Unconventional Mott Transition in $K_xFe_{2-y}Se_2$

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Whether the newly discovered $K_xFe_{2-y}Se_2$ systems are doped Mott or band insulators is key to how superconductivity emerges at lower temperature. With extant theoretical studies supporting conflicting scenarios, a more realistic approach is urgently called for. Here, we use LDA+DMFT to study this issue in $K_xFe_{2-y}Se_2$. We find that the undoped $KFe_{1.6}Se_2$ system is a new kind of Mott-Kondo insulator (MKI). Electron doping this MKI drives a Mott transition to an orbital-selective non-Fermi liquid metal. Good agreement with spectral and transport responses supports our view, implying that superconductivity arises from a doped Mott insulator, as in the high-$T_c$ cuprates.

Recent finding of high-$T_c$ superconductivity (HTSC) with $T_c \gtrsim 30$ K in $K_xFe_{2-y}Se_2$ [1] and $(K,Tl)Fe_{2}Se_2$ [2,3] is remarkable for the following reasons: (i) While its SC $T_c$ is comparable to that for the 1111- and 122-Fe pnictides (FePn), it occurs near antiferromagnet (AF) insulators: Fe composition is the tuning parameter for insulator-metal and SC instabilities. Evidence for Fe vacancy order [4] is interesting: its generic effect is to reduce LDA (local-density-approximation) bandwidths [5]. Theoretically, both Mott [6,7] and band [8] insulating states have been proposed as candidates. (ii) It intensifies the fundamental debate [9] on the degree of electronic correlations in Fe-based SC. Transport and optical data reveal insulating behavior (albeit with small gap) well above $T_N(\delta)$, $\delta$ being the electron doping. However, optical [10] and ARPES [11] studies also clearly show large-scale spectral weight transfer (SWT) (over an energy scale $O(2.0)$ eV) as a function of temperature ($T$) across the magnetic and SC instabilities, a fingerprint of Mottness. Both $\rho(T) \gtrsim T$ and bad metallicity above $T_c$ are features shared along with other non-Landau-Fermi-liquid (NFL) metals close to a Mott or selective-Mott instability [12]. ARPES [11] also seems to show a curious co-existence of Mott- and “band-insulating” spectral features in $K_xFe_{2-y}Se_2$. Large-scale SWT on a scale of $O(2.0)$ eV in response to a small ($k_BT \approx O(10)$ meV) perturbation, however betrays the “hidden” strong correlations. Its implications for SC are intriguing: are the Mott- or “band”-like subsets of the renormalised spectrum relevant for SC that emerges at lower $T_c$? (iii) Finding of large local moment value on Fe, $M_F \approx 3.3 \mu_B$, suggests strong electronic correlations. Even in the more itinerant FePn, relevance of a “dual” picture for the parent magnets is now increasingly recognised [13]: such a dual picture must be even more relevant for $K_xFe_{2-y}Se_2$.

These findings necessitate incorporation of reasonably strong multi-band electronic correlations. All five $d$-bands crossing the Fermi energy ($E_F$) must be kept at a “minimally realistic” level in order to satisfactorily resolve the doped Mott-versus-band insulator issue above. In multi-orbital (MO) systems, sizable correlations also drive new physical effects: they induce orbital selective (OS) bad-metallic states with no LFL coherence, naturally yielding bad metallic resistivity above $T_c$. An OS-Mott scenario also generically “wipes out” a subset of Fermi surface (FS) sheets from the renormalized, correlated band structure (Lifshitz transition). In turn, this can have far-reaching consequences for the symmetry of the SC pair function, $\Delta(k)$: details and presence or absence of gap nodes crucially depends on whether or not $\Delta(k)$ intersects the renormalized FS of such a metal. Ex tant ARPES data show only electron pockets in metallic $K_{0.8}Fe_{1.7}Se_2$ and anisotropic s-wave SC gap [14]. However, NMR $T_1^{-1}(T) \approx T^2$ for $T < \frac{1}{2}T_c$ remains puzzling in this context [13]. This unsettled state of affairs calls for detailed theoretical scrutiny: it must, in view of the fundamentally conflicting views discussed above, base itself on an satisfying description of the “normal” state.

Here, we use state-of-the-art LDA-plus-dynamical mean-field theory (LDA+DMFT) [15] to address these issues. In LDA+DMFT studies of FePn, their degree of correlatedness has been the bone of contention: consensus has fluctuated between weakly [17] to sizably correlated limits [18]. In $K_xFe_{2-y}Se_2$, a perusal of the resistivity data [11] for non-AF but SC samples reveals that these are proximate to a Mott transition: $\rho_{dc}(T)$ immediately above $T_c$ is very bad metallic and quickly crosses over to an insulator-like dependence. In these cases, AF order cannot be responsible for insulating behavior: rather, these data show that destroying AF order reveals underlying Mottness, where (i)-(iii) above can be rationalised naturally. Clearly, electronic correlations can only get stronger as one approaches the insulator, and AF and/or orbital order can naturally arise via spin-orbital superexchange. In this work, we confirm this hypothesis, showing that a strong correlation view achieves good semiquantitative accord with a range of data in both, insulating and metallic phases of $K_xFe_{2-y}Se_2$. In particular, we clarify the co-existing Mott- and “band” insulator-like features in ARPES in a qualitatively new Mott-Hubbard scenario. Armed with these strengths, we qualitatively discuss the constraints our view imposes on mechanisms of (unconventional) SC in $K_xFe_{2-y}Se_2$.

We start with the experimental structure of
KFe$_{1.6}$Se$_2$ [4]. LDA calculations were performed using the linear muffin-tin orbitals (LMTO) [19] scheme, in the atomic sphere approximation: Self-consistency is reached by performing calculations with 242 irreducible k-points. The radii of the atomic spheres were chosen as $r_{\text{Fe}}=2.6$ (Fe), $r_{\text{Se}}=4.25$ (K) and $r_{\text{Se}}=4.25$ (Se) a.u. in order to minimize their overlap. Within LDA, the one-electron part is $H_0 = \sum_{\mathbf{k},a,\sigma} \epsilon_a^{\mathbf{k}} c_{\mathbf{k},a,\sigma}^\dagger c_{\mathbf{k},a,\sigma}$, where $a = \pm x, \pm y, \pm z, 3\pm z^2 - \pm x^2, \pm x^2 - \pm y^2$ label the (diagonalized in orbital basis) five 3d bands. The MO Coulomb interactions (treated within DMFT) constitute the interaction term, which reads $H_{\text{int}} = U \sum_{i,a,b} n_{i\sigma} n_{ia\uparrow} + U' \sum_{i,a\neq b} n_{i\sigma} n_{ib\sigma} - J_H \sum_{i,a,b} S_{ia} \cdot S_{ib}$. We use the MO iterated perturbation theory as an impurity solver for DMFT. Though not numerically exact, it has a proven record of recovering correct LFL metallic behavior [20] and good semiquantitative agreement in a host of real systems.

Fig. 1 shows the LDA DOS for KFe$_{1.6}$Se$_2$, whereby a clear and sizable reduction (O(20\%)) of the average LDA bandwidth ($W_{\text{LDA}}$) relative to that for FeSe, induced by presence of Fe vacancy order, is seen. FeSe is already a bad metal close to a Mott insulator [21], and the significantly smaller $W_{\text{LDA}}$ for KFe$_{1.6}$Se$_2$ then naturally suggests emergence of a Mott insulator in the latter. Indeed, our results show a small but clear insulating gap in DMFT spectra. Several interesting features, especially germane to the above discussion, are now manifest: (i) the Mott gap is clearly orbital-dependent, i.e., intrinsically anisotropic. (ii) Examination of the orbital-resolved (imaginary parts) self-energies reveal a behavior hitherto unique to Fe-based systems. Namely, $\text{Im}\Sigma_{\alpha}(\omega)$ with $\alpha = xz, yz, xy, x^2-y^2$ clearly reveal their Mott insulating character, i.e, a pole at $\omega = E_F(=0)$, as shown in Fig. 2. On the other hand, $\text{Im}\Sigma_{\alpha}(\omega)$ for $\alpha = 3z^2-r^2$ simultaneously shows Kondo insulator features, i.e, $\text{Im}\Sigma_{\alpha}(\omega) = 0$ in the gap region. Thus, remarkably, KFe$_{1.6}$Se$_2$ shows co-existing Mott and Kondo insulating gaps, and we dub this a Mott-Kondo insulator (MKI). One might be tempted to try to link this to the band insulator state found in LDA, especially since a Kondo insulator is an analytically continued version of a band insulator (where sizable correlations do exist above the gap scale). If electron doping would result in a metal where only the $3d_{xy}$ band would be driven metallic, it would necessarily mandate an effective, doped Kondo insulator [22], rather than doped Mott-insulator modelling. However, in ARPES data for the doped metal, the renormalised FS comprises sheets having only $xz, yz$ orbital character, in accord with DMFT results (see below). Thus, the insulator-metal transition must now be viewed in terms of a doped Mott insulator, in accord with earlier model-based work [5,6]. We emphasise that no such insulating state is found for $U \lesssim 3.0$ eV (not shown) in absence of magnetic order.

Electron doping ($n = 6+\delta$, with $\delta > 0$) the MKI leads to a bad-metallic state. Consistent with our results in Fig. 2, we find that only $xz, yz$ orbitals show bad metallic behavior, characterised by absence of sharp LFL resonances at $E_F$, while Mott insulating behavior persists in the $xy, x^2-y^2, 3z^2-r^2$ channels. Thus, we find an OS metal, and strong scattering between the Mott-localised and metallic states leads to complete suppression of the LFL quasiparticles via the Anderson orthogonality catastrophe [23], leading to emergence of anomalously broad
a function of electron doping \( n = 6 + \delta \), clearly showing a low-energy pseudogap associated with bad-metallicity. Low-energy DMFT spectra (inset) in good agreement with ARPES results \[2, 11, 14\].

spectra in DMFT, see Fig. 3. Microscopically, infrared LFL behavior (narrow “Kondo” resonance in DMFT) is killed off by strong scattering between the Mott-localised quasi-itinerant components of the (DMFT) matrix-spectral function, due to sizable and quasi-itinerant components of the DMFT) matrix-spectral function, and is intimately tied with the OS insulator-metal transition in the five-band Hubbard model we use. Thus, our selective-Mott metal is a MO counterpart of the FL* metal [24].

If our proposal is to hold, a range of responses must find a consistent explication without additional assumptions: we now show this is indeed the case. (i) A direct comparison between DMFT spectra and (AR)PES data show very good accord: in addition to describing the overall PES lineshape very well, the DMFT data show very good accord: in addition to describing the overall PES lineshape very well, the DMFT spectra (inset of Fig. 3) also resolve two peaks in the infrared results \[2, 11, 14\].

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(ii) Transport properties in DMFT are directly computable from the full DMFT propagators, since vertex corrections entering the Bethe-Salpeter equation are small enough to be neglected in multi-band cases \[25\]. In Fig. 4 we show the dc resistivity for various \( n \). Clear (Mott) insulating behavior obtains for \( n = 6.0 \), as expected. With \( \delta > 0 \), a doping-dependent crossover from a “high”-\( T \) insulator to a low-\( T \) (lower inset in Fig. 4) bad metal obtains. Ob-
ual, inter-site and inter-orbital (in multi-band systems) two-particle interactions can generate ordered states directly from the bad metal. This is the philosophy used earlier \cite{28} for the 1111-FePn systems. As in earlier work, restricting ourselves to the \(xx, yz\) orbital sector, the effective pair-hopping term (second order in \(t_{ab}\)) is \(H^{(2)} \approx -1/2 \sum_{k,k',a,b} V_{ab}(k,k')c_{ka\sigma}^\dagger c_{k'a\sigma}^\dagger c_{kb\sigma}^\dagger c_{k'b\sigma} + h.c\). Decoupling \(H^{(2)}\) in the particle-hole and particle-particle channels gives \(H^{(2)}_{MF} = \sum_{k,a,b}(\Delta^{(1)}_{ab}(k)c_{ka\sigma}c_{kb\sigma} + h.c) + (\Delta^{(2)}_{ab}(k)c_{ka\uparrow}c_{kb\downarrow} + h.c),\) where \(\Delta^{(1)}_{ab}(k) = \langle \gamma(k)c_{ka\sigma}c_{kb\sigma} \rangle\) and \(\Delta^{(2)}_{ab}(k) = \langle \gamma(k)c_{ka\sigma}c_{kb\sigma} \rangle\) with \(\gamma(k) = \cos k_x + \cos k_y + \cos k_x \cos k_y\) for the frustrated case of Fe-based systems. These represent orbital nematic (with orbital order and lattice distortion) \cite{28} and inter-orbital pairing \cite{28} instabilities. Extending DMFT to study both these orders in \(K_2Fe_2-ySe_2\) is more problematic, however: the large moment, \(\mu_{Fe} = 3.3\mu_B\), the block-spin moment \(M \approx 11\mu_B\), and the block spin-AF order \cite{20} suggests that both, possible orbital order \cite{31} with lattice distortion and subsequent AF, as well as SC instabilities must involve coupling between four-site plaquettes, beyond what our \(H^{(2)}_{MF}\) would give. Since the Mott transition already occurs at high \(T\), a way to proceed might involve using the present DMFT results as a template for deriving an appropriate low-energy, plaquette-centered model using the active \(xx, yz\) orbital states to address these issues as recognised by Baskaran in a different approach \cite{32}. This is currently underway, and will be reported in future.

In conclusion, using LDA+DMFT for a minimally realistic five-band Hubbard model, we resolve the issue of a doped Mott- vs band insulator physics in \(K_2Fe_{2-y}Se_2\) systems in favor of the former, Mott view. Good quantitative accord with key spectral and transport data in a sizably correlated picture confirms this view, and strongly suggests close underlying similarities (in spite of very different chemistry) between SC emerging here from a doped multi-orbital Mott-Kondo-insulating state with \(d\)-wave SC in doped high-\(T_c\) cuprates.

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