Supplemental material to the article

"On the origin of the shallow and "replica" bands in FeSe monolayer superconductors"

In this Supplement we provide computational details and crystallographic data for FeSe based systems under consideration. Also here we present orbital resolved LDA+DMFT calculated quasipartical bands for these materials.

1. Computation details

The LDA′ calculations [1, 2] of KFe_2Se_2 compound were performed using the Linearized Muffin-Tin Orbitals method (LMTO) [3]. The electroning structures of FeSe monolayer and FeSe monolayer on $SrTiO_3$ substrate were calculated within FP-LAPW method [4].

For the DMFT part of LDA+DMFT calculations we employed CT-QMC impurity solver [5, 6]. To define DMFT lattice problem for KFe_2Se_2 compound we used the full LDA Hamiltonian (i.e. without any orbitals downfolding or projecting) same as in Refs. [7, 8]. For isolated FeSe layer and FeSe/STO projection on Wannier functions was done for Fe-3d and Se-4p states (isolated FeSe layer) and for Fe-3d, Se-4p states and O-2p_y states from TiO_2 layer adjacent to $SrTiO_3$ (FeSe/STO). Standard wien2wannier interface [9] and wannier90 projecting technique [10] were applied to this end.

The DMFT(CT-QMC) computations were done at reciprocal temperature $\beta = 40$ ($\sim$290 K) with about 10^8 Monte-Carlo sweeps. Interaction parameters of Hubbard model were taken $U = 5.0\,\mathrm{eV}$, $J = 0.9\,\mathrm{eV}$ for isolated FeSe and FeSe/STO and $U = 3.75\,\mathrm{eV}$, $J = 0.56\,\mathrm{eV}$ for KFe_2Se_2 [11]. We employed the self-consistent fully-localized limit definition of the double-counting correction [2]. Thus computed values of Fe-3d occupancies and corresponding double-counting energies are $E_{dc} = 18.886$, $n_d = 5.79$ ($K_{0.76}Fe_{1.72}Se_2$), $E_{dc} = 31.63$, $n_d = 7.35$ (isolated FeSe layer), $E_{dc} = 30.77$, $n_d = 7.16$ (FeSe/STO).

The LDA+DMFT spectral function maps were obtained after analytic continuation of the local self-energy $\Sigma(\omega)$ from Matsubara frequencies to the real ones. To this end we have applied Pade approximant algorithm [12] and checked the results with the maximum entropy method [13] for Green's function $G(\tau)$.

2. Crystal structure

2.1. FeSe, FeSe/STO

The crystal structure of the bulk FeSe has tetragonal structure with the space group $P4/nmm$ and lattice parameters $a = 3.765$ Å, $c = 5.518$ Å. The experimentally obtained crystallographic positions are the following: Fe(2a) (0.0, 0.0, 0.0), Se(2c) (0.0, 0.5, z_{Se}), $z_{Se} = 0.2343$ [14]. For LDA calculation of isolated FeSe layer the slab technique was applied with these crystallographic parameters.

The FeSe/STO crystal structure was taken from LDA calculation with crystal structure relaxation [15]. FeSe monolayer was placed on three TiO_2-SrO layers to model the bulk $SrTiO_3$ substrate. The FeSe/STO slab crystal structure parameters are $a = 3.901$ Å, Ti-Se distance 3.13 Å, Fe-O distance 4.43 Å, distance between top (bottom) Se ion and the Fe ions plane is 1.41 Å (1.3 Å). Atomic positions are: Sr – $(0.5, 0.5, -1.95\,\mathrm{AA})$, O – $(0.5, 0, 0)$, $(0, 0, -1.95\,\mathrm{Å})$, Ti – $(0, 0, 0)$.

LDA+DMFT calculations of FeSe/STO were performed for doping level of 0.2 electrons per Fe ion.

2.2. KFe_2Se_2

The ideal KFe_2Se_2 compound has tetragonal structure with the space group $I4/mmm$ and lattice parameters $a = 3.9136$ Å and $c = 14.0367$ Å. The crystallographic positions are the following: K(2a) (0.0, 0.0, 0.0), Fe(4d) (0.0, 0.5, 0.25), Se(4e) (0.0, 0.5, z_{Se}) with $z_{Se} = 0.3539$ [16].

Chemical composition $K_{0.76}Fe_{1.72}Se_2$ corresponds to the total number of electrons 26.52 per unit cell (the stoichiometric compound has total number of electrons equal to 29.0). Total number of electrons 26.52 per unit cell corresponds to the doping level of 1.24 holes per Fe ion. This doping level was taken for LDA′+DMFT calculations. Position of corresponding Fermi level at about -0.4 eV is shown on left panel of Fig. 2 (main part of article).

3. LDA+DMFT orbital resolved quasiparticle bands

To show different Fe-3d orbitals contribution to LDA+DMFT spectral functions of FeSe based systems under consideration we present here the corresponding orbital resolved spectral function maps (Figs. 1, 2). In Fig. 1 it is clearly seen that the quasiparticle bands of isolated FeSe monolayer are well defined and have similar shape to the LDA bands except correlation narrowing by the same constant factor for all bands. The quasiparticle bands of FeSe/STO are more broad but still well defined. The main contribution to spectral function near the Fermi level belongs to Fe-3d_{xz}, Fe-3d_{yz} and Fe-3d_{xy} states both for the isolated FeSe layer and FeSe/STO. The spectral function of $K_{0.76}Fe_{1.72}Se_2$ is shown in Fig. 2. Here the bands are

strongly renormalized by correlations not only by the constant scaling factor, but also because of band shapes modifications in comparison to LDA bands. Since electronic correlations are quite strong for $K_{0.76}Fe_{1.72}Se_2$ and bands are rather broadened by lifetime effects we explicitly show here the spectral function maxima positions by crosses. Despite the difference of correlation effects in both systems one can conclude that quasiparticle bands structures around the Fermi level are rather similar.

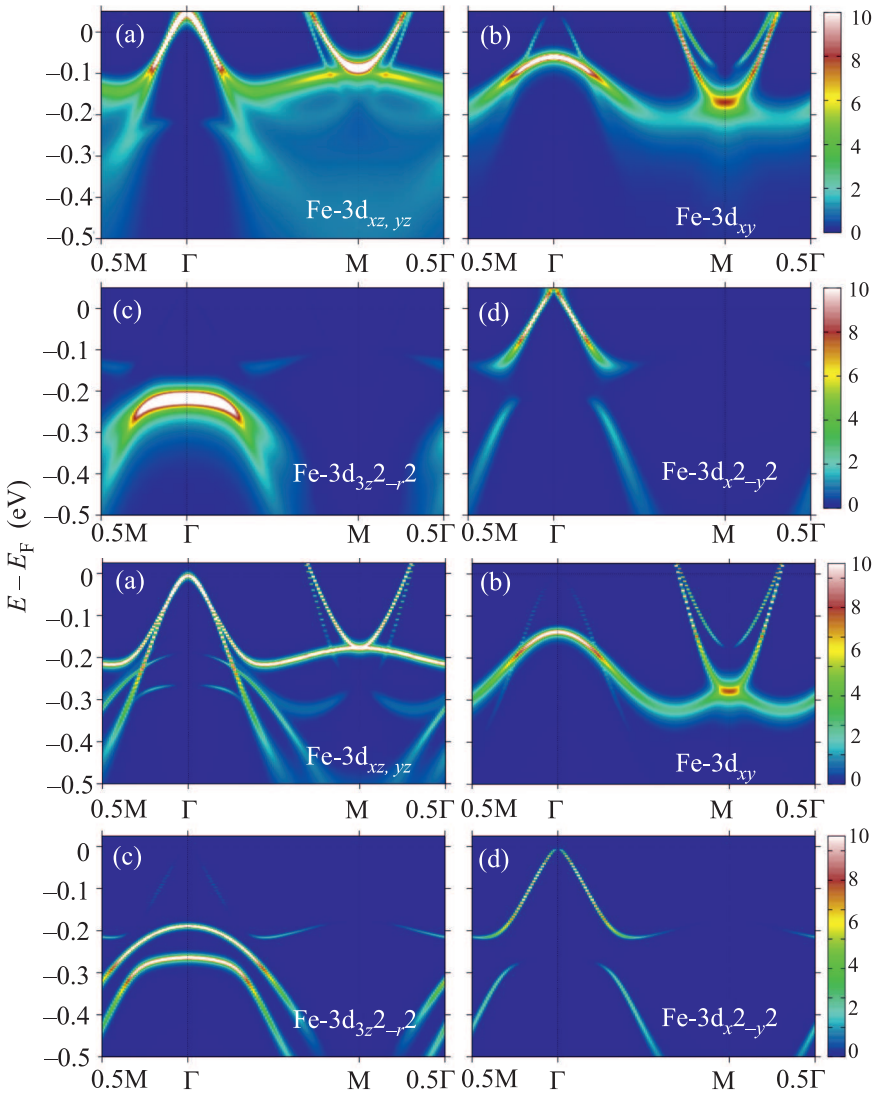

Figure 1: (Color online) LDA+DMFT spectral function map for different Fe-3d orbitals of FeSe monolayer on $SrTiO_3$ substrate (top) and isolated FeSe monolayer (bottom): (a) – Fe-3d$_{xz}$ and Fe-3d$_{yz}$, (b) – Fe-3d$_{xy}$, (c) – Fe-3d$_{3z^2-r^2}$, (d) – Fe-3d$_{x^2-y^2}$. Fermi level is at zero energy

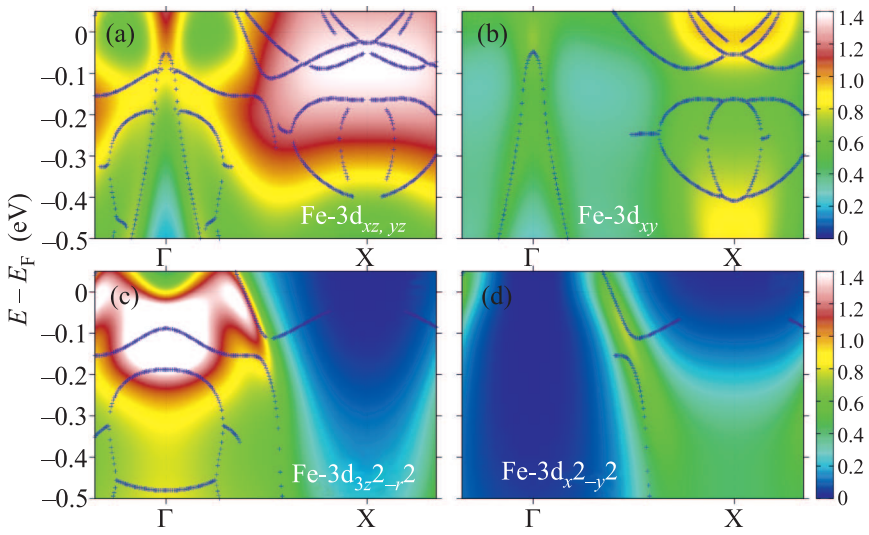

Figure 2: (Color online) LDA′+DMFT spectral function map for different Fe-3d orbitals of $K_{0.76}Fe_{1.72}Se_2$: (a) – Fe-3d$_{xz}$ and Fe-3d$_{yz}$, (b) – Fe-3d$_{xy}$, (c) – Fe-3d$_{3z^2-r^2}$, (d) – Fe-3d$_{x^2-y^2}$. Maxima of the spectral density are shown with crosses. Fermi level is zero energy

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