Microscopic Description of Unconventional Nodal Superconductivity in FeSe

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Finding of unconventional superconductivity (USC) in FeSe in an electronic “normal” state with broken $C_{3v}$ symmetry testifies to the diversity of pairing states in Fe-based superconductors. Moreover, such USC emerges as a direct instability of a normal state without Landau Fermi liquid quasiparticles, increasingly dubbed a “strange” metal. Here, we combine inputs from a first-principles correlated electronic structure method (LDA+DMFT) and symmetry analyses to propose a novel mechanism for unconventional nodal superconductivity as a direct instability of an incoherent bad metal without Landau Fermi-liquid quasiparticles. We find that a ferro-quadrupolar order, with novel spin quadrupolar correlations enhances orbital-selective Mottness in FeSe, and competes with unconventional, nodal superconductivity with $s_d$ pair symmetry. We support our proposal by demonstrating good accord with spectral and magnetic fluctuation data, and rationalize the strain and pressure dependence of $T_c$ by appealing to competition between superconductivity and electronic ferro-quadrupolar order.

I. INTRODUCTION

Unconventional Superconductivity (USC) in Iron-based pnictides and chalcogenides can seemingly host a range of gap function symmetries, a surprising fact that seems to be related to their intrinsically multi-orbital (MO) character (in contrast to cuprates, where $d$-wave SC is universal). The essential idea is that suitable external perturbations sensitively modify the anisotropic band structure in a band-selective way, and it is then conceivable that SC gap nodes may or may not exist, depending ultimately upon whether or not the SC gap function $\Delta(k)$ intersects the multi-sheeted, renormalized Fermi surface. Finally, as in cuprates and some $f$-electron systems, USC is now found to generically arise from a non-Landau Fermi liquid (nLFL) metal near optimal doping, precluding description of USC as a conventional instability of an itinerant LFL metal.

The “simplest” FeSe system is an especially suitable case in point. Additional unique features here are: (i) SC (reaching $T_c = 37$ K under high pressure) and with a record high $T_c \simeq 100$ K in FeSe films deposited on SrTiO$_3$ substrates occurs in a local moment metal without antiferromagnetic (AF) order (though AF order appears at high pressure), in contrast to other cases where SC at ambient pressure always occurs close to AF order. (ii) USC occurs in a “normal” state which is simultaneously an incoherent nLFL metal and breaks the fourfold discrete lattice rotational symmetry, as in some underdoped 122-FeAs systems. Whether (i) this “normal” state thus hosts orbital nematic order, and its relation (competitor or facilitator) to SC, and (ii) do they originate from the same set of effective interactions in the “normal” incoherent metal, are intriguing issues. (iii) Tunneling data reveal nodal-SC for pure FeSe which, remarkably, evolves into nodeless-SC with Te substitution (iv) Tensile strain rapidly suppress Sc, which, at first glance, is not inconsistent with multiple orbitals and gap nodes.

Taken together, (i)–(iv) imply challenging constraints for a theory of SC in FeSe$_{1-x}$Te$_x$: (i) poses an obvious challenge to theories relying on AF quantum criticality, since AF is only found in Te-rich and/or pressurized samples, though this does not exclude strong, short-range dynamic spin fluctuations. As pointed out above, both, the nature and extent of orbital-dependent electronic structure reconstruction (O-ESR) in FeSe is crucial to distinguishing nodeless versus nodal SC. That the observed anisotropy is much greater than its pure structural counterpart favors a predominantly electronic origin for O-ESR. In light of (i)–(ii), understanding (iii) must now involve interplay between the form-factor of $\Delta(k)$ and changes in the anisotropic correlated electronic structure with $x$.

In this work, we develop a theory to address these issues. In a non-trivial extension of earlier work, we study the origin and consequences of the two-fold ($C_{2v}$) symmetry in the normal and SC states in FeSe. In fact, for FeSe, one has to look for USC in the $C_{2v}$ structure, a difference that has important implications for pair symmetry. Earlier (Hartree-Fock) Bogoliubov approach includes $C_{4v} \rightarrow C_{2v}$ symmetry breaking phenomenologically. Our microscopic approach involves deriving both orbital nematic (ON) and USC orders from a specific residual interaction that we argue becomes more relevant than the incoherent one-particle mixing in a nLFL metal.

Specifically, two LDA+DMFT studies find a qualitatively similar “normal” state: an incoherent nLFL metal without LFL quasiparticles, argued to occur either via a lattice orthogonality catastrophe via spin-orbital freezing in a metallic, orbital-selective Mott phase (OSMP) or via the related Hund’s metal route. Already in the “normal” state at high $T$, it is known that orbital-selective Mott correlations selectively localize the $xy$ states. What is important for our purposes here is that the resulting orbital-selective (OS) Mott state and reconstruction of LDA Fermi surfaces (FS) breaks the analytic continuation with any renormalized FL metal that results from the unconstructed LDA Fermi surfaces (which is indeed what would happen if local multi-orbital correlations were weak and selective-Mott localization of $xy$-states would not occur). We argue that the resultant irrelevance of coherent one-electron mixing ($H_{\text{bhop}}^{\text{IL}}$) the OSMP that occurs at intermediate-to-strong coupling precludes the “weak coupling” BCS-like instability to a superconductor. In earlier work, we argued that these now arise via a route first expounded by Anderson in the v
cuprate context: the essential idea is that upon coupling two Luttinger liquids (LLs), coherent one-electron hybridization scales to irrelevance because of spin-charge separation, but, to higher (second) order, spinon or holon pairs can coherently tunnel between two LLs. Since the “normal” state we find is a local non-Landau FL metal, it is the residual inter-site interaction (formally obtained by replacing LL chains in Anderson’s proposal with local sites in our case) which is preferentially relevant relative to the one-electron interband hybridization, and generates the instabilities to ordered states directly from the incoherent metal (see below). In other words, in analogy with Anderson’s idea, irrelevance of $H_{\text{hyb}}^{(1)}$, itself makes the corresponding two-particle hopping (appearing as a residual interaction, $H_{\text{res}}$) more interesting. Interestingly, the same $H_{\text{res}}$ also generates an (electronic) ON instability in the competing particle-hole (p-h) channel, allowing us to study their interplay.

II. THEORY AND RESULTS

The Fermi pockets of tetragonal-FeSe in the local-density approximation (LDA) with $C_d$ symmetry are predominantly composed of the Fe-$d_{zx,zy,zy}$ orbital states: the central hole pocket at $\Gamma = (0,0)$ point is mainly of $yz, xz$ character, while the electron pockets at $M = (\pm \pi, 0), (0, \pm \pi)$ have mainly $xy, xz$ orbital component. For simplicity, we focus on the $xz, yz$ orbitals: due to interband proximity effect, the remainder of the $d$ orbitals will also play a role in reality. We first show how an electronic (orbital) nematic drives FeSe into an OSMP with co-existent localized ($xz$) and bad-metallic ($yz$) states. The one-electron hopping matrix in this orbital sector, formally written as $H_{\text{hyb}} = \sum_{i,j,a,b} t_{ab}(c_i^\dagger \sigma \epsilon_{i,j,a,b} c_j \sigma + H.c.)$, with $i,j$ being nearest (n.n) and next-nearest neighbors (n.n.n), is strongly geometrically frustrated (GF) in the FeAs(Se) systems with a large ratio of n.n.n (diagonal) to n.n hopping strengths: $t_{ab}^{(n.n.n)} / t_{ab}^{(n.n)} \approx 0.7 - 1.0$. As discussed before, residual two-particle interactions, i.e., hopping processes to second-order in $H_{\text{hyb}}$, are more relevant in the non-LFL “normal” state. As derived before, this reads

$$H_{\text{res}} \approx - \frac{1}{U + \gamma H} \sum_{\langle i,j\rangle,a,b,\sigma,\sigma'} t_{ij,a,b} c_i^{\dagger \sigma} \epsilon_{i,j,a,b} c_j^{\sigma'} \epsilon_{i,j,a,b} c_j^{\sigma'} + H.c.) \ .$$

This resembles a generalized effective spin-orbital superexchange, now ipso facto validated by the selective-Mott nature of the “normal” state. It is important to emphasize that this reflects the inherent spin-orbital entanglement in multi-orbital correlated systems, and implies that the spin-fluctuations are intrinsically orbital-selective: in particular, onset of an OSMP will enhance anisotropy of spin fluctuations. Thus, the form of $H_{\text{res}}$ shows that entangled short-ranged dynamical spin-orbital correlations facilitate instabilities of the incoherent “normal” state to competing orders. This is because $H_{\text{res}}$ can now be readily decoupled into (a) particle-hole (p-h) and (b) particle-particle (p-p) mean fields, a procedure exact within dynamical mean-field theory (DMFT), since both scale as $1/D$. Here, (a) corresponds to either ON ($\sigma = \sigma'$) or AF-SDW ($\sigma' = -\sigma$) orders, while (b) represents an USC instability. Since SC in FeSe arises after the lattice distortion (implying ON) has set in, we adopt the stratagem of first studying the ON instability, and then focus on the USC instability, with very interesting consequences. In momentum space, $H_{\text{res}}$ looks very suggestive:

$$H_{\text{res}} \approx - \sum_{k,k',a,b,\sigma,\sigma'} V_{ab} \gamma(k) \gamma(k') c_{k,a,\sigma}^{\dagger} \delta_{k,a,\sigma} \delta_{k',a,\sigma} - c_{-k,b,-\sigma}^{\dagger} c_{-k',b,-\sigma} c_{k',a,\sigma}^{\dagger} c_{k,a,\sigma} + H.c.) \ .$$

where $\gamma(k) = (c_x + c_y) + \alpha c_z$ and $\alpha = \cos k_x, k = x, y$. $\alpha =O(0.7)$ is large, thanks to the strong GF in FeSe, and $V_{ab} = \frac{t_{ab}^{(n.n.n)}}{t_{ab}^{(n.n)}}$. This can be decoupled as

$$H_{\text{res}}^{MF} = - \sum_{k,k',a,b,\sigma,\sigma'} V_{ab} \gamma(k) \gamma(k') \[(1 - \langle n_{k,a,b} \rangle) n_{k,\sigma} + \langle n_{k,a,b} \rangle \langle n_{k,a,b} \rangle - \langle n_{k',a,b} \rangle \langle n_{k',a,b} \rangle - c_{-k,b,-\sigma}^{\dagger} c_{-k',b,-\sigma} c_{k',a,\sigma}^{\dagger} c_{k,a,\sigma} + H.c.) \ .$$

ON order arising as a consequence of ferro-orbital order (FOO) directly arises from the first term in $H_{\text{res}}^{MF}$ above, which pushes the $b$-fermion ($xz$-orbital) states below the $a$-fermion ($yz$-orbital) states by an amount $-\sum_{ab} \gamma(k) \langle n_{k,a,b} \rangle$. The resulting FOO is described by an order parameter, $Q_{xz} = (n_{x,z} - n_{y,z})$, which is precisely the ferro-quadrupolar order (QOO) parameter, couples to an appropriate symmetry-adapted phonon mode and drives $C_{q_y} \rightarrow C_{q_x}$, symmetry breaking and to orthorhombicity: thus, the $T$-dependence of the FOO is reflected in that of the orthorhombicity, $< O > = \frac{Q_{xz}}{Q_{ocz}}$. Furthermore, the fact that it shows a mean-field $T$-dependence is also in accord with our view, since we derive this instability by a mean-field decoupling of $H_{\text{res}}$. USC arises from the third term. The second term in $H_{\text{res}}^{MF}$ is especially interesting: it reads $H_{\text{res,ph}}^{MF} = \sum_{a=x,z} \Delta_{ph}^{a} \sum_{k} \gamma(k) n_{a}^{(k)}$ with $\Delta_{ph}^{a} = \sum_{k} \gamma(k) \langle n_{a}^{(k)} \rangle$, which is precisely the bond-orbital order (BOO) needed to reconcile ARPES dispersions in the electronic nematic phase in FeSe. Thus, remarkably, this specific feature of the ON state in FeSe (and possibly in other Fe pnictides as well) falls out naturally as a consequence of the instabilities arising from residual two-particle (inter-site and intra- as well as inter-orbital) interactions in the incoherent nLFL “normal” state we find. We defer detailed comparison with FS to future work, but emphasize here that our proposed mechanism is naturally consistent with very recent ARPES work. A related proposal has recently been made within a slave-rotor approach who use the same terms as those in our $H_{\text{res}}^{MF}$ phenomenologically. Here, this term (precisely the last term in Eq. (4) of Yu et al) is derived from a leading-order two-particle residual interaction generated from an incoherent “normal” state. That this on-site FOO plus BOO, or a FOO state is the main competitor to USC, at least in FeSe, is also bared by generalizing the argument of Podolski et al. The pseudospin generator $\Delta_{ph}^{a} = \frac{1}{V} \sum_{k,a,x,z} \epsilon_{k,a} c_{k,a,\sigma}^{\dagger} c_{k,a,\sigma}$ rotates the bond-orbital parameter $\Delta_{ph}^{a}$ above the $c_{-}\cdot\cdot$ SC order parameter, $\Delta_{ph}^{a} = \Delta_{ph}^{a} \cdot \cdot$; these orders thus compete (a reflection of both arising from the same $\Delta_{ph}^{a}$).
$H_{\text{rel}}$). Within the $xz-yz$ orbital sector, the above relation also suggests the emergence of a hidden SO(6) invariance at a transition between ON+BOO and USC states. Remarkably, terms which break this emergent symmetry, such as finite chemical potential (doping) and strain (which produces explicit finite orbital polarization) can lead to a transition between ON+BOO and USC orders: these may already have been seen. In FeSe, ON order pushes the $xz$ (b) band below its “normal” state value by $\nu_{ab}$ (first term in $H_{\text{rel}}$ above). The non-existence of the $d_{xz}$ Fermi pocket in ARPES data is a Fermi surface reconstruction arising from a p-h order-induced downshift of the $xz$-band states as above. This also naturally accounts for a Lifshitz transition at $T^\star$. Indeed, many observations have been interpreted in terms of a Lifshitz transition: we posit that this is associated with enhancement of orbital selectivity by orbital nematicity arising from FOO+BOO as above. Very recent ARPES data are completely consistent with this result as well: the $xz$-orbital states are not seen in data and, furthermore, using only the $yz$-Fermi pocket to compute ARPES spectra in the USC state agree well with data as well. In fact, the USC scales with the $d_{xy}$-orbital spectral weight. Again, while this is put in by postulating an orbital polarization, such a term, driving a Lifshitz transition via enhanced orbital selectivity, arises from residual interactions ($H_{\text{rel}}$) in our work, and involves mutually entangled, intersite and interband spin-orbital correlations (not spin-orbit coupling). These features explicitly break SO(6) symmetry stabilizing ON+BOO order (without such symmetry breaking USC would always win over ON+BOO); thus, the onsite FOO, now arising from residual interactions, creates the proper conditions for emergence of BOO that competes with USC. This finding may have much broader applicability: in cuprates, recent work also indicates a bond-modulated order that may be the leading competitor to $d$-wave SC, at least in hole-doped cuprates.

However, since USC now occurs in the ON phase with $C_{2v}$ symmetry, it is clear that the SC pair-symmetry cannot anymore be cleanly labeled by the $A_{1g}$ irreducible representation (irrep) of $C_{4v}$. Instead, since the $A_{1g}$ irrep of $C_{2v}$ transforms like $x^2, y^2, z^2$ it now follows that the $k$-space form-factor of the symmetry-allowed SC-pair-function must rigorously be given by $\Delta_c(k) = \Delta_c(\cos k_x + \eta \cos k_y + 2\Delta \cos k_x \cos k_y)$ with $\eta \neq 1$. Remarkably, this is precisely the “anisotropic” $s_{\pm}$ form assumed phenomenologically. Our central result is that this is derived from a specific residual interaction in the incoherent metal that is also consistent with the spontaneous lowering of crystal symmetry in the “normal” state. Interestingly, for $\alpha \approx 0.7 - 1.0$, the anisotropic SC pair function will intersect the electron Fermi pockets at the $M$-points, accounting for signatures of nodal pairing in FeSe. Finally, examination of LDA FS shows small c-axis warping, so that accidental c-axis gap nodes cannot exist in FeSe. Thus, planar nodal $s_{\pm}$ SC emerges as the most probable in FeSe.

We now show how our results are in good semiquantitative accord with a host of spectral and magnetic data. The first term in $H_{\text{rel}}$ directly leads to $\langle n_{xz} \rangle > \langle n_{yz} \rangle$ by itself, implying a FOO along with a finite orbital nematicity, $\langle N \rangle = n_{xz} - n_{yz} / 2(n_{xz} + n_{yz}) > 0$ as a leading instability of the nLFL metal. $\langle N \rangle > 0$ immediately couples to an appropriate symmetry-adapted phonon mode, lowering the $C_{4v}$ crystal symmetry to $C_{2v}$. This is manifestly done in the orbital-resolved DOS in Fig. 1 which shows an enhanced pseudogap in $\rho_{xz}(\omega)$ at the expense of a reduced one in $\rho_{xy}(\omega)$; i.e., orbital-selective incoherence is enhanced in the ON phase. Though this is already consistent with STM data more studies are needed to put this on surer ground.

USC with a finite $\Delta_{xz} = \langle \psi_{k,s \sigma}^\dagger \psi_{k,-s \sigma} \rangle > 0$ now occurs in the $C_{2v}$ structure. This instability is studied exactly as before by solving $H = H_n + H_{\text{rel}}^{(MF)}$, but where $H_n$ is now the full five-band Hubbard model including ON+BOO order. In particular, the “normal” state Fermi surface is now already reconstructed above $T_c$, and the $xz$-pocket is absent, in accord with very recent ARPES data. In Fig. 2 we show the local spectral function (DOS), now interpreted as usual as the Bogoliubov quasiparticle spectrum in the SC state. A number of features, germane to USC in FeSe, immediately stand out. First, as observed generically across strongly correlated SC, an anomalous “normal” state without LFL quasiparticles [finite $\text{Im} \Sigma_i(\omega = E_F)$ with a quasilinear energy dependence] gives way to a much more coherent low-energy response $V$-shape at low energies in the gap region, strongly suggesting a nodal gap structure. (ii) Spectral-weight redistribution across the SC transition on an energy scale much larger than the gap value (see below), testifying to the strongly correlated nature of the system. Remarkably, opening up of the SC gap also results in orbital-selective sharpening of the low-energy coherent feature just below $E_F$ and to the (self-consistent) development of a pronounced peak-dip-hump structure that should be visible in spectral probes. (iii) That SC occurs in a “normal” state with broken $C_{4v}$ symmetry is manifest in the DOS asymmetry in the normal and USC states. While (i) and (iii) are consistent with STM data, (ii) requires PES experiments to be analyzed over a larger energy window than hitherto done: that this will show up sizable spectral weight transfer and the orbital-selective peak-dip-hump features across $T_c$ is a specific prediction of our work.

The nodes in the gap function, implied by the $V$-shaped DOS in the USC state are rationalized by observing that the SC pair function with anisotropic $s_{\pm}$-symmetry, $\Delta_{xz}(k) = \Delta_1(c_x + \eta c_y) + \Delta_S c_x c_y$ with $c_x = \cos k_x, \cos k_y$ and $\alpha = \Delta_2 / \Delta_1 \simeq 0.7 - 1.0$) will intersect the Fermi pockets in FeSe at the M points. Quite remarkably, the momentum-averaged spectral function (DOS) in the USC state turns out to be in good accord with PES data as shown in Fig. 3. Our computed results are also in fair accord with STM data clearly revealing (i) the $C_{2v}$ lattice symmetry in which SC appears as an asymmetry in $\rho_{xz,yc}(\omega)$, (ii) the overall spectral lineshape, as well as (iii) the clear $V$-shaped gap in the DOS.

Remarkably, our proposal also accounts for NMR data for FeSe as follows. We have studied the T-dependent evolution of local dynamical spin fluctuations in the whole T-range by computing the nuclear magnetic resonance (NMR) spin-relaxation rate $1/T_1(T)$ as a function of $T$ in FeSe by repeating earlier calculations for the 1111-FeAs systems $(T/T_1)^{-1}$ measures the weighted average of the various $q$ modes of low-frequency spin fluctuations. Satisfyingly, as seen in Fig. 4 correct $T$-dependence of $(T/T_1)^{-1}$ over the whole T-range is well captured by DMFT. This clearly marks the strong enhancement of large-$q$ spin fluctuation modes as $T$ is reduced toward $T_c$. In the normal state, the source of the increasing spin correlations lies in the partially unquenched and strongly quantum-mechanically fluctuating local moment in DMFT: this is also what causes bad-metallic normal state conductance in FeSe and $T_c$. A normal state pseudogap is clearly manifested by the fact that $1/T_1$ decreases below $T^* \simeq 25 \text{K}(2T_c)$ in excellent accord with very recent estimates. Below $T_c$, $T_1$ suddenly decreases due to the suppression of low-energy spin fluctuations by SC
(not spin-orbit) dynamical modes in the residual interactions (predominantly short-ranged (near and next-nearest neighbors in spin detailed in Supplementary Information (SI): interestingly, we find that the power-law orthogonality catastrophe accompanying orbital-selective Mottness arising from the Lifshitz transition discussed before. This is by \(1/\text{field}, \ H\), between FQO and SQ-fluctuations naturally arises: observation of critical scaling in \(1/\text{anisotropic extended symmetry down to } C_{2v}\) symmetry down to \(C_{2v}\) by ferro-orbital order (FOO) is visible. Further, the Bogoliubov quasiparticle DOS (lower panels) shows clear orbital-dependent \(V\)-shaped and gapless structure, consistent with in-plane gap nodes in FeSe.

Figure 1] LDA+DMFT many-body density-of-states for \(x_z, y_z\) bands in the orbital nematic (ON) (upper panels) and the orbital-nematic-plus-anisotropic extended \(i_s\)-wave superconducting (ON+SC) state proposed in the text. Clear difference in \(\rho_{xz}(\omega), \rho_{yz}(\omega)\), implying breaking of \(C_{4v}\) symmetry down to \(C_{2v}\).

gap opening. Absence of the Hebel-Slichter (HS) coherence peak is a consequence of a strongly incoherent normal state self-energy: the latter overdamps the coherence peak. Finally, \(T^{-1} \approx T^n\) with \(3 < n < 5\) below \(T_c\), in accord with in-plane SC-gap nodes: this will also result in vanishing of the HS peak. Furthermore, using \(A_k/T \approx 1/T_1T\) \(^{24}\) we also estimate the muon depolarization rate in the \(\mu\) SR study: the inset of Fig. 5 shows \(1/T_1 T\) vs \(T\) on a log-log plot. Clearly, both \(1/T_1 T\) and hence \(A_k/T\) for \(V_{ab} = 0.021\) exhibit approximate power-law behavior with \(T\), i.e., they vary as \(T^{-n}\) with \(0 < n < 1\), in nice qualitative accord with data, though the exponent \(n\) in the \(T^{-n}\) dependence is same for both (probably, a reflection of our use of a \(T\)-independent \(A(q)\) in the calculation). Below \(T^* = 25\) K, the critical scaling behavior is cut-off by the low-energy pseudogap, again in full accord with data. In our theory, this pseudogap (see Fig. 5) emerges from enhancement of orbital-selective Mottness accompanying onset of ON+BOO order as above, and not from preformed Cooper pairs.

These observations show that the “pair glue”, arising from \(H_{res}\), is essentially composed of entangled inter-site spin- and orbital (not spin-orbit) dynamical modes in the residual interactions (\(H_{res}\), even as FeSe does not show AF order. Since SC pairing is predominantly short-ranged (near and next-nearest neighbors in \(H_{res}\)), the pair coherence length will be small and the upper critical field, \(H_c\), will be large (\(H_c = 47\) T under pressure \(^{32}\)) as seen. The “normal” state clearly has critical spin fluctuations, as evidenced by \(1/T_1 T \approx T^{-n}\) for \(40 < T < 80\) K in Fig. 5. We posit that such critical magnetic fluctuations can naturally arise from a lattice orthogonality catastrophe accompanying orbital-selective Mottness arising from the Lifshitz transition discussed before. This is detailed in Supplementary Information (SI): interestingly, we find that the power-law \(T\)-dependencies related to soft dynamical \(\text{spin-quadrupolar (SQ)) fluctuations (see SI), i.e., to a Fourier transform of } S_i^a Q_i^a(\tau); S_i^b Q_i^b(0) >, \text{ with } S_i^a Q_i^a = (c_{i,a} \sigma c_{i,b} - \sigma + h.c), \text{ with } a = x_z, b = y_z \text{ in the } x_z, y_z \text{ orbital sector. Given a finite FQO as derived before from } H_{res}, \text{ a direct symmetry adapted coupling between FQO and SQ-fluctuations naturally arises: observation of critical scaling in } 1/T_1 T \text{ is now directly tied to the latter. Our findings are thus also consistent with a metallic state with FQO and SQ correlations, and not too far from AF order, which actually occurs under pressure.}\(^{32}\) A detailed elucidation of this last point requires a study of FeSe under pressure, and is out of scope of this work.
Figure 2] Orbital-resolved spectral functions (upper panels) and self-energies (lower panels). Direct comparison with normal state results\textsuperscript{17} clearly shows how USC gap opening suppresses normal state incoherence, leading to a much more “coherent” self-energy with negligible damping at low energy. Thus, this shows that the “normal”-USC phase transition is a coherence-restoring one, in analogy with cuprates.

III. DISCUSSION

Given the form of $H_{res}$, the main competitor to SC is an orbital-EN-plus BOO or FQO state that drives a T-O structural transition. Support for this comes from a $T-x$ phase diagram\textsuperscript{31} and ARPES\textsuperscript{21} in the former, SC $T_c$ peaks precisely at a $x_c$ where the T-O transition is suppressed to $T = 0$. This also has other consequences, outlined below. First, replacing Se by Te causes electronic structure changes owing to reduced chalcogen height ($h$) in the unit cell. This parameter has been identified as relevant to the fermiology of FeAs systems\textsuperscript{38} and the sensitivity of LDA FS to $h$ is well known. In FeSe geometry, decreasing $h$ increases the diagonal Fe-Te-Fe hopping relative to the near neighbor Fe-Fe hopping. Translated into dependence of $J_1, J_2$ on $h$ in our strong coupling view, this means an effective change in the effective residual two-particle interaction, pushing the ratio $J_2/J_1$ to values high enough that the SC gap function no longer intersects the renormalized FS in the $C_{2v}$ structure\textsuperscript{30} This will drive nodeless, anisotropic $s_{\pm}$, accounting for the transition from nodal to nodeless $s_{\pm}$ SC as seen\textsuperscript{7,30}. Also, the suppression of SC under strain and its enhancement at high pressure finds a natural explication: strain couples linearly to the (orbital) nematic order parameter, and stabilizes nematicity. Since $\langle N \rangle$ competes with $\Delta_{tot}$, this weakens $s_{\pm}$ SC. And, as is ubiquitous in MO systems, high pressure weakens nematic order by reducing the $d_{xz} - d_{yz}$ splitting, enhancing SC.\textsuperscript{32} Finally, minute impurity content (e.g, excess Fe in Fe$_{1+\delta}$Se with $0 < \delta < 1$) will rapidly suppress nodal $s_{\pm}$ SC (in the parent FeSe) on rather general grounds. Thus, our proposal provides a natural qualitative explication for a wide range of unusual behaviors exhibited in the SC state in Fe$_{1+\delta}$Se$_{1-x}$Te$_x$.

That USC arises in an orbital nematic state with $C_{2v}$ structure is also consistent with the fact that the observed vortex shape anisotropy strongly exceeds values expected on pure structural grounds.\textsuperscript{24} Further, absence of AF order and interband nesting at $Q = \pm (\pi, 0), \pm (0, \pi)$ in the correlated electronic structure presents problems for a magnetic origin for nematicity in FeSe. But orbital EN-plus BOO does not require nesting, and thus emerges as the natural contender for the competing order in FeSe. Given the loss of the $xy$-band states due to the OSMP in the “normal” state already above FQ and SC ordering scales also precludes use of LDA band structures and associated nesting features to rationalize such features.

We emphasize that Cooper pairing naturally turns out to be orbital-selective in our work, since the “normal” state above $T_c$ already has enhanced orbital-selectivity due to finite $< Q^2 >$, i.e, a state with FQO, also viewable as an orbital nematic. This is manifest in our finding of orbital-selective anisotropy in the Bogoliubov DOS in the SC state above, which exhibits marked orbital selective changes with- and without a finite SC pairing, simply because such orbital differentiation is already driven by FQO above
The text is too long to be included here, but it discusses the correlation between electronic structure and superconductivity in Fe-based materials, focusing on the role of orbital-selective pairing and Mott insulating states. It also mentions recent ARPES data and compares its approach with previous studies. The text is technical and requires a deep understanding of condensed matter physics.
Figure 4 | The $T$-dependent NMR spin-lattice relaxation rate for the normal and our proposed anisotropic extended $s_{\pm}$-wave SC in FeSe. Direct comparison with data shows impressive accord over the whole $T$-range. Above $T_c$, $(T/T_c)^{-1}$ shows a $T$-dependent increase, showing progressive build-up of short-range (large-$q$) AF spin correlations. Absence of Hebel-Slichter peak above $T_c$ and power-law fall-off for $T << T_c$ are signatures of strong normal state scattering and nodal SC gap (see text for explanations).

our proposal also naturally involves strongly entangled spin-orbital correlations, and the spin fluctuations are intrinsically orbital-selective. This is explicit in the residual two-particle interaction, $H_{\text{res}}$. The distinguishing features in our approach are (i) strongly enhanced orbital-selectivity leads to an OSMP (loss of $xy$-band states) and leads to breakdown of Landau quasiparticle description already in the “normal” state, in favor of a metallic state with branch-cut responses in the correlation functions, a feature of the “strange” metal, and (ii) SC pairing in our case involves strong short-ranged, spin-orbital fluctuations, and will lead to a short coherence length SC pairing. High $H_{c2}$ values and the fact that FeSe is close to a BCS-BEC crossover support such a view.

In our picture, however, an orbital-selective Mott behavior is the symmetry-unbroken “normal” state, well-documented theoretically, is necessary: (i) lack of hole pockets in the renormalized electronic structure (via the Lifshitz transition at $T^*$ in experiments) leads to breakdown of “adiabatic continuity” with LDA Fermi surfaces. It is then impermissible to use LDA Fermi surfaces to derive instabilities to ordered states, and (ii) it is the vanishing LFL quasiparticle residue in the “normal” state ($\zeta_{FL} = 0$) that leads to preferential relevance of the two-particle residual interaction, $H_{\text{res}}$, relative to the one-electron hybridization between $xz$, $yz$ states, $H_{\text{hyb}}^{(1)}$, in LDA. We have shown that both, FQO and USC orders follow from $H_{\text{res}}$ as competing orders, and discussed how this is qualitatively consistent with indications from a wide variety of spectral probes.

If the normal state were a weakly correlated LFL, $H_{\text{hyb}}^{(1)}$ would necessarily need to be preferentially relevant in FeSe in a situation where FS nesting is absent. This latter view would be applicable if a FL metal underwent conventional BCS-like instabilities to nematic and SC orders. Unambiguous observation of nLFL behavior (bad-metallicity and quasi-linear-in-$T$ resistivity) in the “normal” state in FeSe without any symmetry-breaking supports the approach taken here. Together with earlier DMFT work for Fe-based SC and the good accord with a range of key physical responses in both, normal and USC states, the present approach testifies to the relevance of strong correlation-driven orbital-selective-Mott state and its novel role in fomenting unconventional nematic and superconductive orders in FeSe.

Methods

To study the competing orders in FeSe, we employ the local-density-approximation plus dynamical-mean-field-theory (LDA+DMFT) which takes into consideration the most relevant local multi-orbital and associated Hund correlation effects germane to the strongly correlated metallic state in FeSe. LDA+DMFT implementation used here also correctly describes disorder, pressure and temperature effects in multi-band electronic systems. The LDA+DMFT scheme is an ideal starting point towards description of correlation-driven metal-to-insulator transitions, Fermi and non-Fermi liquid metallic states in quasi-local limits solely due to strong dynamical correlations in idealized many-particle models as well as in real multi-orbital systems. It also provides mean-field descriptions of the competing orders in correlated systems at low temperatures, and of large modification of physical properties in response to external perturbations like pressure, chemical doping, magnetic and electric fields, etc. The one-electron...
LDA density-of-states were computed using the non-fully-relativistic version of the PY-LMTO code. To incorporate the effects of dynamical electronic correlations in FeSe, we use the multi-orbital iterated-perturbation-theory (MO-IPT) as an impurity solver of the many-particle problem in DMFT, as described in detail in Refs. 7.

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Author contributions

M.S.L. conceived the problem. L.C performed DMFT calculations. M.S.L and L.C analyzed the LDA+DMFT results and wrote the manuscript with inputs from B.F. All authors discussed the results and reviewed the manuscript.

Additional information
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IV. SUPPLEMENTARY INFORMATION: UNUSUAL LOCALLY CRITICAL SPIN FLUCTUATIONS FROM ORBITAL SELECTIVE MOTTNES

Here, we detail how correlations in an orbital-selective Mott phase (OSMP) offer a natural insight into the intermediate-in-$T$ power-law behavior of spin fluctuations in FeSe. To this end, we analyze the analytic structure of local, one- and two-particle correlators in the OSMP by appealing to the underlying impurity model of DMFT, where analytic progress is possible. We explicitly show how these are very distinct in the OSMP and non-OSMP phases of the model, the difference arising from the fact that a local orthogonality catastrophe in the OSMP destroys the Landau quasiparticles via a fundamental change in the analytic structure of the one-electron propagator from a pole- to an infra-red singular branch-cut structure at low energy.

In the text, we have shown how onset of an orbital nematic-plus bond-ordered orbital (ON+BOO) state enhances the already substantial orbital-selective correlations in FeSe. Specifically, such correlations push the $xz$-band states below $E_F$ (Lifshitz transition), whence this metal now has co-existent “metallic” ($yz$-band) and Mott localized ($xz$-band) states at low energy. We restrict ourselves to only the $xz, yz$ orbital sector to flush out the essential physics. The multi-orbital interaction terms in the two-band Hubbard model,

$$H_{xz,yz} = U' \sum_{i \sigma} n_{i,xz,\sigma} n_{i,yz,\sigma} - J_H \sum_{i \sigma} S_{i,xz} S_{i,yz}$$

compete with the inter-orbital one-electron hybridization, $H_{hyb}^{(1)} = t_{xz,yz} \sum_{\sigma} (c_{i,xz,\sigma}^\dagger c_{j,yz,\sigma} + h.c.)$ (putting $a = xz, b = yz$ in $H_{hyb}$ in the main text). In the OSMP phase, the interplay between $H_{xz,yz}$ and $H_{hyb}^{(1)}$ leads to emergence of anomalous behavior, which we now discuss.

Were the metallic state to be a Landau Fermi-liquid (LFL) metal, $H_{hyb}^{(1)}$ would be necessarily relevant in the renormalization group sense. This would be the case only as long as no OSMP occurred in the DMFT solution, since a relevant inter-orbital one-electron hybridization requires absence of any Mott-like localization (since otherwise, no coherent one-electron mixing in the $xz - yz$ channel could result). Once the OSMP sets in, this situation undergoes a basic change: In the OSMP, action of $H_{hyb}^{(1)}$ will necessarily mix a metallic band $yz$ electron with a Mott-localized $xz$-band electron. However, in this case, a coherent one-electron mixing would be suppressed to zero, since the lower Hubbard band states in the $xz$-band are now filled with one electron per Fe site, and so cannot recoil, simply because there are no lower-Hubbard band states into which they can do so. In fact, the $xz - yz$ mixing will necessarily generate a doubly occupied state in the $xz$-sector, where a $yz$-band fermion, transferred to the $xz$-orbital, will result in a doubly occupied state on the $xz$-orbital. But this is a high-energy, upper-Hubbard band state, and thus has no one-to-one correspondence with any one-electron state. The upshot is that the corresponding mixing term thus undergoes a radical change, and now reads $H_{hyb}^{(1)} = t_{xz,yz} \sum_\sigma (n_{i,xz,-\sigma} c_{i,xz,\sigma}^\dagger c_{j,yz,\sigma} + h.c.)$, i.e., it describes hybridization of metallic $yz$ states with upper-Hubbard band states in the $xz$ sector, which are projected $X$-fermions. This correlation-induced blocking of the one-electron hybridization thus leads to irrelevance of $H_{hyb}^{(1)}$ and to Kondo destruction, and results in a non-LFL state.

The mechanism of breakdown of Landau FL metallicity is now manifest. The above argument leads us to consider the two-orbital Hubbard model in a regime where $H_{hyb}^{(1)}$ is irrelevant, and so the physics is now controlled by strong scattering between $xz, yz$ states. As stated above, the fact that $xz$-carriers cannot any more recoil during scattering leads to very interesting anomalies. [S1] Specifically, within the local “impurity problem” of the DMFT, both interaction terms in $H_{xz,yz}$ now correspond to strong scattering of $yz$ carriers by recoilless (Mott localized) $xz$-states. This corresponds precisely to the venerated X-ray edge problem, [S1,S2] where the $xz$-fermion propagator as well as the “excitonic” inter-band fluctuation propagator acquires anomalous dimensions, and the one- and two-particle spectra exhibit anomalous branch-cut continua instead of the conventional pole structure of a LFL metal: explicitly,

$$\rho_{xz}(\omega) = \int d\tau e^{i\tau \omega} < T x_{c}^\tau (0) c_{xz}(\tau) c_{xz}^\dagger(0) >= \theta(-\omega)|\omega|^{-(1-\eta)}$$

with $\eta = \frac{1}{2}\tan^{-1}(U'/W)$, where $W$ is an effective one electron band-width. Thus, $0 < \eta < 1$. In Fig. [1] we indeed find that in the ON+BOO phase (with $< N > = 0.2$), the local spectral function of $xz$-states shows a very deep pseudogap (a finite-temperature effect, we would have $\rho_{xz}(\omega = E_F) = 0$ at $T = 0$), accompanied by an anomalously broadened lineshape of half-width $O(1)$ eV, while the corresponding $yz$-spectral function exhibits clear power-law fall-off of the above type with no discernible Landau quasiparticle pole-like peak at $E_F = 0$, in full qualitative accord with the above arguments.

More crucially, in such a “strange” metal, the corresponding local spin fluctuation spectrum measured in NMR relaxation rate via $1/T_1 \gamma_z \tilde{\chi}_{loc}''(\omega)$ with $\chi_{loc}''(\omega)$ being the imaginary part of the local dynamical spin-flip correlator, is also expected to show infra-red singular behavior. This is because the “excitonic” spin-flip correlation function involving “spin excitons” created from the $xz,yz$ fermions, also has an anomalous branch-cut form due to the X-ray-edge mapping:

$$\tilde{\chi}_{loc}''(\omega) = \int d\tau e^{i\tau \omega} < T c_{i,xz,-\sigma}^\dagger c_{i,yz,-\sigma}^\dagger c_{i,yz,-\sigma} c_{i,xz,-\sigma}^\dagger(0) > ~ \simeq |\omega|^{-(2\eta - \nu^2)}$$

Remarkably, this “interband excitonic” correlation function is precisely that of a spin quadrupole, defined as $Q_{i,xz}^{\sigma,-\sigma} = (c_{i,xz,\sigma}^\dagger c_{i,xz,-\sigma}^\dagger + h.c.)/2$. In the $xz,yz$-degenerate orbital sector relevant to undistorted structure of FeSe, it is natural to define orbital pseudospins, $Q_{i,z} = (n_{i,a} - n_{i,b})/2, Q_{i,xz} = (c_{i,xz,a}^\dagger c_{i,xz,b} + h.c.)/2, Q_{i,yz} = (c_{i,yz,a}^\dagger c_{i,yz,b} + h.c.)/2i$, satisfying the usual pseudospin-1/2 SU(2) algebra: the first defines the FOO or FOQO order parameter, while the second and third describe quadrupolar charge and current fluctuations in the FOO state. In fact, it is an orbital bond-localized inter-band quadrupolar pseudospin-1/2 SU(2) algebra, given the $Q_{i,z}^2 = Q_{i,xz}^2 = 0$ for both $xz$ and $yz$ orbital pseudospins, which is the key ingredient for the observed anomalous $\tilde{\chi}_{loc}''(\omega)$ behavior.
above, and fluctuations in the former will inevitably cause SQ fluctuations. In the OSMP which is enhanced in the FOO (FQO) state as shown in the main text, we have thus shown that a lattice orthogonality catastrophe results in singular SQ-fluctuations. Thus, the power-law behavior in $1/T_1 T$ and $\mu$SR relaxation rate arises from such singular correlations in the OSMP. Our finding of a FQO state is also qualitatively consistent with recent Raman scattering work.\(^{15}\)

At finite $T$, this should yield $\omega/T$ scaling. It would be interesting to test whether inelastic neutron scattering lineshapes show approximate $\omega/T$-scaling for FeSe in the range where $1/T_1 T$ exhibits a critical power-law behavior; if these spin fluctuations would be almost momentum independent, this would lend further positive support to our idea. Using the above form for $\chi_{\text{loc}}''(\omega)$ to compute $1/T_1 T$ immediately yields a $T^{-n}$ power-law behavior with $0 < n < 1$, as seen. It is not, however, possible to quantitatively obtain the value of $n$ from this argument, because lattice self-consistency effect will affect the precise values of the above exponents (which correspond to the underlying “impurity” model). Nonetheless, this argument directly rationalizes the power-law behavior of $1/T_1 T$ above the pseudogap scale, and links it to an Anderson-Nozieres orthogonality catastrophe that arises naturally in an OSMP view.

On the other hand, in the non-OSMP state, which occurs in the 2-band Hubbard model whenever the one-electron hybridization is relevant, the $xz$-fermion can recoil during the scattering processes involving $xz$, $yz$ carriers, now simply because absence of Mott localization enables such low-energy recoil to occur. The impurity problem of DMFT for the 2-band Hubbard model is now the above impurity model with a relevant $H_{\text{hyb}}^{(1)} = t_{ab} \sum_{<i,j>,\sigma} (c_{i,xz,\sigma}^\dagger c_{j,yz,\sigma} + h.c.)$. As is well known, a two-stage spin-orbital screening now quenches the local spin-orbital moments via orbital and spin- Kondo effect, leading to a correlated spin-orbital Kondo resonance in both, $G_{xz}(\omega)$ and $G_{yz}(\omega)$ at low energy, reinstating Landau FL quasiparticles and a FL metallic state at low $T$. Thus, within local (DMFT) approaches, observation of non-Landau quasiparticle behavior in the normal state requires an OSMP to occur. Such an OSMP, enhanced by nematicity (or FQO in the main text) in FeSe, is also consistent with very recent QPI and ARPES results in the USC state in FeSe, as emphasized in the main text.

Finally, in the OSMP state, onset of orbital-nematic plus bond-order (ON+BOO) in FeSe further enhances orbital-selective correlations, opening up a low-energy pseudogap in $\rho_{yz}(\omega)$ as shown in the main text. This suppresses the infra-red singular behavior described above, qualitatively accounting for the breakdown of QC scaling below $T^* \approx 25$ K in our LDA+DMFT calculations, again in good accord with data.\(^{24}\)

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