\sqrt{2}} \frac{|\uparrow\uparrow\uparrow\rangle + |\uparrow\downarrow\uparrow\rangle}{\sqrt{2}} \], \quad E_{22} = \frac{U}{2}
\]
\[ |\psi_{3}\rangle = \frac{|\uparrow\uparrow\downarrow\rangle |\uparrow\downarrow\downarrow\rangle + |\uparrow\downarrow\downarrow\rangle |\uparrow\uparrow\uparrow\rangle}{\sqrt{2}} \], \quad E_{3} = 2U \quad (24)
\]

In the following we assume that the fully empty/filled states \(|\psi_{3}\rangle\) can be discarded. This is justified close to the Mott transition.\(^8\) In terms of these, the eigen-functions \(j = 1, 2\) for the sector \(i = 1, 2\) are

\[
\begin{pmatrix}
\hat{\psi}_{11} \\
\hat{\psi}_{12}
\end{pmatrix} = \begin{pmatrix}
\cos \theta_i & \sin \theta_i \\
-\sin \theta_i & \cos \theta_i
\end{pmatrix} \begin{pmatrix}
|\psi_{11}\rangle \\
|\psi_{12}\rangle
\end{pmatrix} \quad (25)
\]

where \(\tan 2\theta_i = 2\alpha_i/(E_{2i} - E_{1i})\) and the eigen-energies

\[
\tilde{E}_{ij} = E_{1i} + E_{2i} - \sigma_j \sqrt{(E_{1i} - E_{2i})^2 + \alpha_i^2} \quad (26)
\]

where \(\sigma_1 = -\sigma_2 = 1\). The level-repulsions are \(\alpha_1 = 2\alpha_1\) and \(\alpha_2 = 2\alpha_1\). The ground state belongs to the sector \(i = 1\).

The correlation functions \(\Pi_{ij}(\tau) \equiv \langle \hat{T}_{\tau} \hat{\psi}_{i} \hat{\psi}_{j} \rangle\) are then

\[
\begin{align*}
\Pi_{11}(\tau) &= Z_1 + (1 - Z_1) e^{-|\tau| E_{12}} \\
\Pi_{22}(\tau) &= e^{-|\tau| E_{21}} \sin^2 (\theta_2 + \theta_1) + e^{-|\tau| E_{22} - E_{11}} \cos^2 (\theta_1 + \theta_2)
\end{align*} \quad (27)
\]

where as before \(Z_1 = \sin^2 (2\theta_1)\).

**Holon-doublon peaks in orbital selective Mott phases**

Eqs. (24) and (27) show that the lowest-energy intermediate state accessed by \(\Pi_{22}\) is the doublon-holon state \(|\psi_{21}\rangle\).

This state is only accessible to the spectral function via the excited state \(|\psi_{12}\rangle\), i.e. when the first (wider) band is metallic. It is instructive to study this effect on a two-site problem where one site is interacting and the other site plays the role of a non-interacting bath.\(^9\) This is studied explicitly in the Appendix A, and here we discuss the main result. Fig. (4) shows the relevant sectors of a two-site Hamiltonian when \(t_2 = 0\) (emulating Mott localization of the second band). The blue and red arrows are the transitions caused by \(d_{1\alpha}^\dagger\) and \(d_{2\alpha}^\dagger\), respectively. The ground state is in the central sector and contains an admixture of the excited state due to the off-diagonal mixing \(2t_1\). The spectral function of the first (delocalized) orbital, probed by the blue arrows, contains renormalized Hubbard peaks as well as a peak near zero frequency which would evolve into an Abrikosov-Suhl resonance when the number of bath sites increases.\(^{25}\) The holon-doublon state \(|02, 2\rangle\), is only accessible via the excited state admixture (shown in bright red) and would disappear for \(t_1 = 0\). In other words, in the fully atomic limit, \(|02, 2\rangle\) is a dark state and is only visible when \(t_1 \neq 0\). The slave-spin method is capable of capturing the correlations discussed above in the two-site problem (Appendix A) as well as the DMFT results of Núñez Fernández et al.\(^9\) for the Hubbard model. Within single-site approximation, the Hamiltonian of the slave-spin sector is the same in all these cases (Eq. (23) and Fig. 3), while the mean-values of the parameters \(\alpha_1\) and \(\alpha_2\) are different. The transitions probed by the \(\tau_{2\alpha}^2\) in the slave-spin Hilbert space are marked by red in Fig. (2). The action of \(\tau_{2\alpha}^2\) on \(|\psi_{11}\rangle\) leads to \(|\psi_{21}\rangle\) whereas its action on \(|\psi_{12}\rangle\) leads to \(|\psi_{22}\rangle\) (bright red). Therefore, only if the ground state contains admixture of \(|\psi_{12}\rangle\), i.e. when \(\theta_1 \neq 0\) and the first band is metallic, do the holon-doublon state appear in the spectral function of the second orbital, or vice versa.

Assuming that the second orbital is in the Mott phase, in the impurity model \(G_{2\alpha}(\tau) = -\delta_{n0} \tau\) and in the lattice model \(G_{2\alpha}(n, \tau) = -\delta_{n0} \tau \rightarrow 2\). Multiplying by \(\Pi_{22}(\tau)\) and Fourier transforming we find

\[
2G_{d2}(z) = \frac{\sin^2 (\theta_2 + \theta_1)}{z - (E_{21} - E_{11})} + \frac{\cos^2 (\theta_1 + \theta_2)}{z - (E_{22} - E_{11})} - (z \rightarrow -z). \quad (28)
\]

The first term is the doublon-holton peak observed by the DMFT\(^9\) in the spectrum of the narrow band in OSM regime. The spectrum of the wider (itinerant) band is not affected and is essentially given by the results of previous section, Eq. (21), up to an enhancement of the effective Coulomb energy \(U/2 \rightarrow U/2 + \Delta\) by the inter-orbital interaction.
The spectrum of wider/narrower channels are shown in blue/red color. (a) In presence inter-orbital interaction $\Delta = 0.3$, $t_2/t_1 = 0.5$ $U = 3$ and $t_1 = 0.5$. (b) A zoom into low frequency part of (a). (c) No inter-orbital interaction $\Delta = 0$, $t_2/t_1 = 0.05$, $U = 3.2$ and $t_1 = 0.5$. (d) A zoom into low frequency part of (c).

### Comparison with DMFT and discussion

Fig. (5) compares the result of calculation of spectrum between slave-spin mean-field and DMFT.\(^9\) Fig. (5)(a) shows a comparison of the spectra in the OSMP regime in presence of inter-orbital interaction $\Delta = 0.3$ and $t_2/t_1 = 0.5$. The incoherent broadening of the Hubbard peaks are not captured in the mean-field theory. However, a zoom into the low-frequency part in Fig. (5)(b) shows that there is a good agreement on the position and the amplitude of the holon-doublon resonance between the two methods.

In spite of the qualitative agreements for $\Delta \neq 0$, there are some disagreements for $\Delta = 0$ between slave-spin mean-field predictions and the DMFT+DMRG numerics. Fig. (5)(c) compares the spectra for $\Delta = 0$ and large anisotropy $t_2/t_1 = 0.05$. The slave-spin mean-field predicts OSMP in this regime, whereas DMFT predicts a finite $Z$. A zoom into low-frequency part of spectra in Fig. (5)(d) shows that in contrast to DMFT result, for $\Delta = 0$ the spectrum of the narrow band remains gapped and the wavefunction renormalization remains zero [also Fig. (5)(a)] in the mean-field solution.

The origin of this gap can be traced back to the pole $z = \tilde{E}_{21} - \tilde{E}_{11}$ in Eq. (28). When $\Delta \to 0$, this gives

$$z \to \sqrt{(U/4)^2 + 4a_1^2} - \sqrt{(U/4)^2 + 2a_1^2}.$$ \hspace{1cm} (29)

If $a_1 \gg U$ this is at $z = (2 - \sqrt{2})a_1$ and for $a_1 \ll U$ is $z \to 4a_1^2/U$, which linearly or quadratically depends on the width of the wider channel (or $T_{K1}$ in the case of impurity). Therefore, the peak is expected to remain at finite frequency and follow the width of the coherent band in the wider channel.

Equations (27) and (28) show that although the total spectral weight of the two orbitals is equal to one, $\sin(2\theta_1) \neq \sin(\theta_1 + \theta_2)$ and thus the weight of two holon-doublon peaks in the second orbital is not equal to the weight of the coherence band in the first orbital, in contrast to the observation of Ref.\(^9\). $\theta_1 \in (0, \pi/4)$ quantifies the admixture of high-energy state in the ground state of block $i$. For $\Delta/U \ll 1$, we have $\theta_2 < \theta_1$ and the coherence peak has higher spectral weight than the doublon-holon peaks. But for $2\Delta/U > 1 - 1/\sqrt{2}$ this trend reverses.

When both bands are metallic, all five states mix to create various eigen-states. When $a_2$ is small, we can assume that the energies of $|\psi_{ij}\rangle$ are only slightly modified from OSMP regime and $|\psi_{11}\rangle$ is still the ground state. However, the low-lying excited state $|\tilde{\psi}_{21}\rangle$ receives some admixture of $|\psi_{12}\rangle$ of $O(a_2)$. Therefore, $\langle \tilde{\psi}_{21}|\tilde{T}_{\text{int}}|\tilde{\psi}_{11}\rangle = O(a_2)$ is non-zero and therefore, a weak resonance feature appears at energy $\tilde{E}_{21}$ in the function $\Pi_1(z)$. This feature is further weakened due to convolution with the coherence band $G_f(z)$ and appears as slight modulation of the coherence band in the wider orbital.

### Further numerical results from Slave-spin

In this section, we summarize the numerical solution to the slave-spin mean-field equations. Fig. (6) shows the evolution of quasi-particle peaks with Hubbard $U$ for $\Delta = 0$, equivalent to zero Hund’s coupling in the Kanamori-Hubbard model. Figs. 6(a,b) show the case of $t_2/t_1 = 0.1$ which contains the OSMP regime (according to mean-field). The wide band spectrum shows the coherent peak as well as renormalized Hubbard peaks. In the Narrow band spectrum the coherent peak disappears at $U/U_{c1} \sim 0.2$ (top inset) while a doublon/holon resonant feature appears at $\omega/t_1 \sim 0.5$ which follows the evolution of the coherent band in the first (wider) orbital, and going to zero when the first orbital enters the Mott phase. Figs. 6(c,d) show the case of $t_2/t_1 = 0.3$ which is the locking regime (bottom inset). The spectrum of the wide band is similar to before, but the narrow band is different in that a) There is a coherent peak at $\omega \sim 0$. Instead of doublon-holon peak, we have a doublon-holon band whose splitting follows the width of coherent band in the first orbital. There are additional fine structures in the Hubbard peaks in this case.

Figs. 7(a,b) show the evolution of quasi-particle peaks in the narrower (second) band in presence of the inter-orbital interaction $\Delta/U_{c1} = 0.05$ for (a) $t_2/t_1 = 0.1$ and (b) $t_2/t_1 = 0.3$ with wavefunction renormalizations shown in the insets. The spectrum of the wider band is similar to the $\Delta = 0$ case. In both cases the doublon/holon quasi-particles are present but they disappear (at the Mott transition of the wider band) while their splitting is still finite.
orbital interaction \( \Delta = 0 \)

FIG. 7. The spectrum of the narrower band in presence of inter-orbital interaction \( \Delta = 0 \). (a,b) OSMP driven by bandwidth anisotropy and \( t_2/t_1 = 0.1 \). The middle inset shows the wavefunction renormalizations vs. \( U/U_{c1} \) for the first (blue) and second (red) bands. (c,d) The locking regime \( t_2/t_1 = 0.3 \). The doublon-holon peaks develop into bands in the spectrum of the narrower band in addition to the coherence band. Middle inset: wavefunction renormalizations.

IV. CONCLUSIONS

We have introduced a general formalism, by which, slave particle methods capture atomic multiplets in the spectrum of nearly Mott localized orbitals. We studied the commutativity of the mean-field decoupling and single-site approximation, showing that the multiplets get renormalized and acquire dispersion within DMFT. We used slave-spins and applied our formalism to reproduce the holon-doublon peak found in the DMFT results of Núñez Fernández et al. 9 for the two-band Hubbard model. Overall there is a good agreement between DMFT and slave-spin mean-field. However, in contrast to DMFT, the splitting between holon-doublon peaks in slave-spin mean-field solution does not go to zero in the limit of zero Hund’s coupling, in consistency with an OSM phase. This raises the question of whether quantum fluctuations beyond mean-field can destroy the OSMP in absence of Hund’s coupling, which we leave for the future.

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APPENDIX A - TWO SITE PROBLEM

1. Single-orbital case

The Hamiltonian is \( H = H_0 + H_T + H_c \) where

\[
H_0 = \frac{U}{2} (n^{\uparrow}_1 + n^{\downarrow}_1)^2, \quad H_T = \sum_\sigma \left( t^{\dagger}_d c^\sigma_+ + c^\sigma_- + \text{h.c.} \right)
\]

In the two site problem, at half-filling we have \( H_c = 0 \). The Hamiltonian has \( SU^{\text{charge}}(2) \otimes SU^{\text{spin}}(2) \) symmetry. Anticipating future symmetry-lowering additions, we use a smaller \( U^{\text{charge}}(1) \otimes U^{\text{spin}}(1) \) symmetry to label states with \( Q^2 \) and \( S^2 \). The distinct atomic states are denoted by filled circles at corresponding energy in Fig. (8), and the transition between them by \( H_T \) are marked in black. As a result, the Hamiltonian is block diagonal and the largest block is \( 2 \times 2 \) corresponding to three two-level systems on the right of Fig. (8). Each group of atomic states connected by lines form a block. The larger the off-diagonal element of the block, the larger is the level-repulsion. Therefore, the ground state is given by the rightmost block:

\[
E_g = U/4 - \sqrt{(U/4)^2 + 4t_1^2}
\]

\[
|\psi_g\rangle = \cos \theta_g \left| \uparrow\downarrow \right\rangle + \sin \theta_g \left| \downarrow\uparrow \right\rangle
\]

with \( \theta_g = \tan^{-1} (E_g/2t_1) \). The Green’s function is

\[
G(\tau) = \left\langle -T d^{\dagger}_\tau (\tau) d^\tau_1 (0) \right\rangle
\]

At zero temperature and positive time \( \tau > 0 \) we have

\[
G_D(\tau) \equiv G(\tau > 0) = -\langle \psi_g | e^{\tau H} d^\tau_1 e^{-\tau H} d^{\dagger}_1 | \psi_g \rangle
\]

The blue lines in Fig. (8) show the transition caused by acting with \( d^\dagger_1 \) on the ground state block. The result is creation of a “doublon” at the impurity site, which belongs to the second rightmost block. The intermediate states in \( \pm \) are

\[
|\psi_{\pm}\rangle = \cos \theta_{\pm} | \sigma, 2 \rangle + \sin \theta | 2, \sigma \rangle
\]
with the energies
\[ E_\pm = U/4 \pm \sqrt{(U/4)^2 + t_1^2} \]  
(36)

where \( \theta_\pm = \tan^{-1}(E_\pm/t_1) \). Therefore, we find
\[ G_d(z) = \frac{1}{2} \sum_{\alpha = \pm} \left[ \sin^2(\theta_\alpha + \theta_\pm) \right] \left[ z - (E_\alpha - E_g) \right] - \left( z \rightarrow -z \right), \]
(37)

where we used that the matrix element is given by \( \langle \psi_d | f_d^\dagger | \psi_g \rangle = \sin(\theta_\alpha + \theta_\pm) \). This spectrum is composed of two resonances symmetric w.r.t. \( \omega = 0 \). The two energies have simple approximations in the limit of large \( U \): one at \( \sim t^2/U \) and the other at \( \sim U/2 \). The low-frequency resonance at \( \omega = E_+ - E_g \) is the single-bath site signature of Abrikosov resonance peak (metallic regime), whereas the high-frequency resonance at \( \omega = E_+ - E_g \) is the renormalized Hubbard peak.

\section*{Slave-spin}

It is remarkable that the same structure comes from slave-spin method. After mean-field decoupling of slave-spins and the quasiparticles we find \( H = H_f + H_z,ss - 2az \) where \( H_f = \sum_\sigma (t \bar{f}_\sigma c_\sigma + h.c.) \) and \( t_\sigma = z_\sigma t \). Diagonalizing \( H_f \) using molecular bonding/anti-bonding states we find
\[ H = i \left( f_\sigma^\dagger f_\sigma + f_\sigma f_\sigma^\dagger \right), \quad \sqrt{2} f_\pm = f \pm c \]  
(38)

The mean-field parameter \( z \) can be worked out from minimizing \( F(a, z) = F_f + E_S(a_s) - \Sigma_\sigma a_\sigma z_\sigma \). Eliminating \( a \) gives
\[ E_S = \frac{U}{4} \left[ 1 - \frac{1}{\sqrt{1 - z^2}} \right] \]  
(39)

and
\[ F(z) = F_f(z) - \frac{U}{4} \left[ 1 - \sqrt{1 - z^2} \right] \]  
(40)

In the present problem \( F_f = -2ztz \). Therefore,
\[ z = \left[ 1 + (U/8t)^2 \right]^{-1/2} \]  
(41)

The Green’s function \( G_f(\tau) = \langle -T f(\tau) f^\dagger \rangle \) for \( \tau > 0 \) is
\[ G_f(\tau) = -\frac{1}{2} \text{sign}\tau e^{-\tilde{\Omega}|\tau|} \]
(42)

Multiplying this by \( \Pi(\tau) \) from Eq. (20) we find
\[ G_d(z) = \frac{1}{2} \left[ \frac{Z}{z - \tau} + \frac{1 - Z}{z - (\tau + D)} \right] \]  
(43)

which again has the two peak structure we saw previously.

Fig. (9) shows a comparison of the exact solution to the one obtained from slave-spin method in the single-orbital two-site problem for \( U = 10t \). It can be shown that when \( U/t \gg 1 \) or \( U/t \ll 1 \) the two plots coincide.
Using Eqs. (32) and (45) we have

\[ m_{n\sigma} = \langle \psi_n | d_{z\uparrow}^\dagger | \psi_g \rangle = \beta_n \cos \theta_g + \frac{\alpha_n + \gamma_n}{\sqrt{2}} \sin \theta_g. \]  

where \( \theta_g \) determines the degree of the admixtures in the ground state.

**Slave-spin**

The diagrammatic representation of the slave-spin Hamiltonian is shown in Fig. (3) with the states listed in Eq. (24). Most generally, at zero temperature the doublon part of \( \Pi_2(z) \) is given by

\[ \Pi_2(z) = \frac{|m_{21,g}|^2}{z - \Delta E_{21}} + \frac{|m_{22,g}|^2}{z - \Delta E_{22}} + \frac{|m_{3,0}|^2}{z - \Delta E_{3}} + (z \rightarrow -z) \]

where \( \Delta E_{21} \equiv E_n - E_g \). The lowest energy resonance is in the first term. Keeping only that term and writing both doublon and holon contribution we have

\[ \Pi(z) = |m_{21,g}|^2 \left[ \frac{1}{z - \Delta E_{21}} - \frac{1}{z + \Delta E_{21}} \right] \]  

**APPENDIX B - DIAGNOLONZING THE SLAVE-SPIN HAMILTONIAN**

Using the notation

\[ |\uparrow_1\rangle = |\uparrow_{1\uparrow} \uparrow_{1\downarrow}\rangle, \quad |\downarrow_1\rangle = |\downarrow_{1\uparrow} \uparrow_{1\downarrow}\rangle, \quad |\downarrow_0\rangle = |\uparrow_{1\uparrow} \downarrow_{1\downarrow}\rangle, \]

a simplified choice of basis for atomic states is given by

\[ |\psi_{11,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_2\rangle}{\sqrt{2}} \]  

\[ |\psi_{13,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_2\rangle}{\sqrt{2}} \]  

\[ |\psi_{12,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_2\rangle}{\sqrt{2}} \]

\[ |\psi_{22,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_0\rangle}{\sqrt{2}} \]  

\[ |\psi_{23,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_2\rangle}{\sqrt{2}} \]

\[ |\psi_{24,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_2\rangle}{\sqrt{2}} \]

\[ |\psi_{21,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_2\rangle}{\sqrt{2}} \]

\[ |\psi_{3,\pm}\rangle = \frac{|\uparrow_1\rangle + |\downarrow_1\rangle}{\sqrt{2}} \pm \frac{|\downarrow_0\rangle}{\sqrt{2}} \]

and their energies are shown in the vertical axis in Fig. (12). The transition caused by acting on these atomic states with \( \tau_{1\downarrow} \) and \( \tau_{2\uparrow} \) are indicated in blue/red, respectively with the indicated amplitudes. Since the kinetic Hamiltonian \( a_r \tau_{r\sigma} \) has
equal amplitudes for $\sigma = \uparrow, \downarrow$, the dashed lines cancel each other and they drop out.

For the purpose of the paper note that when $t_2 = 0$ or $a_2 = 0$, all the red lines (as well as all the blue dashed lines) drop out and the Hamiltonian becomes a doubly degenerate (plus and minus sectors) version of Fig. (3). Moreover, a chemical potential $\mu$ couples the two sectors.

**APPENDIX C - SPECTRAL REPRESENTATION OF $G_d$**

Eq. (6) in the real frequency reads

$$G''_d(\omega) = - \int \frac{dx}{\pi} G''_f(\omega - x) \Pi''(x) [f(\omega - x) + n_B(-x)]$$

where $G''(\omega) \equiv \Im [G(\omega + i\eta)]$. This can be combined with Eq. (8), but to go to zero temperature, we need to separate out the wavefunction normalization part. Assuming that the slave-spin Hamiltonian has a non-degenerate ground state, the resulting spectrum at zero temperature is

$$G''_d(\omega) = Z G''_f(\omega)$$

$$+ \sum_{n \neq g} \left[ G''_f(\omega + \Delta E_n) \theta(-\omega + \Delta E_n) \left| \langle n | z_a | g \rangle \right|^2 
+ G''_f(\omega - \Delta E_n) \theta(\omega + \Delta E_n) \left| \langle g | z_a | n \rangle \right|^2 \right].$$

Here, $\Delta E_n = E_n - E_g$ and the ground state is treated separately and excluded from the summation.

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