On the Spatial Locality of Electronic Correlations in LiFeAs

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We address the question of the degree of spatial non-locality of the self energy in the iron-based superconductors, a subject which is receiving considerable attention. Using LiFeAs as a prototypical example, we extract the self energy from angular-resolved photoemission spectroscopy (ARPES) data. We use two distinct electronic structure references: density functional theory in the local density approximation and linearized quasiparticle self consistent GW (LQSGW). We find that in the LQSGW reference, spatially local dynamical correlations provide a consistent description of the experimental data, and account for some surprising aspects of the data such as the substantial out of plane dispersion of the electron Fermi surface having dominant $xz/yz$ character. Hall effect and resistivity data are shown to be consistent with the proposed description.

Introduction. The origin of superconductivity in the iron pnictides and chalcogenides is an outstanding open problem in condensed matter physics. Two opposite points of view have been presented. In the first one, superconductivity originate{s} from {t}he exchange of spatially non-local antiferromagnetic (AFM) spin fluctuations and non-local correlations are also essential in the normal state. The second one posits a more local pairing due to the Hund’s coupling, which in turns requires a rather local picture of the normal state.

Answering this question requires a proper understanding of the degree of spatial locality of electronic correlations in the normal state. This has been addressed previously by a comparison of theoretical calculations to experiments. Some results favor the local picture, while others support the non-local view.

Here, we take a different approach and address this question by a direct examination of experimental data from angle-resolved photoemission spectroscopy (ARPES). We consider LiFeAs, a prototypical iron-based superconductor which is free from magnetic and nematic instabilities and which has been intensively studied for more than a decade. We focus on the electronic self-energy, which quantifies the electronic correlation effect on emergent quasiparticles. Following a similar procedure to that successfully employed for Sr$_2$RuO$_4$, we use the experimentally measured quasiparticle dispersions for the different Fermi surface (FS) sheets, determine the self-energy and assess its degree of spatial locality.

Our results offer a solution to the local vs. non-local conundrum. We find that the electronic self-energy can be separated, to a good approximation, into a non-local part which is frequency independent, and a dynamical (frequency-dependent) part which is spatially local to a good approximation. The non-local part can be incorporated in the reference Hamiltonian with respect to which the dynamical self-energy is defined, and we show that the quasiparticle GW approximation provides a good starting point to that effect. These findings are in line with previous work by Tomczak et al. but we emphasize that our conclusions are established directly from experimental observations.

These findings also rationalize the previous successes of dynamical mean field theory (DMFT) at describing the physics of iron-based superconductors. In combination with electronic structure methods, DMFT has put forward many early predictions for these compounds, such as a large mass enhancement which was later observed in optics as well as several materials trends including the evolution of the correlation strength and the level of orbital differentiation across multiple families of iron pnictides and chalcogenides. With hindsight, our work, as well as Refs. 21, 27, emphasizes GW+DMFT as a method of choice for investigating this family of materials.

Method. We start by describing the procedure that we use to determine the electronic self-energy and assess its degree of spatial locality, while making a relatively small number of theoretical assumptions. Ignoring photoemission matrix elements, extrinsic and surface effects, we relate the measured photoemission spectra to the spectral function associated with the one particle Green’s function:

$$G(k,\omega) = [\omega - H(k) - \Sigma(k,\omega)]^{-1}_{\sigma}\sigma',$$

(1)
In this expression, $H(k)$ is a reference Hamiltonian matrix, written in a localised basis of orbitals $m, \sigma$ ($\sigma$ is the spin index), $\omega$ is the frequency, and $k$ is the wavevector in the Brillouin zone. $\Sigma(k, \omega)$ is the self-energy matrix for the given reference Hamiltonian $H(k)$. The chemical potential is included in $H(k)$.

We consider two different choices for the reference Hamiltonian $H(k)$. The first is the Kohn-Sham Hamiltonian obtained from density-functional theory in the local density approximation (DFT-LDA), as implemented in the Wien2k software package \cite{25, 26}. The second is the quasiparticle Hamiltonian obtained from the linearized quasiparticle self-consistent GW method (LQSGW), as implemented in the FlapwMBPT code \cite{28, 29}. For the localised basis set $(m\sigma)$, we calculated maximally localized Wannier functions \cite{37, 38} by using the Wannier90 \cite{39}, Wien2Wannier \cite{40} and ComDMFT \cite{41} packages.

They are constructed in a wide energy window including Fe($d$) and As($p$) orbitals, as described in the Supplemental Material (SM) \cite{41}. For simplicity, we take the spin-orbit coupling (SOC) to be local in the chosen basis, and take it to be present only on iron atoms (for an assessment of the quality of this approximation, see SM \cite{41}).

We first discuss the electronic structure associated with $H(k)$, i.e. in the absence of the self-energy. In Fig. 1(a), we compare the FS of DFT-LDA to that of LQSGW. The LQSGW FS clearly displays a significant shrinking of the $xz/yz$ dominated hole/electron pockets, $\alpha'$, $\alpha$, and $\delta$ sheets in comparison to LDA, as pointed out in previous work \cite{21, 27}. This is because non-local electronic interactions are more prominently taken into account in this procedure for extracting the self-energy from ARPES data follows Ref. \cite{26} for Sr$_2$RuO$_4$. From a theoretical viewpoint, the dispersions of the different branches of quasiparticles are the solutions of $\det[\omega - H(k) - \text{Re}\Sigma(k, \omega)] = 0$ (neglecting the lifetime effects associated with $\text{Im}\Sigma$). We use the measured positions of the maximum of the momentum distribution curves (MDC) associated with several quasiparticle bands, for a given binding energy $\omega$, as an input to this equation which is then solved by a numerical root-finding procedure for the real part of the self-energy (for details of the procedure, see SM \cite{41}).

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This procedure requires, of course, that an ansatz is made about the momentum dependence of $\Sigma$. We consider the self-energy in the basis of local orbitals, and assume that it is independent of the out-of-plane momentum $k_z$ and that off-diagonal (inter-orbital) matrix elements are absorbed into the renormalization of the SOC \cite{14, 46}. Two different ansätze are made for the in-plane momentum dependence. (i) The self-energy components are simply assumed to be independent of momentum - we refer to this as the ‘DMFT ansatz’. (ii) The Brillouin zone is divided into two patches, centered around the $\Gamma$- and $M$-points, respectively, as illustrated on Fig. 2 and a more flexible momentum dependence is allowed which is piecewise constant in each patch. We refer to this ansatz as the ‘DCA ansatz’ since it corresponds to a two-site DCA approximation \cite{47}. These

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|c|c|c|c|}
\hline
 & $\alpha'$ & $\alpha$ & $\beta$ & $\gamma$ & $\delta$ & Net.
\hline
LDA & 0.01 & 0.14 & 0.33 & 0.18 & 0.28 & -0.02
\hline
LDA+DCA ansatz & 0.00 & 0.08 & 0.37 & 0.23 & 0.39 & +0.17
\hline
LDA+DMFT ansatz & 0.00 & 0.06 & 0.36 & 0.19 & 0.35 & +0.12
\hline
LQSGW & 0.00 & 0.08 & 0.35 & 0.20 & 0.21 & -0.03
\hline
LQSGW+DCA ansatz & 0.00 & 0.05 & 0.36 & 0.20 & 0.26 & +0.04
\hline
\end{tabular}
\caption{The net Fermi surface volumes, $V_{FS,total}$ (hole + electron) and Fermi surface volumes of each sheet (electrons/unit cell) in (a) the LDA, (b) the LDA+DCA ansatz, ($k_z=0.00$ for fitting of hole bands and $k_z=0.35$ for fitting of electron bands), (c) the LDA+DMFT ansatz, ($k_z=0.00$ for fitting of hole bands and $k_z=0.35$ for fitting of electron bands), (d) the LQSGW, (e) the LQSGW+DCA ansatz, ($k_z=0.00$ for fitting of hole bands and $k_z=0.55$ for fitting of electron bands). 0.02-0.03 (electrons/unit cell) in the net Fermi surface volume is the numerical uncertainty.}
\end{table}
two ansätze thus read (see SM for details [41]):

\[
\begin{align*}
\text{DMFT ansatz: } & \Sigma_m(k, \omega) = \Sigma_m(\omega) \quad (2) \\
\text{DCA ansatz: } & \Sigma_m(k, \omega) = \Sigma_m(\Gamma, \omega) \quad \text{if } k \in \Gamma \\
& = \Sigma_m(M, \omega) \quad \text{if } k \in M \quad (3)
\end{align*}
\]

The components of the self-energy within the DCA ansatz are obtained by fitting the experimental hole bands at K=Γ and electron bands at K=M separately. We also note that this ansatz is physically motivated by the AFM wave-vector of spin fluctuations and corresponding Brillouin zone folding (Fig. 2(b)) [48]. We emphasize that these ansätze are made for the components of the self-energy expressed in the basis of local orbitals. The transformation to the quasiparticle (band) basis is momentum dependent and leads to significant momentum dependence of the self-energy in that basis even if a DMFT ansatz is made (see also Ref. [20]).

The experimentally determined quantities in ARPES are \( \theta_k \) (the emission angle of the electron) and \( E_{\text{kin}} \) (the kinetic energy of the electron). Hence, only the in-plane momentum can be inferred directly according to \( \hbar k || = \sqrt{2mE_{\text{kin}} \sin \theta_k} \), together with the energy \( \omega \). In contrast, the perpendicular component of the momentum \( k_z = k_{\perp Z} \) reads: \( \hbar k_{\perp Z} = \sqrt{2m(E_{\text{kin}} \cos 2\theta_k + V_0)} \), and thus requires the knowledge of the inner potential \( V_0 \) [49]. As a result, experimental uncertainties affect the determination of \( k_z \); in our case, the experimental value of \( k_z \) around the electron pockets is only constrained to be in the interval of \([0.3, 0.7]\). For the hole pockets, there is little uncertainty that the data arise from \( k_z = 0 \) [43]. (See SM [41]). For the electron pockets, we considered two different ways to infer \( k_z \): (i) The first is to require that the Fermi surface volume satisfies Luttinger’s theorem, as obtained by a full Brillouin zone integration and assuming that the self-energy does not depend on \( k_z \). This leads to \( k_z \approx 0.55 \) for electron pockets within both the LDA+DCA and the LQSGW+DCA ansätze. (ii) The second one determines \( k_z \) by requesting that the resulting self energy is as local as possible. This leads to \( k_z=0.35 \) for the LDA+DCA ansatz and \( k_z=0.55 \) for the LQSGW+DCA ansatz (see SM [41]). Note that in that case, Luttinger’s theorem is violated within the LDA+DCA ansatz, while the value \( k_z = 0.55 \) ensures both Luttinger’s theorem and maximal locality when using the LQSGW reference. We explored both possible determinations of \( k_z \), and the overall consistency with the measured three dimensional dispersion of the Fermi surface serves as an additional criterion to ascertain the quality of the approximations. The ARPES data used to obtain these results are those of Ref. [19, 43], also displayed on Fig. 3 [40].

Results. Our main results are summarized in Figs. 1(b-d), 6 and Tables S3 and I. The full frequency dependence of the self-energies extracted from the procedure described above is displayed on Fig. S5 in the supplemental material (SM) [41]. Here, we focus on the key low-energy behaviour of the fitted self-energies, as characterized by the zero-frequency (static) values \( \Sigma_m(0) \), as well as the quasiparticle weights \( Z_m = \frac{1}{1 - \frac{\partial \Sigma_m}{\partial \omega} |_{\omega=0}} \), displayed in Table I. Comparing the values obtained within the DCA ansatz for the \( \Gamma \) and \( M \) BZ patches, we see that, when starting from LDA, the static components of the self-energy are spatially local (momentum independent) to a good approximation for the \( xz/yz \) orbitals, while a higher degree of momentum-dependence holds for the \( xy \) orbital. However, the latter observation is not unambiguous since a large error bar is found for \( \Sigma_{xy}(M,0) \) (of order 0.062 eV, see SM [41] for details about the evaluation of error bars). This is due to the large Fermi velocity of the \( xy \)-orbital driven \( \gamma \) band at \( K = M \).

The quasiparticle weight associated with the \( xy \) orbital is found to be weakly momentum dependent, while stronger momentum dependence is found for the \( xz/yz \) orbital, with also a certain level of ambiguity due to the large error bar at the \( \Gamma \)-point. This strong momentum dependence of the dynamical self-energy of the \( xz/yz \) orbitals has been discussed in Refs. [2, 6, 18] in relation to the strong coupling of the quasiparticles of the \( xz/yz \) driven \( \alpha \) and \( \alpha' \) hole-like FS sheets to the existing AFM correlation in LiFeAs [18]. Indeed, these FS sheets are close to the AFM zone boundary. The values of the quasiparticle weights obtained here, (0.15 (\( \Gamma \)) 0.12 (\( M \)) for \( xy \) and 0.25 (\( \Gamma \)) 0.16 (\( M \)) for \( xz/yz \)), are smaller than that of the computed LDA+DMFT values reported in Refs. [13, 13, 19] (\( Z_{xy}=0.26 \) and \( Z_{xz/yz}=0.34 \)). They are however close to the values (0.17 – 0.19) reported by de Haas-van Alphen experiments [51].

Table I also displays the results obtained by using a DMFT (local) ansatz for the self-energies. As seen there, the values of the quasiparticle weights are interme-
Table II. Zero frequency self-energy ($\Sigma_m(K, 0)$) and quasi-particle residue ($Z_m(K)$) extracted from ARPES data of LiFeAs [14], with the LDA+DCA ansatz, the LQSGW+DCA ansatz, and the LDA+DMFT ansatz. We use $k_z=0.00$ for K-Γ (hole sheets) for both the LDA and the LQSGW references, $k_z=0.35$ for K-M (electron sheets) for the LDA reference, and $k_z=0.55$ for K-M (electron sheets), for the LQSGW reference. Error bars (total) are computed from the peak width of both in plane $k$ and out of plane $k_z$. (See SM for the details on the definition of the error bars [41].)

|                  | LDA+DCA ansatz | LQSGW+DCA ansatz | LDA+DMFT ansatz |
|------------------|----------------|------------------|-----------------|
|                  | $\Sigma_m(\Gamma, 0)$ (eV) | $Z_m(\Gamma)$ | $\Sigma_m(M, 0)$ (eV) | $Z_m(M)$ | $\Sigma_m(\Gamma, 0)$ (eV) | $Z_m(\Gamma)$ | $\Sigma_m(M, 0)$ (eV) | $Z_m(M)$ |
| $xy$             | 0.029±0.025 | 0.15±0.01 | -0.130±0.062 | 0.12±0.01 | 0.062±0.036 | 0.18±0.01 |
| $xz/yz$          | -0.083±0.040 | 0.25±0.13 | -0.113±0.026 | 0.16±0.03 | -0.027±0.003 | 0.38±0.01 |

Data of Refs [19, 52] (see SM [11] for comparison), (ii) a good description of the $k_z$ dependent $\gamma$ FS in ARPES of Refs [12, 53], (iii) and a qualitative description of the $k_z$ dependent $\delta$ FS in ARPES with correct $k_z$ for the crossing of the $\delta$ and $\gamma$ FSs and somewhat larger curvature of the $\delta$ FS near the momentum of A of Ref. [12]. Fig 4(d) implies that for electron bands, the overall amplitude of $k_z$ dependent variation of the $\delta$ and $\gamma$ FSs in the LQSGW+DCA fit is consistent with the ARPES data of Ref. [12].

Summarizing, our results demonstrate that - within the uncertainties associated with experimental error bars - the self-energy is found to be quite local when using the LQSGW reference Hamiltonian, and that this also provides a satisfactory description of FS properties. In contrast, when using LDA as a reference, a significant degree of momentum-dependence/non-locality is found, as well as a too large volume of the $\delta$-FS sheet leading to a violation of Luttinger’s theorem. This conclusion is also apparent on the full frequency dependence of the extracted self-energies reported on Fig S5 in the SM. [11]

We compare in Fig 3(a-c) the experimental ARPES intensity to the fitted hole bands of LiFeAs using different theoretical approaches, (a) the LDA+DMFT ansatz (b) the LDA+DCA ansatz and (c) the LQSGW+DCA ansatz. For the $xy$ dominant $\beta$ band, all schemes compare well with ARPES. In contrast, we observe some differences between the different ansætze (comparable to error bars) for the position of the top of the $\alpha$ band with dominant $xz/yz$ character. The LDA+DMFT ansatz leads to a lower energy than the LDA+DCA ansatz and the LQSGW+DCA ansatz. The splitting of the states with $xz/yz$ character at the $\Gamma$ point is controlled by the SOC and given by $\lambda Z_{xz/yz}$. Its experimental value is 9.5-11.4 meV [54, 55]. The values of $\lambda$ in LDA and LQSGW are 50 meV and 25 meV respectively, which when multiplied by the the extracted $Z$’s from Table II indeed leads to values close to 10 meV in both cases (see SM for details on the effect of SOC in LiFeAs [11]).

We now turn to the electron bands in Fig 3(d-i). Along the $\Gamma - M$ direction, the $\gamma$ band has almost pure $xy$ character, and is seen in $\sigma$ polarized ARPES. This $\gamma$ band is well described by both the DCA ansatz and the DMFT ansatz within both references (LDA and LQSGW), see panels Fig 3(g,h, and i). For the $xz/yz$ dominant $\delta$ band, the LDA+DCA ansatz and the LQSGW+DCA ansatz yield quasiparticle spectra which are consistent with ARPES within error bars as shown in Fig 3(e, and f). However, differences between the fits are seen for the $xz/yz$ driven $\delta$ band with the LDA+DMFT ansatz having a steeper dispersion and a lower bottom than the DCA ansatz, as seen in Fig 3(d, e, and f).

In summary, our analysis demonstrates that an LQSGW reference [27, 33, 56] in combination with quite local self-energies provides a description of the quasiparticle dispersions of LiFeAs in good agreement with ex-
The conclusion of this analysis (see details in SM [41]) is that the $xy$ orbital is found to have a larger scattering rate than the $xz/yz$ one, and that it undergoes a clear crossover at $T \sim 150$ K between a high-$T$ incoherent regime to a low-$T$ coherent one. This is consistent with the LQSGW+DMFT finding that the $xy$ orbital is the more correlated one. At low-$T$ both scattering rates are found to have a Fermi liquid $T^3$ behaviour. As a consistency check, we also obtain satisfactory agreement with the magnetoresistance data. Let us emphasize that, in contrast, studies emphasizing non-locality due to low-energy AFM fluctuations yield a non-Fermi liquid scattering rate of the $xz/yz$ orbital which is larger than that of $xy$ [5, 6].

Several authors have pointed at some discrepancies between experimental data and the predictions of LDA+DMFT, which is usually interpreted as a failure of the DMFT to take into account non-local effects [5, 6]. Here, based on a direct analysis of ARPES experimental data, we presented a very different picture, consistent with the electronic structure+DMFT conceptual framework. We have shown that most of the discrepancies between LDA+DMFT and ARPES measurements arise from the choice of the reference Hamiltonian, and not from low energy dynamical self-energy effects. When the LQSGW Hamiltonian is used as a starting point, we found that the DMFT ansatz describes quite well the available experimental data, including transport measurements.

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![Figure 3. Comparison between the ARPES data of Ref. 19, 43 for LiFeAs (color intensity map) with different polarizations ($\pi$ and $\sigma$) and the quasiparticle dispersions obtained with the different ansätze discussed in the text. The results of LDA+DMFT, LDA+DCA, and LQSGW+DCA ansätze are shown in panels (a, d and g), (b, e and h), and (c, f and i), respectively. The hole pocket data are taken at $k_z=0.00$ and the electron pockets data are taken at a value of $k_z$ within the range of [0.3, 0.7] 14 42. The blue colored ellipses are theoretical error estimates arising from the width of the MDC peak and the uncertainty in $k_z$. For the electron bands, $k_z=0.55$ has been used in the fit using LQSGW as a reference, while $k_z=0.35$ when using LDA as a reference (see main text).](image_url)
slightly above the superconducting transition temperature (18 K) of LiFeAs.\textsuperscript{[24]}

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CONSTRUCTION OF THE REFERENCE HAMILTONIAN OF $H^{LDA}$

In this section, we describe details of the construction of the reference Hamiltonian in the LDA, $H^{LDA}$. We construct maximally localized Wannier function (MLWF) of LiFeAs for the $p-d$ model approach ($Fe(d)$ and $As(p)$) using the Wannier90 and the Wien2Wannier packages. We use the energy window of the interval of -6.0 eV to 3.0 eV for $Fe(d)$ and $As(p)$ bands. We used the experimental crystal structure of LiFeAs of the Ref. 60. For the convergence of the charge density in the LDA, we used a $k$-mesh of 10000, and checked that the charge density and the total energy are converged with criterions of $5 \times 10^{-4}$ (electrons/formula unit) and 0.7 (meV/formula unit), respectively. For the construction of MLWF, we used a $k$-mesh of $11 \times 11 \times 7$. The local axis for the MLWF of $Fe(d)$ is chosen such that (i) $z$ along $c$ of the unit cell, and (ii) $x$ and $y$ axes toward nearest neighboring Fe atoms. For the $p-d$ model, the spread function of the MLWF is converged as (i) 0.985, 1.073, 1.111, and 1.333 ($\AA^2$) for $z^2$, $x^2-y^2$, $xy$, and $xz/yz$ orbitals of Fe, and (ii) 3.197, and 3.301 ($\AA^2$) for $p_z$ and $p_{x,y}$ orbitals of As.

Fig S1 presents Fermi surfaces and low energy band structures of $p-d$ model + $\lambda_{SOC}$ (LDA+$\lambda_{SOC}$) in comparison with LDA plus SOC (LDA+SOC) with $\lambda_{SOC}=50$ meV. This data implies that the electronic structure of the $p-d$ model+$\lambda_{SOC}$ (LDA+$\lambda_{SOC}$) and LDA+SOC are consistent. The factor that the local SOC is implemented only in the $Fe(d)$ orbital in the $p-d$ model+$\lambda_{SOC}$ (LDA+$\lambda_{SOC}$) proves that the SOC in the LDA+SOC is fully $Fe(d)$ orbital driven.

Figure S1. (a) Comparison of Fermi surfaces of the LDA+SOC and the $p-d$ model + $\lambda_{SOC}$ (LDA+$\lambda_{SOC}$). (b) Comparison of band dispersions of the LDA+SOC and the $p-d$ model + $\lambda_{SOC}$ (LDA+$\lambda_{SOC}$). The effective SOC of $Fe(d)$ is $\lambda_{SOC} = 50$ meV. Here we take $k_z=0.00$ for hole bands, and $k_z=0.35$ for electron bands.
CONSTRUCTION OF THE REFERENCE HAMILTONIAN OF $H_{LQSGW}$

In this section, we describe details of the construction of the reference Hamiltonian in the LQSGW, $H_{LQSGW}$. LQSGW calculation is performed by using FlapwMBPT package \cite{S61, S62}, which is based on full-potential linearized augmented plane wave plus local orbital method. For the crystal structure, experimental lattice constants and atomic positions \cite{S25} are used. The Muffin-tin (MT) radius ($R$) is chosen in the following way: 1.74 for Li, 2.27 for Fe, and 2.16 for As in Bohr radius. Wave functions are expanded by spherical harmonics with $l$ up to 3 for Li, 4 for Fe, and 4 for As in the MT spheres. In the interstitial region (IS), it is expanded by plane waves with the cutoff ($K_{cut}$) of $R_{Fe} \times K_{cut} = 4.4$. Product basis set is expanded by spherical harmonics with $l$ up to 4 in the MT spheres and by planewaves with the cutoff ($G_{cut}$) of $R_{Fe} \times G_{cut} = 7.4$ in IS region. All the unoccupied states are taken into account for both polarizability and self-energy calculation. The Brillouin zone is sampled in $6 \times 6 \times 4$ grid.

By using ComWann modules in ComDMFT package \cite{S34} utilizing Wannier90 package \cite{S39}, 42 wannier functions are constructed: Li-p, Fe-s, Fe-p, Fe-d, As-s, As-p, and As-d orbitals. The frozen energy window is set between -9 eV to 7 eV and the disentanglement energy window is between -9 eV to 49 eV. Initial trial orbitals are constructed by using Muffin-tin orbitals with well-defined angular momentum characters. The local axis for the MLWF of Fe($d$) is chosen such that (i) $z$ along $c$ of the unit cell, and (ii) $x$ and $y$ axes toward nearest neighboring Fe atoms. The spread function of the MLWF is converged as (i) 0.497, 0.493, 0.524, and 0.522 ($\text{Å}^2$) for $z^2$, $x^2-y^2$, $xy$, and $xz/yz$ orbitals of Fe, and (ii) 1.499, and 1.724 ($\text{Å}^2$) for $p_z$, and $p_{x,y}$ orbitals of As.

Fig. S2 presents low energy band structures of the LQSGW+$\lambda_{SOC}$ with local SOC for Fe($d$) (with $\lambda_{SOC}=25$ meV) in comparison with the LQSGW plus SOC (LQSGW+SOC). The overall consistency in between the band structure of the LQSGW+SOC and the LQSGW+$\lambda_{SOC}$ implies that the local SOC in Fe($d$) is a good approximation. The splitting of $\alpha$ and $\alpha'$ bands at $\Gamma$ is 35 meV for the LQSGW+SOC and 25 meV for the LQSGW+$\lambda_{SOC}$, respectively (See Table S1). In Fig. S2 it is shown that dispersions of $\alpha$, $\beta$, $\delta$, and $\gamma$ bands are agreement in between the LQSGW+SOC and the LQSGW+$\lambda_{SOC}$. These $\alpha$, $\beta$, $\delta$, and $\gamma$ bands are taken for the extraction of the self-energy.
Figure S3. (a) and (b) comparison of the ARPES data to the (i) LDA reference ($\lambda_{SOC}=50$ meV) and the (ii) LQSGW reference ($\lambda_{SOC}=25$ meV) with $k_z=0.00$ for hole pockets ($\alpha$, $\alpha'$, and $\beta$) and $k_z=0.35$ for electron pockets ($\delta$ and $\gamma$). (c) and (d) same as (a) and (b) with $k_z=0.00$ for hole pockets ($\alpha$, $\alpha'$, and $\beta$) and $k_z=0.45$ for electron pockets ($\delta$ and $\gamma$). (e) and (f) same as (a) and (b) with $k_z=0.00$ for hole pockets ($\alpha$, $\alpha'$, and $\beta$) and $k_z=0.55$ for electron pockets ($\delta$ and $\gamma$).

**ARPES VERSUS BAND STRUCTURES $H(k)$ OF LDA AND LQSGW**

In this section, we describe comparisons of bands in between ARPES and $H(k)$ of LDA and LQSGW to provide background information for the extraction of self-energy.

Fig. S3 presents comparison of ARPES ([S19], see the main text), to the band structures of $H(k)$ of LDA and LQSGW. The case of $k_z=0.00$ is shown for hole pockets for the extraction of self-energy. The case of $k_z=0.35-0.55$ is shown for the electron pockets to show the effect of the choice of $k_z$ for the extraction of the self-energy.

**LDA reference**—For hole pockets with $k_z=0.00$, following factors are shown in Fig. S3(a,c, and e). (i) There is an agreement of the size of the $xy$ driven $\beta$ Fermi surface of $H^{LDA}(k)$ to the ARPES. (ii) There is a significant shrinking of the size of $xz/yz$ driven $\alpha$ and $\alpha'$ Fermi surfaces in the ARPES with respect to those Fermi surfaces.
from $H^{LDA}(k)$. (iii) There are renormalizations of bands in the ARPES from the $H^{LDA}(k)$ for $\alpha$, $\alpha'$, and $\beta$ bands. For electron pockets with $k_z = 0.35$, following factors are shown in Fig S3(b). (i) There is a shrinking of the $xz/yz$ driven $\delta$ band in the ARPES with respect to the band from $H^{LDA}(k)$, with a similar amount with respect to the case of $xz/yz$ driven $\alpha$ and $\alpha'$. (ii) There is a small shrinking of the $xy$ driven $\gamma$ band in the ARPES with respect to the band from $H^{LDA}(k)$. (iii) There are renormalizations of bands in the ARPES from the $H^{LDA}(k)$ for $\delta$ and $\gamma$ bands. For electron pockets with $k_z = 0.55$, the $xy$ driven $\gamma$ band of $H^{LDA}(k)$ have a similar feature with respect to the case with $k_z=0.35$, as shown in Fig S3(b and f). For the electron band $\delta$ with $k_z = 0.55$, for the size of the Fermi momentum, there is an agreement in between the ARPES and the $H^{LDA}(k)$ as shown in Fig S3(f). For $k_z=0.55$ for electron pockets, there are also renormalizations of bands in ARPES with respect to that in the $H^{LDA}(k)$ as shown in Fig S3(f).

$LQSGW$ reference—For hole pockets with $k_z=0.00$, following factors are shown in Fig S3(a,c, and e). (i) There is an agreement of the size of the $xy$ driven $\beta$ Fermi surface of $H^{LQSGW}(k)$ with the ARPES. (ii) There is a small shrinking of the size of $xz/yz$ driven $\alpha$ and $\alpha'$ Fermi surfaces in the ARPES with respect to those Fermi surfaces from $H^{LQSGW}(k)$. (iii) There are renormalizations of bands in the ARPES from the $H^{LQSGW}(k)$ for $\alpha$, $\alpha'$, and $\beta$ bands. For electron pockets with $k_z = 0.55$, following factors are shown in Fig S3(f). (i) There is a small shrinking of the $xz/yz$ driven $\delta$ band in the ARPES with respect to the band from $H^{LQSGW}(k)$, with a similar amount with respect to the case of $xz/yz$ driven $\alpha$ and $\alpha'$. (ii) There is an agreement of the Fermi momentum of the $xy$ driven $\gamma$ band in the ARPES with respect to the band from $H^{LQSGW}(k)$. (iii) There are renormalizations of bands in the ARPES from the $H^{LQSGW}(k)$ for $\delta$ and $\gamma$ bands. For electron pockets with $k_z = 0.35$, following factors are shown in Fig S3(b). (i) There is a significant shrinking of the $xz/yz$ driven $\delta$ band in the ARPES with respect to the band from $H^{LQSGW}(k)$, with a much larger amount with respect to the case of $xz/yz$ driven $\alpha$ and $\alpha'$. (ii) There is an agreement of the Fermi momentum of the $xy$ driven $\gamma$ band in the ARPES with respect to the band from $H^{LQSGW}(k)$. (iii) There are renormalizations of bands in the ARPES from the $H^{LQSGW}(k)$ for $\delta$ and $\gamma$ bands.

**EXTRACTION OF THE SELF-ENERGY**

In this section, we describe the procedure for the extraction of the self-energy of LiFeAs. For $H(k)$, we assume the reference Hamiltonian of LDA+\(\lambda_{SOC}\) (\(\lambda_{SOC}=\) 50 meV) and LQSGW+\(\lambda_{SOC}\) (\(\lambda_{SOC}=\) 25 meV) as described in sections I and II.

In order to extract the self-energy (real part) from the ARPES data, we define the $|d|$ of Eq. S1 from the Green’s function which is defined in the main text. The momentum (k) of the maximum of the momentum distribution curve (MDC) of the ARPES for each band (of index of $\nu$) are noted as $k^M_{\nu,MDC}$ assigned to each frequency of $\omega$ as shown in Eq S1. The quasiparticle spectra of ($\omega,k$) is given by the condition of $det(G^{-1}(k,\omega)) = 0$ with the condition of $\text{Im}\Sigma = 0$. Thus, the variable $|d|$ should be minimized for each frequency. From the defined reference Hamiltonian of $H(k)$, the variable $|d|$ is a function of the self-energy for each frequency. We minimize $|d|$ as a root finding procedure from the self-energy variable for each frequency. From this procedure, the self-energy is extracted.

$$d^2 = \sum_{\nu} |det(G^{-1}(k^M_{\nu,MDC},\omega))|^2$$
$$\nu = (\alpha, \beta, \gamma, \text{and} \delta)$$
$$|d| = f(\Sigma_{xz/yz}(k,\omega), \Sigma_{xy}(k,\omega)) \quad \text{(S1)}$$

Several assumptions for the simplification of self-energy is made to solve Eq S1. The first, for the low energy, most of orbital character is $xz/yz$ and $xy$. Thus, we consider the self-energy of $xz/yz$ and $xy$ of Fe(d) only as shown in Eq S1. The second, we assume that self-energy is $k_z$ independent. The third, for the in-plane momentum dependence of the self-energy, we employed two ansätze, namely, the DMFT ansatz and the DCA ansatz as shown in Eq S2.

$$\Sigma_m^-(k,\omega) = \Sigma_m^-(K,\omega) \text{ DCA ansatz}$$
$$\Sigma_m^-(k,\omega) = \Sigma_m^-(\omega) \text{ DMFT ansatz} \quad \text{(S2)}$$

In the DMFT ansatz, the self-energy is momentum independent, and all bands of $\nu$ index for the given frequency is considered for the minimization of $|d|$ as shown in Eq S2. In the DCA ansatz, the self-energy is a coarse grained constant in the momentum space for $K$ of Brillouin zone (BZ) patch as shown in Eq S2. We used the 2 dimensional BZ patch in the main text, in relation to the existing antiferromagnetic (AFM) correlation with q vector close to
In practice, the coarse grained momentum $K=\Gamma$ includes $k$ for $\alpha$, $\alpha'$ and $\beta$, hole bands, and the coarse grained momentum $K=M$ includes $k$ for $\delta$ and $\gamma$ electron bands. We consider $\alpha$, $\beta$, $\delta$, and $\gamma$ bands for the extraction of the momentum independent self-energies in the DMFT ansatz. In the DCA ansatz, we consider $\alpha$ and $\beta$ bands for $K=\Gamma$, and $\delta$ and $\gamma$ bands for $K=M$, for the extraction of the $K$ dependent self-energy as described in Eq.\[S51\] The description of $\alpha'$ could be achieved from these procedures due to the similar $K$ dependence of self-energy in between $\alpha$ and $\alpha'$ of $xz/yz$ orbital.

From the symmetry of the lattice of LiFeAs, we assumed that self-energy of Fe($d$) is orbitally diagonal. From Refs.\[S44\] and \[S46\] it is shown that for the regime of Hund’s metal, due to the larger energy scale of Kondo screening in the orbital sector ($T^o_{K}$) with respect to the SOC ($\lambda_{SOC}$), $T^o_{K} > \lambda_{SOC}$. SOC is not effective on the orbital diagonal self-energy having a frequency dependent coherence-incoherence crossover.

For orbitally off-diagonal self-energy, in Refs.\[S44\] and \[S45\] it is shown that from the example of Sr$_2$RuO$_4$, this off-diagonal term could be absorbed into the renormalization of SOC. We assume that this orbitally off-diagonal self-energy is zero. We confirmed that this assumption properly describes the SOC effects on the ARPES near the zero frequency. Table \[S1\] summarizes the effective SOC constant $\lambda_{SOC}$ of the reference Hamiltonian, and its comparison with the band splitting energy of $\alpha$ and $\alpha'$ band at $k=\Gamma$, $\Delta$ for ARPES ($\Delta_{ARPES}$), LDA+SOC ($\Delta_{LDA}$), and LQSGW+SOC ($\Delta_{LQSGW}$). The $\Delta$ is effectively equal to $Z\lambda_{SOC}$ ($Z$ is the renormalization constant of the $xz/yz$ orbital at $k=\Gamma$) where $Z=1.0$ if the self-energy vanishes.

Table \[S1\]. The SOC induced splitting (in meV unit) of the $\alpha$ and $\alpha'$ at $k=\Gamma$, $\Delta$, of ARPES ($\Delta_{ARPES}$), LDA+SOC ($\Delta_{LDA}$), and LQSGW+SOC ($\Delta_{LQSGW}$). We have shown the effective SOC ($\lambda_{SOC}$) of the reference Hamiltonian, $H(k)$, of the LDA ($\lambda_{LDA}^{SOC}$) and LQSGW ($\lambda_{LQSGW}^{SOC}$). The $\Delta$ of the LDA+DCA fit ($Z\lambda_{LDA}^{SOC}$) and the LQSGW+DCA fit ($Z\lambda_{LQSGW}^{SOC}$) is also shown, to be compared with its experimental value of $\Delta_{ARPES}$.

| $\Delta_{ARPES}$ | $\Delta_{LDA}$ | $\Delta_{LQSGW}$ | $\lambda_{LDA}^{SOC}$ | $\lambda_{LQSGW}^{SOC}$ | $Z\lambda_{SOC}$ | $Z\lambda_{SOC}$ |
|------------------|----------------|-------------------|-----------------------|-----------------------|-----------------|-----------------|
| 9.5-11.4          | 50             | 35                | 50                    | 25                    | 12.5            | 9.5             |

DETERMINATION OF $k_z$ FOR THE REFERENCE OF ELECTRON SHEETS FOR $H^{LDA}$ AND $H^{LQSGW}$

In this section, we describe the relation in between (i) the variable of $k_z$ for electron pockets for the reference Hamiltonian $H(k)$ and (ii) the spatial locality of self-energy and the validity of the Luttinger’s theorem, as discussed in the main text.

LDA reference - Table \[S2\] presents the self-energy from the LDA+DCA ansatz with $k_z=0.00$ for hole pockets and $k_z=0.55$ for electron pockets. It is shown that the self-energy of this case have a strong spatial non-locality with strong momentum $(K)$ dependence. This feature implies that the LDA+DMFT ansatz is not applicable with $k_z=0.55$ for electron pockets. With this choice of $k_z=0.55$ for electron pockets, the LDA+DCA ansatz fulfills the Luttinger’s theorem as shown in Table \[S3\] and Fig.\[S1\]a). However, in Fig.\[S1\]a), it is shown that the LDA+DCA ansatz with $k_z=0.55$ for electron pockets provides a substantial overestimation of $k_z$ dependent dispersion of the $\delta$ Fermi surface with respect to the ARPES and the LQSGW+DCA ansatz with $k_z=0.55$ for electron pockets (see the main text). \[S4\] In the main text, we have shown that the setting of $k_z=0.35$ for electron pockets provides more local static self-energy ($\Sigma(0)$) but violates the Luttinger’s theorem (see Table \[S3\] and Table \[S5\]).

In summary, for the LDA reference, there is no $k_z$ for electron sheet such that fulfills both the locality of self-energy and the Luttinger’s theorem together.

LQSGW reference - Table \[S4\] presents the self-energy from the LQSGW+DCA ansatz with $k_z=0.00$ for hole pockets and the $k_z=0.35$ for electron pockets. It is shown that the self-energy of this case have a strong spatial non-locality with strong momentum $(K)$ dependence. With this choice of the $k_z=0.35$ for electron pockets, the LQSGW+DCA ansatz violates the Luttinger’s theorem as shown in Table \[S3\] and Fig.\[S1\]b). In the main text, we have shown that for the setting of $k_z=0.55$ for electron pockets, the Luttinger’s theorem is obeyed and the self-energy is spatially local (see Table \[S3\] and Table \[S5\]).

In summary, for the LQSGW reference, there is single value of $k_z=0.55$ for electron pockets which (i) fulfills the Luttinger’s theorem and (ii) provides a validity of the spatially local self-energy.

ERROR BAR IN THE SELF-ENERGY, IN PLANE $k$ ERROR VERSUS OUT OF PLANE $k_z$ ERROR

In this section, we describe the procedure for the extraction of the error bar in the self-energy.
Figure S4. (a) Fermi surfaces for the LDA+DCA ansatz with $k_z=0.00$ for hole bands fit and $k_z=0.55$ for electron bands fit (total Fermi surface volume of -0.01 (electrons/unit cell)). (b) Fermi surfaces for the LQSGW+DCA ansatz with $k_z=0.00$ for hole bands fit and $k_z=0.35$ for electron bands fit (total Fermi surface volume of +0.18 (electrons/unit cell)). The ARPES data for fitting is taken from Ref.S19 (See main text).

Table S2. Zero frequency self-energy ($\Sigma_m(K,0)$) and quasiparticle residue ($Z_m(K)$) from the ARPES of LiFeAs with the LDA reference using the DCA ansatz. We set $k_z=0.00$ for $K=\Gamma$ (for hole sheets) and $k_z=0.55$ for $K=M$ (for electron sheets), for the LDA reference. Error bars (total) are computed from the peak width of both in plane $k$ and out of plane $k_z$. For the peak width, we adapted the resolution in the MDC of LiFeAs, 0.01 ($\pi/a$) (0.008 Å$^{-1}$). Here, $\sqrt{2}a$ is the lattice constant of LiFeAs. This resolution in the MDC is taken from the best resolution limit for the $\beta$ sheet with the resolution limit of the energy of 3 meV.

| $K$ | $\Sigma_m(\Gamma,0)$ (eV) | $Z_m(\Gamma)$ | $\Sigma_m(M,0)$ (eV) | $Z_m(M)$ |
|-----|--------------------------|---------------|---------------------|---------|
| $xy$ | 0.029±0.025 | 0.15±0.01 | -0.153±0.060 | 0.10±0.01 |
| $xz/yz$ | -0.083±0.040 | 0.25±0.13 | 0.042±0.014 | 0.34±0.04 |

In the extraction of error bar of zero frequency self-energy ($\Sigma_m(K,0)$) and quasiparticle residue ($Z_m(K)$) from
Table S3. The Fermi surface volumes (electrons/unit cell) in (a) the LDA, (b) the DCA fitting with the reference of the LDA with \(k_z=0.35\) for electron bands, (c) the DMFT fitting with the reference of the LDA with \(k_z=0.35\) for electron bands, (d) the DCA fitting with the reference of the LDA with \(k_z=0.55\) for electron bands (See Fig S5(a)), (e) the LQSGW, (f) the DCA fitting with the reference of \(S_{QSGW}\) with \(k_z=0.55\) for electron bands, and (g) the results of the \(S_{QSGW}+\)DCA fit with \(k_z=0.35\) for electron bands which violates the Luttinger’s theorem (See Fig S5(b)). We fit the hole bands with \(k_z=0.00\) for both the LDA and the LQSGW references.

|     | \(\alpha\) | \(\alpha'\) | \(\beta\) | \(\gamma\) | \(\delta\) | \(\text{total}\) |
|-----|------------|------------|----------|-----------|----------|----------------|
| LDA | 0.01       | 0.14       | 0.33     | 0.18      | 0.28     | -0.02          |
| LDA+DCA \((k_z=0.35\) for fitting of electron bands) | 0.00        | 0.08      | 0.37     | 0.22      | 0.39     | +0.14          |
| LDA+DMFT \((k_z=0.35\) for fitting of electron bands) | 0.00        | 0.06      | 0.36     | 0.19      | 0.35     | +0.12          |
| LDA+DCA \((k_z=0.55\) for fitting of electron bands) | 0.00        | 0.08      | 0.37     | 0.19      | 0.24     | -0.01          |
| LQSGW | 0.00       | 0.08      | 0.35     | 0.20      | 0.21     | -0.03          |
| LQSGW+DCA \((k_z=0.55\) for fitting of electron bands) | 0.00        | 0.05      | 0.36     | 0.20      | 0.26     | +0.04          |
| LQSGW+DCA \((k_z=0.35\) for fitting of electron bands) | 0.00        | 0.05      | 0.36     | 0.23      | 0.31     | +0.18          |

Table S4. Zero frequency self-energy \(\Sigma_m(K,0)\) and quasiparticle residue \(Z_m(K)\) from the ARPES of LiFeAs with the LQSGW reference using the DCA ansatz. We set \(k_z=0.00\) for \(K=\Gamma\) (for hole sheets) and \(k_z=0.35\) for \(K=M\) (for electron sheets), for the LDA reference. Error bars (total) are computed from the peak width of both in plane \(k\) and out of plane \(k_z\). For the peak width, we adapted the resolution in the MDC of LiFeAs, 0.01 \((\pi/a) (0.008 \text{ Å}^{-1})\). Here, \(\sqrt{2\alpha}\) is the \(a_{\text{lattice}}\), where \(a_{\text{lattice}}\) is the lattice constant of LiFeAs. This resolution in the MDC is taken from the best resolution limit for the \(\beta\) sheet with the resolution limit of the energy of 3 meV.

\[
\begin{array}{cccccc}
\Sigma_m(1,0) & Z_m(1) & \Sigma_m(M,0) & Z_m(M) \\
xy & 0.002\pm0.014 & 0.21\pm0.01 & 0.041\pm0.033 & 0.21\pm0.01 \\
xyz & -0.027\pm0.063 & 0.38\pm0.01 & -0.188\pm0.021 & 0.16\pm0.01 \\
\end{array}
\]

Table S5. Zero frequency self-energy \(\Sigma_m(K,0)\) and quasiparticle residue \(Z_m(K)\) from the ARPES of LiFeAs with the LDA reference using the DCA ansatz. We set \(k_z=0.00\) for \(K=\Gamma\) (for hole sheets) and \(k_z=0.35\) for \(K=M\) (for electron sheets), for the LDA reference. Error bars are computed from the peak width of in-plane \(k\) (parentheses is out of plane \(k_z\)). For the peak width, we adapted the resolution in the MDC of LiFeAs, 0.01 \((\pi/a) (0.008 \text{ Å}^{-1})\). Here, \(\sqrt{2\alpha}\) is the \(a_{\text{lattice}}\), where \(a_{\text{lattice}}\) is the lattice constant of LiFeAs. This resolution in the MDC is taken from the best resolution limit for the \(\beta\) sheet with the resolution limit of the energy of 3 meV.

\[
\begin{array}{cccc}
\Sigma_m(1,0) & Z_m(1) & \Sigma_m(M,0) & Z_m(M) \\
xy & 0.029\pm0.025(0.000) & 0.15\pm0.01(0.00) & -0.130\pm0.062(0.004) & 0.12\pm0.01(0.01) \\
xyz & -0.083\pm0.040(0.000) & 0.25\pm0.13(0.01) & -0.113\pm0.013(0.023) & 0.16\pm0.01(0.02) \\
\end{array}
\]

Table S6. Zero frequency self-energy \(\Sigma_m(K,0)\) and quasiparticle residue \(Z_m(K)\) from the ARPES of LiFeAs with the LQSGW reference using the DCA ansatz. We set \(k_z=0.00\) for \(K=\Gamma\) (for hole sheets) and \(k_z=0.55\) for \(K=M\) (for electron sheets), for the LQSGW reference. Error bars are computed from the peak width of in-plane \(k\) (parentheses is out of plane \(k_z\)). For the peak width, we adapted the resolution in the MDC of LiFeAs, 0.01 \((\pi/a) (0.008 \text{ Å}^{-1})\). Here, \(\sqrt{2\alpha}\) is the \(a_{\text{lattice}}\), where \(a_{\text{lattice}}\) is the lattice constant of LiFeAs. This resolution in the MDC is taken from the best resolution limit for the \(\beta\) sheet with the resolution limit of the energy of 3 meV.

\[
\begin{array}{cccc}
\Sigma_m(1,0) & Z_m(1) & \Sigma_m(M,0) & Z_m(M) \\
xy & 0.022\pm0.014(0.000) & 0.21\pm0.01(0.00) & 0.044\pm0.036(0.001) & 0.18\pm0.00(0.01) \\
xyz & -0.027\pm0.003(0.000) & 0.38\pm0.01(0.00) & -0.051\pm0.006(0.114) & 0.30\pm0.03(0.03) \\
\end{array}
\]

The ARPES, we consider two independent sources of error (i) in plane \(k\) error and (ii) out of plane \(k_z\) error, with consideration of the MDC peak width of 0.01 \((\pi/a) (0.008 \text{ Å}^{-1})\). There is an intrinsic uncertainty in the value of \(k_z\) as the surface breaks translation symmetry and therefore, the \(k_z\) is not a good quantum number. As shown in Fig S8, the most dominant \(k_z\) dependent variation is in the \(xyz/yz\) orbital dominant \(\delta\) band placed at \(K=M\), and other bands have a smaller \(k_z\) dependency. As a result, the most of the error bars are from the in plane \(k\) error, and a sizable \(k_z\) peak width induced error is only at the \(K=M\) for \(xyz/yz\) orbital (contribute to the \(\delta\) band), for both the LDA reference and the LQSGW reference, as shown in Table S7 and Table S8.
LOCALITY OF DYNAMICAL SELF-ENERGY

In this section, we present dynamical part self-energies in the LDA reference and LQSGW reference, in the main text, with \( k_z = 0.35 \) for electron bands fit in the LDA reference and \( k_z = 0.55 \) for electron bands fit in the LQSGW reference.

The full frequency dependence of the extracted self-energies is presented on Fig. S5, which displays the dynamical part \( \text{Re} \Sigma_m(K, \omega) - \text{Re} \Sigma_m(K, 0) \) for the different schemes considered in this work. By comparing the \( K = \Gamma \) and \( K = M \) data, it is immediately apparent from this figure that the fits based on the LQSGW reference lead to a much higher degree of locality for both orbital components than those based on the LDA reference. These factors imply that the self-energy from the LQSGW reference is spatially local, confirms the validity of the LQSGW+DMFT ansatz for the description of the quasiparticle of LiFeAs.

![Figure S5](image)

**Figure S5.** (a) and (b) Dynamical part of the self energies, \( \Sigma_m(K, \omega) - \Sigma_m(K, 0) \) in the LQSGW+DCA ansatz for \( xz/yz \) orbital and \( xy \) orbital, respectively. (c) and (d) Dynamical part self energy of \( xz/yz \) and \( xy \) orbitals, in the LDA+DCA ansatz and in the LDA+DMFT ansatz. Here, we use \( k_z = 0.00 \) for hole bands for both the LDA and the LQSGW references. We use \( k_z = 0.35 \) for electron bands for the LDA references. We use \( k_z = 0.55 \) for electron bands for the LQSGW references (see the main text and the Section V).

VALIDITY OF THE LQSGW+DCA ANSATZ FOR ARPES

To show the validity of the self-energy of the LQSGW+DCA ansatz, we compared spectra from the LQSGW+DCA ansatz to the ARPES of Ref. [S19] in Fig. S6 and the ARPES of Ref. [S52] in Fig. S7 with different \( k \) path (\( \Gamma \)-\( M \)) and \( k_z \) values. Fig. S6 and Fig. S7 implies that present LQSGW+DCA ansatz provides a good description of the quasiparticle spectra for multiple examples of the \( k \) path and the \( k_z \).
Figure S6. (a) Comparison of band dispersions of LiFeAs of Ref.[S10] from the ARPES (color contour) and the LQSGW+DCA ansatz for hole pockets ($\alpha$ and $\beta$) in the $\Gamma$-M $k$ path ($k_z=0.00$) from $\sigma$ polarized light. (b) Same as (a) for $\alpha'$ from $\pi$ polarized light.[S10]

Figure S7. (a) Comparison of band dispersions of LiFeAs of Ref.[S52] from the ARPES (red circle) and the LQSGW+DCA ansatz (black dots) for $k_z=0.00$ for hole pockets ($\alpha$, $\alpha'$, and $\beta$) in the $\Gamma$-M $k$ path ($k_z=0.00$). (b) Same as (a) for $k_z=0.50-0.70$. (c) Same as (a) for $k_z=1.00$. [S52]

**SPIN ORBIT COUPLING OF LIFEAS**

In this section we estimate the admixture of $xz/yz$ and $xy$ orbital character near the Fermi level near the $\Gamma$ point. Fig[S8] presents single particle $t_{2g}$ stages in the presence of a tetragonal crystal field ($\varepsilon$) and SOC ($\lambda$) as defined by the one site Hamiltonian in Eq[S3].

$$H = \lambda (l \cdot s) t_{2g} + \frac{1}{2} \varepsilon (c_{xy,\uparrow}^\dagger c_{xy,\uparrow} + c_{xy,\downarrow}^\dagger c_{xy,\downarrow} - c_{xz,\uparrow}^\dagger c_{xz,\uparrow} - c_{xz,\downarrow}^\dagger c_{xz,\downarrow} - c_{yz,\uparrow}^\dagger c_{yz,\uparrow} - c_{yz,\downarrow}^\dagger c_{yz,\downarrow})$$ (S3)
Figure S8. (a) Multiplet states in the presence of strong tetragonal crystal field ($\varepsilon_t$), with small spin orbit coupling ($\lambda$). (b) Multiplet states in the presence of strong spin orbit coupling ($\lambda$) small tetragonal crystal field ($\varepsilon_t$). A, B, and C Kramers doublets are continuously connected from the regime of (a) to the regime of (b), presented in Eq.S5 and Eq.S6. (c) Orbital contents of A, B, and C Kramers doublet with variation of $\lambda/\varepsilon_t$. $\psi$ is the Kramer’s doublet A, B, and C of Eq.S3. $\chi_{m,\sigma}$ is the atomic $t_{2g}$ orbital ($m$: orbital, $\sigma$: spin). From the tetragonal symmetry, the $yz$ orbital have a same projection weight with respect to the $xz$ orbital. The regime for non-degenerate bands of LiFeAs is denoted from yellow color.

The $\lambda(l \cdot s)_{t_{2g}}$ of Eq.S3 is given by Eq.S4 in the order of $xz, \uparrow$, $yz, \uparrow$, $xy, \uparrow$, $xz, \downarrow$, $yz, \downarrow$, and $xy, \downarrow$ states.

$$\lambda(l \cdot s)_{t_{2g}} = \begin{pmatrix}
0 & -i\frac{\lambda}{2} & 0 & 0 & 0 & i\frac{\lambda}{2} \\
n\frac{\lambda}{2} & 0 & 0 & 0 & 0 & -\frac{\lambda}{2} \\
0 & 0 & 0 & -i\frac{\lambda}{2} & \frac{\lambda}{2} & 0 \\
0 & 0 & i\frac{\lambda}{2} & 0 & i\frac{\lambda}{2} & 0 \\
0 & 0 & i\frac{\lambda}{2} & -i\frac{\lambda}{2} & 0 & 0 \\
-\frac{\lambda}{2} & -\frac{\lambda}{2} & 0 & 0 & 0 & 0
\end{pmatrix} \tag{S4}$$

The strong and weak $\lambda$ cases are shown in Fig.S8(b) and Fig.S8(a), respectively. The eigenvectors of the Hamiltonian Eq.S3 are Kramer’s doublets A, B, and C. Their orbital content is shown schematically as a function of $\lambda/\varepsilon_t$ in Fig.S8.

$$|m_z|s_z, s_z = \uparrow = -\frac{1}{\sqrt{2}}(|xz, \uparrow> + i|yz, \uparrow>)$$

$$|m_z|s_z, s_z = \downarrow = \frac{1}{\sqrt{2}}(|xz, \downarrow> - i|yz, \downarrow>)$$

$$|m_z|s_z, s_z = \pm \downarrow = -\frac{1}{\sqrt{2}}(|xz, \downarrow> + i|yz, \downarrow>)$$

$$|m_z|s_z, s_z = \pm \uparrow = \frac{1}{\sqrt{2}}(|xz, \uparrow> - i|yz, \uparrow>)$$

$$|xy, \uparrow>$$

$$|xy, \downarrow>$$

$$|x, \uparrow>$$

$$|x, \downarrow>$$
$$|j_{\text{eff}} = 1/2, j_z = 1/2 > = - \frac{1}{\sqrt{3}} (|xy, \uparrow> + |yz, \downarrow> + i |xz, \downarrow>)$$

$$|j_{\text{eff}} = 1/2, j_z = -1/2 > = \frac{1}{\sqrt{3}} (|xy, \downarrow> - |yz, \uparrow> + i |xz, \uparrow>)$$

$$|j_{\text{eff}} = 3/2, j_z = 1/2 > = \frac{1}{\sqrt{6}} (2 |xy, \uparrow> - |yz, \downarrow> - i |xz, \downarrow>)$$

$$|j_{\text{eff}} = 3/2, j_z = -1/2 > = \frac{1}{\sqrt{6}} (2 |xy, \downarrow> + |yz, \uparrow> - i |xz, \uparrow>)$$

$$|j_{\text{eff}} = 3/2, j_z = 3/2 > = - \frac{1}{\sqrt{2}} (|yz, \uparrow> + i |xz, \uparrow>)$$

$$|j_{\text{eff}} = 3/2, j_z = -3/2 > = \frac{1}{\sqrt{2}} (|yz, \downarrow> - i |xz, \downarrow>)$$

(S6)

The single particle states A, B, and C of Eq. S5 and Eq. S6 is connected with the variable of $\lambda/\epsilon_t$. Fig. S8(c) presents orbital contents in the $t_{2g}$ orbital in the presence of the tetragonal crystal field ($\epsilon_t$) and SOC ($\lambda$), as a function of $\lambda/\epsilon_t$.

In the region of parameters that correspond to the $\Gamma$ point of LiFeAs, $\epsilon_t$ correspond to $\sim 250$ meV $\lambda_{SOC}=50$ meV, within LDA, hence the ratio $\lambda_{SOC}/\epsilon_t$ is less than 0.2 for LiFeAs. This estimate of the upper limit of $\lambda_{SOC}/\epsilon_t$ also holds for LQGW+$\lambda_{SOC}$.

**TRANSPORT PROPERTIES OF LIFEAS AND THE LQGW+DMFT ANSatz**

In this section, we analyze transport data to show that a spatially local scattering rate of LiFeAs, motivated by the LQGW+DMFT ansatz, provides a natural explanation for the transport experiments of Ref. [S57]. We can view this as an independent corroboration, that the scattering rate is spatially local, while strongly orbital dependent can be taken to be the same in the electron and hole pockets.

Fig. S9 presents the experimental temperature dependent resistivity ($\rho$) and the experimental temperature dependent Hall coefficient adapted from Ref. [S57]. This data shows that the Hall coefficient is electron like, most prominently around the temperature of $\sim 100$ K. With lowering of the temperature below $\sim 100$ K, this electron like Hall conductivity is reduced. Around the temperature of $\sim 100$ K, the resistivity data shows that upon cooling from this temperature, a Fermi liquid behaviour is obtained as $\rho$ proportional to $T^2$.

Figure S9. Experimental temperature dependent resistivity ($\rho$) of LiFeAs (left panel), and experimental temperature dependent Hall coefficient ($R_H$) of LiFeAs (right panel), adapted from Ref. [S57].
Table S7. Mass of the bare band structure \((m_\nu, \nu=\alpha, \beta, \gamma, \text{and } \delta)\) of LQSGW+\(\lambda_{SOC}\) in the unit of the free electron mass \((m_e)\), and the charge carrier number \((n_\nu)\) of the LQSGW+DCA ansatz from Table S3 \((k_z=0.55\) for electron bands fit).

| \(m_\alpha\) | \(m_\beta\) | \(m_\gamma\) | \(m_\delta\) |
|-------------|-------------|-------------|-------------|
| 1.73\(m_e\) | 1.36\(m_e\) | 0.79\(m_e\) | 1.20\(m_e\) |
| \(n_\alpha\) | \(n_\beta\) | \(n_\gamma\) | \(n_\delta\) |
| 0.05        | 0.36        | 0.20        | 0.26        |

We use a very simplified model of the transport summarized in Eq S7 expressing the resistivity and the Hall coefficient in terms of the mobility \(\mu\), a charge carrier number \(n\) and a scattering rate \((1/\tau)\) with the mobility \(\mu\) of each FS pocket given by Eq S8.

Assuming that the scattering rate \((1/\tau)\) depends on the orbital but not on whether the pocket is electron or hole like, we apply the scattering rate, \((1/\tau)\), of the \(xz/yz\) to \(\alpha\) and \(\delta\) Fermi surfaces, while the scattering rate of the \(xy\) is applied to \(\beta\) and \(\gamma\) Fermi surfaces. \(m_\nu\) is the bare mass of each FS which we estimate from our LQSGW reference as shown in Table S7. \(m_\alpha\) and \(m_\beta\) are estimated from the \(\Gamma-X\) \(k\)-path \((k_z=0.00)\), while \(m_\gamma\) is estimated from \(\Gamma-M\) \(k\)-path \((k_z=0.55)\). While \(m_\delta\) has substantial \(k_z\) dependence as shown in Fig S3, transport quantities involve a weighted average of occupation and mobility. In our simplified model, we use an average value 1.20\(m_e\) as summarized in Table S7.

We checked that variations in \(m_\delta\) in the range of \([1.00m_e, 1.80m_e]\) gives rise to similar trends for the extracted scattering rate, and the converted transport data.

For the carrier number of each FS, we take the value from the LQSGW+DCA ansatz as shown in Table S7.

\[
\rho^{-1}(T) = \sum_\nu n_\nu e \mu_\nu
\]

\[
R_H(T) = \left(\sum_\nu n_\nu e \mu_\nu^2 \times \text{sgn}(\nu)\right)/\sigma^2, \quad \text{sgn}(\nu)=+1 \text{ for holes, -1 for electrons}
\]

(S7)

\[
\mu_\alpha = \frac{eT_{xz/yz}}{m_\alpha}\]

\[
\mu_\beta = \frac{eT_{xy}}{m_\beta}\]

\[
\mu_\gamma = \frac{eT_{xy}}{m_\gamma}\]

\[
\mu_\delta = \frac{eT_{xz/yz}}{m_\delta}
\]

(S8)

Fig S10 presents extracted temperature dependent scattering rate \((1/\tau)\) of the \(xz/yz\) and the \(xy\) orbitals from the experimental data of \(\rho\) and \(R_H\) of Ref. S57. It is shown that the \(xz/yz\) orbital is more coherent than the \(xy\) orbital. The scattering rate of the \(xz/yz\) orbital is nearly proportional to \(T^2\) from 0 K \((0 K^2)\) to 265 K \((70000 K^2)\), implying that a Fermi liquid behaviour for this temperature range. The scattering rate of the \(xy\) orbital is nearly proportional to \(T^2\) from 0 K \((0 K^2)\) to 100 K \((10000 K^2)\), and there is a coherence-incoherence crossover around the temperature of 150 K. This result of the scattering rate (Fig S10) from the transport data of Fig S9 implies that (i) the \(xy\) orbital is more incoherent than the \(xz/yz\) orbital, and (ii) there is no anomalous behaviour for the \(xz/yz\) orbital.

To check the validity of the orbital dependent spatially local scattering rate in Fig S10, we compute magnetoresistance (MR) coefficient, \(a\), from the scattering rate using the formulation of Eq S9. Fig S11 compares the computed MR coefficient with its experimental value of Ref. S57. It is shown that the computed MR coefficient is consistent with the experimental MR coefficient, validates the local ansatz for the orbital dependent scattering rate.
Figure S10. Extracted scattering rate \(1/\tau\) of \(xz/yz\) and \(xy\) orbitals from the data of Fig. S9 [S57] and using Eq. S7, Eq. S8, and Table S7.

Figure S11. Computed temperature dependent magnetoresistance coefficient \(\alpha\) from Eq. S9 and Eq. S8, using the extracted scattering rate \(1/\tau\) of Fig. S10 with Table S7 of the LQSGW+DCA ansatz (blue line). The computed \(\alpha\) is compared with its experimental value adapted from Ref. [S57] (red dots).

\[
\alpha(T) = \frac{\delta \rho(T, H)}{\rho(T, 0) H^2} = \frac{\delta \sigma(T, H)}{\sigma(T, 0)} - \left( \frac{\sigma_{xy}}{\sigma_{xx}} \right)^2 \sigma_{xy} = H \sigma_{xx}^2 R_H
\]

\[
\left( \frac{\sigma_{xy}}{\sigma_{xx}} \right)^2 = H^2 \sigma_{xx}^2 R_H^2 = H^2 \sigma^2 R_H^2
\]

\[
- \frac{\delta \sigma(T, H)}{\sigma(T, 0)} = \left( \sum_\nu n_\nu e^2 \mu_\nu^3 / |\sigma| H^2 \right)
\]

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