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Normal-state correlated electronic structure of tetragonal FeSe superconductor

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Abstract. Tetragonal FeSe, a prototype iron-chalcogenide superconductor, shows signatures of a strange incoherent normal state. Motivated thereby, we use LDA+DMFT to show how multi-band correlations generate a low-energy pseudogap in the normal state, giving an incoherent metal in good semi-quantitative agreement with observations. Anomalous responses in the normal state, including orbital-dependent effective mass enhancement and photoemission lineshape, are consistently understood.

1. Introduction

High temperature superconductivity in the Iron pnictides and chalcogenides compounds [1] is the latest surprise among a host of correlated electron materials. While unconventional superconductivity sets in close to the border of a frustration-induced [2] striped-spin-density-wave state with doping in the so-called 1111-pnictides, no magnetic long range order is seen in the tetragonal phase of Iron Selenide (FeSe) [3] and FeSe$_{1-x}$Te$_x$ [4], labelled 11 systems, for small $x$ in ambient conditions. Undoped FeSe exhibits superconductivity with $T_c = 9$ K: upon tuning the carrier concentration of single-layer films $T_c$ rises to 65 K [5]. Superconductivity is sensitive to stoichiometry - minute non-stoichiometry in Fe$_{1+y}$Se destroys the superconducting state [6]. Unconventional superconductivity at $T_c = 34$ K is even observed in the high pressure orthorhombic structure in FeSe [7] in contrast to the 1111-pnictides, where it is stable in the tetragonal structure. Interestingly, a two-step increase in $T_c$ as a function of pressure (with a large $dT_c/dP$ beyond $P_{c1} = 1.5$ GPa) is observed [8]. In contrast, superconductivity in FeSe is suppressed under tensile strain [9]. Moreover, extant experiments for the normal state show electron correlation fingerprints. Photoemission (PES) experiments [10, 11] show evidence of an incoherent, pseudogapped metallic state [10] in FeSe, instead of a narrow Landau quasiparticle peak at the Fermi level, $E_F$. Ab initio band structure calculations [12] compare poorly with PES data, as is checked by direct comparison (see below). In addition, the ultrahigh-resolution PES spectra show a low energy kink at $\approx 8$ meV [11]. Finally, an ARPES [13] study shows appreciable, orbital-dependent effective mass enhancement (16 – 21) in the normal state of FeSe$_{0.42}$Te$_{0.58}$, directly testifying sizable correlations in this system. As in 1111-compounds [14], the kink in PES sharpens with cooling, and evolves smoothly across $T_c$. Depending upon $x$, superconductivity in Fe(Se$_{1-x}$Te$_x$) either arises from an insulator-like normal state, or from a bad metal with $\rho_{dc}(T) \propto T$ [15]. Finally, a minute amount of alloying by Cu drives FeSe
to a Mott-Anderson insulator [16]. Thus, FeSe is close to a metal-insulator transition, i.e, to Mottness [17]. Needless to say, a proper microscopic understanding of the coupled orbital-spin [18] correlations manifesting in such anomalous behavior in Fe(Se,Te) systems is a basic prerequisite for understanding how superconductivity emerges from such a normal state.

In this work we undertake a systematic local-density approximation plus dynamical mean-field theory (LDA+DMFT) [19] study of tetragonal FeSe. Sizable electronic correlations are shown to be necessary for gaining proper insight into the anomalous normal state responses in this system. Good semi-quantitative agreement with PES [10] supports our description.

2. Results and discussion

In our numerical simulation we start with the tetragonal (space group: $P4/nmm$) structure of FeSe with lattice parameters derived by Hsu et al. [20]. One-electron band structure calculations based on local-density-approximation (LDA) were performed for FeSe using the linear muffin-tin orbitals (LMTO) [21] scheme. Our LDA results for the total density of states (DOS) is shown in Fig. 1 (dotted line). Similar total DOS were also obtained by other groups [12], showing that the electronic states relevant to Fe-superconductors are Fe-$d$-band states. As found in previous calculations, the Fe-$d$ bands hybridize with Se-$p$ bands around -3.8 eV, giving rise to a small, separated band of $d$ character below 3 eV binding energy. Interestingly, the resulting “gap” at high energy is not seen in PES experiments [10, 11], which show only a broad continuum in this energy range. As discussed below, this discrepancy is resolved by dynamical spectral weight transfer (SWT) which originates from sizable electronic correlations in FeSe.

**Figure 1.** Comparison between the LDA (dotted) and LDA+DMFT (solid, dot-dashed and long-dashed) density-of-states (DOS) for the Fe $d$-orbitals in FeSe. Large-scale transfer of spectral weight from low energy to the Hubbard bands with increasing $U$ is visible. Also clear is the destruction of the low-energy Fermi liquid (FL) quasiparticle peak at $U = 4$ eV.
Figure 2. Comparison between the LDA+DMFT result for FeSe and angle-integrated photoemission (PES, triangles) [10]. Good semiquantitative agreement is seen for $n = 5.8$. In particular, the low-energy energy spectrum (up to 0.1 eV binding energy) and the peak at $-0.17$ eV in PES is resolved in the DMFT spectrum with $U = 4.0$ eV and $J_H = 0.7$ eV. (The inset shows the total LDA+DMFT spectral functions. LDA result is shown for comparison.)

Though LDA provides reliable structural information on a one-electron level, it generically fails to capture the ubiquitous dynamical correlations in $d$-band compounds, and so cannot access normal state incoherence in $d$-band systems. Combining LDA with dynamical-mean-field-theory (DMFT) is the state-of-the-art prescription for remedying this deficiency [19]. Within LDA, the one-electron part for tetragonal FeSe is $H_0 = \sum_{k,a,\sigma} \epsilon_a(k)c_{k,a,\sigma}^\dagger c_{k,a,\sigma}$, where $a = x^2 - y^2, 3z^2 - r^2, xz, yz, xy$ label the diagonalized, five $d$ bands. In light of the correlation signatures cited above full, multi-orbital (MO) Coulomb interactions must be included. These constitute the interaction term, which reads $H_{\text{int}} = U \sum_{i,a} n_{ia\uparrow} n_{ia\downarrow} + U' \sum_{i,a\neq b} n_{ia\uparrow} n_{ib\uparrow} - J_H \sum_{i,a\neq b} S_{ia} S_{ib}$. To pinpoint the relevance of MO electronic interactions in the system, we present LDA+DMFT results for $U = 2, 3, 4$ eV, $U' = U - 2J_H$, and fixed $J_H = 0.7$ eV. In this sense, our study is not \textit{ab initio}, but should be looked upon as a realistic correlated model and numerical simulation for FeSe. To solve the MO-DMFT equations, we use the MO iterated-perturbation-theory as an impurity solver [22].

Fig. 1 shows how LDA+DMFT modifies the LDA band structure. MO dynamical correlations arising from $U, U'$ and $J_H$ lead to spectral weight redistribution over large energy scales and the formation of lower- (LHB) and upper-Hubbard (UHB) bands. As seen, the UHB at 2.4 eV for $U = 2$ eV (and, $U' = 0.6$ eV) moves to higher energies with increasing $U$. The LHB is not clearly resolved for $U \leq 2$ eV. Indeed, we observe a relatively sharp and quasi-coherent
Figure 3. Orbital-resolved LDA (dotted) and LDA+DMFT (with $U = 4.0$ eV, $U' = 2.6$ eV and $J_H = 0.7$ eV) DOS for the Fe $d$-orbitals in FeSe for three doping values. Large-scale dynamical spectral weight transfer occurring hand-in-hand with orbital selective incoherence is visible.

low-energy peak, with a prominent shoulder feature instead of the LHB at $\omega \simeq -1.0$ eV. Similar features are visible in other results [23] for similar $U$ values. Correlation effects, however, become more visible at $U \geq 3$ eV. In contrast to the $U = 2$ eV result, a LHB at 2.8 eV binding energy is clearly resolved with $U = 3$ eV. With increasing $U$, the LHB is shifted toward energies where the Se-$p$ bands occur in the LDA: this superposition of the $pd$-band and LHB for $U = 4$ eV makes difficult to observe the LHB experimentally. Fig. 1 also shows that the DOS at $E_F$ is pinned to its LDA value for $U \leq 3$ eV. This is the expected behavior for a Fermi liquid (FL) metal. With increasing $U$, however, our LDA+DMFT results show drastic modification of the spectral functions near $E_F$. Revealingly, in addition to large-scale SWT, we find that the FL-like pinning of the LDA+DMFT DOS to its LDA value, found for small $U$, is lost for $U = 4$ eV. Instead, the metallic state shows a clear pseudogap at $E_F$, with no Landau FL quasiparticles.

In Fig. 2, we compare our $U = 4$ eV (and, $U' = 2.6$ eV) results with PES for doped FeSe$_{1-x}$ [10]. Good semiquantitative agreement with experiment is visible for $n = 5.8$, where $n$ is the total band filling of the iron $d$ shell. In particular, the broad peak at $\approx -0.17$ eV as well as the detailed form of the lineshape in PES is well reproduced by LDA+DMFT results for the hole doped case. This may suggest that the experiment could have been done on a tetragonal sample with small Selenium excess (we recall that exact stoichiometry is a sensitive issue in the FeSeTe alloys) [24]. For comparison, the computed LDA+DMFT spectra for the undoped ($n = 6.0$) and electron doped ($n = 6.1$) cases show progressively more disagreement with PES at low energies. However, the overall lineshapes, along with the peak around $-0.2$ eV and the low-energy pseudogap remain robust features in the DMFT calculation. In contrast to this, the correlated spectral functions close to $E_F$ are insensitive to small changes in the electron (hole) concentration: we predict that combined PES/XAS on doped samples might show this in future.

We now focus on orbital resolved spectral functions of FeSe. Clear orbital-selective (OS)
Figure 4. Orbital-resolved LDA+DMFT self-energies for electron-doped FeSe. Upper panel: Real parts showing a low-energy kink feature, at about 15 meV below $E_F$, in $\text{Re}\Sigma_a(\omega)$ with $a = xy, xz, yz, x^2 - y^2$. In the inset, we show the computed orbital-dependent effective masses. These are sizably enhanced relative to LDA values, in good quantitative accord with ARPES data [13]. Lower panel: The corresponding imaginary parts, showing clear sub-linear ($xy, xz, yz, x^2 - y^2$) and almost quadratic ($3z^2 - r^2$) $\omega$-dependence for $\omega \leq E_F$.

...incoherence is visible in Fig. 3: a low-energy pseudogap is visible in the $xz, yz, x^2 - y^2$ DOS, and only the $xy, 3z^2 - r^2$ DOS show very narrow FL-like resonances at $E_F$. Examination of the self-energies in Fig. 4 shows that, for $n = 5.8$, only $\text{Im}\Sigma_{3z^2-r^2}(\omega) \simeq -a\omega^2$ for $\omega < E_F(= 0)$. Using the Kramers-Krönig relation, it follows that the Landau FL quasiparticle residue, $Z$ vanishes near-identically for the $xz, yz, x^2 - y^2$ band carriers [from $\text{Re}\Sigma(E_F)$], direct numerical evaluation gives $Z_{xz,yz} = 0.046, Z_{x^2-y^2} = 0.059$. This translates into an effective mass enhancement $[m^* / m \equiv \frac{1}{Z} = 1 - \frac{d}{d\omega} \text{Re}\Sigma(\omega)|_{\omega=0}]$ of 21.5 for the $xz, yz$ carriers and 17.0 for the $x^2 - y^2$ carriers, as shown in the inset of Fig. 4. This is in good accord with values estimated by an ARPES study on FeSe$_{0.42}$Te$_{0.58}$ superconductor [13], confirming the hypothesis about electronic correlations in FeSe made in that work. In our LDA+DMFT, these orbital-selective mass enhancements point toward the relevance of sizable MO electronic correlations in FeSe. However, we also notice that $d\Sigma / d\omega$ has appreciable frequency dependence at low energy: for a Landau FL metal, this quantity should be constant. Our finding of a frequency dependence in $d\Sigma / d\omega$ is thus fully consistent with a pseudogapped, incoherent metallic state as found above.

Finally, we shall point out that recent studies seem to be converging toward an intermediate-to-strong correlation scenario for the 122-Fe arsenides and chalcogenides [25, 26] as well as the 11-Fe selenides [27]. Semiquantitative agreement with the details of the PES lineshape along with specific description of transport [17] lends further credence to our view, which places the FeSe(Fe) in the incoherent, bad-metallic regime of a sizably correlated MO Hubbard model. In
earlier LDA+DMFT studies for the 1111-Fe pnictides [14] and 122-selenides [26], we found an incoherent metal normal state similar in many respects to the one shown here. Our study thus shows that sizable $d$-band electronic correlations are generic to the Fe-based superconductive materials.

3. Conclusion

To conclude, based on a five-orbital LDA+DMFT study, we have shown that orbital-selective incoherence characterizes the normal metallic phase in tetragonal FeSe. Good semiquantitative agreement with photoemission spectra and rationalization of a variety of unusual observations in a single picture lend support for our proposal. Sizable multi-orbital correlations are shown to be necessary to derive this orbital-selective incoherent metal. Emergence of superconductivity at low $T$, along with extreme sensitivity of the ground state(s) to minute perturbations in FeSe$_{1-x}$Te$_x$ compounds should thus be considered as manifestations of the myriad possible instabilities of such an incoherent non-Fermi liquid metal in close proximity to Mottness.

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