Time-Reversal Symmetry Breaking Superconductivity in the Coexistence Phase with Magnetism in Fe Pnictides

Alberto Hinojosa, Rafael M. Fernandes, and Andrey V. Chubukov

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Time-reversal symmetry breaking superconductivity in the coexistence phase with magnetism in Fe-pnictides

Alberto Hinojosa,[1] Rafael M. Fernandes,[2] and Andrey V. Chubukov[1]

[1]*Department of Physics, University of Wisconsin, Madison, WI 53706, USA*

[2]*School of Physics and Astronomy, University of Minnesota, Minneapolis, MN 55455, USA*

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We argue that superconductivity in the coexistence region with spin-density-wave (SDW) order in weakly doped Fe-pnictides differs qualitatively from the ordinary s^{+-} state outside the coexistence region as it develops an additional gap component which is a mixture of intra-pocket singlet (s^{++}) and inter-pocket spin-triplet pairings (the $t-$state). The coupling constant for the $t-$channel is proportional to the SDW order and involves interactions that do not contribute to superconductivity outside of the SDW region. We argue that the s^{+-} and $t-$type superconducting orders coexist at low temperatures, and the relative phase between the two is in general different from 0 or π, manifesting explicitly the breaking of the time-reversal symmetry promoted by long-range SDW order. We argue that time-reversal may get broken even before true superconductivity develops.

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Introduction Iron-based superconductors (FeSCs) have been the subject of intense study since 2008[1]. Their rich phase diagram includes the regions of superconductivity (SC), spin density wave (SDW), nematic order, and a region where SDW, SC, and nematic order coexist[2–4]. Outside the SDW/nematic region, SC develops in the spin-singlet channel and in most of Fe-based superconductors it has $s-$wave symmetry with a π phase shift between the SC order parameters on hole and on electron pockets (s^{+-} gap structure)[5,6].

It has been recently argued by several groups that the multiband structure of FeSCs allows for superconducting states with more exotic properties[7–21]. Of particular interest are SC states that break time-reversal symmetry (TRS), as such states have a plethora of interesting properties like, e.g., novel collective modes[15,20,22–24]. TRS-broken states emerge when the phase differences ψ_i between SC order parameters on different Fermi surfaces (FS) are not multiples of π.

The two current proposals for TRS breaking in FeSCs are $s + id$[7,11–13,19] and $s + is$ states[10,15,20,21]. The first emerges when attractions in the $d-$wave and $s-$wave channels are of near-equal strength. The second emerges when there is a competition between different s^{+-} states favored by inter-pocket and intra-pocket interactions. Both of these proposals were, however, argued to be applicable only to strongly hole or electron-doped FeSCc. For weakly/moderately doped FeSCs the common belief is that s^{+-} superconductivity is robust.

In this communication we argue that an exotic state which breaks TRS can emerge already at low doping, in a range where SC is known[4,25–32] to emerge from a pre-existing SDW state. We show that SDW order induces attraction in another pairing channel, for which the order parameter is an admixture of spin-singlet and spin-triplet components (the two are mixed in the SDW state since spin rotational symmetry is broken). Because a triplet component is involved, we call this a t-state. In the absence of nesting, s^{+-} and $t-$ components are linearly coupled, and the development of s^{+-} SC order at T_c triggers an immediate appearance of $t-$order component with the same phase ($s + t$ state or $s - t$ state, depending the sign of the bilinear coupling). Such a state has been discussed in the SDW/SC coexistence region of the cuprates, organic and heavy fermion materials[33–39] and the Fe-pnictides, in the context of nodeless superconductivity immediately below $T_c < T_{sdw}$ (Refs. [40–47]).

In this letter we show that the $s \pm t$ state exists only near T_c, while at a lower T there is a phase transition into a state where a relative phase between the two SC components is different from 0 or π, i.e., the order parameter has a $s + e^{i\theta} t$ form. This order parameter does not transform into itself under TRS, unlike $s + t$ order. As a result, the order parameter manifold contains an additional Z_2 Ising degree of freedom, which gets broken by selection of $+\theta$ or $-\theta$. The TRS broken state emerges via a phase transition inside a superconductor, which should have experimental manifestations. We note in this regard that, although the TRS of the system is formally broken already at the SDW transition temperature $T_N > T_c$, the TR operation transforms one magnetic state into another state from the same $O(3)$ manifold, i.e., there is no additional Z_2 degree of freedom which one could associate with TRS. Only when θ becomes different from 0 or π, does the order parameter manifold acquire an additional Z_2 degree of freedom associated with TRS.

We show that the $s + e^{i\theta} t$ state emerges already in the minimal three-band model of one circular hole pocket and two symmetry-related elliptical electron pockets[40,42,48]. The presence of the other hole pockets complicates the analysis but does not lead to new physics. We argue that, when the original 4-fermion interactions are rewritten in terms of a and b fermions, which describe states near the two reconstructed FSs (Fig. 2) and projected onto the particle-particle subset, the two different pairing channels emerge. One is the usual spin-singlet s^{+-} channel, for which the SC order parameter is $\Delta_1 \propto \sum_{\mathbf{k}} i\sigma^y_{\alpha\beta} \left[\langle a_{\mathbf{k}\alpha} a_{-\mathbf{k}\beta}\rangle - \langle b_{\mathbf{k}\alpha} b_{-\mathbf{k}\beta}\rangle\right]$.

The second pairing channel, with order parameter Δ_2, has two contributions. One is a spin-triplet inter-pocket term $\sum_{\mathbf{k}} \sigma^x_{\alpha\beta}\langle a_{\mathbf{k}\alpha} b_{-\mathbf{k}\beta}\rangle)$ (hence the name $t-$state), and the other is a spin-singlet s^{++} type term $\sum_{\mathbf{k}} i\sigma^y_{\alpha\beta}\left[\langle a_{\mathbf{k}\alpha} a_{-\mathbf{k}\beta}\rangle + \langle b_{\mathbf{k}\alpha} b_{-\mathbf{k}\beta}\rangle\right]$. The presence of the s^{++} component in Δ_2 is crucial as with it the kernel in the gap equation for Δ_2 is logarithmic (as it is for Δ_1), implying that even a weak attraction gives rise to superconductivity. We emphasize that the triplet component of Δ_2, $\langle a_{\mathbf{k}\alpha} b_{-\mathbf{k}\beta}\rangle$, would not spontaneously emerge by itself because the FSs for a and b fermions are disconnected and appears only because it couples linearly to the s^{++} component $\langle a_{\mathbf{k}\alpha} a_{-\mathbf{k}\beta}\rangle + \langle b_{\mathbf{k}\alpha} b_{-\mathbf{k}\beta}\rangle$. A similar situation emerges in Fe-pnictides with only electron pockets[49].

The structure of Δ_1 and Δ_2 is shown in Figs. 1a and 1b. Our analysis of the non-linear gap equations for Δ_1 and Δ_2 shows that the two SC orders coexist in some parameter range, and the relative phase between the two is different from 0 or π in the general case when the two orders are linearly coupled in the Ginzburg-Landau (GL) functional, and equals to $\pm\pi/2$ for the special case when linear coupling is absent (Fig. 3).

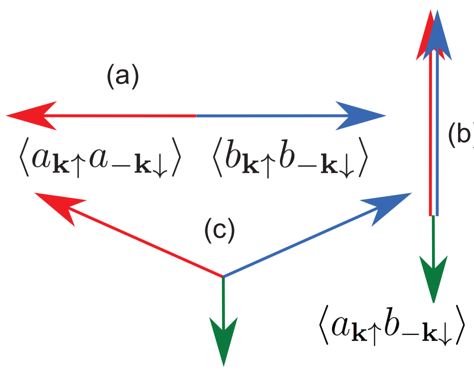

Figure 1: The structure of gap functions in different SC states: (a) pure s^{+-} state, (b) pure $t-$ state, (c) $s{+}it$ state with $\pm\pi/2$ phase difference between s^{+-} and $t-$ components. Operators a and b describe fermions near the reconstructed FSs.

The model. We consider a three band model with c fermions with momenta near the hole pocket at $(0,0)$ and f fermions with momenta near the electron pockets centered at $(0,\pi)$ and $(\pi,0)$ in the 1-Fe Brillouin zone (Fig. 2a)[48,50]. The c and f fermions form circular and elliptical FSs, respectively, with dispersions given by $\xi^c_{\mathbf{k}} = \mu_c - \frac{\mathbf{k}^2}{2m_c}$ and $\xi^f_{\mathbf{k}} = -\mu_f + \frac{k_x^2}{2m_x} + \frac{k_y^2}{2m_y}$. Since the SDW state picks an ordering vector $\mathbf{Q}$, which is either $(0,\pi)$ or $(\pi,0)$, one of the electron pockets does not participate in this order. We choose $\mathbf{Q}=(0,\pi)$ without loss of generality and effectively reduce the model to two bands. We follow earlier works[51,52] and consider five possible repulsive interactions in the band basis: inter-pocket, density-density, exchange, pair hopping, and intra-pocket interactions. The corresponding couplings are U_1, U_2, U_3, and $U_4 = U_5$, respectively. We present the interaction Hamiltonian in the Supplementary material (SM). All couplings are assumed to be already renormalized from their bare values by fermions with energies larger than the upper energy cutoff Λ. Without SDW, SC in this model arises only in the s^{+-} channel. The corresponding coupling is $U_3 - U_4$, and we assume that it is positive (attractive). The couplings U_1 and U_2 do not participate in SC pairing, but U_1 contributes to the coupling in the SDW channel $U_1 + U_3 > 0$, which for $U_i > 0$ is larger than in SC channels. RG studies found that the SC interaction gets larger as energy decreases in the RG flow[51–54]. Yet, at low doping, the SDW order comes first and SC develops in the coexistence region with magnetism.

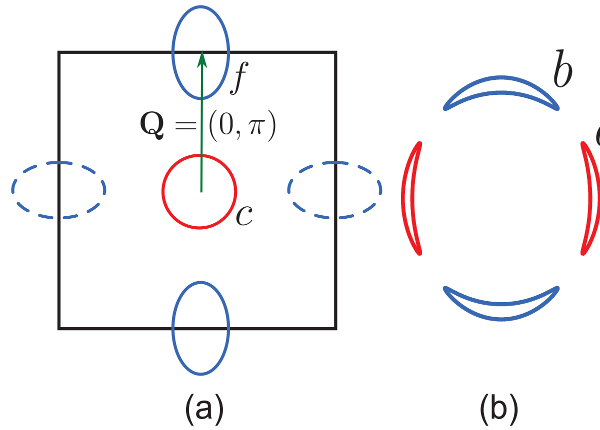

Figure 2: Fermi surfaces in (a) the paramagnetic state, (b) the SDW state.

We approximate the interactions U_i as angle independent although in general they do contain symmetry-imposed angular dependencies along the FSs, associated with the orbital content of the FSs. These angular dependencies give rise to angular variations of the s^{+-} gaps in the absence of SDW and in some cases lead to accidental gap nodes, e.g., in P-doped materials[6,55]. In the coexistence regime, there is an additional angular variation of the pairing interactions, imposed by the angle dependence of the SDW coherence factors which dress the bare interactions U_i[41,42,44,47]. Because the orbital content and the SDW coherence factors lead to similar angular dependence of the pairing interactions, we treat the original vertices as constants but keep the SDW-induced angular dependencies. The momentum dependence of the original interactions also leads to angular dependence of the SDW gap which actually vanishes along particular directions, at least in a three-pocket model[56]. This angular dependence is important at strong coupling, where it preserves a small but finite FS, but not at moderate coupling because even a constant SDW gap does not completely gap the FS[57].

The self-consistent equation for the SDW order parameter M and the reconstructed fermionic dispersions in the SDW state have been obtained before[48]. The quadratic

Hamiltonian in terms of the quasiparticles a and b is

$$\mathcal{H}_0 = \sum_{\mathbf{k}} \left[\xi^a_{\mathbf{k}} a^\dagger_{\mathbf{k}\alpha} a_{\mathbf{k}\alpha} + \xi^b_{\mathbf{k}} b^\dagger_{\mathbf{k}\alpha} b_{\mathbf{k}\alpha}\right], \qquad (1)$$

where $\xi^{a,b}_{\mathbf{k}} = \delta_{\mathbf{k}} \mp \sqrt{\xi^2_{\mathbf{k}} + M^2}$ and we have expressed the original dispersions in terms of the linear combinations $\delta_{\mathbf{k}} = \frac{\xi^f_{\mathbf{k}} + \xi^c_{\mathbf{k}}}{2}$ and $\xi_{\mathbf{k}} = \frac{\xi^f_{\mathbf{k}} - \xi^c_{\mathbf{k}}}{2}$. In general $\delta_{\mathbf{k}} = \delta_0 + \delta_2 \cos 2\theta$, where the first term measures the doping level ($\delta_0 = 0.5 v_F (k^c_F - k^f_F)$) and the second one accounts for the (weak) ellipticity of the electron pocket (Ref.[40]). The coherence factors $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ are expressed in terms of these parameters as $u_{\mathbf{k}} = \sqrt{\frac{1}{2}\left(1 + \frac{\xi_{\mathbf{k}}}{\sqrt{\xi^2_{\mathbf{k}} + M^2}}\right)}$, $v_{\mathbf{k}} = \operatorname{sgn} M \sqrt{\frac{1}{2}\left(1 - \frac{\xi_{\mathbf{k}}}{\sqrt{\xi^2_{\mathbf{k}} + M^2}}\right)}$ (see SM). The FSs for a and b fermions are shown in Fig. 2b.

Superconductivity. Re-writing the pairing interactions in terms of the new fermions, we find conventional pairing terms like $a^\dagger_{\mathbf{k}\uparrow} a^\dagger_{-\mathbf{k}\downarrow} a_{-\mathbf{p}\downarrow} a_{\mathbf{p}\uparrow}$ or $a^\dagger_{\mathbf{k}\uparrow} a^\dagger_{-\mathbf{k}\downarrow} b_{-\mathbf{p}\downarrow} b_{\mathbf{p}\uparrow}$, and anomalous terms like $a^\dagger_{\mathbf{k}\uparrow} a^\dagger_{-\mathbf{k}\downarrow} (a_{-\mathbf{p}\downarrow} b_{\mathbf{p}\uparrow} + a_{-\mathbf{p}\uparrow} b_{\mathbf{p}\downarrow})$. To solve for the SC order parameter, we then need to introduce both spin-singlet pairings $i\sigma^y_{\alpha\beta}\langle a_{\mathbf{k}\alpha} a_{-\mathbf{k}\beta}\rangle$ and $i\sigma^y_{\alpha\beta}\langle b_{\mathbf{k}\alpha} b_{-\mathbf{k}\beta}\rangle$ between fermions belonging to the same pocket, and spin triplet pairing $\sigma^x_{\alpha\beta}\langle a_{\mathbf{k}\alpha} b_{-\mathbf{k}\beta}\rangle$ between fermions belonging to different pockets.

The full pairing Hamiltonian in the BCS approximation has the form

$$\mathcal{H}_\Delta = \frac{1}{2} i\sigma^y_{\alpha\beta} \sum_{\mathbf{p}} \left[\Delta_{aa}(\mathbf{p}) a^\dagger_{\mathbf{p}\alpha} a^\dagger_{-\mathbf{p}\beta} + \Delta_{bb}(\mathbf{p}) b^\dagger_{\mathbf{p}\alpha} b^\dagger_{-\mathbf{p}\beta}\right] + \frac{1}{2}\sum_{\mathbf{p}} \Delta_{ab}(\mathbf{p}) \sigma^x_{\alpha\beta} [a^\dagger_{\mathbf{p}\alpha} b^\dagger_{-\mathbf{p}\beta} - b^\dagger_{\mathbf{p}\alpha} a^\dagger_{-\mathbf{p}\beta}] + \text{H.c.} \qquad (2)$$

Because there are three different anomalous terms, the diagonalization of the pairing Hamiltonian leads to a set of three coupled equations for Δ_{aa}, Δ_{bb}, and Δ_{ab}. Parameterizing Δ_{ij} as

$$\Delta_{aa,bb}(\mathbf{p}) = \pm\Delta_1 + \Delta_2(2u_{\mathbf{p}}v_{\mathbf{p}}) + \Delta_3(u^2_{\mathbf{p}} - v^2_{\mathbf{p}}), \qquad (3)$$

$$\Delta_{ab}(\mathbf{p}) = \Delta_2(u^2_{\mathbf{p}} - v^2_{\mathbf{p}}) - \Delta_3(2u_{\mathbf{p}}v_{\mathbf{p}}), \qquad (4)$$

we express the equations for SC order parameters as

$$\Delta_1 = \frac{U_3 - U_4}{2} \sum_{\mathbf{k}} \left[\langle aa\rangle - \langle bb\rangle\right], \qquad (5)$$

$$\Delta_2 = (U_2 - U_1) \sum_{\mathbf{k}} \left[u_{\mathbf{k}} v_{\mathbf{k}} (\langle aa\rangle + \langle bb\rangle) + (u^2_{\mathbf{k}} - v^2_{\mathbf{k}})\langle ab\rangle\right]$$

$$\Delta_3 = -\frac{U_3 + U_4}{2} \sum_{\mathbf{k}} \left[(u^2_{\mathbf{k}} - v^2_{\mathbf{k}})(\langle aa\rangle + \langle bb\rangle) - 4u_{\mathbf{k}} v_{\mathbf{k}} \langle ab\rangle\right]$$

where $\langle aa\rangle \equiv i\sigma^y_{\alpha\beta}\langle a_{-\mathbf{k}\beta} a_{\mathbf{k}\alpha}\rangle$, $\langle bb\rangle \equiv i\sigma^y_{\alpha\beta}\langle b_{-\mathbf{k}\beta} b_{\mathbf{k}\alpha}\rangle$, $\langle ab\rangle \equiv \sigma^x_{\alpha\beta}\langle b_{-\mathbf{k}\beta} a_{\mathbf{k}\alpha}\rangle$. Each average is in turn expressed in terms of Δ_i ($i = 1, 2, 3$), i.e. Eqs. (5) represent the set of three coupled non-linear equations for the SC order parameters in the presence of SDW order.

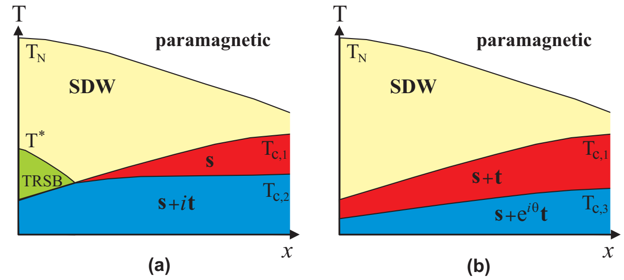

Figure 3: Schematic phase diagram of a superconductor in coexistence with SDW. (a) The special case when s and t order parameters do not couple linearly (nested FSs). (b) The generic case when s and t superconducting components couple linearly (non-nested FSs). In the $s + e^{i\theta}t$ and $s + it$ phases ($\theta = \pi/2$), the relative phase between the s and t components is frozen at $0 < \theta < \pi$ and TRS is broken, together with the $U(1)$ symmetry of the global phase. In the TRSB phase, only TRS is broken. This phase is likely present in the generic case but its boundaries are not known and we do not show it.

We see from (5) that three combinations of the interactions U_i appear in the pairing channel. Two have familiar forms[51]: $U_3 - U_4$ and $-(U_3 + U_4)$ are the couplings in the s^{+-} and s^{++} channels, respectively, in the absence of SDW order. A non-zero M couples the s^{+-} and s^{++} channels, but since the coupling in the s^{++} channel is strongly repulsive, the SDW-induced mixing of s^{+-} and s^{++} channels should not lead to any new physics. The third coupling $U_2 - U_1$, on the other hand, does not contribute to SC in the absence of SDW order. Its presence in Eq. (5) implies that SDW order not only modifies the two existing pairing channels, but also generates a new channel of fermionic pairing.

We present the full expressions for $\langle ij\rangle_{\mathbf{k}}$ in the SM and here focus on the linearized gap equations, valid at the corresponding $T_{c,i}$. Expanding the r.h.s. of (5) to first order in Δ_{ij} we obtain

$$\langle aa\rangle_{\mathbf{k}} \pm \langle bb\rangle_{\mathbf{k}} = \frac{\Delta_{aa}(k)}{2\xi^a_k} \tanh\frac{\xi^a_k}{2T} \pm \frac{\Delta_{bb}(k)}{2\xi^b_k} \tanh\frac{\xi^b_k}{2T}$$

$$\langle ab\rangle_{\mathbf{k}} = \frac{\Delta_{ab}(k)}{2(\xi^a_k + \xi^b_k)} \left(\tanh\frac{\xi^a_k}{2T} + \tanh\frac{\xi^b_k}{2T}\right) \qquad (6)$$

where Δ_{ij} are expressed via Δ_i by Eq. (4). Substituting (6) into the r.h.s. of (5) we obtain the set of three coupled linearized Eqs. on Δ_i which can be easily solved.

To understand the physics, we first focus on the case of "maximally-nested" FSs, where $\delta_0 = 0$ but $\delta_2 \neq 0$, i.e. $\xi^b_{\mathbf{k}}$ becomes $-\xi^a_{\mathbf{k}}$ under a rotation by 90 degrees. We found that this symmetry decouples the three linearized gap equations for Δ_i, which become

$$\Delta_1 \left[1 - \frac{U_3 - U_4}{2} N_F \int X_{\mathbf{k}}\right] = 0 \tag{7}$$
$$\Delta_2 \left[1 - (U_2 - U_1) N_F \int \left(u_{\mathbf{k}}^2 v_{\mathbf{k}}^2 X_{\mathbf{k}} + (u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2)^2 Y_{\mathbf{k}}\right)\right] = 0$$
$$\Delta_3 \left[1 + \frac{U_3 + U_4}{2} N_F \int \left((u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2)^2 X_{\mathbf{k}} + 8 u_{\mathbf{k}}^2 v_{\mathbf{k}}^2 Y_{\mathbf{k}}\right)\right] = 0$$

where N_F is the density of states at the FS, $\int = \int d\xi \frac{d\varphi}{2\pi}$, $u_{\mathbf{k}} v_{\mathbf{k}} = M/(2\sqrt{M^2 + \xi_{\mathbf{k}}^2})$, $u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2 = \xi_{\mathbf{k}}/\sqrt{M^2 + \xi_{\mathbf{k}}^2}$, and

$$X_{\mathbf{k}} = \frac{\tanh \frac{\xi_{\mathbf{k}}^a}{2T_c}}{\xi_{\mathbf{k}}^a}, \quad Y_{\mathbf{k}} = \frac{\tanh \frac{\xi_{\mathbf{k}}^a}{2T_c} + \tanh \frac{\xi_{\mathbf{k}}^b}{2T_c}}{2(\xi_{\mathbf{k}}^a + \xi_{\mathbf{k}}^b)} \tag{8}$$

The first and the last Eqs. (7) have familiar forms for s^{+-} and s^{++} superconductivity. For positive U_i, the s^{++} channel is repulsive, but s^{+-} superconductivity develops at $T = T_{c,1}$ if $U_3 - U_4$ is positive. The momentum integral $\int X_{\mathbf{k}}$ is logarithmically singular, as expected in BCS theory, hence $T_{c,1}$ is non-zero already at weak coupling. The second Eq. in (7) is the gap equation in the new pairing channel. In the presence of SDW the kernel in this channel is also logarithmically singular due to the contribution from $\langle aa\rangle_{\mathbf{k}} + \langle bb\rangle_{\mathbf{k}}$. Hence, if $U_2 - U_1$ is positive, the $t-$channel becomes unstable towards pairing at a non-zero $T_{c,2}$. Once Δ_2 becomes non-zero, it induces a non-zero inter-pocket pairing component $\langle ab\rangle_{\mathbf{k}}$.

$s + it$ state with broken time-reversal symmetry As it is customary for competing SC orders, the order which develops first tends to suppress the competitor by providing negative feedback to the gap equation for the competing order[20]. Yet, if the repulsion between the competing SC orders is not too strong, the two orders coexist at low enough temperatures. The issue then is what is the relative phase between the two $U(1)$ order parameters Δ_1 and Δ_2. To address this issue we derived by standard means[50,58] the GL Free energy, $\mathcal{F}(\Delta_1, \Delta_2)$ (see SM). To fourth order in $\Delta_{1,2}$ we obtained

$$\mathcal{F}(\Delta_1, \Delta_2) = \alpha_1 |\Delta_1|^2 + \alpha_2 |\Delta_2|^2 + \beta_1 |\Delta_1|^4 + \beta_2 |\Delta_2|^4 + 2\gamma_1 |\Delta_1|^2 |\Delta_2|^2 + \gamma_2 \left(\Delta_1^2 (\Delta_2^*)^2 + (\Delta_1^*)^2 \Delta_2^2\right) \tag{9}$$

where β_1 and β_2 are positive. The two orders coexist when $\beta_1 \beta_2 > (\gamma_1 - |\gamma_2|)^2$. This condition can be satisfied in the presence of disorder[59,60]. The relative phase θ between $\Delta_1 = |\Delta_1| e^{i\psi + \theta/2}$ and $\Delta_2 = |\Delta_2| e^{i\psi - \theta/2}$ is determined by the sign of the γ_2 term in (9). We found that γ_2 is positive:

$$\gamma_2 = \sum_{\mathbf{k}} (2 u_{\mathbf{k}} v_{\mathbf{k}})^2 \left[\frac{1}{|\xi_{\mathbf{k}}^a|^3} + \frac{1}{|\xi_{\mathbf{k}}^b|^3}\right]. \tag{10}$$

Minimization of Eq. (9) then shows that $\theta = \pm\pi/2$. Because $\theta = \pi/2$ and $\theta = -\pi/2$ are different states, the system spontaneously breaks the Z_2 TRS [61]. In the TRS-broken state, the phases of the order parameters $\langle aa\rangle_{\mathbf{k}}$ and $\langle bb\rangle_{\mathbf{k}}$ are φ and $\pi - \varphi$, where $0 < \varphi < \pi/2$. The third gap, which is generally required to satisfy the set of complex gap equations in the TRS-broken state is provided by $\langle ab\rangle_{\mathbf{k}}$, whose phase in this situation is $-\pi/2$. We show the gap structure schematically in Fig. 1 where we associated $\langle ij\rangle_{\mathbf{k}}$ with vectors, whose directions are set by the phases. We also performed Hubbard-Stratonovich analysis beyond mean-field level[50], by allowing the phases of $\Delta_{1,2}$ to fluctuate, and found (see SM) that when $T_{c,2} \approx T_{c,1} \equiv T_c$, the system breaks TRS and sets the relative phase $\theta = \pm\pi/2$ at a temperature $T^* > T_c$. In between T^* and T_c, TRS is broken, but the $U(1)$ symmetry associated with the global phase of Δ_1 and Δ_2 remains intact. Such a state is typical for systems whose order parameter manifold contains both continuous and discrete symmetries [2,23,62,63]. At T_c, the global phase is broken and both SC orders develop simultaneously. A schematic phase diagram is shown in Fig. 3a.

$s + e^{i\theta} t$ state So far we considered the "maximally-nested" case, with $\delta_0 = 0$. For the more generic case $\delta_0 \neq 0$ we find that the GL functional (9) contains a bilinear coupling between the two SC states, i.e. a term $\alpha_3 (\Delta_1 \Delta_2^* + \Delta_1^* \Delta_2)$ with $\alpha_3 < 0$ (details in the SM). In this situation, the onset of the s^{+-} state at $T_{c,1}$ necessarily triggers the emergence of a t state. The relative phase between the two order parameters at $T \le T_{c,1}$ is $\theta = 0$, i.e., the state is $s+t$. Yet, the SC state still breaks TRS at a lower temperature $T_{c,3} < T_{c,1}$. Indeed, comparing the $\alpha_3 (\Delta_1 \Delta_2^* + \Delta_1^* \Delta_2)$ and $\gamma_2 \left(\Delta_1^2 (\Delta_2^*)^2 + (\Delta_1^*)^2 \Delta_2^2\right)$ terms in the GL functional we immediately see that $\theta = 0$ only as long as $\Delta_1 \Delta_2 < \alpha_3 / 4\gamma_2$. Once the temperature is reduced and $\Delta_{1,2}$ grow, this condition breaks down at $T = T_{c,3}$, and at lower T the minimum of the GL functional shifts to $\theta \neq 0$. At a lower T, the SC state becomes $s + e^{i\theta} t$ and TRS gets broken (see Fig. 3b). This GL analysis attests the generality of our results. In particular, the momentum dependence of the interactions, introduced by the orbital content of the FS (which we neglected), would only change the GL parameters, but not the GL form and hence would not invalidate our conclusion that an $s + e^{i\theta} t$ state emerges at low T.

Conclusions In this paper we argued that a SC state, which explicitly breaks TRS, appears when SC emerges from a pre-existing SDW-ordered state. We found that in the presence of SDW, the spin-triplet channel with inter-pocket pairing couples to spin-singlet intra-pocket pairings on the reconstructed FSs. This leads to the emergence of a new pairing channel, which we labeled as $t-$pairing to emphasize that it involves spin-triplet. We analyzed the interplay between s^{+-} and $t-$ SC orders and showed that they coexist at low T with a relative phase $0 < \theta < \pi$. As a result, the phases of the gaps on different FSs differ by less than a multiple of π. Such a state breaks time-reversal symmetry and has been long sought in the studies of FeSCs. We argued that in a generic case TRS gets broken in the SC manifold at temperatures lower than T_c. This should give rise to features in experimentally probed thermodynamic quantities.

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