

**Orbital-selective Mott phase in multiorbital models for iron pnictides and chalcogenides**Rong Yu<sup>1,2</sup> and Qimiao Si<sup>3</sup><sup>1</sup>*Department of Physics, Renmin University of China, Beijing 100872, China*<sup>2</sup>*Department of Physics and Astronomy, Shanghai Jiao Tong University, Shanghai 200240, China and Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, China*<sup>3</sup>*Department of Physics & Astronomy, Rice University, Houston, Texas 77005, USA*

(Received 13 May 2017; published 7 September 2017)

There is increasing recognition that the multiorbital nature of the  $3d$  electrons is important to the proper description of the electronic states in the normal state of the iron-based superconductors. Earlier studies of the pertinent multiorbital Hubbard models identified an orbital-selective Mott phase, which anchors the orbital-selective behavior seen in the overall phase diagram. An important characteristics of the models is that the orbitals are kinetically coupled, i.e., hybridized, to each other, which makes the orbital-selective Mott phase especially nontrivial. A  $U(1)$  slave-spin method was used to analyze the model with nonzero orbital-level splittings. Here we develop a Landau free-energy functional to shed further light on this issue. We put the microscopic analysis from the  $U(1)$  slave-spin approach in this perspective, and show that the intersite spin correlations are crucial to the renormalization of the bare hybridization amplitude towards zero and the concomitant realization of the orbital-selective Mott transition. Based on this insight, we discuss additional ways to study the orbital-selective Mott physics from a dynamical competition between the interorbital hybridization and collective spin correlations. Our results demonstrate the robustness of the orbital-selective Mott phase in the multiorbital models appropriate for the iron-based superconductors.

DOI: [10.1103/PhysRevB.96.125110](https://doi.org/10.1103/PhysRevB.96.125110)**I. INTRODUCTION**

In many strongly correlated systems, superconductivity is closely connected to a strongly correlated bad-metal normal state and a nearby antiferromagnetic order. As such, there has been considerable effort devoted to the understanding of the electron correlation effects and the associated magnetism. In the case of the iron-based superconductors (FeSCs) [1–6], recent developments have further highlighted the importance of electron correlations. For instance, angle-resolved photoemission spectroscopy (ARPES) has found an orbital-selective Mott phase (OSMP) in the iron chalcogenides [7–10]. The OSMP phase arises in multiorbital Hubbard models of the FeSCs, which contain both Hubbard interactions and Hund's coupling [11,12]. Evidence for the OSMP has also come from a variety of other measurements [13–16]. A complementary approach to the electron correlations of the FeSCs describes the localization-delocalization phenomena in the form of an orbital differentiation and a coherence-incoherence crossover [17,18], though OSMP is not explicitly invoked. In addition, recent experiments have identified a Mott insulating system in the copper-doped iron pnictides [19], which accompanies the earlier observations of Mott insulating states in the iron chalcogenides [20–22]. These observations considerably expand on the bad-metal behavior known, since early on in the field, through other properties of the iron pnictides [23–25]. For example, the room-temperature electrical resistivity is large (reaching the Mott-Ioffe-Regel limit, as defined by the normalized mean free path  $k_F\ell$  being of order unity), in contrast to good metals such as Cr (for which  $k_F\ell$  at room temperature is much larger than 1). Moreover, the Drude optical weight is much reduced [23].

The parent systems of FeSCs have the iron valence +2, with  $n = 6$  electrons occupying the  $3d$  orbitals. Correspondingly, multiple  $3d$ -electron orbitals are important [24–30]. In order to address the bad-metal physics, an important question is how

the itinerancy of the electronic states is reduced with increasing Coulomb interactions. The appropriate multiorbital Hubbard models include the intra- and interorbital Hubbard interactions ( $U$  and  $U'$ ) and the Hund's coupling ( $J_H$ ).

The OSMP appears in the paramagnetic solution to the multiorbital Hubbard models [11]. In this phase, the nondegenerate  $3d$   $xy$  orbital loses its coherent spectral weight at the Fermi energy, while this weight remains nonzero for the other orbitals (including the degenerate  $3d$   $xz/yz$  orbitals). These features have been clearly identified in the ARPES measurements [7–10].

From a theoretical perspective, it is important to stress that, the orbitals are kinetically coupled, i.e., hybridized, to each other in these models. In particular, the bare kinetic hybridization between the  $3d$   $xy$  orbital and the other  $3d$  orbitals is nonzero. In the OSMP phase, this hybridization is renormalized to zero and the  $3d$   $xy$  orbital is no longer mixed with the other orbitals in the low-energy electronic excitations.

More generally, orbital selectivity has been discussed in the correlation effects; see, for example, Refs. [17,31–35]. It can be defined in terms of the mass enhancement ( $m^*/m_b$ , the ratio of the effective mass observed experimentally to that of the noninteracting band dispersion) being different for the electronic states with predominantly different orbital contents. In practice,  $m^*/m_b$  for electronic states with predominantly  $3d$   $xy$  orbital character is much larger (reaching 10–20 in the iron selenides) than that for the electronic states with predominantly  $3d$   $xz/yz$  (typically 3–4) and other orbital characters [4,10].

Strictly speaking, the orbital selectivity is in itself not precisely defined, because the hybridization mixes the different orbitals in the electronic band states. However, it becomes sharply defined as a precursor to the OSMP [11]. In other words, the OSMP anchors the notion of orbital selectivity in general. Note that the picture of the orbitally differentiated

coherence-incoherence crossover has also yielded the  $3d$   $xy$  orbital to be localized at sufficiently high temperatures, while itinerant at zero temperature [36,37]. Hence the generalization of the notion of OSMP is relevant in this case as well. These highlight the conceptual importance of the OSMP in characterizing the orbital selectivity in general.

### Orbital vs band basis

OSMP for the multiorbital Hubbard model has been discussed in several contexts. Most of these studies start from multiple inequivalent bands, and define the Hubbard and Hund's interactions in the band states [38–43]. By definition, the band basis diagonalizes the noninteracting part of the Hamiltonian. In other words, kinetically, they are decoupled from each other. In the presence of interactions, the weight of the coherent electrons near the Fermi surface will be renormalized below 1 in a band-dependent way. An OSMP corresponds to the regime where the renormalized coherent spectral weights for some of the bands vanish while those for the others remain nonzero.

For the FeSCs, multiorbital Hubbard models are defined in the orbital basis [12,24–35]. The definition of the nondegenerate orbitals is unambiguous. For the degenerate  $3d$   $xz/yz$  orbitals, there can be alternative definitions, but the degeneracy ensures that the different definitions are equivalent to each other.

In the orbital basis, the kinetic part of the Hamiltonian is not diagonal. Therefore, in the Hamiltonian, the orbitals are kinetically coupled, i.e., hybridized, to each other. In the OSMP solution of Ref. [11], the destruction of the quasiparticle spectral weight  $Z_{xy}$  is accompanied by the suppression of the renormalized interorbital kinetic hybridization between the  $3d$   $xy$  orbital and the other ones.

The purpose of this paper is to clarify how the above happens. We do so by formulating a Landau free-energy functional, which also demonstrates the robustness of the

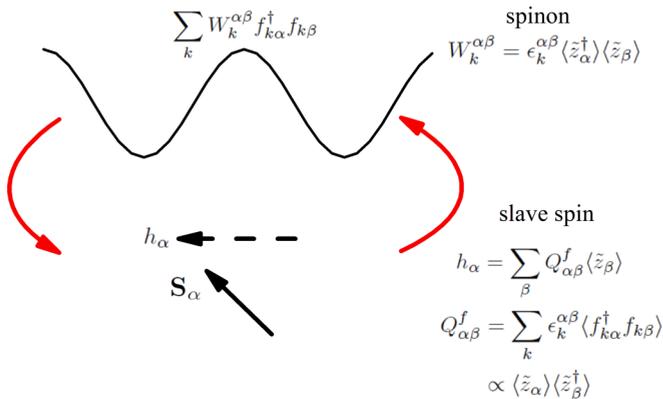


FIG. 1. Illustrating the effect of the interorbital kinetic hybridization in the  $U(1)$  slave-spin theory. The black curve shows the effective spinon dispersion, which is generated by  $W_k^{\alpha\beta} f_{k\alpha}^\dagger f_{k\beta}$ . (The physical spin index  $\sigma$  is suppressed in the figure legends.) Meanwhile, the slave-spin  $S_\alpha$  experiences a local field  $h_\alpha = \sum_\beta Q_{\alpha\beta}^f \langle \tilde{z}_\beta \rangle$ , where  $Q_{\alpha\beta}^f \propto \langle \tilde{z}_\alpha \rangle \langle \tilde{z}_\beta^\dagger \rangle$ . The red arrows indicate the self-consistency between  $W_k^{\alpha\beta}$  and  $h_\alpha$ , which results in a biquadratic interorbital coupling as shown in Eqs. (19) and (21).

OSMP. Viewed from this perspective, we show the crucial role that the intersite spin correlations play in generating the OSMP (cf. Fig. 1) within the the microscopic  $U(1)$  slave-spin approach [11,32].

The remainder of the paper is organized as follows. In Sec. II we define the model and summarize the  $U(1)$  slave-spin approach. Section III is devoted to the formulation of the Landau free-energy functional in terms of the orbital-dependent quasiparticle weight and how the OSMP appears as a distinct phase permitted by the Landau functional. We then address, in Sec. IV, the OSMP as derived by the microscopic  $U(1)$  slave-spin approach from the perspective of the Landau free-energy functional. Landau analysis has been useful in clarifying the Mott transition in the dynamical mean-field theory (DMFT) context [44]. Section V discusses the implications of the insight gained in the present work for further studies on the OSMP, and Sec. VI summarizes the paper. Finally, in Appendixes A through C, we present further details on the saddle-point equations of the  $U(1)$  slave-spin approach.

## II. MULTIORBITAL HUBBARD MODEL AND THE $U(1)$ SLAVE-SPIN APPROACH

We now define the model and, to facilitate the analysis in the next section, summarize the microscopic approach [32] based on a  $U(1)$  slave-spin representation.

### A. Multiorbital Hubbard model

The multiorbital Hubbard model for the FeSCs takes the following form:

$$H = H_0 + H_{\text{int}}. \quad (1)$$

$H_0$  contains the tight-binding parameters among the multiple orbitals,

$$H_0 = \frac{1}{2} \sum_{ij\alpha\beta\sigma} t_{ij}^{\alpha\beta} d_{i\alpha\sigma}^\dagger d_{j\beta\sigma} + \sum_{i\alpha\sigma} (\Delta_\alpha - \mu) d_{i\alpha\sigma}^\dagger d_{i\alpha\sigma}, \quad (2)$$

where  $d_{i\alpha\sigma}^\dagger$  creates an electron in orbital  $\alpha$  ( $= 1, \dots, 5$ ) with spin  $\sigma$  at site  $i$ ,  $\Delta_\alpha$  refers to the energy level associated with the crystal field splitting (which is diagonal in the orbital basis), and  $\mu$  is the chemical potential. In general,  $t_{ij}^{\alpha\beta} \neq 0$  for  $\alpha \neq \beta$ , corresponding to a nonzero kinetic hybridization between the different orbitals. For latter references, we note that the on-site energy for the  $3d$   $xy$  orbital is different from any of the other four  $3d$  orbitals: for any orbital  $\beta \neq xy$ , the level splitting  $\Delta_{xy,\beta} \equiv \Delta_{xy} - \Delta_\beta \neq 0$ . The on-site interaction  $H_{\text{int}}$  reads

$$H_{\text{int}} = \frac{U}{2} \sum_{i,\alpha,\sigma} n_{i\alpha\sigma} n_{i\alpha\bar{\sigma}} + \sum_{i,\alpha<\beta,\sigma} \{U' n_{i\alpha\sigma} n_{i\beta\bar{\sigma}} + (U' - J_H) n_{i\alpha\sigma} n_{i\beta\sigma} - J_H (d_{i\alpha\bar{\sigma}}^\dagger d_{i\alpha\bar{\sigma}} d_{i\beta\bar{\sigma}}^\dagger d_{i\beta\bar{\sigma}} + d_{i\alpha\sigma}^\dagger d_{i\alpha\sigma} d_{i\beta\sigma}^\dagger d_{i\beta\sigma})\}. \quad (3)$$

where  $n_{i\alpha\sigma} = d_{i\alpha\sigma}^\dagger d_{i\alpha\sigma}$ . Here  $U$ ,  $U'$ , and  $J_H$ , respectively, denote the intraorbital repulsion, the interorbital repulsion, and the Hund's rule exchange coupling. In the following, we will take  $U' = U - 2J_H$  [45].

### B. $U(1)$ slave-spin theory

The metal-insulator transition in the model has been studied by using a  $U(1)$  slave-spin theory, which was introduced in Ref. [32]. Here we summarize the approach to set the stage for our consideration of the OSMF in the next two sections. For further details, we refer to Ref. [32] as well as Appendixes A and B. In addition, we refer to Appendix C for a comparison with the  $Z_2$  slave-spin theory of Ref. [46] (see also Refs. [47,48]).

In the  $U(1)$  slave-spin formulation, the  $XY$  component of a quantum  $S = 1/2$  spin operator ( $S_{i\alpha\sigma}^+$ ) is used to represent the charge degree of freedom of the electron at each site  $i$ , for each orbital  $\alpha$ , and each spin flavor  $\sigma$ . Correspondingly, a fermionic ‘‘spinon’’ operator ( $f_{i\alpha\sigma}^\dagger$ ) is used to carry the spin degree of freedom. The electron creation operator is then represented as follows:

$$d_{i\alpha\sigma}^\dagger = S_{i\alpha\sigma}^+ f_{i\alpha\sigma}^\dagger. \quad (4)$$

This is implemented by a constraint,

$$S_{i\alpha\sigma}^z = f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma} - \frac{1}{2}, \quad (5)$$

which restricts the Hilbert space to the physical one.

This representation contains a  $U(1)$  gauge redundancy corresponding to  $f_{i\alpha\sigma}^\dagger \rightarrow f_{i\alpha\sigma}^\dagger e^{-i\theta_{i\alpha\sigma}}$  and  $S_{i\alpha\sigma}^+ \rightarrow S_{i\alpha\sigma}^+ e^{i\theta_{i\alpha\sigma}}$ . Therefore, the slave spins carry the  $U(1)$  charge, similarly as in the slave-rotor approach [49].

To ensure that the saddle point captures the correct quasiparticle spectral weight in the noninteracting limit (being equal to 1), we define a dressed operator in the Schwinger boson representation of the slave spins (in a way similar to the standard slave-boson theory [50]):

$$\hat{z}_{i\alpha\sigma}^\dagger = P_{i\alpha\sigma}^+ a_{i\alpha\sigma}^\dagger b_{i\alpha\sigma} P_{i\alpha\sigma}^-, \quad (6)$$

where  $P_{i\alpha\sigma}^\pm = \sqrt{1/2 + \delta \pm (a_{i\alpha\sigma}^\dagger a_{i\alpha\sigma} - b_{i\alpha\sigma}^\dagger b_{i\alpha\sigma})/2}$ , and  $\delta$  is an infinitesimal positive number to regulate  $P_{i\alpha\sigma}^\pm$ .

Here  $a_{i\alpha\sigma}$  and  $b_{i\alpha\sigma}$  are Schwinger bosons representing the slave-spin operators:  $S_{i\alpha\sigma}^+ = a_{i\alpha\sigma}^\dagger b_{i\alpha\sigma}$ ,  $S_{i\alpha\sigma}^- = b_{i\alpha\sigma}^\dagger a_{i\alpha\sigma}$ , and  $S_{i\alpha\sigma}^z = (a_{i\alpha\sigma}^\dagger a_{i\alpha\sigma} - b_{i\alpha\sigma}^\dagger b_{i\alpha\sigma})/2$ . They satisfy an additional constraint,

$$a_{i\alpha\sigma}^\dagger a_{i\alpha\sigma} + b_{i\alpha\sigma}^\dagger b_{i\alpha\sigma} = 1. \quad (7)$$

In other words, they are hard-core bosons. In this representation, the constraint in Eq. (5) becomes

$$a_{i\alpha\sigma}^\dagger a_{i\alpha\sigma} - b_{i\alpha\sigma}^\dagger b_{i\alpha\sigma} = 2f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma} - 1. \quad (8)$$

At the same time, Eq. (4) becomes

$$d_{i\alpha\sigma}^\dagger = \hat{z}_{i\alpha\sigma}^\dagger f_{i\alpha\sigma}^\dagger. \quad (9)$$

The Hamiltonian, Eq. (1), can then be effectively rewritten as

$$H = \frac{1}{2} \sum_{ij\alpha\beta\sigma} t_{ij}^{\alpha\beta} \hat{z}_{i\alpha\sigma}^\dagger \hat{z}_{j\beta\sigma} f_{i\alpha\sigma}^\dagger f_{j\beta\sigma} + \sum_{i\alpha\sigma} (\Delta_\alpha - \mu) f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma} - \lambda_{i\alpha\sigma} \left[ f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma} - \frac{1}{2} (\hat{n}_{i\alpha\sigma}^a - \hat{n}_{i\alpha\sigma}^b) \right] + H_{\text{int}}^S. \quad (10)$$

Here  $\lambda_{i\alpha\sigma}$  is a Lagrange multiplier to enforce the constraint in Eq. (8). In addition,  $H_{\text{int}}^S$  is the interaction Hamiltonian, Eq. (3),

rewritten in the slave-spin representation  $H_{\text{int}} \rightarrow H_{\text{int}}(\mathbf{S})$  [32], and subsequently with the slave-spin operators substituted by the Schwinger bosons. The quasiparticle spectral weight

$$Z_{i\alpha\sigma} = |z_{i\alpha\sigma}|^2 \equiv |\langle \hat{z}_{i\alpha\sigma} \rangle|^2. \quad (11)$$

A metallic phase corresponds to  $Z_{i\alpha\sigma} > 0$ , and a Mott insulator corresponds to  $Z_{i\alpha\sigma} = 0$  in all orbitals with a gapless spinon spectrum.

After decomposing the boson and spinon operators and treating the constraint on average, we obtain two saddle-point Hamiltonians for the spinons and the Schwinger bosons, respectively:

$$H_f^{\text{mf}} = \sum_{k\alpha\beta} [\epsilon_k^{\alpha\beta} \langle \hat{z}_\alpha^\dagger \rangle \langle \hat{z}_\beta \rangle + \delta_{\alpha\beta} (\Delta_\alpha - \lambda_\alpha + \tilde{\mu}_\alpha - \mu)] f_{k\alpha}^\dagger f_{k\beta}, \quad (12)$$

$$H_S^{\text{mf}} = \sum_{\alpha\beta} \left[ Q_{\alpha\beta}^f (\langle \hat{z}_\alpha^\dagger \rangle \langle \hat{z}_\beta \rangle + \langle \hat{z}_\beta \rangle \langle \hat{z}_\alpha^\dagger \rangle) + \delta_{\alpha\beta} \frac{\lambda_\alpha}{2} (\hat{n}_\alpha^a - \hat{n}_\alpha^b) \right] + H_{\text{int}}^S, \quad (13)$$

where  $\delta_{\alpha\beta}$  is Kronecker’s delta function,  $\epsilon_k^{\alpha\beta} = \frac{1}{N} \sum_{ij\sigma} t_{ij}^{\alpha\beta} e^{ik(r_i - r_j)}$ , with  $N$  being the number of sites in the lattice, and

$$Q_{\alpha\beta}^f = \sum_{k\sigma} \epsilon_k^{\alpha\beta} \langle f_{k\alpha\sigma}^\dagger f_{k\beta\sigma} \rangle / 2, \quad (14)$$

$$\hat{z}_\alpha^\dagger = \langle P_\alpha^+ \rangle a_\alpha^\dagger b_\alpha \langle P_\alpha^- \rangle. \quad (15)$$

In addition,  $\tilde{\mu}_\alpha$  is an effective on-site potential defined as

$$\tilde{\mu}_\alpha = 2\bar{\epsilon}_\alpha \eta_\alpha, \quad (16)$$

where

$$\bar{\epsilon}_\alpha = \sum_{\beta} (Q_{\alpha\beta}^f \langle \hat{z}_\alpha^\dagger \rangle \langle \hat{z}_\beta \rangle + \text{c.c.}) \quad (17)$$

and

$$\eta_\alpha = (2n_\alpha^f - 1) / [4n_\alpha^f (1 - n_\alpha^f)], \quad (18)$$

with  $n_\alpha^f = \frac{1}{N} \sum_k \langle f_{k\alpha}^\dagger f_{k\alpha} \rangle$ .

Equations (12) and (13) represent the main formulation of the  $U(1)$  slave-spin approach at the saddle-point level. Note that the slave-spin part is single site in nature. By contrast, the pseudofermion part must contain intersite couplings, which will play an important role in the analysis (see next section). We study the metal-to-insulator transitions in the paramagnetic phase preserving the translational symmetry. These allow us to drop the spin and/or site indices of the Schwinger bosons (slave spins) and the Lagrange multiplier  $\lambda_\alpha$  in the above saddle-point equations. We refer to Appendixes A and B for a detailed derivation of these saddle-point Hamiltonians. The parameters  $z_\alpha$  and  $\lambda_\alpha$  are solved self-consistently. The parameter  $\tilde{\mu}_\alpha$  introduced above is crucially important to ensuring that the noninteracting limit is properly captured (with  $Z_\alpha = |\langle \hat{z}_\alpha \rangle|^2 = |z_\alpha|^2 = 1$  and correct electron dispersion) regardless of whether the system is at or away from half-filling (see Appendix C for more details). By contrast, in the  $Z_2$  slave-spin formulation, the parameter  $\tilde{\mu}_\alpha$  is absent in the saddle-point

equations, and the proper noninteracting limit cannot be easily recovered for the generic case of multiple nondegenerate orbitals away from half-filling; see Appendix C for further discussions on this point as well.

### III. LANDAU FREE-ENERGY FUNCTIONAL AND THE ORIGIN OF THE ORBITAL-SELECTIVE MOTT TRANSITION

In a multiorbital system, an OSMP may exist besides the metallic and the Mott insulating phases. In an OSMP, some of the orbitals are Mott localized and the others are still metallic; the quasiparticle spectral weight  $Z$  vanishes for the former orbitals and remains nonzero for the latter ones. In this section we clarify how an OSMP can arise in the slave-spin approach and develop a Landau theory to describe the orbital-selective Mott transition (OSMT).

We start from the two saddle-point Hamiltonians, Eqs. (12) and (13). Consider first Eq. (12), where the kinetic hybridization between two different orbitals  $\alpha \neq \beta$  is  $W_k^{\alpha\beta} f_{k\alpha}^\dagger f_{k\beta}$ , with  $W_k^{\alpha\beta} = \epsilon_k^{\alpha\beta} \langle \tilde{z}_\alpha^\dagger \rangle \langle \tilde{z}_\beta \rangle \propto \langle \tilde{z}_\alpha^\dagger \rangle \langle \tilde{z}_\beta \rangle$ . Recall that  $\langle f_{k\alpha}^\dagger f_{k\beta} \rangle$  is determined by an averaging of  $f_{k\alpha}^\dagger f_{k\beta}$  with respect to  $H_f^{\text{mf}}$ , and it is nonzero in response to an effective ‘‘field’’  $W_k^{\alpha\beta}$  applied to the kinetic hybridization operator  $f_{k\alpha}^\dagger f_{k\beta}$  in  $H_f^{\text{mf}}$ . For the case we consider, with a nonzero orbital level difference,  $\Delta_{\alpha,\beta} \equiv \Delta_\alpha - \Delta_\beta \neq 0$ , the susceptibility describing the linear response of  $\langle f_{k\alpha}^\dagger f_{k\beta} \rangle$  to  $W_k^{\alpha\beta}$  will be finite, leading to  $\langle f_{k\alpha}^\dagger f_{k\beta} \rangle \propto W_k^{\alpha\beta} \propto \langle \tilde{z}_\alpha \rangle \langle \tilde{z}_\beta^\dagger \rangle$ ; this is illustrated in Fig. 1, top panel. As a result, the kinetic hybridization of the spinons is

$$\langle H_f^{\text{mf}} \rangle_{\alpha\beta} = \sum_k \epsilon_k^{\alpha\beta} \langle \tilde{z}_\alpha^\dagger \rangle \langle \tilde{z}_\beta \rangle \langle f_{k\alpha}^\dagger f_{k\beta} \rangle \propto |\langle \tilde{z}_\alpha \rangle|^2 |\langle \tilde{z}_\beta \rangle|^2. \quad (19)$$

Consider next Eq. (13), which shows that the slave-spin operator  $\tilde{z}_\alpha$  for orbital  $\alpha$  experiences an effective field of  $h_\alpha = \sum_\beta Q_{\alpha\beta}^f \langle \tilde{z}_\beta \rangle$ , where  $Q_{\alpha\beta}^f$  is defined in Eq. (14). Similar reasoning as in the previous paragraph gives rise to

$$Q_{\alpha\beta}^f \propto \langle \tilde{z}_\alpha \rangle \langle \tilde{z}_\beta^\dagger \rangle, \quad (20)$$

for  $\alpha \neq \beta$  and  $\Delta_{\alpha,\beta} \neq 0$ . In other words,  $h_\alpha \propto \langle \tilde{z}_\alpha \rangle |\langle \tilde{z}_\beta \rangle|^2$ , as illustrated in Fig. 1. Taking the expectation value of  $\tilde{z}_\alpha^\dagger$  with respect to  $H_S^{\text{mf}}$  then yields the following component of the free energy from  $H_S^{\text{mf}}$ :

$$\langle H_S^{\text{mf}} \rangle_{\alpha\beta} \rightarrow |\langle \tilde{z}_\alpha \rangle|^2 |\langle \tilde{z}_\beta \rangle|^2. \quad (21)$$

From Eqs. (19) and (21), we see that for both the spinons and the slave spins, the interorbital correlations appear as a biquadratic coupling  $|z_\alpha|^2 |z_\beta|^2$ . This biquadratic interaction is a natural result of a self-consistent solution of the two saddle-point equations, Eqs. (12) and (13) (Fig. 1). It is crucial to the stabilization of an OSMP. As  $z_\alpha$  approaches zero, so does  $Q_{\alpha\beta}^f$ ; correspondingly, the effective field acting on the slave spin  $h_\alpha$  also goes to zero in spite of a nonzero  $z_\beta$ , making the OSMP an internally consistent solution.

To see how the OSMP arises more explicitly, we can construct a Landau free-energy functional in terms of the quasiparticle weights  $z_\alpha$ . For simplicity of notation, we take the  $3d$   $xy$  and another  $3d$  orbital as orbitals 1 and 2, but our

analysis straightforwardly applies to the case of more than two orbitals. The free-energy density reads

$$f = \sum_{\alpha=1,2} (r_\alpha |z_\alpha|^2 + u_\alpha |z_\alpha|^4) + v |z_1|^2 |z_2|^2, \quad (22)$$

in which the biquadratic coupling  $v$  term comes from the kinetic hybridization as discussed above. The quadratic terms  $r_\alpha |z_\alpha|^2$  arise from the kinetic energy of the saddle-point Hamiltonian in Eq. (13) [as well as in Eq. (12)]. For example, since  $\sum_k \langle f_{k\alpha}^\dagger f_{k\alpha} \rangle$  is the spinon density in orbital  $\alpha$ , which is of order  $O(1)$  even when  $z_\alpha$  approaches zero; thus,  $Q_{\alpha\alpha}^f \sim O(1)$ , and gives rise to the quadratic terms ( $r_\alpha |z_\alpha|^2$ ) in the Landau free-energy density. Taking the derivatives with respect to  $|z_\alpha|$ , we obtain

$$\frac{\partial f}{\partial |z_1|} = |z_1| (2r_1 + 4u_1 |z_1|^2 + 2v |z_2|^2) = 0, \quad (23)$$

$$\frac{\partial f}{\partial |z_2|} = |z_2| (2r_2 + 4u_2 |z_2|^2 + 2v |z_1|^2) = 0. \quad (24)$$

There could then be three solutions:

- (1)  $r_1 + 2u_1 |z_1|^2 + v |z_2|^2 = r_2 + 2u_2 |z_2|^2 + v |z_1|^2 = 0$ , which yields  $|z_1| \neq 0$ ,  $|z_2| \neq 0$ , corresponding to a metallic phase;
- (2)  $|z_1| = |z_2| = 0$ , corresponding to a Mott insulator;
- (3)  $|z_1| = 0$ ,  $|z_2| = \sqrt{-\frac{r_2}{2u_2}}$  (or  $|z_2| = 0$ ,  $|z_1| = \sqrt{-\frac{r_1}{2u_1}}$ ), corresponding to an OSMP.

### IV. THE ORBITAL-SELECTIVE MOTT TRANSITION IN A FIVE-ORBITAL MODEL FOR IRON CHALCOGENIDES AS AN ILLUSTRATION OF THE LANDAU THEORY

In the previous section we have constructed a Landau theory, and shown that an OSMP is an allowed solution to the free-energy functional. Strictly speaking, the Landau theory works only when  $z_\alpha$  is sufficiently small for each orbital. In the more realistic situation, such as in the five-orbital model for iron chalcogenides,  $z_{xz/yz}$  may still be sizable across the OSMT for the localization of the  $3d$   $xy$  orbital. Therefore, it is instructive to show that the general consideration obtained in the previous section is valid in the five-orbital model.

For this reason, we revisit the OSMT in the five-orbital model for  $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$  system [11]. For an illustrative purpose, we consider the case without iron-vacancy order and with the electron filling  $n = 6$ . For definiteness, we fix the ratio of the Hund’s rule coupling to the intraorbital Hubbard interaction to  $J_H/U = 0.25$ . We have calculated the evolution of the quasiparticle spectral weights  $Z_\alpha = |z_\alpha|^2$  with increasing  $U$  for the  $3d$   $xz/yz$  and  $xy$  orbitals. The results are summarized in Fig. 2(a). An OSMT takes place, at which  $Z_{xy}$  vanishes, where  $Z_{xz}$  (as well as  $Z_{yz} = Z_{xz}$ ,  $Z_{x^2-y^2}$  and  $Z_{3z^2-r^2}$ ) remain nonzero.

On approach of the OSMT, we can investigate how the renormalization factor for the interorbital hybridization  $Q_{xz,xy}^f$  (and  $Q_{yz,xy}^f$ , which is equal to  $Q_{xz,xy}^f$  as dictated by the  $C_4$  symmetry) behaves. We see that for  $U$  smaller than 4 eV, where the system is sufficiently away from the OSMT,  $Q_{xz,xy}^f$  decreases relatively slowly. By contrast, as  $U$  gets closer to  $U_{\text{OSMT}}$ , where the system enters the linear-response

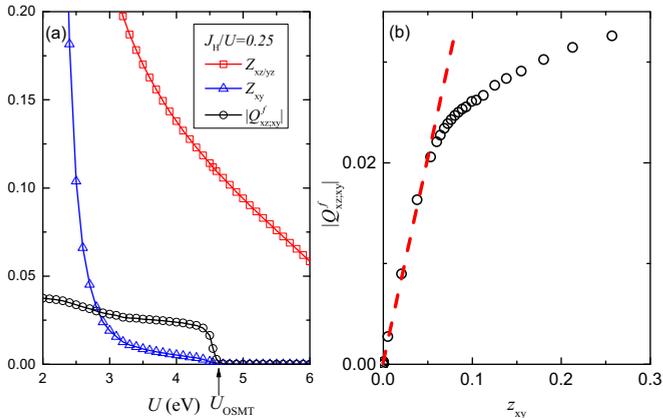


FIG. 2. (a) The evolution of the quasiparticle spectral weights  $Z_{xy}$ ,  $Z_{xz/yz}$ , and the renormalization factor for the interorbital hybridization  $|Q^f_{xz/yz,xy}|$  as a function of  $U$  in a five-orbital model for  $K_x\text{Fe}_{2-y}\text{Se}_2$  (without ordered iron vacancies), with  $J_H/U = 0.25$  and electron filling  $n = 6$ . An OSMT occurs at  $U = U_{OSMT}$ . (b)  $|Q^f_{xz/yz,xy}|$  vs  $z_{xy}$ . The linear dependence expected from the Landau analysis is shown by a linear fit (dashed line). The model parameters are the same as in (a).

regime,  $Q^f_{xz,xy}$  rapidly decreases. This behavior implies that suppressing the interorbital hybridization is key to the OSMT. Indeed, it follows from the general analysis in the previous section that  $Q^f_{xz/yz,xy}$  would vanish as the OSMT is reached. The relationship  $Q^f_{xz,xy} \propto z_{xz}z_{xy}$  would be expected if both  $z_{xy}$  and  $z_{xz}$  were small. In our case, since  $z_{xz}$  is still sizable across the OSMT, the relationship  $Q^f_{xz,xy} \propto z_{xz}$  would acquire sizable corrections; such corrections will not affect the existence or critical behavior of the OSMT. On the other hand, near the transition,  $Q^f_{xz,xy}$  is expected to be linearly proportional to  $z_{xy}$ . This proportionality is indeed satisfied, as shown in Fig. 2(b).

## V. DISCUSSIONS

We have analyzed the OSMT in multiorbital models pertinent to the FeSCs. From a Landau free-energy functional, the OSMP can be realized because the effective coupling between the quasiparticle weights of the different orbitals is biquadratic (instead of bilinear). In our analysis, we have emphasized the role of a nonzero orbital level splitting between the  $3d$   $xy$  orbital and the other  $3d$  orbitals (particularly the  $xz/yz$  orbitals).

We have clarified how the crucial feature of the Landau analysis, namely the absence of the bilinear coupling between the quasiparticle weights of the different orbitals, arises within the  $U(1)$  slave-spin approach. Crucial to this feature is the coupling of the pseudofermions at different sites. At the saddle point level, the slave spins are treated through a local description, with its condensate capturing the quasiparticle weight. By contrast, the effective Hamiltonian of the pseudofermions, which is at least bilinear, must contain intersite coupling. Because the slave spins have a  $U(1)$  symmetry, we can construct the saddle-point equations in the gauge with the slave spins alone describing the charge degrees freedom. Correspondingly, the physical spin degrees of freedom are

entirely captured by the pseudofermions. Thus, the intersite pseudofermion coupling reflects the intersite coupling of the physical spin degrees of freedom. In this way, the analysis here highlights the importance of the intersite spin correlations in renormalizing the interorbital kinetic hybridization to zero, as the OSMP is realized.

This insight suggests complementary means of studying the OSMP. In terms of physical variables, another setting in which interorbital hybridization has been shown to be dynamically suppressed is in the heavy fermion systems [51–53]. There, the RKKY spin-exchange interactions compete against the Kondo hybridization between the  $f$  and conduction electrons, and the resulting destruction of the Kondo hybridization can be viewed as an OSMT of  $f$  electrons. One of the means that captures this interplay between the hybridization and collective spin correlations is the extended dynamical mean field theory (EDMFT) [54]. In the EDMFT approach, such intersite spin correlations are treated in terms of physical spins (instead of the auxiliary fermions).

For the multiorbital Hubbard Hamiltonian, Eq. (1), the effect of intersite spin correlations can be taken into account by adding an explicit exchange interaction  $J_{ij}$  between the spins at the different sites. The total Hamiltonian is then the multiorbital analog of the one-band Hubbard-Heisenberg model [55]. (Alternatively, such a term can be considered as being effectively generated at an intermediate energy scale, by integrating out the incoherent part of the slave-spin fluctuation spectrum.) Within the EDMFT, the effects of such intersite correlations are studied dynamically through a bosonic bath, whose spectrum is determined self-consistently. This coupling of the bosonic bath to the local spin degrees of freedom competes with the hybridization term. When the competition is strong enough, it can drive the hybridization to zero, thereby realizing an OSMP. For the Anderson lattice model relevant to heavy fermion systems, this method has already shown that an OSMP arises, even when an on-site hybridization is present. This line of study holds a clear promise to bring about further new insights into the orbital-selective Mott physics in multi-orbital models pertinent to the iron-based superconductors.

We close this section by noting that the orbital-selective Mott physics is also of interest in a variety of other contexts, such as  $\text{VO}_2$  [56] and multiorbital systems at low dimensions [57–59].

## VI. CONCLUSION

In this paper we have revisited on the orbital-selective Mott phase identified in previous studies in the multiorbital Hubbard models for iron pnictides and iron chalcogenides. This type of models contains kinetic hybridization between the  $3d$  electron orbitals. We have constructed a Landau free-energy functional in terms of the quasiparticle renormalization factor  $z_\alpha$ . We have shown that a kinetic hybridization between the different orbitals introduces biquadratic couplings between the quasiparticle renormalization  $z$  factors of the different orbitals. The absence of bilinear couplings is a property that is generic to multiorbital models with nonzero orbital level splittings. It makes the orbital-selective Mott phase possible.

Within the microscopic  $U(1)$  slave-spin approach, the absence of the bilinear coupling among the  $z$ 's can be traced

to the intersite spin correlations. This amounts to a linear relationship between the renormalized interorbital kinetic hybridization and the quasiparticle weight for the  $3d\ xy$  orbital (when the latter is sufficiently small). Such a linear relationship is shown to be satisfied in the previously identified solution near the orbital-selective Mott transition.

More generally, our analysis here illustrates that intersite spin correlations are important in renormalizing the interorbital kinetic hybridization to zero, thereby generating the OSMP. This insight suggests additional means of studying the orbital-selective Mott physics in the multiorbital models. In particular, an extended dynamical mean field theory allows the study of the dynamical competition between the interorbital hybridization and collective spin correlations. This method has been used to demonstrate an OSMP in the case of Anderson lattice model, which contains an on-site hybridization. It would be a promising way to gain further insights into the dynamical suppression of the hybridization effect for the orbital-selective Mott phase in multiorbital models pertinent to the iron-based superconductors. Given the growing recognition that the orbital selectivity plays an important role in the iron pnictides and iron chalcogenides, such studies are clearly worth the efforts.

In short, through a Landau analysis, we have demonstrated the robustness of the orbital-selective Mott phase in multiorbital models pertinent to the iron-based superconductors. We have also suggested means for further theoretical studies of the orbital-selective Mott physics, which may be relevant to bad metals in a variety of correlated electron systems.

*Note added.* After completing this manuscript, we became aware of another work [60] which studied a particle-hole-symmetric multiorbital Hubbard model with an interorbital kinetic and/or on-site hybridization and without an orbital-level splitting. The two works reached consistent conclusions where there is overlap.

#### ACKNOWLEDGMENTS

We are grateful to E. Abrahams, W. Hu, L. de' Medici, V. Dobrosavljević, Y. Komijani, and G. Kotliar for useful discussions, and to G. Kotliar for a careful reading of the manuscript and the constructive comments to it. This work has in part been supported by the National Science Foundation of China Grants No. 11374361 and No. 11674392 and Ministry of Science and Technology of China Grant No. 2016YFA0300504 (R.Y.), and by the Department of Energy Office of Basic Energy Sciences and the Robert A. Welch Foundation Grant No. C-1411 (Q.S.). R.Y. acknowledges the hospitality of Rice University. Q.S. acknowledges the hospitality of University of California at Berkeley, as well as the Aspen Center for Physics (NSF Grant No. PHY-1607611) during the 2016 summer program where part of this work was discussed.

#### APPENDIX A: DERIVATION OF THE SADDLE-POINT EQUATIONS

To facilitate the detailed derivations and analyses presented in the next two appendixes, we summarize here the derivation [32] of the equations Eqs. (12) and (13) from Eq. (10). By decomposing the slave (Schwinger) boson—or, equivalently, the slave spin—and the pseudofermion operators in Eq. (10),

we obtain

$$H_f^{\text{mf}} = \frac{1}{2} \sum_{ij\alpha\beta\sigma} t_{ij}^{\alpha\beta} \langle \hat{z}_{i\alpha\sigma}^\dagger \hat{z}_{j\beta\sigma} \rangle f_{i\alpha\sigma}^\dagger f_{j\beta\sigma} + \sum_{i\alpha\sigma} (\Delta_\alpha - \lambda_{i\alpha\sigma} - \mu) f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma}, \quad (\text{A1})$$

$$H_S^{\text{mf}} = \frac{1}{2} \sum_{ij\alpha\beta\sigma} t_{ij}^{\alpha\beta} \langle f_{i\alpha\sigma}^\dagger f_{j\beta\sigma} \rangle z_{i\alpha\sigma}^\dagger z_{j\beta\sigma} + \sum_{i\alpha\sigma} \frac{\lambda_{i\alpha\sigma}}{2} (\hat{n}_{i\alpha\sigma}^a - \hat{n}_{i\alpha\sigma}^b) + H_{\text{int}}^S, \quad (\text{A2})$$

where  $\langle \dots \rangle$  denotes the averaging taken with respect to these Hamiltonians, and  $\hat{n}_{i\alpha\sigma}^a = a_{i\alpha\sigma}^\dagger a_{i\alpha\sigma}$ . Here  $H_S^{\text{mf}}$  has an internal  $U(1)$  symmetry of the bosons. For the single-orbital case, it is a Bose-Hubbard model for two species of hard-core bosons (or, equivalently, a model for interacting  $XY$  spins), and it possesses a phase transition from a bosonic Mott insulator to a superfluid. More generally, we start from the side with a Bose condensation in the composite boson field  $z_{i\alpha\sigma}$ . The leading term is captured by a single-site decomposition in Eqs. (A1) and (A2), with  $\hat{z}_{i\alpha\sigma}^\dagger \hat{z}_{j\beta\sigma} \approx \langle \hat{z}_{i\alpha\sigma}^\dagger \rangle \hat{z}_{j\beta\sigma} + \hat{z}_{i\alpha\sigma}^\dagger \langle \hat{z}_{j\beta\sigma} \rangle - \langle \hat{z}_{i\alpha\sigma}^\dagger \rangle \langle \hat{z}_{j\beta\sigma} \rangle$ . We focus on the paramagnetic phase with the translational symmetry preserved, in which the spin and site indices can be dropped without causing ambiguity. The boson Hamiltonian then reads

$$H_S^{\text{mf}} \approx \sum_{\alpha\beta} Q_{\alpha\beta}^f (\langle \hat{z}_{i\alpha\sigma}^\dagger \rangle \hat{z}_\beta + \langle \hat{z}_\beta \rangle \hat{z}_{i\alpha\sigma}^\dagger) + \sum_\alpha \frac{\lambda_\alpha}{2} (\hat{n}_\alpha^a - \hat{n}_\alpha^b) + H_{\text{int}}^S. \quad (\text{A3})$$

In Eq. (A3) we Taylor-expand  $\hat{z}_\alpha$  and  $\hat{z}_\alpha^\dagger$  in terms of  $\hat{A} - \langle \hat{A} \rangle$  (where  $\hat{A} = \hat{n}^a, \hat{n}^b, a^\dagger b$ ), and keep up to the linear terms in  $\hat{A} - \langle \hat{A} \rangle$ . This leads to

$$\hat{z}_\alpha^\dagger \approx \bar{z}_\alpha^\dagger + \langle \bar{z}_\alpha^\dagger \rangle \eta_\alpha [\hat{n}_\alpha^a - \hat{n}_\alpha^b - (2n_\alpha^f - 1)], \quad (\text{A4})$$

where  $\bar{z}_\alpha^\dagger = \langle P_\alpha^+ \rangle a_\alpha^\dagger b_\alpha \langle P_\alpha^- \rangle$ . The details involved in the derivation of Eq. (A4) is given in Appendix B. Note that  $n_\alpha^f = \langle \hat{n}_\alpha^a \rangle = 1 - \langle \hat{n}_\alpha^b \rangle$  from the constraints. With this, we find that  $\langle \bar{z}_\alpha \rangle = \langle \bar{z}_\alpha \rangle$ , which is defined as  $z_\alpha$  [cf. Eq. (11)]. Using Eq. (A4), the saddle-point Hamiltonian given in Eq. (A3) becomes

$$H_S^{\text{mf}} \approx \sum_{\alpha\beta} Q_{\alpha\beta}^f (\langle \bar{z}_\alpha^\dagger \rangle \bar{z}_\beta + \langle \bar{z}_\beta \rangle \bar{z}_\alpha^\dagger) + \sum_\alpha \left( \frac{\lambda_\alpha}{2} + \bar{\epsilon}_\alpha \eta_\alpha \right) (\hat{n}_\alpha^a - \hat{n}_\alpha^b) + H_{\text{int}}^S. \quad (\text{A5})$$

Further using the constraint Eq. (5), we can redefine  $\lambda_\alpha$  to move the term proportional to  $\eta_\alpha$  to  $H_f^{\text{mf}}$  by introducing an effective on-site potential  $\tilde{\mu}_\alpha$ . We then arrive at the two saddle-point Hamiltonians, Eqs. (12) and (13). Recognizing the hard-core nature of the bosons (or, equivalently, recognizing that they can be transformed back to  $XY$  spins), we can exactly diagonalize the Hamiltonian in Eq. (13) even though it contains quartic terms of the boson operators in  $H_{\text{int}}^S$ .

**APPENDIX B: DERIVATION OF EQ. (A4)**

Starting from the definition of the projectors  $P_{i\alpha\sigma}^{\pm} = 1/\sqrt{1/2 + \delta \pm (\hat{n}_{i\alpha\sigma}^a - \hat{n}_{i\alpha\sigma}^b)/2}$ , we expand  $\hat{n}_{i\alpha\sigma}^a - \hat{n}_{i\alpha\sigma}^b$  about its saddle-point value and obtain

$$P_{\alpha}^{+} = \frac{1}{\sqrt{\left[\frac{1}{2} + \frac{1}{2}(\langle \hat{n}_{\alpha}^a \rangle - \langle \hat{n}_{\alpha}^b \rangle) + \delta\right] \left[1 + \frac{\Delta \hat{n}}{1 + \langle \hat{n}_{\alpha}^a \rangle - \langle \hat{n}_{\alpha}^b \rangle + 2\delta}\right]}} \approx \frac{1}{\sqrt{\frac{1}{2} + \frac{1}{2}(\langle \hat{n}_{\alpha}^a \rangle - \langle \hat{n}_{\alpha}^b \rangle) + \delta}} \left\{ 1 - \frac{\Delta \hat{n}}{2(1 + \langle \hat{n}_{\alpha}^a \rangle - \langle \hat{n}_{\alpha}^b \rangle + 2\delta)} \right\}, \quad (\text{B1})$$

where  $\Delta \hat{n} = (\hat{n}_{\alpha}^a - \hat{n}_{\alpha}^b) - (\langle \hat{n}_{\alpha}^a \rangle - \langle \hat{n}_{\alpha}^b \rangle)$ . Here we have again dropped the site and spin indices for simplicity. By using the constraint in Eq. (5) we have  $\langle \hat{n}_{\alpha}^a \rangle - \langle \hat{n}_{\alpha}^b \rangle = 2n_{\alpha}^f - 1$ . This further simplifies Eq. (B1) to

$$P_{\alpha}^{+} \approx \frac{1}{\sqrt{n_{\alpha}^f + \delta}} \left( 1 - \frac{\Delta \hat{n}}{4(n_{\alpha}^f + \delta)} \right). \quad (\text{B2})$$

Similarly, we have

$$P_{\alpha}^{-} \approx \frac{1}{\sqrt{1 - n_{\alpha}^f + \delta}} \left( 1 + \frac{\Delta \hat{n}}{4(1 - n_{\alpha}^f + \delta)} \right). \quad (\text{B3})$$

Inserting these into the definition of  $\hat{z}_{\alpha}^{\dagger}$  in Eq. (6), we obtain

$$\begin{aligned} \hat{z}_{\alpha}^{\dagger} &\approx \langle P_{\alpha}^{+} \rangle \left( 1 - \frac{\Delta \hat{n}}{4(n_{\alpha}^f + \delta)} \right) [\langle a_{\alpha}^{\dagger} b_{\alpha} \rangle + (a_{\alpha}^{\dagger} b_{\alpha} - \langle a_{\alpha}^{\dagger} b_{\alpha} \rangle)] \langle P_{\alpha}^{-} \rangle \left( 1 + \frac{\Delta \hat{n}}{4(1 - n_{\alpha}^f + \delta)} \right) \\ &\approx \langle P_{\alpha}^{+} \rangle a_{\alpha}^{\dagger} b_{\alpha} \langle P_{\alpha}^{-} \rangle + \langle P_{\alpha}^{+} \rangle \langle a_{\alpha}^{\dagger} b_{\alpha} \rangle \langle P_{\alpha}^{-} \rangle \frac{(2n_{\alpha}^f - 1)\Delta \hat{n}}{4n_{\alpha}^f(1 - n_{\alpha}^f) + \delta} + O(\Delta \hat{n}^2), \end{aligned} \quad (\text{B4})$$

where we have defined  $\langle P_{\alpha}^{+} \rangle = 1/\sqrt{n_{\alpha}^f + \delta}$  and  $\langle P_{\alpha}^{-} \rangle = 1/\sqrt{1 - n_{\alpha}^f + \delta}$ . Using the definition of  $\tilde{z}_{\alpha}^{\dagger}$  given in Eq. (15) and that of  $\eta_{\alpha}$  given in Eq. (18), we arrive at

$$\hat{z}_{\alpha}^{\dagger} \approx \tilde{z}_{\alpha}^{\dagger} + \langle \tilde{z}_{\alpha}^{\dagger} \rangle \eta_{\alpha} \Delta \hat{n}, \quad (\text{B5})$$

which, then, yields Eq. (A4).

**APPENDIX C: RECOVERY OF THE PROPER NONINTERACTING LIMIT IN THE  $U(1)$  SLAVE-SPIN APPROACH**

In this section we show how the saddle-point equations of the  $U(1)$  slave-spin theory recovers the correct noninteracting ( $U = J_H = 0$ ) limit for a general multiorbital Hubbard model. In the noninteracting limit, the quasiparticle spectral weight of the itinerant electrons is not renormalized by the electron correlations; therefore

$$Z_{\alpha} = 1 \quad (\text{C1})$$

for each orbital. In addition, the spinon dispersion should be identical to the original tight-binding dispersion of the physical  $d$  electrons. Both features would be captured if Eq. (C1) is accompanied by

$$\lambda_{\alpha} = \tilde{\mu}_{\alpha}. \quad (\text{C2})$$

Indeed, in this case Eq. (12) becomes

$$H_f^{\text{mf}} = \sum_{k\alpha\beta} \{ \epsilon_k^{\alpha\beta} + \delta_{\alpha\beta}(\Delta_{\alpha} - \mu) \} f_{k\alpha}^{\dagger} f_{k\beta}. \quad (\text{C3})$$

This generates exactly the same dispersion as for the original tight-binding model of the  $3d$  electrons in Eq. (2).

We now show that Eqs. (C1) and (C2) indeed solve the saddle-point equations in the noninteracting case. Our strategy is to show that  $\lambda_{\alpha} = \tilde{\mu}_{\alpha}$  leads to  $Z_{\alpha} = 1$  in this case. With  $\lambda_{\alpha} = \tilde{\mu}_{\alpha}$ , the Hamiltonian  $H_S^{\text{mf}}$  at  $U = J_H = 0$  in Eq. (13) becomes

$$\begin{aligned} H_S^{\text{mf}} &= \sum_{\alpha\beta} \left\{ Q_{\alpha\beta}^f (\langle \tilde{z}_{\alpha}^{\dagger} \rangle \tilde{z}_{\beta} + \langle \tilde{z}_{\beta} \rangle \tilde{z}_{\alpha}^{\dagger}) + \delta_{\alpha\beta} \frac{\tilde{\mu}_{\alpha}}{2} (\hat{n}_{\alpha}^a - \hat{n}_{\alpha}^b) \right\} \\ &= \sum_{\alpha} \{ h_{\alpha} \tilde{z}_{\alpha}^{\dagger} + h_{\alpha}^* \tilde{z}_{\alpha} + (h_{\alpha} \langle \tilde{z}_{\alpha}^{\dagger} \rangle \eta_{\alpha} + h_{\alpha}^* \langle \tilde{z}_{\alpha} \rangle \eta_{\alpha}) (\hat{n}_{\alpha}^a - \hat{n}_{\alpha}^b) \} = \sum_{\alpha} (a_{\alpha}^{\dagger}, b_{\alpha}^{\dagger}) \mathcal{H}_{S_{\alpha}}^{\text{mf}} \begin{pmatrix} a_{\alpha} \\ b_{\alpha} \end{pmatrix}, \end{aligned} \quad (\text{C4})$$

where  $h_\alpha = \sum_\beta Q_{\alpha\beta}^f \langle \tilde{z}_\beta \rangle$  and  $\mathcal{H}_{S_\alpha}^{\text{mf}}$  is a  $2 \times 2$  matrix,

$$\mathcal{H}_{S_\alpha}^{\text{mf}} = \begin{pmatrix} \frac{(2n_\alpha^f - 1)(h_\alpha \langle \tilde{z}_\alpha \rangle + \text{c.c.})}{4n_\alpha^f(1 - n_\alpha^f)} & \frac{h_\alpha}{\sqrt{n_\alpha^f(1 - n_\alpha^f)}} \\ \frac{h_\alpha^*}{\sqrt{n_\alpha^f(1 - n_\alpha^f)}} & \frac{-(2n_\alpha^f - 1)(h_\alpha \langle \tilde{z}_\alpha \rangle + \text{c.c.})}{4n_\alpha^f(1 - n_\alpha^f)} \end{pmatrix}. \quad (\text{C5})$$

Without losing generality, we take  $\langle \tilde{z}_\alpha \rangle$  to be real, in which case  $h_\alpha$  is also real. By diagonalizing  $\mathcal{H}_{S_\alpha}^{\text{mf}}$ , we obtain that, at  $T = 0$ ,

$$|\langle \tilde{z}_\alpha \rangle| = \frac{1}{2\sqrt{(n_\alpha^f - 1/2)^2 |\langle \tilde{z}_\alpha \rangle|^2 + n_\alpha^f(1 - n_\alpha^f)}}. \quad (\text{C6})$$

This equation has two solutions, either  $|\langle \tilde{z}_\alpha \rangle| = 1$  or  $|\langle \tilde{z}_\alpha \rangle| = -1/(2n_\alpha^f - 1)^2 < 0$ . Because the second solution is unphysical, we have  $|\langle \tilde{z}_\alpha \rangle| = 1$ , corresponding to  $Z_\alpha = 1$  for each orbital  $\alpha$ .

Therefore, in the noninteracting case,  $\lambda_\alpha = \tilde{\mu}_\alpha$ , and the saddle-point equations of the  $U(1)$  slave-spin theory recover the correct noninteracting limit of the multiorbital Hubbard model. In the case of degenerate orbitals at half-filling, this result is straightforward. On the other hand, for the generic case of nondegenerate orbitals, when the electron density of orbital  $\alpha$  (which is proportional to  $\langle \hat{n}_\alpha^a \rangle - \langle \hat{n}_\alpha^b \rangle$ ) is away from half-filling,  $\lambda_\alpha$  is generally nonzero, and a nonzero  $\tilde{\mu}_\alpha$  is crucial for the recovery of the proper noninteracting limit.

### Comparison with the $Z_2$ slave-spin theory

We now make a comparison between the  $U(1)$  and the  $Z_2$  slave-spin theory. In the  $Z_2$  formulation, the field  $\tilde{\mu}_\alpha$  is absent. Therefore, for multiorbital models with nondegenerate orbitals away from half-filling, the saddle-point equations of the  $Z_2$  formulation do not correctly capture the limit of zero interactions. It has been shown in Ref. [32] that by choosing a particular projector, the saddle-point Hamiltonian  $H_S^{\text{mf}}$  of the  $U(1)$  theory—which does incorporate the parameters  $\tilde{\mu}_\alpha$ —can be written in a form appropriate for the  $Z_2$  slave-spin theory. This suggests a route to remedy the  $Z_2$  formulation in the generic case of multiorbital models with nondegenerate orbitals away from half-filling, such that the  $\tilde{\mu}_\alpha$  parameters be introduced; whether this can be done in a natural way remains unclear. Note that, even if this is achieved, the agreement between the saddle-point results of the  $U(1)$  and  $Z_2$  formulations only applies to the metallic phase where the slave spins are ordered. The  $Z_2$  theory is insufficient to describe a Mott insulating state, due to the fact that the pseudofermions must carry not only the physical spin degrees of freedom but also the physical charge degrees of freedom [48].

- 
- [1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, Iron-based layered superconductor  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$  ( $x = 0.05\text{--}0.12$ ) with  $T_c = 26$  K, *J. Am. Chem. Soc.* **130**, 3296 (2008).
- [2] D. C. Johnston, The puzzle of high temperature superconductivity in layered iron pnictides and chalcogenides, *Adv. Phys.* **59**, 803 (2010).
- [3] P. Dai, Antiferromagnetic order and spin dynamics in iron-based superconductors, *Rev. Mod. Phys.* **87**, 855 (2015).
- [4] Q. Si, R. Yu, and E. Abrahams, High temperature superconductivity in iron pnictides and chalcogenides, *Nat. Rev. Mater.* **1**, 16017 (2016).
- [5] P. J. Hirschfeld, Gap symmetry and structure to reveal the pairing mechanism in Fe-based superconductors, *C. R. Phys.* **17**, 197 (2016).
- [6] F. Wang and D.-H. Lee, The electron-pairing mechanism of iron-based superconductors, *Science* **332**, 200 (2011).
- [7] M. Yi, D. H. Lu, R. Yu, S. C. Riggs, J.-H. Chu, B. Lv, Z. K. Liu, M. Lu, Y. T. Cui, M. Hashimoto, S.-K. Mo, Z. Hussain, C. W. Chu, I. R. Fisher, Q. Si, and Z.-X. Shen, Observation of Temperature-Induced Crossover to an Orbital-Selective Mott Phase in  $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$  ( $\text{A} = \text{K, Rb}$ ) Superconductors, *Phys. Rev. Lett.* **110**, 067003 (2013).
- [8] M. Yi, Z.-K. Liu, Y. Zhang, R. Yu, J.-X. Zhu, J. J. Lee, R. G. Moore, F. T. Schmitt, W. Li, S. C. Riggs, J.-H. Chu, B. Lv, J. Hu, T. J. Liu, M. Hashimoto, S.-K. Mo, Z. Hussain, Z. Q. Mao, C. W. Chu, I. R. Fisher, Q. Si, Z.-X. Shen, and D. H. Lu, Observation of universal strong orbital-dependent correlation effects in iron chalcogenides, *Nat. Commun.* **6**, 7777 (2015).
- [9] Y. J. Pu, Z. C. Huang, H. C. Xu, D. F. Xu, Q. Song, C. H. P. Wen, R. Peng, and D. L. Feng, Temperature-induced orbital selective localization and coherent-incoherent crossover in single-layer  $\text{FeSe/Nb : BaTiO}_3/\text{KTaO}_3$ , *Phys. Rev. B* **94**, 115146 (2016).
- [10] M. Yi, Y. Zhang, Z.-X. Shen, and D. H. Lu, Role of the orbital degree of freedom in iron-based superconductors, [arXiv:1703.08622](https://arxiv.org/abs/1703.08622).
- [11] R. Yu and Q. Si, Orbital-Selective Mott Phase in Multiorbital Models for Alkaline Iron Selenides  $\text{K}_{1-x}\text{Fe}_{2-y}\text{Se}_2$ , *Phys. Rev. Lett.* **110**, 146402 (2013).
- [12] R. Yu, J.-X. Zhu, and Q. Si, Orbital-dependent effects of electron correlations in microscopic models for iron-based superconductors, *Curr. Opinion Solid State Mater. Sci.* **17**, 65 (2013).
- [13] Z. Wang, M. Schmidt, J. Fischer, V. Tsurkan, M. Greger, D. Vollhardt, A. Loidl, and J. Deisenhofer, Orbital-selective

- metal-insulator transition and gap formation above  $T_c$  in superconducting  $\text{Rb}(1-x)\text{Fe}(2-y)\text{Se}_2$ , *Nat. Commun.* **5**, 3202 (2014).
- [14] X. Ding, Y. Pan, H. Yang, and H.-H. Wen, Strong and nonmonotonic temperature dependence of Hall coefficient in superconducting  $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$  single crystals, *Phys. Rev. B* **89**, 224515 (2014).
- [15] W. Li, C. Zhang, S. Liu, X. Ding, X. Wu, X. Wang, H.-H. Wen, and M. Xiao, Mott behavior in  $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$  superconductors studied by pump-probe spectroscopy, *Phys. Rev. B* **89**, 134515 (2014).
- [16] P. Gao, R. Yu, L. Sun, H. Wang, Z. Wang, Q. Wu, M. Fang, G. Chen, J. Guo, C. Zhang, D. Gu, H. Tian, J. Li, J. Liu, Y. Li, X. Li, S. Jiang, K. Yang, A. Li, Q. Si, and Z. Zhao, Role of the 245 phase in alkaline iron selenide superconductors revealed by high-pressure studies, *Phys. Rev. B* **89**, 094514 (2014).
- [17] Z. P. Yin, K. Haule, and G. Kotliar, Kinetic frustration and the nature of the magnetic and paramagnetic states in iron pnictides and iron chalcogenides, *Nat. Mater.* **10**, 932 (2011).
- [18] C. Aron and G. Kotliar, Analytic theory of Hund's metals: A renormalization group perspective, *Phys. Rev. B* **91**, 041110(R) (2015).
- [19] Y. Song, Z. Yamani, C.-D. Cao, Y. Li, C. L. Zhang, J. Chen, Q. Z. Huang, H. Wu, J. Tao, Y. Zhu, W. Tian, S. X. Chi, H. Cao, Y.-B. Huang, M. Dantz, T. Schmitt, R. Yu, A. H. Nevidomskyy, E. Morosan, Q. Si, and P. Dai, A Mott insulator continuously connected to iron pnictide superconductors, *Nat. Commun.* **7**, 13879 (2016).
- [20] M.-H. Fang, H.-D. Wang, C.-H. Dong, Z.-J. Li, C.-M. Feng, J. Chen, and H. Q. Yuan, Fe-based superconductivity with  $T_c = 31$  K bordering an antiferromagnetic insulator in  $(\text{Ti}, \text{K})\text{Fe}_x\text{Se}_2$ , *Europhys. Lett.* **94**, 27009 (2011).
- [21] J.-X. Zhu, R. Yu, H. Wang, L. L. Zhao, M. D. Jones, J. Dai, E. Abrahams, E. Morosan, M. Fang, and Q. Si, Band Narrowing and Mott Localization in Iron Oxychalcogenides  $\text{La}_2\text{O}_2\text{Fe}_2\text{O}(\text{Se}, \text{S})_2$ , *Phys. Rev. Lett.* **104**, 216405 (2010).
- [22] M. Wang, M. Yi, H. Cao, C. de la Cruz, S. K. Mo, Q. Z. Huang, E. Bourret-Courchesne, P. Dai, D. H. Lee, Z. X. Shen, and R. J. Birgeneau, Mott localization in a pure stripe antiferromagnet  $\text{Rb}_{1-\delta}\text{Fe}_{1.5-\sigma}\text{S}_2$ , *Phys. Rev. B* **92**, 121101 (2015).
- [23] M. M. Qazilbash, J. J. Hamlin, R. E. Baumbach, L. Zhang, D. J. Singh, M. B. Maple, and D. N. Basov, Electronic correlations in the iron pnictides, *Nat. Phys.* **5**, 647 (2009).
- [24] Q. Si and E. Abrahams, Strong Correlations and Magnetic Frustration in the High  $T_c$  Iron Pnictides, *Phys. Rev. Lett.* **101**, 076401 (2008).
- [25] K. Haule and G. Kotliar, Coherence-incoherence crossover in the normal state of iron-oxypnictides and importance of the Hund's rule coupling, *New J. Phys.* **11**, 025021 (2009).
- [26] K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kontani, and H. Aoki, Unconventional Pairing Originating from the Disconnected Fermi Surfaces of Superconducting  $\text{LaFeAsO}_{1-x}\text{F}_x$ , *Phys. Rev. Lett.* **101**, 087004 (2008).
- [27] Q. Si, E. Abrahams, J. Dai, and J.-X. Zhu, Correlation effects in the iron pnictides, *New J. Phys.* **11**, 045001 (2009).
- [28] S. Graser, T. A. Maier, P. J. Hirschfeld, and D. J. Scalapino, Near-degeneracy of several pairing channels in multi-orbital models for the Fe pnictides, *New J. Phys.* **11**, 025016 (2009).
- [29] C. Cao, P. J. Hirschfeld, and H.-P. Cheng, Proximity of antiferromagnetism and superconductivity in  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$ : Effective Hamiltonian from *ab initio* studies, *Phys. Rev. B* **77**, 220506(R) (2008).
- [30] P. A. Lee and X.-G. Wen, Spin-triplet  $p$ -wave pairing in a 3-orbital model for iron pnictide superconductors, *Phys. Rev. B* **78**, 144517 (2008).
- [31] R. Yu and Q. Si, Mott transition in multi-orbital models for iron pnictides, *Phys. Rev. B* **84**, 235115 (2011).
- [32] R. Yu and Q. Si,  $U(1)$  Slave-spin theory and its application to Mott transition in a multi-orbital model for iron pnictides, *Phys. Rev. B* **86**, 085104 (2012).
- [33] E. Bascones, B. Valenzuela, and M. J. Calderón, Orbital differentiation and the role of orbital ordering in the magnetic state of Fe superconductors, *Phys. Rev. B* **86**, 174508 (2012).
- [34] L. de' Medici, G. Giovannetti, and M. Capone, Selective Mott Physics as a Key to Iron Superconductors, *Phys. Rev. Lett.* **112**, 177001 (2014).
- [35] Y.-Z. Zhang, H. Lee, H.-Q. Lin, C.-Q. Wu, H. O. Jeschke, and R. Valentí, General mechanism for orbital selective phase transitions, *Phys. Rev. B* **85**, 035123 (2012).
- [36] Z. P. Yin, K. Haule, and G. Kotliar, Fractional power-law behavior and its origin in iron-chalcogenide and ruthenate superconductors: Insights from first-principles calculations, *Phys. Rev. B* **86**, 195141 (2012).
- [37] H. Miao, Z. P. Yin, S. F. Wu, J. M. Li, J. Ma, B.-Q. Lv, X. P. Wang, T. Qian, P. Richard, L.-Y. Xing, X.-C. Wang, C. Q. Jin, K. Haule, G. Kotliar, and H. Ding, Orbital-differentiated coherence-incoherence crossover identified by photoemission spectroscopy in  $\text{LiFeAs}$ , *Phys. Rev. B* **94**, 201109 (2016).
- [38] V. I. Anisimov, I. A. Nekrasov, D. E. Kondakov, T. M. Rice, and M. Sigríst, Orbital-selective Mott-insulator transition in  $\text{Ca}_{2-x}\text{Sr}_x\text{RuO}_4$ , *Eur. Phys. J. B* **25**, 191 (2002).
- [39] A. Koga, N. Kawakami, T. M. Rice, and M. Sigríst, Orbital-Selective Mott Transitions in the Degenerate Hubbard Model, *Phys. Rev. Lett.* **92**, 216402 (2004).
- [40] P. Werner and A. J. Millis, High-Spin to Low-Spin and Orbital Polarization Transitions in Multi-orbital Mott Systems, *Phys. Rev. Lett.* **99**, 126405 (2007).
- [41] P. Werner, E. Gull, and A. J. Millis, Metal-insulator phase diagram and orbital selectivity in 3-orbital models with rotationally invariant Hund coupling, *Phys. Rev. B* **79**, 115119 (2009).
- [42] L. de' Medici, S. R. Hassan, M. Capone, and X. Dai, Orbital-Selective Mott Transition out of Band Degeneracy Lifting, *Phys. Rev. Lett.* **102**, 126401 (2009).
- [43] L. de' Medici, Hund's coupling key role in multi-orbital correlations, *Phys. Rev. B* **83**, 205112 (2011).
- [44] G. Kotliar, Landau theory of the Mott transition in the fully frustrated Hubbard model in infinite dimensions, *Eur. J. Phys. B* **11**, 27 (1999).
- [45] C. Castellani, C. R. Natoli, and J. Ranninger, Magnetic structure of  $\text{V}_2\text{O}_3$  in the insulating phase, *Phys. Rev. B* **18**, 4945 (1978).
- [46] L. de' Medici, A. Georges, and S. Biermann, Orbital-selective Mott transition in multiband systems: Slave-spin representation and dynamical mean-field theory, *Phys. Rev. B* **72**, 205124 (2005).
- [47] A. Rüegg, S. D. Huber, and M. Sigríst,  $Z_2$ -slave-spin theory for strongly correlated fermions, *Phys. Rev. B* **81**, 155118 (2010).
- [48] R. Nandkishore, M. A. Metlitski, and T. Senthil, Orthogonal metals: The simplest non-Fermi liquids, *Phys. Rev. B* **86**, 045128 (2012).

- [49] S. Florens and A. Georges, Slave-rotor mean-field theories of strongly correlated systems and the Mott transition in finite dimensions, *Phys. Rev. B* **70**, 035114 (2004).
- [50] G. Kotliar and A. E. Ruckenstein, A New Functional Integral Approach to Strongly Correlated Fermi Systems: The Gutzwiller Approximation as a Saddle Point, *Phys. Rev. Lett.* **57**, 1362 (1986).
- [51] Q. Si, S. Rabello, K. Ingersent, and J. L. Smith, Locally-critical quantum phase transitions in strongly correlated metals, *Nature (London)* **413**, 804 (2001).
- [52] P. Coleman, C. Pépin, Q. Si, and R. Ramazashvili, How do Fermi liquids get heavy and die? *J. Phys.: Condens. Matter* **13**, R723 (2001).
- [53] C. Pépin, Kondo Breakdown as a Selective Mott Transition in the Anderson Lattice, *Phys. Rev. Lett.* **98**, 206401 (2007).
- [54] Q. Si, J. H. Pixley, E. Nica, S. J. Yamamoto, P. Goswami, R. Yu, and S. Kirchner, Kondo destruction and quantum criticality in Kondo lattice systems, *J. Phys. Soc. Jpn.* **83**, 061005 (2014).
- [55] S. Daul, D. J. Scalapino, and S. R. White, Pairing Correlations on  $t$ - $U$ - $J$  Ladders, *Phys. Rev. Lett.* **84**, 4188 (2000).
- [56] S. Mukherjee, N. F. Quackenbush, H. Paik, C. Schlueter, T.-L. Lee, D. G. Schlom, L. F. J. Piper, and W.-C. Lee, Tuning a strain-induced orbital selective Mott transition in epitaxial VO<sub>2</sub>, *Phys. Rev. B* **93**, 241110 (2016).
- [57] J. Rincón, A. Moreo, G. Alvarez, and E. Dagotto, Exotic Magnetic Order in the Orbital-Selective Mott Regime of Multiorbital Systems, *Phys. Rev. Lett.* **112**, 106405 (2014).
- [58] G. Liu, N. Kaushal, S. Li, C. B. Bishop, Y. Wang, S. Johnston, G. Alvarez, A. Moreo, and E. Dagotto, Orbital-selective Mott phases of a one-dimensional three-orbital Hubbard model studied using computational techniques, *Phys. Rev. E* **93**, 063313 (2016).
- [59] S. Li, N. Kaushal, Y. Wang, Y. Tang, G. Alvarez, A. Nocera, T. A. Maier, E. Dagotto, and S. Johnston, Non-local correlations in the orbital selective Mott phase of a one dimensional multi-orbital Hubbard model, *Phys. Rev. B* **94**, 235126 (2016).
- [60] Y. Komijani and G. Kotliar, Analytical slave-spin mean-field approach to orbital selective Mott insulators, *Phys. Rev. B* **96**, 125111 (2017).