Supplemental Material: Formation of Hubbard-like bands as a fingerprint of strong electron-electron interactions in FeSe

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Complete set of high-symmetry cuts at 56 eV

Here we show a complete set of high-symmetry cuts obtained at 56 eV, showing both the quasiparticle bands and the high energy spectral weight in both linear light polarizations. Here, and in the main text, we refer to "Linear Vertical" (LV) and "Linear Horizontal" (LH) polarizations of the incident photon beam, which correspond to s and p polarizations respectively at beamline I05-ARPES at Diamond Light Source (which has a horizontal scattering plane). The data obtained support the conclusion in the main text that the presence of incoherent spectral weight around 1-2.5 eV is a general feature of FeSe.

Fig. SM1(b,c) corresponds to the orientation of Fig. 1 in the main text, where below the d_{yz} quasiparticle band, a rather broad but dispersive feature corresponding to incoherent d_{yz} spectral weight is observed at $\sim$1-2 eV binding energy. Whereas, for the same cut in LH (p) polarisation in Fig. SM1(e,f), the spectrum is dominated by a band of bright intensity at a binding energy of $\sim$ 0.22 eV which can be ascribed to bands with d_{z^2} orbital character; some incoherent excitations of d_{z^2} character are also observed around 2 eV. The cuts through the M point in k,l) also reveal incoherent spectral weight and the formation of Hubbard bands as described in the main text. We also rotated the sample by 45° to access the X-Γ-X direction, where different matrix elements will apply, and the orbital character is also more mixed. However, once again features are observed in the range of 1-2.5 eV binding energy, which are notably much broader than the Se band observed below 3 eV. These cuts in different geometries combined with the photon energy-dependence in Fig. 1 and the further selected cuts in Fig. 2, demonstrate that the peak-dip-hump structure shown in Fig. 1(j) is a general feature of the spectral function of FeSe, and is a signature of strong local electronic correlations.

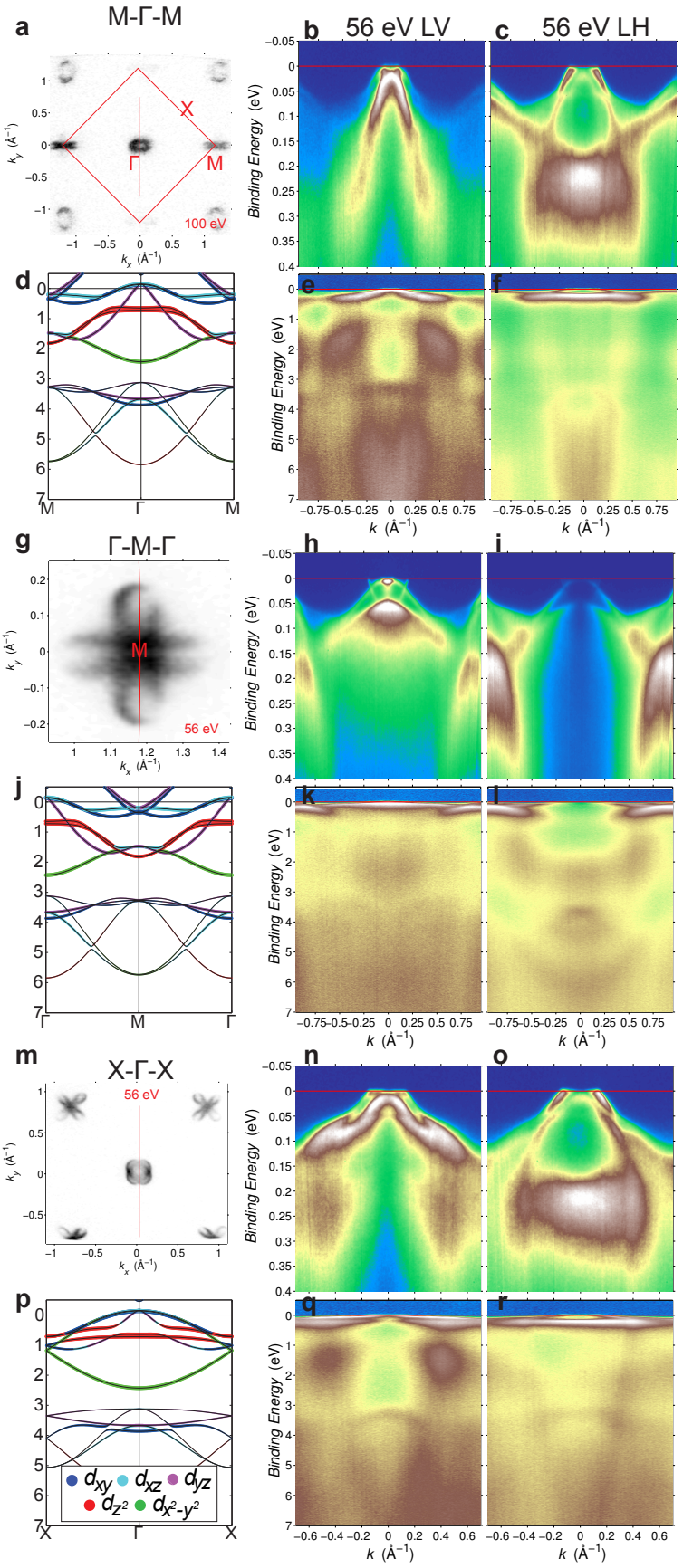

FIG. SM1. Comparison of ARPES data at 56 eV in LV (s) and LH (p) polarisations for a complete set of 3 different high-symmetry cuts in the Brillouin zone. a,g,m) Fermi surface maps, indicating the orientation of the high-symmetry cuts. d,j,p) DFT calculations for the comparable cuts. b,c,h,i,n,o) Low-energy ARPES cuts showing the quasiparticle bands, and e,f,k,l,q,r) corresponding measurements extending to high binding energies. Note that the photon energy of 56 eV, the high-symmetry points are close Z-A-R and not the $k_z = 0$ Γ-M-X labels used here for simplicity.

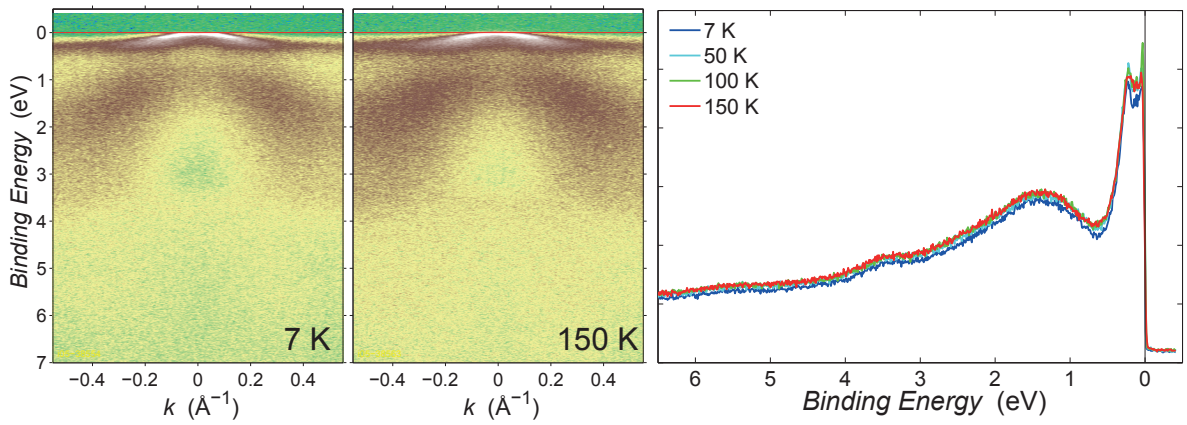

FIG. SM2. Temperature-dependence of high energy ARPES measurements. Data were obtained using and incident photon energy of 37 eV and LV polarisation, comparable to Fig. 1b) of the main text.

Temperature dependence of high energy spectra

Here we substantiate the claim in the main text that no significant temperature dependence of the high energy features is observed, at least up to 150 K. In Fig. SM2 we show high energy ARPES measurements at 7 K and 150 K. These data were obtained on a different sample (and with lower statistics) but the same measurement geometry compared to Fig. 1b) of the main text. Note that in this measurement geometry and section of the Brillouin zone probed, the broad peak from the Hubbard band is found at a lower binding energy compared to other cuts or the summation in Fig. 3b of the main text. No noticeable changes are observed, and the integrated spectral weight (integrating the total intensity in the plot windows) in the right panel shows no significant quantitative difference. Therefore, at least for this temperature range, and for these features of mainly d_{yz} orbital character, the high energy features are temperature-independent. Moreover these data also indicate that the onset of nematic order at T_s=90 K plays no role in the overall high energy features observed by ARPES.

Details of DFT+DMFT calculation

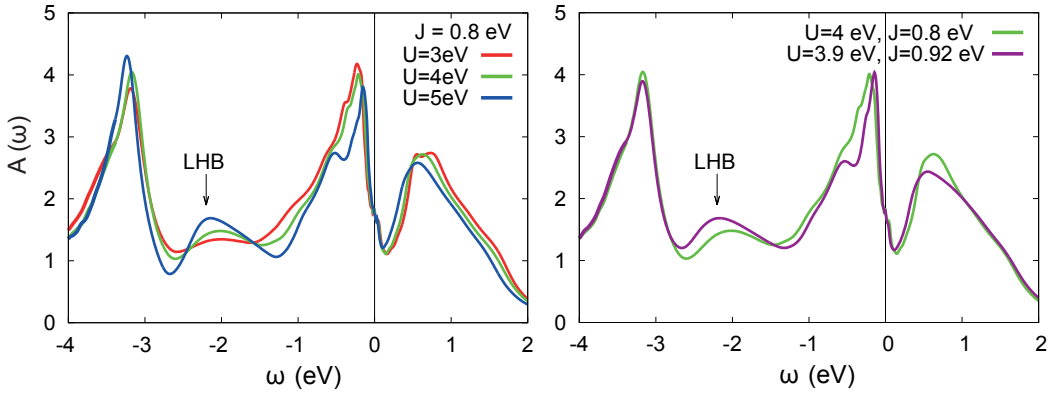

FIG. SM3. a) Integrated spectral weight $A(\omega)$ from DFT+DMFT calculations using different values of U, with J_H fixed at 0.8 eV. b) Comparison with the cRPA values of U,J_H recently determined by [1].

In the main text we focused a DFT+DMFT calculation performed using the parameters U = 4 eV, J_H = 0.8 eV, with particular emphasis on high energy features around 2 eV which we ascribed to a Hubbard-like band. Here we test the robustness of the conclusions against the variation of the interaction parameters. In Fig. SM3(a) we present calculations performed with different values of U, for a fixed value J_H = 0.8 eV. The feature associated with the lower Hubbard-like band gains intensity and is shifted to slightly higher binding energy as a function of increasing U. This behavior is reminiscent of the single-band Hubbard model, further supporting our description that this feature in a multi-band system is "Hubbard-like", as was also concluded in Ref. [2]. However in the Fe-based superconductors, the Hund's coupling J_H is at least as important as U. In Fig. SM3(b) we present the comparison of our usual calculation parameters with U = 3.9 eV, J_H = 0.92 eV recently found by ab-initio constrained RPA techniques [1]. Here it can be seen that the peak associated with the Hubbard-like band is rather sensitive to the value of J_H, as also found in other iron-based superconductors [3]; increasing J_H shifts the Hubbard-like feature to higher binding energy, as well as increasing the total fraction of the incoherent spectral weight. Nevertheless, we find that the Hubbard-like feature is robust over a reasonably wide range of interaction parameters.

TABLE SM1. Quasiparticle weights Z, mass enhancements m^*/m_{LDA} and scattering rates $-Im\Sigma(i0+)$ for the Fe 3d orbitals from the DFT+DMFT calculation ($U = 4$ eV, $J_H = 0.8$ eV).

Orbital	d_{z^2}	$d_{x^2-y^2}$	d_{xy}	$d_{xz/yz}$
Z	0.601	0.621	0.439	0.479
m^*/m_{LDA}	1.66	1.61	2.27	2.09
$-Im\Sigma(i0+)$ [meV]	1.21	1.62	4.49	3.11

In Table SM1 we summarise orbitally-resolved parameters of interest from the DMFT calculation. We average the d_{xz} and d_{yz} values as they differ only very slightly in our calculation in the orthorhombic phase. The effective mass renormalisations and quasiparticle weights are indications of the strength of correlations in the theoretical calculations. Here the calculated mass enhancements underestimate the experimental quasiparticle renormalisations estimated from heat capacity, quantum oscillations and ARPES [4]. Therefore our parameters U=4 eV, J_H=0.8 eV give a slightly less correlated state than is found experimentally. However, since for the purposes of this paper we are largely interested in only making qualitative comparison with of high energy features with ARPES, and also given that the experimental and DMFT Fermi surfaces will also disagree substantially no matter which interaction parameters are used, we do not attempt to use the experiment results to refine our interaction parameters.

[1] Ambroise van Roekeghem, Loïg Vaugier, Hong Jiang, and Silke Biermann, "Hubbard interactions in iron-based pnictides and chalcogenides: Slater parametrization, screening channels and frequency dependence," Phys. Rev. B **94**, 125147 (2016).

[2] Markus Aichhorn, Silke Biermann, Takashi Miyake, Antoine Georges, and Masatoshi Imada, "Theoretical evidence for strong correlations and incoherent metallic state in FeSe," Phys. Rev. B **82**, 064504 (2010).

[3] Steffen Backes, Harald O. Jeschke, and Roser Valentí, "Microscopic nature of correlations in multiorbital AFe2As2 (A=K,Rb,Cs): Hund's coupling versus Coulomb repulsion," Phys. Rev. B **92**, 195128 (2015).

[4] M. D. Watson, T. K. Kim, A. A. Haghighirad, N. R. Davies, A. McCollam, A. Narayanan, S. F. Blake, Y. L. Chen, S. Ghannadzadeh, A. J. Schofield, M. Hoesch, C. Meingast, T. Wolf, and A. I. Coldea, "Emergence of the nematic electronic state in FeSe," Phys. Rev. B **91**, 155106 (2015).