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Normal-state correlated electronic structure of tetragonal FeSe superconductor

L. Craco[1], M.S. Laad[2], S. Leoni[3]

[1] Instituto de Física, Universidade Federal de Mato Grosso, 78060-900, Cuiabá, MT, Brazil
[2] Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany
[3] School of Chemistry, Cardiff University, Cardiff, CF10 3AT, UK

E-mail: lcraco@fisica.ufmt.br

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 ≈ 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

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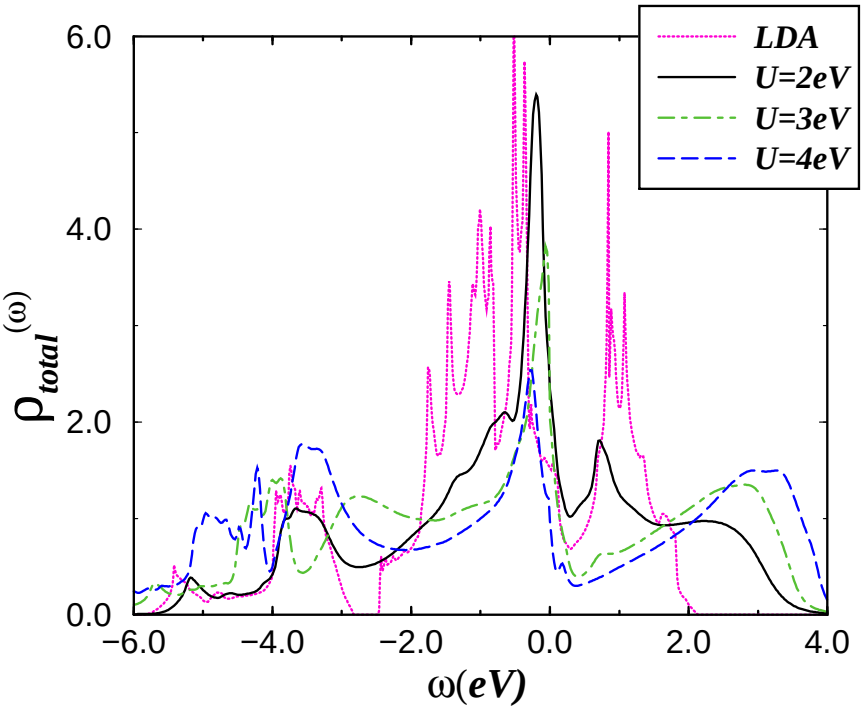

Figure 1. Comparison between the LDA (dotted) and LDA+DMFT (solid, dot-dashed and long-dashed) density-of-states (DOS) for the Fe d-orbitals in FeSe. Large-scale transer of spectral weight from low energy to the Hubbard bands with increasing U is visible. Also clear is the destruction of the low-energy Fermi liquid (FL) quasiparticle peak at $U = 4$ eV.

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.

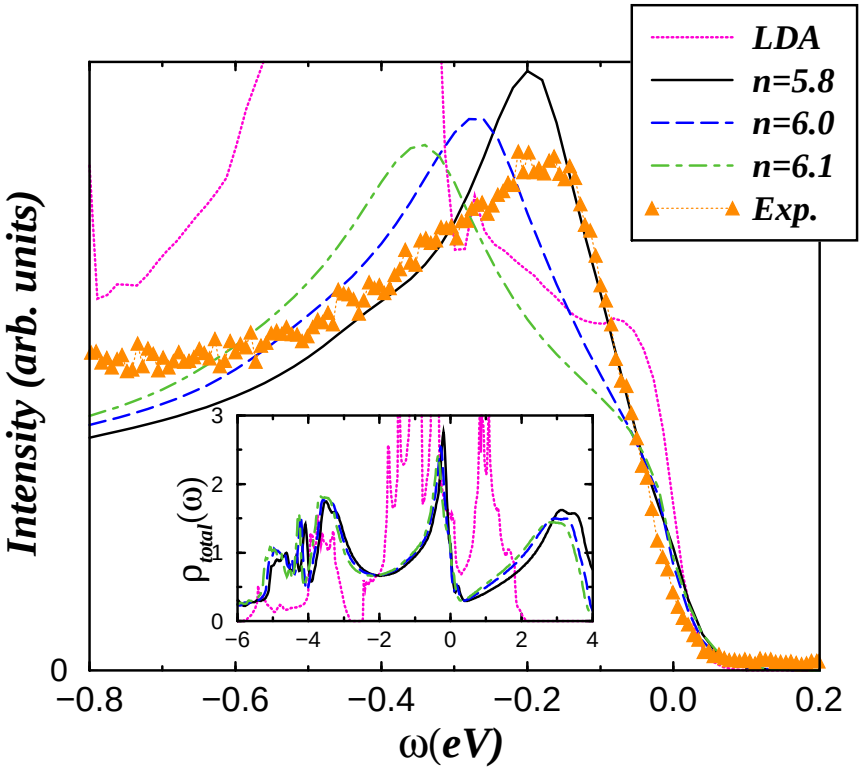

Figure 2. Comparison between the LDA+DMFT result for FeSe and angle-integrated photoemission (PES, triangles) [10]. Good semiquantitative agreement is seen for $n = 5.8$. In particular, the low-energy energy spectrum (up to 0.1 eV binding energy) and the peak at -0.17 eV in PES is resolved in the DMFT 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 $H_0 = \sum_{\mathbf{k},a,\sigma} \epsilon_a(\mathbf{k}) c^\dagger_{\mathbf{k},a,\sigma} c_{\mathbf{k},a,\sigma}$, 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 $H_{int} = U \sum_{i,a} n_{ia\uparrow} n_{ia\downarrow} + U' \sum_{i,a\neq b} n_{ia} n_{ib} - J_H \sum_{i,a\neq b} \mathbf{S}_{ia}.\mathbf{S}_{ib}$. 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 *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

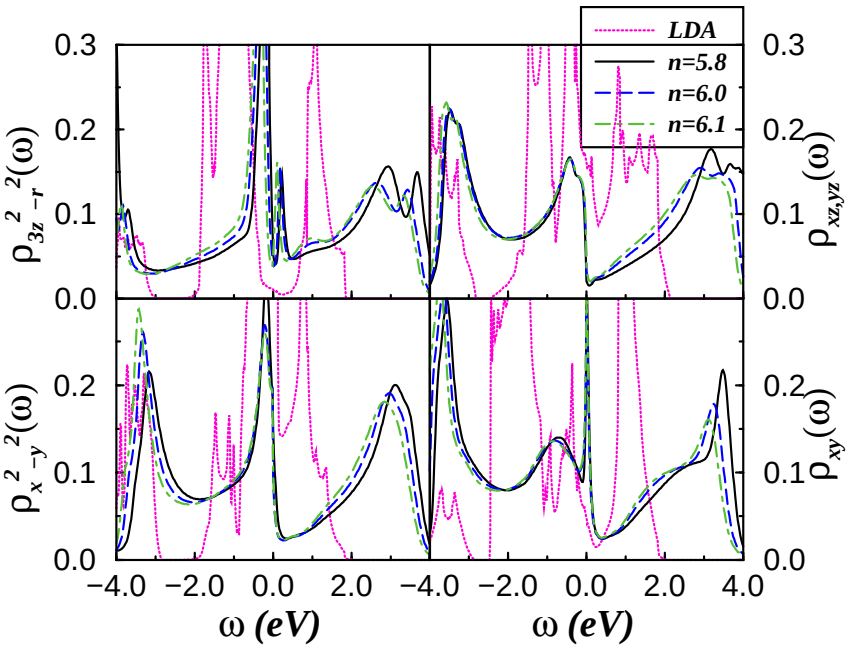

Figure 3. Orbital-resolved LDA (dotted) and LDA+DMFT (with $U = 4.0$ eV, $U' = 2.6$ eV and $J_H = 0.7$ eV) DOS for the Fe d-orbitals in FeSe for three doping values. Large-scale dynamical spectral weight transfer occouring hand-in-hand with orbital selective incoherence is visible.

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 ≈ -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)

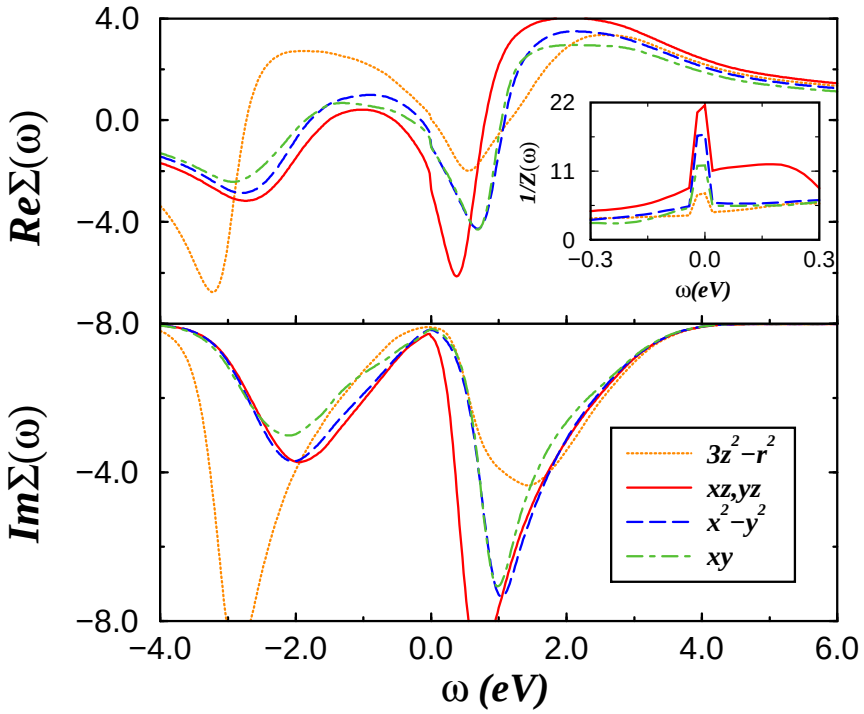

Figure 4. Orbital-resolved LDA+DMFT self-energies for electron-doped FeSe. Upper panel: Real parts showing a low-energy kink feature, at about 15 meV below E_F, in $\mathrm{Re}\Sigma_a(\omega)$ with $a = xy, xz, yz, x^2 - y^2$. In the inset, we show the computed orbital-dependent effective masses. These 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 $(3z^2 - r^2)$ in-ω 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 $\mathrm{Im}\Sigma_{3z^2-r^2}(\omega) \simeq -a\omega^2$ 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 $\mathrm{Re}\Sigma(E_F)$], direct numerical evaluation gives $Z_{xz,yz} = 0.046, Z_{x^2-y^2} = 0.059$). This translates into an effective mass enhancement $[\frac{m^*}{m} \equiv \frac{1}{Z} = 1 - \frac{d}{d\omega} Re\Sigma(\omega)|_{\omega=0}]$ 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(Te) 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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