Band structure parameterization

A tight-binding parameterization of the DFT band structure, e.g. the one by Eschrig and Koepernik [1], carries the advantage of including all five 3d orbitals of Fe. It does not, however, completely agree with the available experimental data. In particular, the Fermi pocket shapes are not in complete agreement with STM or ARPES [2-34] measurements, and it predicts a larger Fermi velocity (by a factor of ~3) compared to what is measured by STM or ARPES [2-34], in addition to different Fermi pocket shapes. While this should not influence in a qualitative manner the mode-coupling signature produced by the model \( E_g \) phonon, which has no momentum structure, the signature produced by coupling to AFSF depends crucially on such details of the band structure. We have thus constructed a two-dimensional tight-binding model, focusing mostly on the Fermi surface shape and the Fermi velocities of the \( \gamma \) pocket and the two electron pockets. The dispersion of each of the bands is fit to the following form:

\[
E^k = \sum_{n,m=0}^{2} t_{nm}^k \cos(nk_x) \cos(nk_y),
\]

where \( k \) is the band index. The parameters \( t_{nm}^k = t_{mn}^k \) are presented in Table. S1. By treating each of the bands independently, we have sacrificed the orbital character information. The Fermi surface and dispersion along high symmetry points are presented in Fig. S1.
**Figure S1.** (a) Fermi surface of our parameterized tight binding model in two-Fe Brillouin zone. (b) The dispersion along high symmetry points.

<table>
<thead>
<tr>
<th>$t^k_{nm}$ (eV)</th>
<th>Band $k = 1$ (Hole pocket)</th>
<th>Band $k = 2$ (Electron pockets)</th>
<th>Band $k = 3$</th>
</tr>
</thead>
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<tr>
<td>$t^k_{00}$</td>
<td>-0.0816</td>
<td>0.115</td>
<td>0.136</td>
</tr>
<tr>
<td>$t^k_{01}$</td>
<td>0.1046</td>
<td>0.065</td>
<td>0.057</td>
</tr>
<tr>
<td>$t^k_{02}$</td>
<td>-0.0350</td>
<td>0.025</td>
<td>0</td>
</tr>
<tr>
<td>$t^k_{11}$</td>
<td>-0.0610</td>
<td>-0.070</td>
<td>-0.074</td>
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<tr>
<td>$t^k_{12}$</td>
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<td>0</td>
</tr>
<tr>
<td>$t^k_{22}$</td>
<td>-0.0230</td>
<td>-0.020</td>
<td>0</td>
</tr>
</tbody>
</table>

**Table S1.** Tight binding parameters for the band parameterization.
(II) Perturbative calculation of the self-energy in the presence of electron-boson coupling in the superconducting phase.

The electron-AFSF coupling in a multi-orbital system takes the form

\[
H_{e-b}^{\text{AFSF}} = \frac{1}{\sqrt{N}} \sum_{k,q} g_{k+q,k}^{\alpha\beta} (\vec{\sigma}_q + \vec{\sigma}^+_q) \cdot (c_{k+q,\alpha s}^\dagger \tilde{s}_{ss'}^c c_{k,\beta s'}),
\]  

(S2)

where \(\vec{S}_q\) represents a spin fluctuation of momentum \(q\) and the operator \(c_{k,\alpha s}^\dagger\) creates an electron in the orbital \(\alpha\) with momentum \(k\) and spin \(s\). \(N\) is the number of sites in the system, and \(\vec{\sigma} = \sigma_x \vec{\sigma} + \sigma_y \vec{\gamma} + \sigma_z \vec{\zeta}\) is the Pauli vector. Here and in the following, repeated orbital and spin indices should be summed over. Note that the coupling function \(g_{k+q,k}^{\alpha\beta}\) is a matrix in orbital space. The full self-energy due to this coupling can be formally represented in Nambu space as

\[
\hat{\Sigma}_{\alpha\beta}(k, i\omega_n) = -\frac{T}{N} \sum_{q, i\nu_n} D(q, i\nu_n) \hat{g}^{\alpha\gamma}
\]

\[
\times \hat{G}_{\gamma\delta}(k + q, i\omega_n + i\nu_n) \tilde{\Lambda}^{\delta\beta}(k, k; i\omega_n + i\nu_n, i\omega_n),
\]  

(S3)

where \(T\) is temperature, and \(D(q, i\nu_n)\) is the dressed Boson propagator with momentum \(q\) and bosonic Matsubara frequency \(\nu_n\). \(\hat{g}_{k+q,k}^{\alpha\beta} = g_{k+q,k}^{\alpha\beta} \hat{\sigma}_0\) and \(\tilde{\Lambda}^{\alpha\beta}(k + q, k; i\omega_n + i\nu_n, i\omega_n)\) are bare and dressed vertex functions, respectively [Fig. S2(a)].

The electron-phonon coupling takes the form of

\[
H_{e-b}^{\text{ph}} = \frac{1}{\sqrt{N}} \sum_{k,q} g_{k+q,k}^{\alpha\beta} (a_q + a_{-q}^\dagger) c_{k+q,\alpha s}^\dagger c_{k,\beta s'},
\]  

(S4)

where \(a_{-q}^\dagger\) is a phonon creation operator and the structure of the coupling matrix \(g_{k+q,k}^{\alpha\beta}\) is given by the symmetry of the phonon mode. This leads to the same form of the self-energy

![Diagram](image)

**Figure S2.** (a) Diagrammatic representation of the exact self-energy and (b) the (lowest-order) approximation used in this work.
as Eq. (S3), except that the coupling constant in Nambu space now becomes \( g^{\alpha\beta}_{k+q,k} = g^{\alpha\beta}_{k+q,k} \hat{\gamma}_2 \).

For conventional strong-coupling superconductors, it was possible to reveal the signatures of electron-phonon coupling through tunneling spectroscopy and to confirm the coupling as a driver for superconductivity through a quantitative fit of the tunneling data to the Eliashberg theory \([5]\). Using the traditional Migdal-Eliashberg treatment of phonon-mediated superconductivity, the Dyson equation \([\text{Eq. (1)}\) main text\] is solved self-consistently, ignoring vertex corrections due to the separation of scales between the electron mass and the atomic mass.

However, attempting to use this approach for Fe-pnictides presents two issues: First, the absence of a separation of scales for spin-fluctuation-driven superconductivity makes it challenging to carry out an analogous theoretical procedure. Second, even without vertex corrections, the self-consistent evaluation of Eq. (S3) for a multi-orbital system requires heavy numerical calculations \([6, 7]\). Interpreting and distilling the obtained results into experimentally distinguishable features with simple insight can be a challenging task.

Instead, we take a perturbative approach of computing the self-energy to the lowest order:

\[
\Sigma_{\alpha\beta}(k, i\omega_n) = -\frac{T}{N} \sum_{q, i\nu_n} D^0(q, i\nu_n) \hat{\gamma}^{\alpha\gamma} \hat{\gamma}^{\beta\delta}_{k+q}(k + q, i\omega_n + i\nu_n) \hat{\gamma}^{\delta\gamma},
\]  

(S5)

using the free Bogoliubon propagator \( \hat{\gamma}^0_{k+q}(k, i\omega_n) \) and free boson propagator \( D^0(q, i\nu_n) \) based on experimental input \([\text{Fig. S2(b)}]\). \( \hat{\gamma}^0_{k+q}(k, i\omega_n) \) can be written as

\[
[\hat{\gamma}^0_{k}(k, i\omega_n)]^{-1} = i\omega_n \hat{\gamma}_0^0 - \Delta_k \hat{\gamma}_1^1 - H_k^0 \hat{\gamma}_3^3,
\]  

(S6)

where \( H_k^0 \) and \( \Delta_k \) are the kinetic energy and gap function of non-interacting Bogoliubons with momentum \( k \). For simplicity, we consider the coupling within the Holstein model, i.e. \( g^{\alpha\beta}_{k+q,k} = g^{\alpha\beta} \). Note, however, that the momentum structures of the two bosons we consider here are distinctly different. \( D^0(q, i\nu_n) \) is peaked around a nonzero momentum \( Q \) for the AFSF, while it is momentum independent to a good approximation for the optical \( E_g \) phonon. For notational simplicity, we omitted the superscript 0 for the bosonic propagator in the main text. However, the bosonic propagator used for the main results were bare propagators in the sense that they do not include effects of interaction with fermions.

One should not think of this approach as double counting the effect of a bosonic mode, or attributing superconductivity and high-energy kink feature to two distinct modes.
Rather, we are separating the role of electron-mode coupling into two. The superconductivity, which possibly is driven by the mode of interest, is taken into account with BCS theory. Being a mean-field approach, BCS theory by itself cannot account for the dynamic properties of the mediating boson, and thus by design cannot include the effect of mode-coupling at high energy. Dressing the Bogoliubov quasiparticle with a bosonic mode can then produce the kink feature. Figure S3 shows the density of states calculated with this approach for the case of a conventional s-wave superconductor. It is in very good agreement with experiments on planar Pb-MgO-Mg tunnel junctions [8]. A similar perturbative approach has been used previously for electron-mode-coupling studies of cuprate superconductors [9]. One shortcoming of this approach is that we cannot test whether the given boson can give rise to superconductivity of the observed transition temperature, as we are starting with the BCS superconducting state. However, we aim at finding features in the renormalization of the electronic spectral properties that can serve as fingerprints of each electron-boson coupling, and gain simple insight into the problem. As apparent from Eq. (S5), the non-trivial orbital dependence can affect these features through the structure of the coupling-constant matrix and the resulting features will be sensitive to dispersions of both bosons and fermions. In particular, the energy at which an observable feature appears may be non-trivially related to the mode energy, when the mode couples to the Bogoliubon propagator connecting different pockets.

Figure S3. Density-of states of a typical s-wave superconductor with pairing $\Delta$ coupled to optical phonon of frequency $\Omega$, computed with perturbative approach.
(III) Models for bosonic modes

(a) Resonant Antiferromagnetic Spin Fluctuation

In superconducting Fe-pnictides with s± gap structure, inter-band scattering can lead to resonant antiferromagnetic spin fluctuation (AFSF) peaked at \( \mathbf{q} = \mathbf{Q} = (\pi, \pi) \) [10, 11]. This mode has been identified in inelastic neutron-scattering measurements, where magnetic correlations peaked near \( \mathbf{Q} \) and energy 6—10meV have been observed [12, 13]. To model a bosonic mode with finite correlation length \( \xi \) centered at \( \mathbf{Q} \), one would typically write down a propagator such as \( D(\mathbf{q}, \omega) \propto (\omega^2 + \nu^2 (\mathbf{q} - \mathbf{Q})^2 + \nu^2 / \xi^2) \). This however, introduces a dispersion to the mode, and integrating over the energies of the mode, which is required, is computationally costly. In order to capture the effects of electron-mode coupling at low energy, especially focusing on the lowest energy where it starts to appear, we instead model the resonant mode with a simple phenomenological model which is peaked at \( (\mathbf{Q}, \Omega) \) with \( \Omega = 6 \text{meV} \) whose spectral weight is \( B(\mathbf{q}, \omega) = \chi(\mathbf{q}) (\delta(\omega - \Omega) + \delta(\omega + \Omega)) \). We assume a gaussian distribution in the momentum space

\[
\chi(\mathbf{q}) = \frac{\exp\left(-\frac{1}{2} \xi^2 (\mathbf{q} - \mathbf{Q})^2 \right)}{\frac{1}{N} \sum \exp\left(-\frac{1}{2} \xi^2 (\mathbf{q} - \mathbf{Q})^2 \right)},
\]

and corresponding propagator in terms of Matsubara frequency is given by

\[
D_{AFSF}^{0}(\mathbf{q}, i\nu_n) = -\frac{2\Omega}{\nu_n^2 + \Omega^2} \chi(\mathbf{q}).
\]

The resulting lowest-order self energy is then

\[
\left[ \Sigma_{AFSF}^{(1)}(\mathbf{k}, i\omega_n) \right]^{\alpha\beta} = g^{\alpha\mu} \left[ \frac{T}{N} \sum_{q, \nu_n} G_{\mu\upsilon}^{0}(\mathbf{k} + \mathbf{q}, i\omega_n + i\nu_n) \chi(\mathbf{q}) \frac{2\Omega}{\nu_n^2 + \Omega^2} \right] g^{\upsilon\beta}.
\]

Many theories predict that the coupling matrix is nearly diagonal in the orbital basis. However, orbital character mixing present in all pockets results in finite amount of inter-band coupling. Because of the sharpness of the momentum dependence of this model, the self-energy effect on the h3 pocket is dominated by this finite inter-band scattering due to the bosonic mode between the h3 pocket and the electron pockets, and the structure of the coupling matrix plays little role in the qualitative feature of the self-energy effect. Therefore we use the simplest model that does not distinguish different bands, and set the coupling constant to be \( g^{\alpha\beta} = g \) for all \( \alpha, \beta \). Because the points at which the self-energy effect is significant is determined by the kinematics constraint (Eq. (2) main text) and occurs at energy higher than the gap scale in our model, the anisotropy of the s± gap does not affect the anisotropy of the self-energy.

(b) Optical \( E_g \) phonon
The optical $E_g$ phonon associated with Fe vibration plays a key role in the orbital-fluctuation-driven superconductivity scenario by Kontani and Onari [14]. Following their approach, we model the phonon as an Einstein mode with the propagator

$$D_{ph}^0(q, i\nu_n) = -\frac{2\Omega}{\nu_n^2 + \Omega^2}. \quad (S10)$$

The non-trivial $E_g$ symmetry of the mode that strongly couples to orbital fluctuations dictates the coupling matrix $g^{\alpha\beta}$, which we take to be momentum-independent in the orbital basis. Note, however, that due to the changing orbital character of an electron in each band, the coupling matrix has a non-trivial momentum dependence in the band basis. The self-energy in Eq. (S5) becomes

$$\left[\Sigma^{(1)}_{ph}(k, i\omega_n)\right]^{\alpha\beta} = g^{\alpha\mu} \left[\frac{T}{N} \sum_{k',\nu_n} G_{\mu\nu}^0(k', i\nu_n + i\nu_n) \frac{2\Omega}{\nu_n^2 + \Omega^2}\right] g^{\nu\beta}. \quad (S11)$$

The resulting self-energy has no explicit momentum dependence, but is a function only of energy and the connected orbitals. For pockets with mixed orbital character, the resulting renormalization effect will still show anisotropic features as the orbital character varies around the Fermi surface. For the $\gamma$ pocket with almost completely $d_{xy}$-orbital character [15], the self-energy effect due to this phonon should be angle independent. This supplements the finding from the kinematic constraint perspective that not only the self-energy features show up at energy $\min(\Delta + \Omega)$ independent of the momentum, but also with uniform strength on the $\gamma$ pocket.
The self-energy effects on the electron spectral function due to the $E_g$ phonon and AFSF

In this section we discuss how the self-energy renormalizes the spectral weight in a multi-orbital system. Treating the orbitals equally and tracing over the orbital indices, the spectral weight $A(k, \omega)$ can be written in terms of the ‘11’-component of the renormalized Nambu Green’s function as

$$A(k, \omega) = -2 \text{Im} \, \text{tr} \left[ \tilde{G}(k, i\omega_n) \right]_{i\omega_n \rightarrow \omega + i0^+}^{11} \quad (S12)$$

where the trace runs over all orbital indices. The superscript indicates a component in Nambu space. The spectral weight has the advantage that it can be straightforwardly related to the self-energy. Direct measurement of electronic spectral weight using ARPES is however limited to the negative energy part of the spectra.

The negative-energy spectral functions including the self-energy effects due to AFSF and $E_g$ phonon discussed in the SI section (II) are presented in Fig. S4 through waterfall plots along two cuts of the $\gamma$ band. With both bosons the cuts along the Fe-As direction [Fig. S4(a)(d)] and along the Fe-Fe direction [Fig. S4(c)(f)] do not show a clear difference in terms of detectable kink features. While the anisotropy in the bare dispersion is visible for both cases, there is no observable anisotropy in the kink features for different reasons. For AFSF (Fig. S4 a-c), the self-energy effect is strongly suppressed, due to the absence of points which satisfy the kinematics condition Eq. (2). For phonons(Fig. S4 d-f), the kink should be present but isotropic due to the uniform orbital character of the $h_3$ band.

Figure S4 shows how anisotropic self-energy would not be detectable in the negative energy range on the $h_3$ pocket of LiFeAs, even if the system has strong electron-mode coupling. Other bands could still exhibit features due to mode coupling at negative energy, which would appear in the average tunneling spectrum. Indeed the average tunneling spectrum shown in Fig 4b shows features at both positive and negative energy. However the average tunneling spectrum is insufficient to extract details of the bosonic mode. Hence we focused on the Fermi surface specific features in the positive energy range where AFSF is predicted to give rise to anisotropic kink features.
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**Figure S4.** Momentum distribution curves (MDC) at different energies along three different cuts on the $\gamma$ band at negative energies. The angle for each cut is measured from Fe-As direction. (a-c) are calculated in the presence of coupling to the AFSF described in Eq. (S9) with energy 6meV using the band structure given in the SI section (III). (d-f) are calculated in the presence of coupling to the $E_g$ phonon described in Eq. (S11) with energy 15meV using the orbital based band structure in Ref. 1. The red dot on each curve indicates the peak position. Note that the band structure we used for the $E_g$ phonon was taken from Ref.[1] in order to reflect the orbital specific coupling, on the other hand the band structure we used for the coupling to AFSF was fitted to the experimental band structure. The dispersion around $h_3$ pocket is different for each of these band structures. In order to compare the results of mode-coupling, we have plotted the MDC curves over different ranges of momenta.
In this section, we describe in detail how the theoretical prediction of the QPI pattern using the standard T-matrix formalism is carried out \([16, 17]\). Within this formalism, a \(\delta\)-function scatterer at \(\mathbf{x} = 0\) with strength \(\hat{\mathcal{V}}\) causes an off-diagonal (in momentum) correction \(\delta \hat{G}_{\alpha\beta}(\mathbf{k}, \mathbf{k}', \omega)\) to the Bogoliubon propagator given by

\[
\delta \hat{G}_{\alpha\beta}(\mathbf{k}, \mathbf{k}', \omega) = \hat{G}_{\alpha\gamma}(\mathbf{k}, \omega) \hat{T}_{\mathbf{k}, \mathbf{k}'}^{\gamma\delta} \hat{G}_{\delta\beta}(\mathbf{k}', \omega),
\]

with the T-matrix

\[
[\hat{T}_{\mathbf{k}, \mathbf{k}'}(\omega)]^{-1} = \hat{\mathcal{V}}^{-1} - \sum_{\mathbf{k}} \hat{G}(\mathbf{k}, \omega).
\]

We further assume weak scattering and take the Born approximation. In principle, full orbital information of the scatterer is needed, which is especially difficult to obtain when working with a parameterized band structure. Therefore, we consider the simplest case in which the scattering is equal in all channels, i.e. \(\hat{T}_{\mathbf{k}, \mathbf{k}'}^{\alpha\beta} = \mathcal{T}\) for all \(\alpha\) and \(\beta\). This approximation enables us to find the correction to the Bogoliubon propagator due to impurities either in orbital or in band basis. We thus find for the change in the Green’s function

\[
\delta \hat{G}_{\alpha\beta}(\mathbf{k}, \mathbf{k}', \omega) = \sum_{\mathbf{k}} \hat{G}_{\alpha\gamma}(\mathbf{k}, \omega) \mathcal{T}_{\gamma\delta} \hat{e}_{\delta\beta}(\mathbf{k}', \omega).
\]

The resulting Fourier components of the density of states read

\[
\delta n(\mathbf{q}, \omega) \propto \text{Im} \sum_{\mathbf{k}} [\hat{G}_{\alpha\beta}(\mathbf{k}, \omega) \mathcal{T}_{\beta\gamma} \hat{e}_{\gamma\alpha}(\mathbf{k} + \mathbf{q}, \omega)]^{11}.
\]

Note that the frequently used approximation of convoluting two spectral functions (imaginary parts of Green functions) amounts to substituting the real part of the response function by the imaginary part (see Eq. S9 in Ref 18 for instance). We have not used such approximation. Instead, we have carried out the full T-matrix calculation to the leading order.
(VI) Measurement of $g(r, \omega)$ and $g(q, \omega)$

This section gives a brief overview of the experimental methods used to obtain the local density of states data, $g(r, \omega)$, and $g(q, \omega) = \text{FFT}[g(r, \omega)](q, \omega)$. Quasiparticle interference (QPI) imaging technique with spectroscopic-imaging scanning tunneling spectroscopy (SI-STS) has been recently used to study the electronic structures of a number of Fe-pnictide compounds [4, 19-202122 23].

LiFeAs crystals were prepared in N$_2$ atmosphere, transported to the cryostat, and cleaved in-situ at around 10K, before cooling down to the base temperature of 1.2K. The cleaved surfaces are atomically flat and exhibit the $a_0=0.38$nm periodicity of either the As or Li layer. $g(r, \omega)$ data is taken using a lock-in method with typical set-up conditions of $(I_{\text{set}}, V_{\text{set}}) = (10\mu A, 10mV)$. Data was taken on a large (~90nm) square fields of view, required to achieve sufficient $q$-space resolution.

Momentum space data is extracted from the Fourier transform $g(q, \omega) = \text{FFT}[g(r, \omega)](q, \omega)$. The data is then 4-fold symmetrized and has the low $q$ features suppressed as in Ref. 12. For comparison, we show data in Fig. S5.
(VII) QPI data above $T_c$ and below the Fermi level

(a) Quasiparticle interference at elevated temperatures

We measured the QPI dispersion not only deep within the superconducting phase at 1.2K, but also at temperatures above $T_c$. However, interpretations of QPI data above $T_c$ (Figure S6) are quite challenging. Due to the inherent energy resolution of all STM data ($\Delta E(T) \approx 3.7 k_BT$), a feature like the kink is extremely difficult to detect due to thermal smearing of more than 3.5meV at 15K=$T_c$.
(VII) QPI data above $T_c$ and below the Fermi level

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Figure S5. $g(r,\omega)$ and $g(q,\omega)$ data. Left column and center column, sequence of $g(r,\omega)$ and $g(q,\omega)$ data as in Fig. 2 in the main text. Right column, raw FFT;$g(r,\omega)(q,\omega)$. The setup conditions were $(I_{\text{set}},V_{\text{set}}) = (10pA,10mV)$.

Figure S6: Dispersion of the QPI intensity (c.f. main text). The blue line is from data at 1.2K, the red line is from data at $T_c$. Error bars denote the thermal resolution of STM, which is around 3.7k_B T (smaller than the diameter of the data points for the blue points).

(b) QPI at negative energies

At negative energies, the $h_3$ QPI signal starts overlapping with an electron-hole interband scattering signal particularly along the Fe-Fe direction (See Figure R3). This in combination with diminishing signal amplitude prohibits an assumption-free analysis of the data along the lines carried out at positive energies.

Figure S7 QPI signal at -8meV. At negative energies the $h_3$ signal starts to overlap with signal originating from electron-hole interband scattering (red circle). This makes clean data analysis as used for positive energies very challenging.

(VIII) Extraction of peak maxima and peak width in the q-$\omega$ plane

The dispersions shown Fig. 5c of the main text can be extracted from the $g(|q|,\omega)$ cuts. Each cut is obtained by using data from the 3-dimensional data set $g(q,\omega)$ but only along a specific direction $|q|$. We then azimuthally average the data within an angle $\pm 10^\circ$ around the direction of $|q|$. The peak positions corresponding to the band dispersion is obtained by fitting line-cuts with a polynomial background plus a Gaussian, as shown in Fig. S8.
Figure S8. The peak positions (blue dots) described in the main text and shown in Fig. 4 are extracted by fits to the $g(|q|, \omega)$ curves (black markers) in different directions (a-e). The data is fitted to a Gaussian plus a 2nd order polynomial background (red curve). The blue dots are the positions of the fitted Gaussian peak, and sometimes lie slightly offset of the maximal value. To avoid compromising the signal with the low-$q$ background stemming from long-range modulations and impurity-atoms, we only fit the data in a range surrounding the peak maximum. The inset in (a) shows an example of a fit with the contribution of only the Gaussian in green.
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Bibliography