Orbital-dependent effects of electron correlations in microscopic models for iron-based superconductors

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A B S T R A C T

The bad metal behavior in the normal state of the iron-based superconductors suggests an intimate connection between the superconductivity and a proximity to a Mott transition. At the same time, there is strong evidence for the multiorbital nature of the electronic excitations. It is then important to understand the orbital-dependent effects of electron correlations. In this paper we review the recent theoretical progresses on the metal-to-insulator transition in multiorbital models for the iron-based superconductors. These include studies of models that contain at least the $3d\,xy$ and $3d\,xz/yz$ models, using a slave-spin technique. For commensurate filling corresponding to that of the parent iron pnictides and chalcogenideds, a Mott transition generally exists in all these models. Near the Mott transition, a strongly correlated metal exhibiting bad metal features and strong orbital selectivity is stabilized due to the interplay of Hund's coupling and orbital-degeneracy breaking. Particularly for the alkaline iron selenides, the ordered vacancies effectively reduce the kinetic energy, thereby pushing the system further into the Mott-insulating regime; in the metallic state, there exists an orbital-selective Mott phase in which the iron $3d\,xy$ orbital is Mott localized while the other $3d$ orbitals are still itinerant. An overall phase diagram for the alkaline iron selenides has been proposed, in which the orbital-selective Mott phase connects between the superconducting phase and the Mott-insulating parent state.

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1. Introduction

Triggered by the seminal discovery of superconductivity at 26 K in F-doped LaFeAsO [1], the study on the iron-based high-temperature superconductors has recently become an important field in condensed matter physics. The iron-based superconductors consist of two major families of compounds, iron pnictides and iron chalcogenides. They share a common layered structure, with a square lattice of iron ions. The electronic properties of the iron-based superconductors are closely associated with the iron $3d$ orbitals. Typically, the parent compounds have an antiferromagnetically ordered ground state [2], and superconductivity emerges within a certain range of chemical doping. The parent systems have the iron valence +2, corresponding to 6 electrons occupying the $3d$ orbitals. In the iron pnictides and some iron chalcogenide compounds, the parent antiferromagnet is metallic. This metallic behavior and the existence of the hole and electron Fermi pockets [3] raise a central question on the role of the electron correlations. On the one hand, the antiferromagnetic order and superconductivity may arise from a Fermi surface nesting mechanism of a weak coupling theory [4–6]. On the other hand, many experiments imply that the parent

pnictides and selenides are bad metals with considerable electron correlations. This is shown by a large value of the room-temperature electrical resistivity, which corresponds to a mean-free path of the order of only k_F^{-1} Ref. [7], and a sizable reduction of the Drude weight in the optical conductivity [8–12]. Indeed, the latter implies that the majority of the electron excitations lie in the incoherent part away from the Fermi energy. Furthermore, measurements of the dynamical spin susceptibility yield an integrated spin spectral weight on the order of $3\mu_B^2$ per Fe in the parent iron pnictides [13]. This size is consistent with quasi-local moments generated by the incoherent electronic excitations, but is too large to account for by the nesting picture of particle-hole excitations from the electrons and holes in the Fermi pockets. ARPES measurements have identified considerable mass renormalization [3,14,15]. All these suggest that the ratio of the Coulomb interactions U (more precisely, a combination of the intra-orbital and inter-orbital Coulomb interactions and the Hund's coupling, see below) to the characteristic bandwidth W is close to U_c/W, the threshold for a Mott transition [16,17]. This incipient Mott picture [16,18,19] expands around the Mott transition in terms of the quasiparticle spectral weight w, which measures the electron correlations in the metallic phase. It describes a bad metal when $w \sim (U_c - U)/W$ is small. The antiferromagnetic order in the bad metal is associated with the incoherent electronic excitations,

which can be modeled by a J_1–J_2 model of quasi-localized magnetic moments. Upon doping, the superconductivity arises out of the bad metal on the verge of the Mott localization. This can take place when the parent compound is either a bad metal (as for iron pnictides) or a Mott insulator (as for alkaline iron selenides). A number of studies on the related strong coupling approach have been carried out in understanding the electronic properties of iron-based superconductors [20–34].

To explore the effect of electron correlations further, it is important to recognize that the electronic structure of the iron-based superconductors involves multiple 3d Fe orbitals. Such multiple orbitals were considered from the beginning [4,16,18,35,36,17,37], and their involvement in the electronic structure near the Fermi energy have been established by ARPES measurements [3]. The tetragonal structure protects only the degeneracy between the 3d xz/yz orbitals, but breaks the degeneracy between these orbitals and the remaining three 3d orbitals. It is therefore instructive to study the orbital-dependent effects of the electron correlations. The appropriate model will include both the intra- and inter-orbital Coulomb interactions, as well as the Hund's coupling.

In this paper we report the recent theoretical progress in understanding the bad metal behavior and the associated metal-to-insulator transition (MIT) in multiorbital models for iron-based superconductors. We study both the bad-metal behavior and the metal-to-Mott-insulator transitions in various multiorbital Hubbard models for the iron pnictides, showing that the proximity to the Mott transition becomes strongly orbital-dependent even for orbitally-independent interactions [38,39]. Our emphasis is on the interplay among the Hubbard interactions, Hund's coupling and the orbital non-degeneracy. For the alkaline iron selenides, we treat the ordered vacancies in terms of a reduction of the bandwidth, which implies an enhanced U/W ratio [40]. The corresponding multiorbital Hubbard models, both in the presence and in the absence of the ordered vacancies, are shown to contain an orbital-selective Mott phase (OSMP) [41]. The latter is characterized by localized orbitals (3d xy) coexisting with itinerant ones, as originally proposed for Ca$_{2-x}$Sr$_x$RuO$_4$ [42]. The OSMP phase provides a natural explanation of the strongly orbital- and temperature-dependent spectral weight observed in the ARPES measurements of the superconducting alkaline iron selenides [43]. It also provides the understanding of an intermediate phase identified through transport measurements in the insulating alkaline iron selenides under a large pressure [44]. We note that theoretical studies on related orbital-dependent aspects of electron correlations are being pursued from a variety of other perspectives and approaches [45–49].

The rest of the paper is organized as follows: In Section 2 we briefly introduce the multiorbital Hubbard model for iron-based superconductors, as well as the slave-spin method [39,50–52] used for studying the MITs in this model. In Section 3 we consider and compare the results of metal-to-Mott-insulator transition in various multiorbital models for iron pnictides. We then proceed to discuss some recent theoretical studies on alkaline iron selenides in Section 4. We address how the ordered iron vacancies enhance the electron correlations and push the system into a Mott insulator in the parent alkaline iron selenides, and discuss the theoretical and experimental studies on the OSMP in these systems. In Section 5, we propose an overall phase diagram to understand the combined effects of tuning both the vacancy order and carrier doping. Some general considerations, particularly on what happens when the electron occupancy moves away from 6, are given in Section 6.

2. Multiorbital Hubbard model and the slave-spin method

We consider a multiorbital Hubbard model for iron-based superconductors. The general form of the Hamiltonian reads

$$H = H_0 + H_{\text{int}}. \tag{1}$$

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

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

where $d_{i\alpha\sigma}^\dagger$ creates an electron in orbital α with spin σ at site i, Δ_α is an on-site energy reflecting the crystal field splitting, and μ is the chemical potential. H_{int} contains on-site Hubbard interactions

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

where $n_{i\alpha\sigma} = d_{i\alpha\sigma}^\dagger d_{i\alpha\sigma}$. The parameters U, U', and J respectively denote the intraorbital repulsion, the interorbital repulsion, and the Hund's rule exchange coupling. They are taken to satisfy $U' = U - 2J$ [53].

The electron correlation effects in the multiorbital Hubbard model is studied via the slave-rotor and the slave-spin methods [39,50–52], which can be viewed as multiorbital versions of the more standard slave-boson theory [54]. In the slave-spin representation, the electron creation operator is rewritten as $d_{i\alpha\sigma}^\dagger = S_{i\alpha\sigma}^+ f_{i\alpha\sigma}^\dagger$. Here, $S_{i\alpha\sigma}^+$ is the ladder operator of a quantum $S = 1/2$ slave spin that carries the electric charge for each orbital and spin flavor, and $f_{i\alpha\sigma}^\dagger$ creates an auxiliary fermion (the spinon) that carries the spin of the electron. The restriction of the Hilbert space to the physical subspace is accomplished by enforcing a constraint, $S_{i\alpha\sigma}^z = f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma} - 1/2$, on each site. A mean-field theory can be obtained by first rewriting the multiorbital Hamiltonian in terms of the slave spins and spinons, then decoupling them. Further performing a single-site approximation and taking into account the translational symmetry of the system in the paramagnetic phase, the resulting mean-field Hamiltonians read:

$$H_f^{\text{mf}} = \sum_{k\alpha\beta}\left[\epsilon_k^{\alpha\beta}\langle O_\alpha^\dagger\rangle\langle O_\beta\rangle + \delta_{\alpha\beta}(\Delta_\alpha - \lambda_\alpha + \tilde\mu_\alpha - \mu)\right]f_{k\alpha}^\dagger f_{k\beta}, \tag{4}$$

$$H_S^{\text{mf}} = \sum_{\alpha\beta}\left[\epsilon^{\alpha\beta}\left(\langle O_\alpha^\dagger\rangle O_\beta + \langle O_\beta\rangle O_\alpha^\dagger\right) + \delta_{\alpha\beta}\lambda_\alpha S_\alpha^z\right] + H_{\text{int}}(\mathbf{S}). \tag{5}$$

Here the spin index σ has been dropped for simplicity. $\epsilon_k^{\alpha\beta} = \frac{1}{N}\sum_{ij} t_{ij}^{\alpha\beta} e^{ik(r_i - r_j)}$, $\epsilon^{\alpha\beta} = \sum_{ij\sigma} t_{ij}^{\alpha\beta} \langle f_{i\alpha\sigma}^\dagger f_{j\beta\sigma}\rangle/2$, $\delta_{\alpha\beta}$ is the Kronecker's delta function, and λ_α is the Lagrange multiplier to handle the constraint. In addition, $\tilde\mu_\alpha$ is an effective chemical potential which is introduced to recover the correct noninteracting limit [41], $H_{\text{int}}(\mathbf{S})$ refers to the interaction Hamiltonian in the slave-spin representation [38], and $O_\alpha^\dagger = \langle P_\alpha^+\rangle S_\alpha^+\langle P_\alpha^-\rangle$, where $P_\alpha^\pm = 1/\sqrt{1/2 + \delta \pm S_\alpha^z}$, with δ being an infinitesimal positive number to regulate $P_\alpha^\pm$. The quasiparticle spectral weight $Z_\alpha = |\langle O_\alpha\rangle|^2$.

The formulism of the slave-rotor theory is very similar, but the slave particle that carries the charge degree of freedom in this case is a quantum $O(2)$ rotor. It is efficient if the interaction has an $SU(2M)$ symmetry for M degenerate orbitals, i.e. when $J = 0$ in Eq. 3. On the other hand, the slave-spin method is more convenient to handle systems with finite Hund's rule coupling and those exhibiting a strong orbital dependence.

Both the slave-rotor and slave-spin theories can describe the MIT in the multiorbital Hubbard model. In particular, the metallic phase corresponds to the state in which the slave particles are Bose condensed ($Z > 0$), so that charge excitations are gapless along with the spin excitations. The Mott insulator corresponds to the state in which the slave particles are disordered ($Z = 0$); here the charge excitations are gapped, while the spin excitations remain gapless.