

Microscopic description of localization-delocalization transitions in BaFe_2S_3

L. Craco^{1,2} and S. Leoni³

¹*Instituto de Física, Universidade Federal de Mato Grosso, 78060-900, Cuiabá, Mato Grosso, Brazil*

²*IFW Dresden, Institute for Solid State Research, P.O. Box 270116, D-01171 Dresden, Germany*

³*School of Chemistry, Cardiff University, Cardiff CF10 3AT, United Kingdom*



(Received 8 June 2018; revised manuscript received 22 October 2018; published 5 November 2018)

We present a microscopic description of electronic reconstruction in BaFe_2S_3 , a system which undergoes a pressure-induced insulator-metal transition followed by a superconducting phase at 24 K. We stress the importance of multiorbital electron-electron interactions for a consistent understanding of its intrinsic Mott-insulating and pressurized, orbital-selective metallic normal states. We explain the first-order nature of the Mott transition, showing that it is driven by dynamical spectral weight transfer in response to changes in the on-site Coulomb interaction to bandwidth ratio. As a by-product of this analysis, we unearth how dynamical correlations underpin spectroscopy and resistivity responses, in good agreement with experiment. Upon electron/hole doping, carrier localization is found to persist because the chemical potential lies in a gap structure with vanishing states near the Fermi energy. We detail the implications of our microscopic analysis for the underlying physics which emerges in the normal state of a compressed BaFe_2S_3 superconductor.

DOI: [10.1103/PhysRevB.98.195107](https://doi.org/10.1103/PhysRevB.98.195107)

I. INTRODUCTION

The discovery of the F-doped LaOFeAs superconductor [1] led to extensive experimental and theoretical study regarding the interplay between electronic structure, magnetism, and unconventional high-temperature (high- T_c) superconductivity [2]. Although many families of Fe-based superconductors have been discovered, the majority of these superconducting materials have a two-dimensional Fe square lattice tetrahedrally coordinated by chalcogen or pnictogen ions. The Fe atoms are nominally divalent (Fe^{2+}) in almost all parent compounds, which usually undergo magnetic or nematic phase transitions at low temperatures [3]. Superconductivity with s_{\pm} symmetry appears when the striped-type antiferromagnetic ordered state is suppressed by pressure or addition of electron/hole carriers via chemical substitution. In spite of the fact that, experimentally, various physical responses show distinct features associated with correlated electron physics, there is still significant disagreement concerning the degree of electron correlation effects in the Fe-based superconductors [2,3]. This has resulted in a situation where multiorbital (MO) electronic structure calculations that explicitly take sizable electron-electron interactions into account were implemented to resolve this crucial issue of fundamental importance [4]. The issue is crucial because the nature and mechanism of the instability of a metal (strange or not) to a superconductor hinge on whether the normal metal state is a conventional Fermi-liquid metal or highly unconventional non-Fermi-liquid metal in close proximity to Mott localization.

In this context, the discovery of BaFe_2S_3 , in which superconductivity emerges in the vicinity of the Mott-insulating state [5,6], is relevant since it confirms previous theoretical suggestions that Mott-insulating parent compounds in the Fe superconducting systems could be found by tuning the ratio of the interaction to bandwidth (U/W) below the critical value for a Mott insulator-metal transition [7]. Pressure-induced

superconductivity in the Fe ladder compounds BaFe_2X_3 ($X = \text{S}, \text{Se}$) [5,6,8] thus focuses the fundamental debate [9] on the degree of electronic correlations in unconventional (non-BCS) superconductors in general. Of particular interest here is BaFe_2S_3 , a narrow-band-gap semiconductor [10] with striped-type magnetic ordering below 120 K at ambient pressures, which undergoes a pressure-induced Mott transition followed by a superconducting phase at a critical pressure P_c of 10.9 GPa [6].

Before delving into the correlated electronic state that emerges in BaFe_2S_3 , we recall that the proximity to a Mott metal-insulator transition [11] (Mottness) is a clear manifestation of dynamical many-body effects in correlated electrons. The charge gap at the Mott transition (MT) jumps discontinuously from a finite value to zero. Mott's original idea [12] was to tune the ratio U/W between the on-site Coulomb interaction U and the one-particle bandwidth W (i.e., the kinetic energy of the electrons) which defines the phase boundary between the metallic and gapped (semiconducting) phases. This scenario holds true for MO systems with similar W values and reduced crystal-field splitting within the active orbitals, but deviations are expected to occur in more general cases where an orbital-selective Mott phase is the precursor to the all-electron Mott insulator [13]. In this work we use local-density approximation plus dynamical mean-field theory (LDA+DMFT) [14] to confirm that both localized and itinerant $3d$ electrons coexist [15] in pressurized BaFe_2S_3 .

The pressure-induced insulator-metal transition in BaFe_2S_3 has been characterized as a bandwidth-controlled Mott transition [5,6]. Up to 10.1 GPa the dc resistivity shows semiconductinglike behavior, and superconductivity at P_c arises directly from a normal state with an insulatinglike resistivity upturn. Such a resistivity upturn, which is followed by a sharp superconducting drop in BaFe_2S_3 [6], is a two-particle manifestation of selective localization

[16] seen in the Fe chalcogenide superconductors [17]. Increasing pressure above 11.6 GPa, the resistivity curves show Fermi-liquid-like T^2 behavior characteristic of a good metal up to about 100 K [6]. However, it turned out that the emergence of superconductivity is sensitive to nominal composition and growth conditions and that the superconducting signal appears to be maximally enhanced at a slightly off-stoichiometric $\text{BaFe}_{2.1}\text{S}_3$ compound [18]. Thus, it is plausible to assume that the superconducting state observed in BaFe_2S_3 emerges from a combination of bandwidth-controlled Mott transition [5,6] and doping of electrons into the iron network [19]. With this in mind, in this work we explore the role played by these two cooperative antilocalization effects [11], showing that the Mott-insulating state is rather robust against small changes in electron concentration of the Fe 3d shell.

II. METHOD

Within the orthorhombic (space group $Cmcm$) crystal structure (see Fig. 1) and using lattice constants and atomic positions at ambient pressure conditions [18], one-electron band structure LDA calculations were performed for BaFe_2S_3 using the linear muffin-tin orbital scheme in the atomic sphere approximation [20]: Self-consistency is reached by performing calculations with 657 irreducible \mathbf{k} points. The radii of the atomic spheres were chosen as $r = 4.95$ a.u. (Ba), $r = 2.57$ a.u. (Fe), and $r = 2.7$ a.u. (S) in order to minimize their overlap. Consistent with previous density functional calculations for the BaFe_2S_3 parent compound [21], our LDA results in Fig. 2 confirm that the active electronic states in this 123-spin-ladder compound involve the Fe 3d carriers, where all d bands have appreciable weight near E_F . From LDA, the one-electron part of the MO Hamiltonian for BaFe_2S_3 is $H_0 = \sum_{\mathbf{k},a,\sigma} \epsilon_a(\mathbf{k}) c_{\mathbf{k},a,\sigma}^\dagger c_{\mathbf{k},a,\sigma}$, where $a = (x^2-y^2, 3z^2-r^2, xz, yz, xy)$ denote its diagonalized 3d orbitals

and $\epsilon_a(\mathbf{k})$ is the corresponding band dispersion, which encodes details of the one-electron (LDA) band structure. These five Fe 3d bands are the relevant one-particle inputs for MO-DMFT which generate a first-order Mott-Hubbard-insulating state at $U = 4.5$ eV as shown below. Similar to tetragonal Fe-based superconductors [4], the correlated many-body Hamiltonian for BaFe_2S_3 reads $H_{\text{int}} = U \sum_{i,a} n_{i,a,\uparrow} n_{i,a,\downarrow} + U' \sum_{i,a \neq b} n_{i,a} n_{i,b} - J_H \sum_{i,a \neq b} \mathbf{S}_{i,a} \cdot \mathbf{S}_{i,b}$. Here U is the on-site Coulomb interaction, $U' = U - 2J_H$ is the interorbital Coulomb interaction term, and J_H is the Hund's coupling. We evaluate the many-particle Green's functions $G_{a,\sigma}(\mathbf{k}, \omega)$ of the MO Hamiltonian $H = H_0 + H_{\text{int}}$ within LDA+DMFT [14], using MO iterated perturbation theory as the impurity solver [22]. The DMFT solution involves replacing the lattice model by a self-consistently embedded MO Anderson impurity model and the self-consistency condition requiring the local impurity Green's function to be equal to the local Green's function for the lattice. The full set of equations for the MO case can be found in Ref. [22].

III. RESULTS AND DISCUSSION

In Fig. 2 we display the effect of moderate [15,21] electron-electron interactions on the orbital-resolved spectral functions of the BaFe_2S_3 parent compound within the d^6 electronic configuration. At $U = 2.0$ eV the system is a Fermi-liquid metal, where all orbitals display appreciable spectral weight at E_F . In spite of this small U value, lower (LHB) and upper (UHB) Hubbard bands are visible in all orbitals, suggesting an intrinsic tendency towards localization in this and related ladder compounds. In the range $3.1 < U < 3.2$ eV (and $J_H = 0.7$ eV) a first-order phase transition from a metallic to an orbital-selective Mott state [13] takes place, where the $3z^2-r^2, xz, xy$ orbitals are Mott localized, whereas yz, x^2-y^2 remain metallic. Strong electronic reconstruction sets in within the selective-Mott-localized orbitals, with the concomitant formation of localized moments (LHB) at energies below -2.5 eV. Both the energy position of the

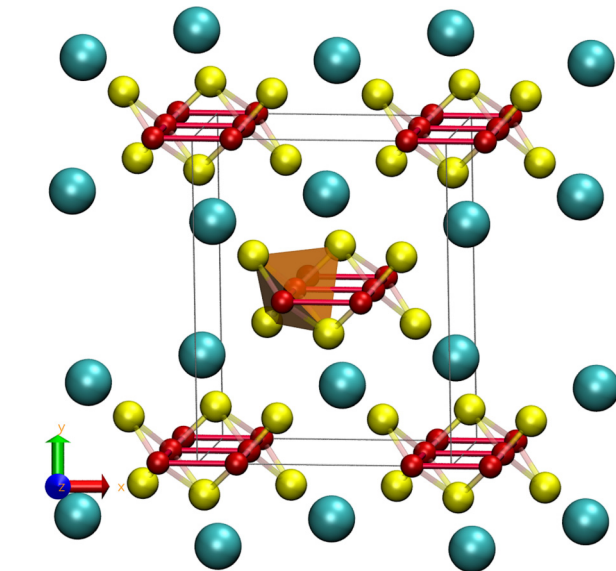


FIG. 1. Crystal structure of BaFe_2S_3 . Large, medium, and small spheres represent, respectively, Ba, S, and Fe atoms.

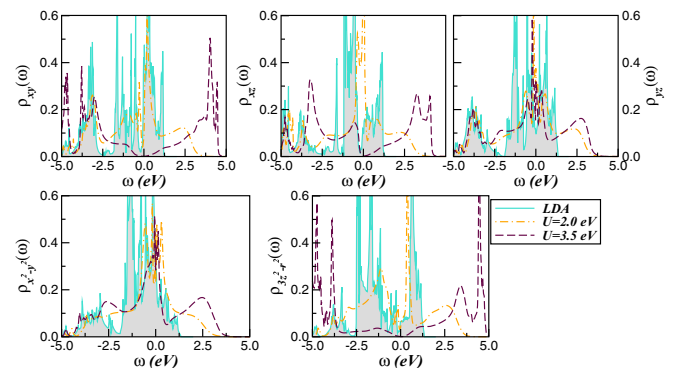


FIG. 2. Orbital-resolved LDA and LDA+DMFT density of states for the Fe 3d orbitals of paramagnetic BaFe_2S_3 , showing that all d bands in LDA span over the Fermi level ($E_F = \omega = 0$). This confirms that the electronic states relevant to orthorhombic Fe ladder compounds are Fe 3d states. The electronic reconstruction with increasing on-site Coulomb interaction and the orbital-selective Mott-Hubbard-insulating state with coexisting metallic and insulating bands at E_F for $U = 3.5$ eV are noteworthy.

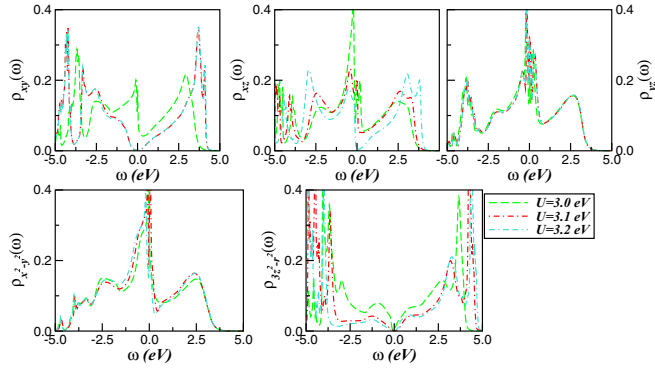


FIG. 3. Orbital-resolved LDA+DMFT DOS for the Fe 3d orbitals across the orbital-selective Mott transition of BaFe₂S₃. Large dynamical spectral weight transfer along with abrupt Mott-Hubbard localization is visible in the $3z^2 - r^2$, xz , xy LDA+DMFT spectral functions.

Hubbard bands and the size of the Mott gap are found to be orbital dependent, and this anisotropic electronic state could be probed in future transport and spectroscopy experiments of pressurized BaFe₂S₃ with partially quenched electron correlation effects. Taken together, our results in Fig. 2 confirm that itinerant and Mott-localized electrons coexist in the orbital-selective Mott state [23] of BaFe₂S₃.

For a more detailed analysis of dynamical MO electronic interaction effects near the orbital-selective phase transition of BaFe₂S₃ at moderate U values, we show the orbital-resolved spectral functions in Fig. 3. Electronic correlations lead to interesting modifications of the correlated spectra. Below the critical value for the orbital-selective insulating phase ($3.1 < U_{OS} < 3.2$ eV) the many-body spectra describe the coexistence of metallic (yz , x^2-y^2) and Mott-localized ($3z^2-r^2$, xz , xy) electronic states. The abrupt electron localization within the xy , xz orbitals and its coexistence with a quasicohherent x^2-y^2 , yz electronic fluid are noteworthy. Although near U_{OS} only gradual changes are seen in the spectral function of the x^2-y^2 , yz orbitals, the LDA+DMFT DOS of the $3z^2-r^2$, xz , and xy orbitals show large changes in spectral weight transfer (SWT) at the orbital-selective critical phase boundary in BaFe₂S₃: The concomitant formation of incipient (x^2-y^2 , yz) and narrow (xy , xz) Hubbard bands at energies above 2.0 eV in Fig. 3 is a one-particle fingerprint of the selective Mottness intrinsic to correlated multiorbital systems.

Like in other Fe chalcogenide systems [13,24], a first-order transition from an orbital-selective metallic state to a correlated Mott insulator (MI) with increasing U is obtained for BaFe₂S₃ (Fig. 4). At $U_{MT} = 4.5$ eV (and $J_H = 0.7$ eV) BaFe₂S₃ is a MO Mott-Hubbard insulator with an orbital-dependent band gap. Hubbard bands are visible, albeit to a different extent in all orbital-resolved spectral functions. As seen in Fig. 4, in this orbital-blocked Mott state the x^2-y^2 , $3z^2-r^2$ orbitals show stronger correlation effects with pronounced Hubbard bands, while the xy , xz , yz orbitals have less tendency towards local moment formation. The appearance of one-electron dispersion at low binding energies within the xy , xz , yz orbitals at U_{MT} , a fingerprint of distinct electronic

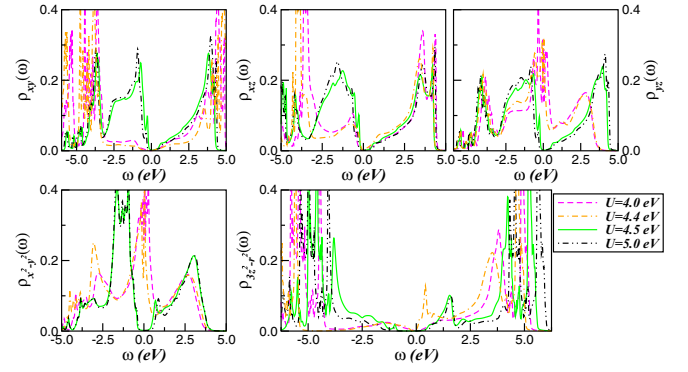


FIG. 4. Orbital-resolved LDA+DMFT DOS for the Fe 3d orbitals of BaFe₂S₃ across the second first-order, orbital-selective metal-insulator transition. Large dynamical spectral weight transfer along with Mott-Hubbard localization on all active orbitals is visible in the LDA+DMFT spectral functions at the critical phase boundary, $4.4 < U_{MT} < 4.5$ eV.

anisotropies within the Fe 3d shell [25], is also noteworthy. Large-scale changes in SWT thus characterizes the Mott transition in BaFe₂S₃. The orbital-resolved LDA+DMFT spectral functions are predicted to be highly reshaped by electron-electron interactions, and future photoemission spectroscopy and x-ray absorption spectroscopy experiments could verify this aspect. Relevant as well are our results at intermediate U values, i.e., below the all-electron Mott transition where metallic (yz , x^2-y^2) and localized (xy , xz , $3z^2-r^2$) electronic states coexist in nature. This remarkable situation is intrinsically tied to the orbital-selective Mott state [26], a state in which electronic localization is restricted to a particular subset of the active orbitals present in the problem. This state usually has strongly incoherent or Mott-localized electrons at a subset of orbitals coexisting with delocalized quasiparticle states at the less correlated orbitals [27].

What happens with the orbital-blocked phase in Fig. 4 upon doping? Even though data exist [18], the robustness of the Mott-insulating state and the instabilities of such a state to unconventional order and, in particular, to high- T_c superconductivity make this an important question to inquire about. In Fig. 5 we show the changes in the correlated electronic structure upon doping ($n_t \equiv 6 \pm 0.2$) the Mott insulator. Consistent with extant transport data [18], the localization-delocalization transition does not occur at small doping. What is the origin of this persistent localized behavior in doped BaFe₂S₃? In Mott-localized Fe chalcogenide systems [29] strong incoherent scattering between different carriers in orbital states split relative to each other due to the specific crystal field leads to two main effects: It leads, via static-Hartree contributions (from the static part of the orbital-dependent self-energies), to orbital-dependent shifts of the 3d bands relative to each other (see Fig. 6), and strong dynamical correlations due to sizable U and U' cause appreciable spectral weight transfer over large energy scales, from high to low energy, upon carrier doping. This second feature leads to a drastic modification of the spectral line shape of the $3z^2-r^2$ orbital, as shown in Fig. 5.

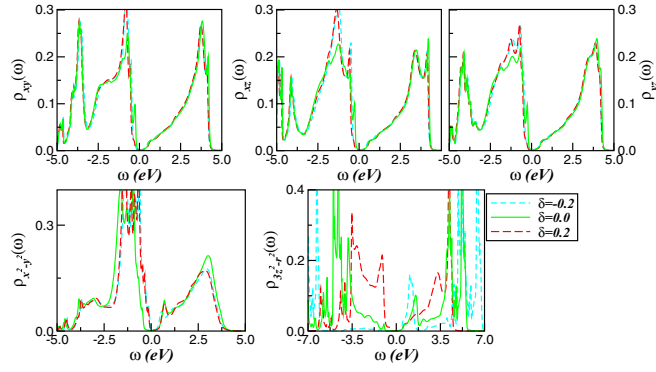


FIG. 5. Evolution of the LDA+DMFT ($U = 4.5$ eV, $J_H = 0.7$ eV) orbital-resolved spectral functions of BaFe_2S_3 for different values of the total band filling of the Fe 3d shell, $n_t = 6 \pm \delta$. Notice the modification of the $3z^2-r^2$ density of states due to large-scale dynamical spectral weight transfer with δ . While all orbitals remain Mott localized, incoherent valence-band states with x^2-y^2 orbital character almost touch E_F due to doping-induced selective-orbital reconstruction in BaFe_2S_3 .

Finally, motivated by the fact that the superconducting state observed in BaFe_2S_3 seems to emerge from a combination of bandwidth-control Mott transition [5,6] and doping of electrons into the iron network [18,19], in Fig. 7 we display the averaged occupation number of each orbital, $n_{a,\sigma} \equiv \langle n_{a,\sigma} \rangle$, computed using the LDA+DMFT orbital-resolved spectral functions of BaFe_2S_3 for $U = 4.5$ eV and $J_H = 0.7$ eV. Our results suggest strong orbital differentiation [30] between the (xy, xz, yz) and $(x^2-y^2, 3z^2-r^2)$ orbitals and enhanced charge carrier concentration within the $x^2-y^2, 3z^2-r^2$ electronic channel with increasing δ . This response is

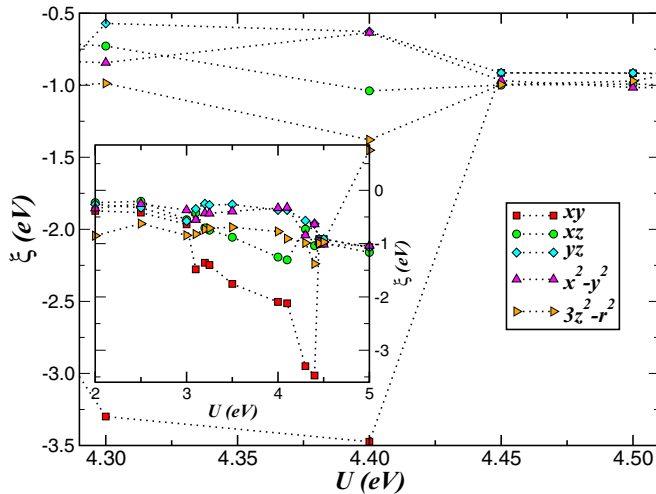


FIG. 6. U dependence of the gravity centers for the different LDA+DMFT bands, showing that xy is the ground-state orbital of metallic BaFe_2S_3 at the border of the pressure-induced Mott transition. Particularly interesting features are the changes in the renormalized orbital splittings due to large spectral weight transfer and the twofold (yz, x^2-y^2) orbital degeneracy at $U = 4.4$ eV. The latter suggests a two-orbital pairing mechanism near P_c similar to tetragonal Fe superconductors [28].

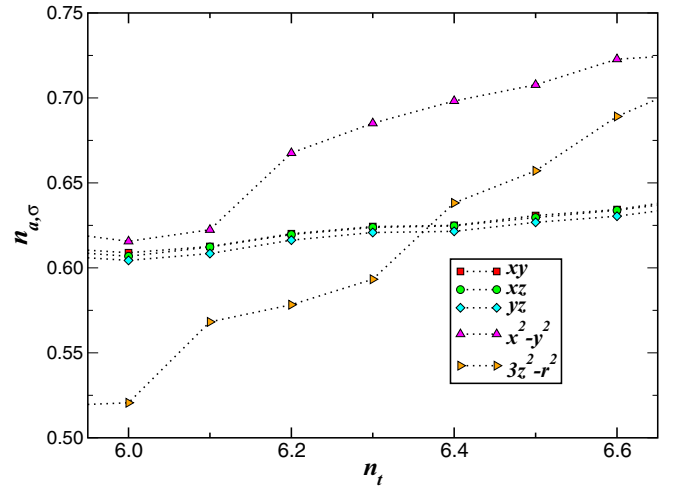


FIG. 7. Evolution of the Fe 3d orbital occupations $n_{a,\sigma}$ with increasing total Fe band filling ($n_t = 6 + \delta$) for $U = 4.5$ eV and $J_H = 0.7$ eV, showing enhanced charge carrier concentration within the $x^2-y^2, 3z^2-r^2$ orbitals upon electron doping of the parent compound.

characteristic of strongly correlated materials, where the orbital differentiation is linked to charge and orbital excitations [31] in the reconstructed electronic state at low temperatures. Here it is noteworthy that in this selective regime some of the active orbitals might get decoupled from the others [13], as seen in our results for the $3z^2-r^2$ orbital in Figs. 3 and 4. Interestingly, this marked behavior is consistent with model calculations for alkaline Fe selenides [13], for which the xy orbital of $\text{K}_{1-x}\text{Fe}_{2-y}\text{Se}_2$ was shown to be the most localized since it was close to half filling as in the case of the $3z^2-r^2$ orbital (see Fig. 7) for $n_t \approx 6.0$.

To visualize how the SWT evolves with electron doping more globally, in Fig. 8 we show our LDA+DMFT results for $\delta = 0.2, 0.4, 0.6$ and fixed $U = 4.5$ eV. As seen in Fig. 8 the UHB of the xy, xz, yz orbitals is robust and almost unaffected by doping. The lack of SWT from UHB to LHB suggests that intra- and interorbital charge fluctuations are

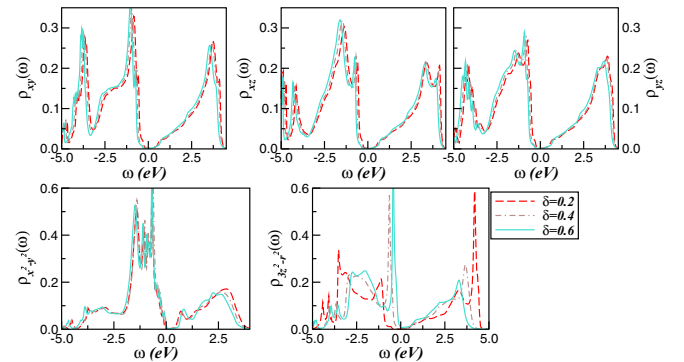


FIG. 8. Orbital-resolved LDA+DMFT ($U = 4.5$ eV) DOS for doped BaFe_2S_3 as a function of electron doping δ , showing the stability of the insulating phase. An important feature is the spectral weight transfer within the $3z^2-r^2$ orbital from the conduction to the valence band and the formation of a sharp peak below E_F with increasing δ .

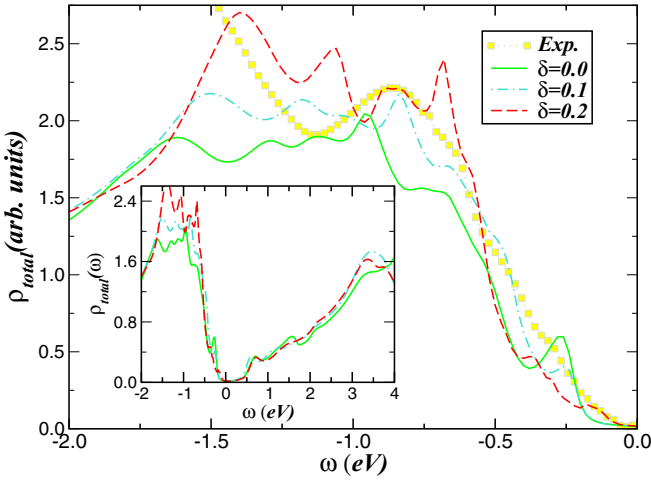


FIG. 9. Comparison between the LDA+DMFT result for pure and doped BaFe₂S₃ and UPS (squares) taken from Ref. [15]. Good quantitative theory-experiment agreement up to 1.5 eV binding energy is visible. In particular, the low-energy line shape and the bump feature around -0.85 eV are accurately resolved in the LDA+DMFT spectrum for electron-doped ($\delta = 0.2$) BaFe₂S₃. The inset shows the doping dependence of LDA+DMFT conduction band states which might be probed in future inverse photoemission spectroscopy experiments.

greatly suppressed, leading to an orbital decoupling phenomenon [27] near the all-electron Mott transition. On the other hand, the canonical SWT expected in electron-doped systems is more clearly visible within the x^2-y^2 orbital, where the spectral weight from the UHB is dynamically transferred to the valence-band states with increasing the total electron concentration of the Fe 3d shell. More interesting, however, is the electronic reconstruction found in the $3z^2-r^2$ orbital, where a sharp peak develops below E_F . This peak structure carries a large amount of the total spectral weight, which as a consequence enhances the electron occupancy in this orbital sector, as shown in Fig. 7. Thus, like for KFe_{1.6}Se₂ [32], strong scattering between the Mott-localized and bad-metallic states leads to complete suppression of the Fermi-liquid quasiparticles via the Anderson orthogonality catastrophe [33], leading to the emergence of an anomalous orbital-blocked state in DMFT, as shown in Fig. 8. Microscopically, infrared Fermi-liquid behavior is completely extinguished by strong scattering between the Mott-localized and incoherent low-energy components of the matrix spectral function due to sizable U' and is intimately tied to the orbital-selective insulator-metal transition [26] in the five-band Hubbard model we use. Remarkably, our results in Fig. 8 suggest that a novel pressure-induced confined metal [34] with mostly $3z^2-r^2$ orbital character could be observed in future experiments of overdoped BaFe₂S₃. Such an unusual phenomenon is predicted to have characteristics akin to the strange metal phase of overdoped cuprate superconductors [35].

In Fig. 9 we compare our LDA+DMFT result with extant ultraviolet photoemission spectra (UPS) spectroscopy data [15]. Since only the five 3d bands have been included in the LDA+DMFT treatment, we restrict ourselves to the energy window up to 1.5 eV binding energy, where only the five 3d

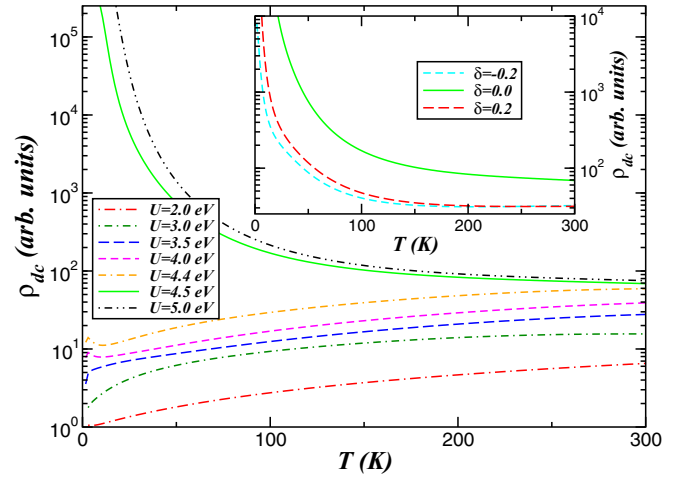


FIG. 10. T dependence of electrical resistivities for Mott-insulating and metallic BaFe₂S₃, showing qualitative good agreement with transport data [5,6]. Particularly interesting is the first-order metallic-to-semiconducting behavior for $4.4 \text{ eV} \leq U \leq 4.5 \text{ eV}$ consistent with normal-state data of Ref. [6]. The inset displays resistivity curves for $U_{\text{MT}} = 4.5 \text{ eV}$ and different total band fillings, showing that the Mott-insulating state is rather robust against small electron/hole doping.

bands dominate in LDA. Good quantitative agreement with the UPS result is obtained at low energies. In particular, the bump feature at -0.85 eV and the low-energy line shape are faithfully reproduced for electron-doped ($\delta = 0.2$) BaFe₂S₃. Taken together with our earlier results [29,32], this confirms that iron 3d states with sizable to strong MO correlations are a universal feature among Mott-localized Fe chalcogenides. Hence, our results for the total spectral function shown in the inset of Fig. 9 constitute a quantitative rationalization of basic one-particle responses of doped 123-spin-ladder Fe chalcogenides and serve as a basis to explore the electronic reconstruction upon doping [18,19] the parent compound.

In earlier works we showed that the MO-correlated nature of different Fe chalcogenide systems [24,29,32] can be semi-quantitatively understood using LDA+DMFT with sizable to strong d -band correlations. Here we extend this aspect to characterize the electrical properties of BaFe₂S₃ and its link to orbital-selective Mottness under pressure. Specifically, we study the T dependence of the dc resistivity and correlate it with the orbital-reconstruction scenario derived above. Given the correlated spectral functions $A_a(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im} G_a(\mathbf{k}, \omega)$, the (static) dc conductivity $\sigma_{\text{dc}}(T)$, computed within the DMFT formalism [36], can be expressed as $\sigma_{\text{dc}}(T) = \frac{\pi}{T} \sum_a \int d\epsilon \rho_a^{(0)}(\epsilon) \int d\omega A_a^2(\epsilon, \omega) f(\omega) [1 - f(\omega)]$. In this expression, $\rho_a^{(0)}(\epsilon)$ is the LDA DOS of the a bands (Fig. 2), and $f(\omega)$ is the Fermi function.

In Fig. 10 we display the T dependence of electrical resistivity [$\rho_{\text{dc}}(T) \equiv 1/\sigma_{\text{dc}}(T)$] with increasing U , computed using the LDA+DMFT orbital-resolved spectral functions for $\sigma_{\text{dc}}(T)$. Various interesting features immediately stand out. First, $\rho_{\text{dc}}(T \rightarrow 0)$ is large with clear insulatinglike behavior at $U \geq 4.5 \text{ eV}$, in accordance with the Mott-insulating description above. This Mott-localized behavior is first order suppressed in the orbital-selective metallic phase at

$U = 4.4$ eV. From this critical value to 4.0 eV the resistivity curves show a small insulating upturn below 10 K. While this resistivity upturn appears around 80 K in pressurized BaFe_3S_3 [5,6], the detailed T dependence in Fig. 10 resembles the one seen in experiment, showing a first-order insulator-metal transition [6] with increasing pressure from ambient to 10.9 GPa. With further reducing the effective U/W ratio or by increasing the one-particle bandwidth W with pressure [11], a crossover from a bad-metallic to a Fermi-liquid behavior (at $U = 2.0$ eV) is observed in $\rho_{dc}(T)$. Finally, in the inset of Fig. 10 we display the effect of doping the parent compound, showing reduced resistivity, albeit still insulating, consistent with experimental observations in electron- and hole-doped BaFe_3S_3 . A particularly interesting feature is the reduced resistivity for hole doping compared to electron doping, which nicely agrees with experimental observations [18].

To rationalize the overall bad-insulating behavior, which is characterized by a small resistivity upturn at low temperatures at the border of the bandwidth-controlled Mott transition [5,6] in pressurized BaFe_2S_3 , the selective localization of $3z^2-r^2$, xz , xy electronic states below U_{MT} in Fig. 4 implies that these orbitals act like an intrinsic source of electronic disorder in the system. This implies that an intrinsic disorder potential, arising from orbital-selective Mott physics, exists in BaFe_2S_3 at pressures close to P_c . Remarkably, such behavior results from strong scattering between effectively Mott localized and itinerant components of the full DMFT propagators. This is intimately linked to orbital-selective Mott-like physics within DMFT [26]. We note that such orbital-selective physics was also proposed earlier for hexagonal iron chalcogenides [37] as well as on phenomenological grounds for iron pnictide superconductors [38]. The qualitative agreement between our resistivity results close to P_c and that seen in experiments [6] suggests appreciable orbital reconstruction at low binding energies and persistent Mott localization within the $3z^2-r^2$ orbital sector, and future theoretical and experimental work to corroborate our prediction is called for. Here we emphasize that coexistence of insulating ($3z^2-r^2$, xz , xy) and metallic (yz , x^2-y^2) states at $3.2 < U_{OS} < 4.4$ eV is the key mechanistic step which allows us to understand the strange metallic

behavior observed at the pressure range between 11.6 and 13.4 GPa in BaFe_2S_3 [5,6]. Hence, it would be interesting to see whether this selective orbital mechanism with quasi-particle resonances coexisting with Mott-localized electronic states near E_F would be observable in angle-resolved photoemission spectroscopy of pressurized BaFe_2S_3 .

IV. CONCLUSION

In summary, we have used LDA+DMFT on a five-band Hubbard model to derive a correlation-induced orbital reconstruction in the spin-ladder compound BaFe_3S_3 . We have analyzed its Mott-insulating nature, revealing it as an effect of multiorbital dynamical correlations. Upon consideration of reduced correlations effects (due to broadening of the electronic bandwidth by pressure) [21] we observe two distinct first-order phase transitions. The first, which is relevant to experiments at moderate pressure conditions, is shown to be of the selective Mott type for which localized and itinerant electrons coexist in the nonmagnetically ordered normal state. Approaching the weak-coupling regime, we predict an abrupt transition from an orbital-selective Mott phase to a Fermi-liquid metal, which is accompanied by large changes in dynamical spectral weight transfer at high and low energies.

ACKNOWLEDGMENTS

L.C.'s work is supported by CNPq (Grant No. 304035/2017-3). S.L. acknowledges support from the UK Research Council for using work in the paper that was undertaken by a student under Project No. EP/M50631X/1 as well as the DFG for support under the priority project SPP1415 and for a personal Heisenberg grant. S.L. also thanks ARCCA Cardiff for computational resources. Via S.L.'s membership of the UK's HPC Materials Chemistry Consortium, which is funded by EPSRC (No. EP/L000202), this work made use of the facilities of ARCHER, the UK's National High-Performance Computing Service, which is funded by the Office of Science and Technology through EPSRC's High End Computing Programme.

-
- [1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, *J. Am. Chem. Soc.* **130**, 3296 (2008).
 - [2] D. C. Johnston, *Adv. Phys.* **59**, 803 (2010); G. R. Stewart, *Rev. Mod. Phys.* **83**, 1589 (2011); Q. Si, R. Yu, and E. Abrahams, *Nat. Rev. Mater.* **1**, 16017 (2016).
 - [3] M. Yi, Y. Zhang, Z.-X. Shen, and D. Lu, *Quantum Mater.* **2**, 57 (2017).
 - [4] K. Haule, J. H. Shim, and G. Kotliar, *Phys. Rev. Lett.* **100**, 226402 (2008); L. Craco, M. S. Laad, S. Leoni, and H. Rosner, *Phys. Rev. B* **78**, 134511 (2008).
 - [5] H. Takahashi, A. Sugimoto, Y. Nambu, T. Yamauchi, Y. Hirata, T. Kawakami, M. Avdeev, K. Matsubayashi, F. Du, C. Kawashima, H. Soeda, S. Nakano, Y. Uwatoko, Y. Ueda, T. J. Sato, and K. Ohgushi, *Nat. Mater.* **14**, 1008 (2015).
 - [6] T. Yamauchi, Y. Hirata, Y. Ueda, and K. Ohgushi, *Phys. Rev. Lett.* **115**, 246402 (2015).
 - [7] G. Baskaran, *J. Phys. Soc. Jpn.* **77**, 113713 (2008); R. Yu, J.-X. Zhu, and Q. Si, *Phys. Rev. Lett.* **106**, 186401 (2011).
 - [8] J. Ying, H. Lei, C. Petrovic, Y. Xiao, and V. V. Struzhkin, *Phys. Rev. B* **95**, 241109(R) (2017).
 - [9] Q. Si, *Nat. Phys.* **5**, 629 (2009); M. M. Qazilbash, J. J. Hamlin, R. E. Baumbach, L. Zhang, D. J. Singh, M. B. Maple, and D. N. Basov, *ibid.* **5**, 647 (2009).
 - [10] W. M. Reiff, I. E. Grey, A. Fan, Z. Eliezer, and H. Steinfink, *J. Solid State Chem.* **13**, 32 (1975); Z. S. Gönen, P. Fournier, V. Smolyaninova, R. Greene, F. M. Araujo-Moreira, and B. Eichhorn, *Chem. Mater.* **12**, 3331 (2000).
 - [11] M. Imada, A. Fujimori, and Y. Tokura, *Rev. Mod. Phys.* **70**, 1039 (1998).
 - [12] N. F. Mott, *Metal-Insulator Transitions* (Taylor and Francis, London, 1974).
 - [13] R. Yu and Q. Si, *Phys. Rev. Lett.* **110**, 146402 (2013).

- [14] G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, *Rev. Mod. Phys.* **78**, 865 (2006).
- [15] D. Ootsuki, N. L. Saini, F. Du, Y. Hirata, K. Ohgushi, Y. Ueda, and T. Mizokawa, *Phys. Rev. B* **91**, 014505 (2015).
- [16] L. Craco, M. S. Laad, and S. Leoni, *Europhys. Lett.* **91**, 27001 (2010); L. Craco, *Solid State Commun.* **253**, 14 (2017).
- [17] T. J. Liu, X. Ke, B. Qian, J. Hu, D. Fobes, E. K. Vehstedt, H. Pham, J. H. Yang, M. H. Fang, L. Spinu, P. Schiffer, Y. Liu, and Z. Q. Mao, *Phys. Rev. B* **80**, 174509 (2009); Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi, and Y. Takano, *J. Phys. Soc. Jpn.* **78**, 074712 (2009); M. H. Fang, J. H. Yang, F. F. Balakirev, Y. Kohama, J. Singleton, B. Qian, Z. Q. Mao, H. D. Wang, and H. Q. Yuan, *Phys. Rev. B* **81**, 020509 (2010).
- [18] Y. Hirata, S. Maki, J.-I. Yamaura, T. Yamauchi, and K. Ohgushi, *Phys. Rev. B* **92**, 205109 (2015).
- [19] Y. Zhang, L. Lin, J.-J. Zhang, E. Dagotto, and S. Dong, *Phys. Rev. B* **95**, 115154 (2017).
- [20] O. K. Andersen, *Phys. Rev. B* **12**, 3060 (1975); For the implementation of the linear muffin-tin orbital scheme see V. Antonov, B. Harmon, and A. Yaresko, *Electronic Structure and Magneto-optical Properties of Solids* (Kluwer, Dordrecht, 2004).
- [21] R. Arita, H. Ikeda, S. Sakai, and M.-T. Suzuki, *Phys. Rev. B* **92**, 054515 (2015).
- [22] L. Craco, *Phys. Rev. B* **77**, 125122 (2008).
- [23] M. Mourigal, S. Wu, M. B. Stone, J. R. Neilson, J. M. Caron, T. M. McQueen, and C. L. Broholm, *Phys. Rev. Lett.* **115**, 047401 (2015).
- [24] B. Freelon, Y. H. Liu, J.-L. Chen, L. Craco, M. S. Laad, S. Leoni, J. Chen, L. Tao, H. Wang, R. Flauca, Z. Yamani, M. Fang, C. Chang, J.-H. Guo, and Z. Hussain, *Phys. Rev. B* **92**, 155139 (2015); L. Craco and S. Leoni, *Sci. Rep.* **7**, 46439 (2017).
- [25] K. Takubo, Y. Yokoyama, H. Wadati, S. Iwasaki, T. Mizokawa, T. Boyko, R. Sutarto, F. He, K. Hashizume, S. Imaizumi, T. Aoyama, Y. Imai, and K. Ohgushi, *Phys. Rev. B* **96**, 115157 (2017).
- [26] A. Liebsch, *Phys. Rev. B* **70**, 165103 (2004); S. Biermann, L. de' Medici, and A. Georges, *Phys. Rev. Lett.* **95**, 206401 (2005); M. S. Laad, L. Craco, and E. Müller-Hartmann, *Phys. Rev. B* **73**, 045109 (2006); L. de' Medici, S. R. Hassan, M. Capone, and X. Dai, *Phys. Rev. Lett.* **102**, 126401 (2009).
- [27] A. Kostin, P. O. Sprau, A. Kreisel, Y. X. Chong, A. E. Böhmer, P. C. Canfield, P. J. Hirschfeld, B. M. Andersen, and J. C. Sámul Davis, *Nat. Mater.* **17**, 869 (2018), and references therein.
- [28] M. S. Laad and L. Craco, *Phys. Rev. Lett.* **103**, 017002 (2009).
- [29] L. Craco, M. S. Laad, and S. Leoni, *J. Phys.: Condens. Matter.* **26**, 145602 (2014).
- [30] Z. Wang, M. Schmidt, J. Fisher, V. Tsurkan, M. Greger, D. Vollhardt, A. Loidl, and J. Deisenhofer, *Nat. Commun.* **5**, 3202 (2014), and references therein.
- [31] N. D. Patel, A. Nocera, G. Alvarez, A. Moreo, S. Johnston, and E. Dagotto, *arXiv:1807.10419*.
- [32] L. Craco, M. S. Laad, and S. Leoni, *Phys. Rev. B* **84**, 224520 (2011).
- [33] P. W. Anderson, *The Theory of Superconductivity in the High- T_c Cuprates*, Princeton Series in Physics (Princeton University Press, Princeton, NJ, 1997).
- [34] Y. Ding, L. Yang, C.-C. Chen, H.-S. Kim, M. J. Han, W. Luo, Z. Feng, M. Upton, D. Casa, J. Kim, T. Gog, Z. Zeng, G. Cao, H.-K. Mao, and M. van Veenendaal, *Phys. Rev. Lett.* **116**, 216402 (2016).
- [35] P. W. Anderson, *Science* **256**, 1526 (1992).
- [36] K. Haule and G. Kotliar, *New J. Phys.* **11**, 025021 (2009).
- [37] L. Craco and S. Leoni, *Europhys. Lett.* **92**, 67003 (2010); L. Craco, J. L. B. Faria, and S. Leoni, *Mater. Res. Express* **4**, 036303 (2017).
- [38] A. Hackl and M. Vojta, *New J. Phys.* **11**, 055064 (2009).