

Landau Theory for Pair Density Modulation in Fe(Te,Se) Flakes

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Motivated by recent scanning tunneling microscopy (STM) experiments reporting a pair-density modulation (PDM) in flakes of FeTe_{0.55}Se_{0.45}, we develop a Landau theory to elucidate its physical origin. We analyze the PDM in terms of screw and glide symmetries, interpreting it as a hybridized state of two order parameters with opposite glide and screw parity. To explain the absence of PDM in the bulk, we argue that the breaking of glide symmetry at the surface allows nematic order to selectively stabilize the PDM in thin flakes. From these symmetry constraints, we show that the opposing glide and screw parities of the condensate favor a site-based, rather than bond-based, pairing mechanism. Thus, the discovery of PDM in superconducting flakes suggests that the pairing in iron-based superconductors is local to the iron atoms, possibly driven by Hund's coupling. We argue that the mismatch in Knight shift between the even and odd parity order parameters will lead to a magnetic-field enhancement of the PDM at low fields, and a reentrant triplet phase at high fields that can be tested in STM experiments.

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Introduction—Iron-based superconductors [1–6] provide a fertile platform for exploring a wide variety of correlated electronic phases. Numerous experiments point toward an unconventional origin for the superconductivity in these materials [5,7,8]. A new kind of modulated superconducting state was discovered in recent scanning tunneling microscopy (STM) experiments [9–12] on ultrathin flakes of FeTe_{0.55}Se_{0.45}, which reveal [11] that the superconducting gap differs by up to 40% on the two iron sublattices, forming a “pair density modulated state” (PDM). Two sequential superconducting transitions are observed on cooling [11]: first, uniform superconductivity develops at 11 K, then a second transition into the PDM state occurs at 9 K [see Fig. 1(a)]. In the PDM, the gap, defined by one half the peak-to-peak separation in the density of states, becomes different at the two iron sites, *A* and *B*, in the unit cell. The gap modulation ($(|\Delta_A| - |\Delta_B|)/(|\Delta_A| + |\Delta_B|) = f$ is 20% in the sample principally studied in experiments, but rises to 40% in the thinnest flakes, while entirely vanishing in the thickest samples. Moreover, the modulated gap structure is also accompanied by a nematic order, forming domains in which the square iron plaquettes undergo a rhombohedral compression [see Fig. 2(c)] of about 1% along their diagonals.

The PDM is intriguing because it resembles a pair-density-wave (PDW) state [7] in which the superconducting order parameter also develops spatial modulation. However, unlike the PDW, the PDM does not break translational symmetry: in this sense it is the superconducting counterpart of the altermagnet [13], for both preserve lattice translational symmetry, because of an underlying nonsymmorphic crystal structure with two equivalent magnetic or superconducting atoms per unit cell. A second distinct feature of the PDM is that it develops spontaneously, in the absence of a preexisting broken symmetry in the normal state, such as a Zeeman split Fermi surface [14,15] or charge density-wave order [16,17] that often accompany PDWs.

In bulk FeSe_{1-x}Te_x the iron atoms form stacked square lattice layers with the chalcogenide Se or Te atoms alternately located above or below neighboring iron plaquettes. This gives rise to a nonsymmorphic structure described by space group *P4/nmm* (No. 129) [18,19] with two iron atoms per unit cell. The structure is invariant under a glide (*G_z*) [20] or a screw operation (*C₄*):

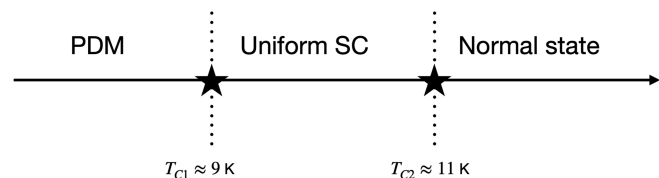


FIG. 1. Schematic illustration of the two-step phase transition. The ground state evolves from PDM state into a uniform superconducting order and then into a normal state. The two critical temperatures are $T_{c1} \approx 9$ K and $T_{c2} \approx 11$ K [11].

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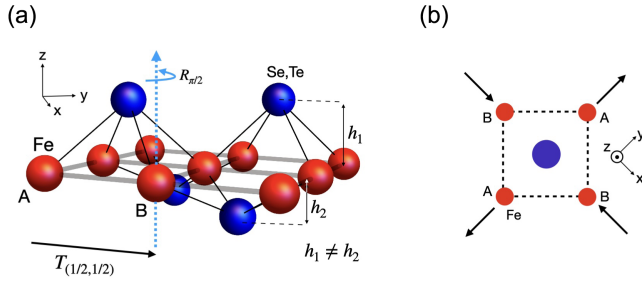


FIG. 2. (a) Structure of single-layer Fe(Te,Se) showing the different distance of the upper and lower chalcogenide atoms from the iron plane. (b) Atomic displacements associated with nematic order, which change sign under a π rotation.

$$G_z = M_z T_{(1/2,1/2)}, \quad \tilde{C}_4 = R_{\pi/2} T_{(1/2,1/2)} \quad (1)$$

corresponding to half-lattice translations $T_{(1/2,1/2)}$ followed by a reflection in the xy plane (M_z) or a $\pi/2$ rotation about the z axis ($R_{\pi/2}$) [see Fig. 2(a)]. However, in thin flakes, the chalcogens lie at different distances above and below the iron plane (see Fig. 2), removing the mirror symmetry [11], leaving just the screw symmetry to protect the equivalence of the two iron atoms. It is this symmetry that is broken by the PDM order. Many experimental observations have reported the existence of nematic order Φ [8,21] in Fe(Te,Se) or monolayer FeSe, which also breaks the screw symmetry which must therefore be considered as an important element in the development of the PDM.

In a recent paper, Papaj, Kong, Nadj-Perge, and Lee (PKPL) [22] propose a BCS description of the PDM state based on the loss of the glide symmetry. In their model, the development of a hybrid order parameter $s_{\pm} + d$ between an extended s - and d -wave superconducting phase removes the equivalence between the two lattice sites and giving rise to a modulated gap structure. While the PKPL theory provides an illustration of a PDM, the role of the symmetries is hidden within the structure of the internal tight-binding model and the assumed BCS couplings. In particular, the screw symmetry of the flakes does not play an explicit role in their theory.

Here, we develop a complementary description of the PDM based on Landau theory: our philosophy is to use the experimental properties of the PDM to gain insight into the microscopic physics. Landau theory allows us to observe the role of order-parameter symmetry and its interplay with nematic order; in particular, the screw symmetry of the flakes plays a central role. We are particularly interested in understanding why the PDM is present in thin flake samples, yet absent in the bulk. Moreover, our approach allows us to consider the constraints on the underlying pairing mechanism that are imposed by the properties of the PDM.

On rather general grounds, we expect that the order parameters which mix to produce the PDM are representations of the screw symmetry, with a well-defined

TABLE I. Symmetry of the three order parameters under fourfold screw operation $\tilde{C}_4$.

Order parameter	Φ	Δ_+	Δ_-
$\tilde{C}_4$ eigenvalues	-1	+1	-1

eigenvalue under the screw operation. Since the formation of PDM requires screw symmetry broken, it is natural to treat the PDM phase as a hybridization of two order parameters Δ_+ and Δ_- of opposite screw parity. Generically, two different order parameters of this kind are separated by a first order phase transition. We argue that their coupling to nematicity only arises at the surface, and this stabilizes the hybridized PDM in the thin flakes. This places some rather interesting constraints on the pairing that can be further tested by experiment.

Under the screw rotation $\tilde{C}_4$, the superconducting order parameter generally transforms as $\tilde{C}_4^{-1} \Delta \tilde{C}_4 = e^{im\pi/2} \Delta$, where m is the angular momentum quantum number. In our discussion, we focus on the time-reversal-symmetric cases with $m = 0, 2$, for which $\tilde{C}_4^{-1} \Delta_{\pm} \tilde{C}_4 = \pm \Delta_{\pm}$. The transformation properties of Δ_+ , Δ_- , and Φ are listed in Table I.

We can use symmetry to relate the gap parameters $\Delta_{A,B}$ on sublattice A and B to the order parameters $\Delta_{\pm}$. To leading order the onsite gap parameter Δ_A can be expressed as a linear combination $\Delta_A = \alpha \Delta_+ + \beta \Delta_-$. Under a screw rotation, $C_4^{-1} \Delta_A C_4 = \Delta_B$, so $\Delta_B = \alpha \Delta_+ - \beta \Delta_-$ picks up a relative minus sign. Adjusting the normalization of the order parameters so that $\Delta_A = \Delta_+ + \Delta_-$ and $\Delta_B = \Delta_+ - \Delta_-$. It follows that once the two order parameters coexist, screw symmetry is broken and the gap amplitudes develop a modulation between neighboring iron sites.

Landau free energy—To describe the PDM, we introduce a Landau free energy that is a function of nematic order Φ and two superconducting order parameters of opposing parity Δ_+, Δ_- ,

$$F = F_{\Delta} + F_{\Delta\Phi}, \quad (2)$$

where

$$F_{\Delta} = a[(T - T_c^+) \Delta_+^2 + (T - T_c^-) \Delta_-^2] + \frac{1}{2}(u_+ \Delta_+^4 + u_- \Delta_-^4 + 2u_{\pm} \Delta_+^2 \Delta_-^2),$$

$$F_{\Delta\Phi} = \alpha \Phi^2 + \lambda \Phi \Delta_+ \Delta_- \quad (3)$$

Here, F_{Δ} is the superconducting (SC) part of the free energy, while $F_{\Delta\Phi}$ describes the nematic order and its coupling to the superconductivity. T_c^+ and T_c^- are the intrinsic transition temperatures of the two superconducting order parameters, tuned by an external field g , so that $T_c^{\pm} = T_0 \pm g$ where $g = 0$ defines a multicritical point where

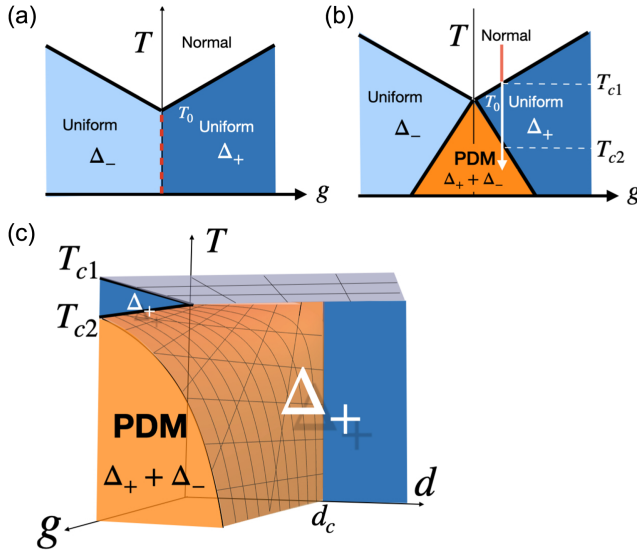


FIG. 3. Phase diagram of the Landau theory showing (a) when $u_{\pm} \geq U_c = \sqrt{u_+ u_-}$, where the coexistence region is absent and (b) where $u_{\pm} < U_c = \sqrt{u_+ u_-}$ and a hybridized phase develops beneath a tetracritical point. (c) Representative 3D phase diagram assuming a nematic coupling to the order parameters, exponentially dependent on flake thickness d , calculated using $u_{\pm} = 8$, $u_+ = u_- = 2$, $\lambda_0 = 7$, $\alpha = 1$, $\xi = 1$, $T_0 = 1$.

these transition temperatures are equal. $a > 0$ and u_- , $u_{\pm}$, and u_+ define the repulsion between the SC order parameters. Quartic terms that contain an odd power of Δ_+ or Δ_- are not invariant under the screw symmetry and are excluded. Finally, note that we assume that the two order parameters are phase locked, with a single overall phase that can be factored out of the free energy, so that the $\Delta_{\pm}$ are real. (See Ref. [23] for more details).

The second term $F_{\Delta\Phi}$ describes the nematic order and its coupling to the superconductivity. $\text{FeTe}_{0.55}\text{Se}_{0.45}$ lies in close proximity to a nematic quantum critical point [21,25], but does not spontaneously develop nematic order outside the PDM, permitting us to approximate the nematic component by a simple quadratic function $\alpha\Phi^2$, where $\alpha > 0$ denotes the distance from the nematic critical point. The minimal symmetry-allowed coupling $\Phi\Delta_+\Delta_-$ is the only term permitted by the screw symmetry, given Φ changes sign under screw rotations.

Our Landau theory highlights the importance of nematic fluctuations. Generically, the phase boundary between the even and odd-parity phases, depends on the strength $u_{\pm}$ of the repulsion between the order parameters (a detailed derivation is provided in Supplemental Material [24]): if $u_{\pm} > U_c$ exceeds the critical value

$$U_c = \sqrt{u_+ u_-}, \quad (4)$$

the transition between the two phases is first order as shown in Fig. 3(a), but if $u_{\pm}$ is less than this value, a tetracritical

point is formed and a hybridized coexistence region opens up within the phase diagram, as shown in Fig. 3(b). Now if we “integrate out” the nematic degree of freedom, rewriting

$$F_{\Delta\Phi} \rightarrow \alpha \left(\Phi + \frac{\lambda}{2\alpha} \Delta_+ \Delta_- \right)^2, \quad (5)$$

$$u_{\pm} \rightarrow u_{\pm}^* = u_{\pm} - \frac{\lambda^2}{4\alpha},$$

we see that nematic fluctuations lower the repulsion between the Δ_+ and Δ_- degrees of freedom, while also predicting that nematic order will develop as a secondary order parameter in a PDM with $\Phi = -(\lambda/2\alpha)\Delta_+\Delta_-$ [26].

This suggests a natural explanation for the absence of the PDM in bulk $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is that the nematic $\Phi\Delta_+\Delta_-$ is forbidden by the additional symmetries of the bulk. In the bulk, both glide-mirror and inversion symmetries are restored and Φ is even parity under both operations, so if $\Delta_{\pm}$ have *opposite* parities under either symmetry, the nematic coupling will vanish in the bulk. Provided that the system is close enough to the nematic critical point, (small α), so that

$$\frac{\lambda^2}{4\alpha} > u_{\pm} - \sqrt{u_+ u_-} > 0, \quad (6)$$

then by (5), $u_{\pm} > U_c$ in the bulk, but acquires the nematically suppressed value $u_{\pm}^* < U_c$ in the flakes, promoting PDM order. As the flakes get thicker, glide symmetry will be restored; modeling the dependence of the nematic coupling λ on thickness d as $\lambda(d) = \lambda_0 e^{-d/\xi}$, where ξ is the structural healing length, then using (6) we can identify an upper-critical flake thickness for the PDM

$$d_c = \frac{\xi}{2} \ln \left[\frac{\lambda_0^2}{4\alpha(u_{\pm} - \sqrt{u_+ u_-})} \right]. \quad (7)$$

Figure 3(c) shows the associated phase diagram. In principle, we could have added gradient terms $|\nabla\Delta_{\pm}|^2$ and $(\nabla\Phi)^2$ to the free energy (Ginzburg-Landau theory). Our simplified Landau theory describes the limit in which the superconducting and nematic coherence lengths are short compared with the healing length ξ . While gradient terms are needed to describe details of the domain walls and spatial structure, we do not expect them to change the basic phase diagram and symmetry constraints we have derived.

Discussion—Let us comment on the possible superconducting pairing configurations that can give rise to the pair-density modulated state. One candidate [22] is a bond-based pairing, in which Cooper pairs consist of electrons mainly residing at different lattice sites. Representative bond-based order parameters are extended s and d wave

TABLE II. Bulk glide G_z and spatial parity eigenvalues P for bond-based and local pairing scenarios, showing that a bulk nematic coupling is forbidden for local pairing.

(Δ_+, Δ_-)	G_z	P	Bulk $\Phi\Delta_+\Delta_-$ symmetry allowed?
$(s_\pm \text{ wave}, d \text{ wave})$	(1,1)	(+, +)	✓
(Uniform, staggered)	(1, -1)	(+, -)	×

$$\Delta(k) = \begin{cases} \Delta_+(\cos k_x + \cos k_y) & s_\pm \\ \Delta_-(\cos k_x - \cos k_y) & d, \end{cases} \quad (8)$$

where the x and y axes lie along the *diagonal* of the iron plaquettes [22]. However, in the bulk, these Cooper pair symmetries transform *identically* under inversion or glide mirror, so a nematic coupling $\lambda\Phi\Delta_+\Delta_-$ is permitted in both flakes and the bulk. Thus, bond-based pairing, though widely used in spin-fluctuation theories of iron-based superconductors [2,5,8], does not provide a natural explanation for the marked absence of PDM in bulk samples (see Table II).

An intriguing alternative is local pairing [27–31], in which pairs develop at the iron sites. Coulomb repulsion between electrons on the iron sites tends to rule out local s -wave pairing, so the alternative is triplet pairing, likely driven by the strong Hund's coupling inside the iron atoms. In this case, a minimal description involves a competition between a uniform (in-phase) and a staggered (out-of-phase) pairing state [27,32] within the 2-Fe unit cell. These have opposite glide and screw parities, restricting nematic coupling to the flakes where glide symmetry is absent. Representative order parameters take the form

$$\underline{\Delta}(\vec{x}) = \begin{cases} \vec{\Delta}_+ \cdot \vec{\sigma}(\sigma_2\lambda_2) & \text{uniform} \\ \vec{\Delta}_- \cdot \vec{\sigma}(\sigma_2\lambda_2)e^{i\vec{Q}\cdot\vec{x}} & \text{staggered,} \end{cases} \quad (9)$$

where $\vec{x}$ is the spatial coordinate of the iron sites, $\vec{\Delta}_{+,-}$ are the d vectors for the uniform and staggered triplet pairing, $(\lambda_2)_{\alpha\beta} = -(\lambda_2)_{\beta\alpha}$ is the antisymmetric operator acting on t_{2g} orbital states, e.g., $\alpha, \beta \in (d_{zx}, d_{zy})$, while $\vec{Q} = (2\pi, 0) \equiv (0, 2\pi)$ is the wave vector for the PDM. We note that the reestablishment of an inversion center between the Fe atoms in the bulk ensures that in the presence of spin-orbit coupling, on the Fermi surface, the uniform Δ_+ phase is an even parity singlet, while the bulk staggered Δ_- phase is an odd-parity triplet [27] (see Table II).

One of the most striking aspects of the PDM implied by Landau theory, is its vicinity to a tetracritical point. While this might reflect an accidental degeneracy between two unrelated order parameters, the symmetry-based local pairing perspective provides a more natural explanation. In this case we would expect that the BCS coupling constants of the two phases to originate from the same

local atomic interactions, so the tetracritical point can be understood as the point where their pair susceptibilities are equal.

We end with a discussion of the predicted effect of magnetic field on the PDM in $\text{FeSe}_{1-x}\text{Te}_x$. Here, an interesting precedent is set by the heavy fermion compound CeRh_2As_2 , which exhibits a magnetic field-tuned phase transition from a singlet to a triplet superconductor [33,34], thought to involve a change from uniform to staggered superconducting order [34,35]. If we regard CeRh_2As_2 as iron selenide without the nematicity, then we can use its example to anticipate the behavior of thin flakes of $\text{FeSe}_{1-x}\text{Te}_x$ in a magnetic field. The suppression χ_+ of the spin susceptibility or Knight shift of the singlet Δ_+ phase is expected to be substantially larger than the suppression χ_- in the Δ_- triplet phase. We can then model this physics by adjusting the quadratic terms of the Landau theory (3), writing

$$T_c^\pm(B) = T_0 \pm g - \chi_\pm B^2, \quad (10)$$

where $\chi_+ \gg \chi_-$. This corresponds to a mapping $T_0 \rightarrow T_0 - \frac{1}{2}(\chi_+ + \chi_-)B^2$ and $g \rightarrow g - \frac{1}{2}(\chi_+ - \chi_-)B^2$: in other words, a differential Knight shift will mean the magnetic field acts as the tuning parameter. The corresponding field-temperature phase diagram is then a conformal transformation of Fig. 3(b) which preserves its structure and tetracritical point, as shown in Fig. 4 (see Supplemental Material for details [24]). At large fields, the Pauli limitation of the singlet Δ_+ phase induces a second-order transition into the uniform triplet

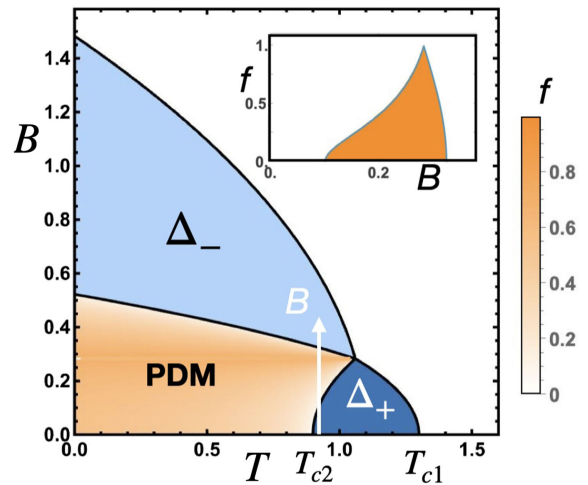


FIG. 4. Schematic field-temperature phase diagram predicted from Landau theory, according to (10), showing field stabilization of gap uniform, Δ_+ phase. Orange density labels the gap modulation parameter $f = (|\Delta_A| - |\Delta_B|)/(|\Delta_A| + |\Delta_B|)$. Inset: field dependence of f in PDM for field labeled by white arrow. Parameters used were $u_+ = u_- = 2, u_\pm = 1, T_0 = 1.2, g = 0.1, T_{c1} = 1.3, T_{c2} = 0.9, \chi_+ = 3; \chi_- = 0.5$.

Δ_- phase. Moreover, since $\Delta_{A,B} = \Delta_+ \pm \Delta_-$, the gap modulation parameter $f = (|\Delta_A| - |\Delta_B|) / (|\Delta_A| + |\Delta_B|) = \min[(\Delta_-/\Delta_+), (\Delta_+/\Delta_-)]$ is predicted to increase with field (see Fig. 4 inset), peaking in the center of the PDM phase, before returning to zero in the reentrant triplet phase. Of course, our treatment gives us no information about the direction of the d vector of the underlying order, so the relevant direction for applying the field cannot be inferred from the Landau theory. However, the main expectations are a robust prediction of the local triplet scenario for $\text{FeSe}_{1-x}\text{Te}_x$ (9) that can be tested in future STM measurements.

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Data availability—Numerical data used to generate the figures in our paper are available in Ref. [36].

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