Nematicity from mixed $S_\pm + d_{x^2-y^2}$ states in iron-based superconductors

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We demonstrate that in iron-based superconductors (SC), the extended $S_\pm$ SC state coexists with the $d_{x^2-y^2}$ state under generic conditions. The mixed $S_\pm + d_{x^2-y^2}$ SC is a natural nematic state in which the tetragonal symmetry $C_4$ is broken to $C_2$ explaining puzzling findings of nematic SC in FeSe films $[1]$. Moreover, we report the possibility of a first order transition at low-T from the nematic $S_\pm + d_{x^2-y^2}$ state to the pure $d_{x^2-y^2}$ state induced by the Zeeman magnetic field proposing an original experimental strategy for identifying our mixed nematic state in FeSe films. Extrapolating our findings, we argue that nematicity in non superconducting states of underdoped and undoped pnictides may reflect mixed $S_\pm + d_{x^2-y^2}$ Density Wave states.

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Iron-based superconductors (SC) support high critical temperatures and unconventional superconducting states like cuprates. However, while in high-T$_c$ hole doped cuprates the gap is certainly $d$-wave, in iron-based materials the situation is much more complex and still under investigation. In a series of materials like Ba$_{1-x}$K$_x$Fe$_2$As$_2$, BaFe$_{2-y}$Co$_y$As$_2$, FeTe$_{1-x}$Se$_x$ and K$_x$Fe$_2$Se$_2$ there is experimental evidence for nodeless superconductivity $[2]$. Apparently, this is not the usual isotropic s-wave gap but it is instead the extended s-wave $S_{text{ext}} \equiv S_\pm$ gap changing sign between the electron and hole Fermi surfaces $[3]$. On the other hand, in materials like LiFeP, LaOFeP, BaFe(As$_{1-x}$P$_x$)$_2$ and BaFe$_{2-d}$Ru$_d$As$_2$ experiments as diverse as NMR, STM, thermal conductivity and penetration depth measurements all point to the presence of gap nodes on the FS $[4, 5]$. On the theoretical side, unconventional either nodeless or nodal gap structures are usually regarded as evidence of non-phononic mechanisms $[3, 4]$. Nonetheless, we have reported recently $[7]$ that electron-phonon interaction dominated by small-q processes produces nodeless $S_\pm$ as well as nodal SC states depending on the doping, including a phonon-driven triplet $p$-wave state $[7]$ possibly observed recently in LiFeAs $[8]$.

Among the iron-based SC, FeSe is the simplest compound representing a prototype for this class of materials on which ideas may be tested. Recent scanning tunneling microscopy and spectroscopy measurements on high quality FeSe films by Song et al. $[9]$ reveal an astonishing feature. Apparently, the tetragonal symmetry $C_4$ is broken to $C_2$ pointing to a nematic SC state, that can certainly not be attributed to the tiny orthorhombic distortion of the FeSe lattice. Nematicity in iron pnictides is already a highly debated issue following reports for a possible electronic nematic phase transition $[4]$ that often coincides and possibly drives $[10, 11]$ the orthorhombic distortion which accompanies the antiferromagnetic transition in undoped and underdoped iron pnictides. Because of the coincidence of nematicity and antiferromagnetism, it has been suggested that the magnetism itself drives an electronic nematic phase transition $[12, 13]$.

However, in the case of FeSe there are no such antiferromagnetic phases involved. As a matter of fact, there have been also proposals for antiferromagnetism-independent-nematicity such as ferro-orbital nematic ordering $[14]$. In the present Letter we introduce a novel approach to the phenomenon of nematicity in pnictides. We demonstrate that $S_\pm$ SC coexists under generic conditions with $d_{x^2-y^2}$ SC and the mixed $S_\pm + d_{x^2-y^2}$ SC state is a prominent nematic state capable of explaining the reports of nematic SC in FeSe films $[1]$. Moreover, we point out that at low temperatures, a Zeeman field can induce a first order transition from the nematic $S_\pm + d_{x^2-y^2}$ state to the pure $d_{x^2-y^2}$ state. We therefore propose an experimental strategy for identifying the mixed $S_\pm + d_{x^2-y^2}$ character of the nematic SC state by repeating the experiments of Song et al. $[1]$ in the presence of in-plane magnetic fields in which case the Zeeman effect will dominate if the FeSe films are sufficiently thin $[15]$. Quite remarkably, we were able to produce self-consistently a Zeeman Field - Temperature phase diagram associated with the mixed nematic phase that exhibits three distinct SC regions and a tetrcritical point reminding the one observed in UPt$_3$ $[16]$. Finally, we show that singlet mixed SC states that violate time-reversal symmetry ($T$) are also accessible, exhibiting Zeeman field-induced transitions within the SC phase.

For our generic discussion, we can model qualitatively iron-based SC like FeSe with a minimal and sufficient two-band model exhibiting a hole pocket around the $\Gamma(0,0)$-point and an electron pocket around $M(\pi, \pi)$-point: $\varepsilon_x(k) = t_1 (\cos k_x + \cos k_y) - t_2 \cos k_x \cos k_y + C - \mu$ and $\varepsilon_y(k) = t_1 (\cos k_x + \cos k_y) + t_2 \cos k_x \cos k_y - C - \mu$ where $\mu$ is the chemical potential and we set $t_1 = 1$, $t_2 = 0.5$, $C = 2$. The present dispersions capture the necessary ingredients allowing for the $S_\pm$ state to emerge, namely well separated electron and hole Fermi surface (FS) sheets. Moreover, we have verified that the results that we report here are independent of any further band structure details. In fact, we keep our analysis as generic as possible by adopting a separable potentials approach for the effective interactions, allowing for a broad discussion concerning a large number of iron-based SC.
For this two band system we can write

\[ \mathcal{H} = \sum_{k, \sigma} \left[ \varepsilon_e(k)c_{k, \sigma}^\dagger c_{k, \sigma}^\dagger + \varepsilon_h(k)d_{k, \sigma}^\dagger d_{k, \sigma}^\dagger \right] - \frac{2}{N} \sum_{k, k'} V(k, k') \left( c_{k, \uparrow}^\dagger c_{k', \downarrow}^\dagger + d_{k, \downarrow}^\dagger d_{k', \uparrow}^\dagger \right) \left( c_{-k', \downarrow}^\dagger c_{-k, \uparrow}^\dagger + d_{-k', \uparrow}^\dagger d_{-k, \downarrow}^\dagger \right), \quad (1) \]

where \( c_{k, \sigma}^\dagger \) and \( d_{k, \sigma}^\dagger \) are annihilation (creation) operators for the electron \( \varepsilon_e(k) \) and hole band \( \varepsilon_h(k) \) bands respectively of spin projection \( \sigma = \uparrow, \downarrow \) and \( N \) the number of lattice points. Notice that for the specific choice of the interaction, where intraband and interband interaction strengths are equal and intraband and hole bands share the same superconducting order parameter \( \Delta(k) \), which is defined in the following manner

\[ \Delta(k) = -\frac{2}{N} \sum_{k'} V(k, k') \left( c_{-k', \downarrow} c_{k', \uparrow} + d_{-k', \uparrow} d_{k', \downarrow} \right) \]

\[ \Delta^*(k) = -\frac{2}{N} \sum_{k'} V(k, k') \left( c_{k', \uparrow} c_{-k', \downarrow} + d_{k', \downarrow} d_{-k', \uparrow} \right) \quad (2) \]

The order parameter \( \Delta(k) = \sum_n \Delta_n f_n(k) \) consists of irreducible representations (IR) \( f_n(k) \) of the point group \( D_{4h} \), that includes \( C_4 \) as a subgroup, and is appropriate for describing the normal (tetragonal) phase of strongly two-dimensional pnictide compounds. Here we shall restrict our analysis to the following IRs: \( f_3(k) = 1 \) \((A_{1g})\), \( f_{S_{\pm}}(k) = \cos k_x + \cos k_y \) \((A_{1g})\) and \( f_{d_{x^2-y^2}}(k) = \cos k_x - \cos k_y \) \((B_{1g})\). All of them are even under inversion as it is required for singlet intraband superconductivity and connect up to nearest neighbors. Note that by Fourier transforming the corresponding effective interaction field \( V(k, k') = \sum_n V_n f_n(k) f_n(k') \) with \( n = S, S_{\pm}, d_{x^2-y^2} \) one can see that in the context of real space extended Hubbard models this would correspond to on-site interactions of the form \( V_S \equiv U \delta_{i,j} \) and nearest neighbor interactions of the form \( V_{S_{\pm}}, V_{d_{x^2-y^2}} \equiv V_{i,j}(\delta_{i,j}+\delta_{i,j}(0,1)) \), where \( i \) and \( j \) are real-space lattice points indices. Nevertheless extended Hubbard models are not the unique option since also a small-q phonon-mediated pairing potential leads to a plethora of non s-wave IRs such as \( S_{\pm} \) or \( d_{x^2-y^2} \). With this reasoning we conclude that by studying here the SC phase competition via separable potentials we report generic results that are universally relevant, and independent of the exact microscopic mechanism of SC.

Within the aforementioned subspace of IRs, there are only two minimal schemes to achieve a nematic state. These are the mixed states \( S + d_{x^2-y^2} \) and \( S_{\pm} + d_{x^2-y^2} \). Of course, the cases \( S + S_{\pm} + d_{x^2-y^2}, iS + S_{\pm} + d_{x^2-y^2}, S + iS_{\pm} + d_{x^2-y^2} \) are also possible but not minimal. In all these symmetry breaking patterns the subgroup \( C_4 \) reduces to \( C_2 \). Notice that for a minimal nematic phase, the two IRs involved must lock in the same phase. If the two phases lock in phases with \( \pi/2 \) difference then the mixed state leads to broken \( T \) but unbroken \( C_4 \). These states are also important and for completeness we shall also discuss features of their phase diagram and their phenomenology.

We consider first the minimal configurations in which nematicity emerges and \( T \) is preserved. In these cases the order parameter \( \Delta(k) \) is the sum of two order parameters \( \Delta_{1,2}(k) \) corresponding to the two coexisting IRs. For simplicity we shall consider that \( \Delta_{1,2}(k) \) are real. At this point, we introduce the spinor \( \Psi_k = (c_{k, \uparrow}^\dagger d_{k, \downarrow}^\dagger c_{-k, \downarrow}^\dagger d_{-k, \uparrow}^\dagger) \) and employ the usual Pauli matrices \( \tau, \rho \). The mean field Hamiltonian can be written in the following compact form

\[ \mathcal{H} = \sum_k \Psi_k^\dagger \begin{pmatrix} \varepsilon_e(k) & 0 \\ 0 & \varepsilon_h(k) \end{pmatrix} - \mu \tilde{\tau}_3 \tilde{\rho}_0 - B \tilde{\tau}_0 \tilde{\rho}_0 + \Delta_1(k) \tilde{\tau}_1 \tilde{\rho}_0 + \Delta_2(k) \tilde{\tau}_1 \tilde{\rho}_0 \end{pmatrix} \Psi_k \quad (3) \]

where we also incorporated the effect of a Zeeman field \( B \). With usual techniques we calculate Green’s functions that exhibit four quasiparticle branches and coupled self-consistent equations that provide the two gaps \( \Delta_{1,2}(k) \).

To illustrate the fact that achieving mixed states is not trivial, we start with the competition between the isotropic S IR and the \( d_{x^2-y^2} \) IR. As expected, s and d-wave SC phases are highly competitive and coexistence

![FIG. 1: (color online) Typical temperature induced first order transition from S (red) to \( d_{x^2-y^2} \) SC (green) for various chemical potentials \( \mu \) obtained when \( V_S < V_{d_{x^2-y^2}} < 1.5V_S \). Free energy calculations not reported here verify the reality of this transition.](image_url)
Here, by verifying that it minimizes the corresponding free energy, we lower the temperature (Fig. 1). We confirm the realization of this transition, as we did for all results reported here, by verifying that it minimizes the corresponding free energy.

While the isotropic s-wave SC state cannot coexist with the d-wave state, the extended s-wave SC state that is widely considered relevant for iron-based superconductors coexists with $d_{x^2-y^2}$ over a wide range of the effective potentials (Fig. 2a). We report in Fig. 2b a typical solution in the mixed $S_\pm + d_{x^2-y^2}$ state plotted solely on the Fermi surface for a hole doped system $\mu = -0.4$. The emergence of nematicity is directly evident from Fig. 2b. In Fig. 2c we present free energy calculations, exhibiting four degenerate (because of symmetry) minima corresponding to the mixed nematic SC state. We insist that all effective potentials and dispersions used in our self-consistent calculations preserve tetragonal symmetry. Only because $S_\pm$ and $d_{x^2-y^2}$ coexist, tetragonal symmetry is broken and nematicity emerges. The essential ingredients leading to the nematic $S_\pm + d_{x^2-y^2}$ state is on one hand the well separated electron and hole pockets that favor the stabilization of the $S_\pm$ IR and on the other, some weak tendency towards the formation of the d-wave that is further assisted by the presence of $S_\pm$. The detailed characteristics of the Fermi surface topology are not crucial for the formation of the nematic state but mainly determine the exact balance of the $S_\pm$ and $d_{x^2-y^2}$ OP's, that controls the nodal or nodeless type of the quasiparticle excitation spectrum.

To demonstrate that our qualitative findings are not peculiar to details of the considered dispersion or to the separable potentials character of the above analysis, we also report in Fig. 3 a self-consistently obtained nodal nematic $S_\pm + d_{x^2-y^2}$ state using a fully momentum dependent small-q phonon-mediated interaction $V(k, k') = V_{Cph} - V_{ph}(q_f^2 + |k - k'|^2)^{-1}$ ($V_{Cph} \approx 0.1 V_{ph}$ and $q_f = \pi/6$) and an accurate hole doped ($\mu = -0.6$) four band model for high-$T_c$ iron pnictides described in Ref. 7. Naturally, also in this case both the dispersion and the interaction used in our calculations preserve the tetragonal symmetry, and only the resulting self-consistent solution depicted in Fig. 3 exhibits nematicity. Note that small-q phonon driven unconventional SC has also been considered in the past for high-$T_c$ cuprates [22, 23], heavy fermion [24], organic...
and cobaltite SC and is known to produce a loss of rigidity of the gap function in momentum space called momentum decoupling thus allowing for gap symmetry transitions. Note also that depending on the relative magnitude of the two order parameters, which in turn depends on the effective potentials, the mixed nematic state $S_{\pm} + d_{x^2-y^2}$ can be either nodeless as in the example of Fig. 2b or nodal as in Fig. 3. The small-q results are just a particular case confirming that the findings of the separable potentials analysis is generic. The nematic $S_{\pm} + d_{x^2-y^2}$ SC state is indeed a model independent phenomenon likely to be behind the puzzling reports of nematicity in FeSe films.

Moreover, the qualitative behavior of the mixed $S_{\pm} + d_{x^2-y^2}$ state in the presence of a Zeeman field may allow its firm identification from the experiments. At low temperatures, we obtain a remarkable first-order field-induced transition from the nematic $S_{\pm} + d_{x^2-y^2}$ state to the pure $d_{x^2-y^2}$ state (Fig. 4). The transition exists only at sufficiently low temperatures. We therefore propose an experimental approach for identifying the $S_{\pm} + d_{x^2-y^2}$ state by applying in-plane magnetic fields to the FeSe films instead of perpendicular fields as in [1]. Exploring the higher temperatures in the presence of the field allows to construct with self-consistent calculations Field - Temperature phase diagrams. Quite remarkably, for the zero field state shown in Fig. 2b, we obtain a Field-Temperature phase diagram exhibiting three distinct SC regions and a tetracritical point (Fig. 4) in analogy to the well known phase diagram of UPt$_3$ obtained there as well in the presence of in-plane fields [19]. In our case there is a simple understanding of this diagram as follows: The $S_{\pm}$ state is stronger in this example owing the higher $T_c$ at zero field. On the other hand, in the presence of a large Zeeman field, the $d_{x^2-y^2}$ state with nodes on the FS is energetically more favorable than a nodeless SC state [23], like $S_{\pm}$, which exhibits a higher critical field at zero temperature. Therefore, the reason for such a complicated phase diagram lies in the extraordinary fact that at zero field, a fully gapped state like $S_{\pm}$ allows at lower temperatures its coexistence with an emergent nodal $d$-wave state.

Mixed states may also lead to $T$ breaking despite the singlet character of the condensates. This naturally happens if the two coexisting order parameters lock in a $\pi/2$ phase difference. As we have already mentioned, when two different IRs coexist with this type of phase locking $C_4$ symmetry is preserved. The mean field Hamiltonian describing such a situation has the following form

$$\mathcal{H} = \sum_k \Psi_k \left\{ \tilde{\tau}_3 \left( \begin{array}{cc} \varepsilon_c(k) & 0 \\ 0 & \varepsilon_h(k) \end{array} \right) \right\} - \mu \tilde{\tau}_3 \hat{\rho}_0 - B \tilde{\tau}_0 \hat{\rho}_0 + \Delta_1(k) \tilde{\tau}_1 \hat{\rho}_0 - \Delta_2(k) \tilde{\tau}_2 \hat{\rho}_0 \right\} \Psi_k \quad (4)$$

leading to a different system of coupled self-consistent gap equations, compared to the previously examined case. We briefly report some results in Fig. 5 that will help the interpretation of eventually observed in-plane field-induced first order transitions in the experiments that we suggest in the previous paragraph. While the state $S + d_{x^2-y^2}$ is not accessible, breaking of $T$ due to $\pi/2$ phase locking, allows for sufficiently large $d$-wave potentials ($V_{4d_{x^2-y^2}} > 1.2V_0$) self-consistent solutions of the $S + id_{x^2-y^2}$ (or equivalently $iS + d_{x^2-y^2}$) type. Remarkably, here as well, we obtain at zero temperature a first order Zeeman field-induced transition from the
mixed $S + i d_{x^2-y^2} (iS + d_{x^2-y^2})$ state to the pure $id_{x^2-y^2}$ ($d_{x^2-y^2}$) state (Fig 5) that exhibits a qualitatively similar behavior to the field-induced transition from the $S_k + d_{x^2-y^2}$ state to the pure $d_{x^2-y^2}$ discussed in the previous paragraph. Nevertheless, the mixed $S + id_{x^2-y^2}$ state preserves the tetragonal symmetry and therefore a first order Zeeman field-induced transition within the SC phase does not necessarily imply the presence of a nematic SC state. The Zeeman field - temperature phase diagram depicted in Fig. 5 exhibits now only two distinct SC regions because the nodal $d_{x^2-y^2}$ state with the higher critical field is now stronger at zero field than the $S_k$ state having the higher critical temperature as well. On the other hand, for the competition of $d_{x^2-y^2}$ ($id_{x^2-y^2}$) with $iS_k (S_k)$ we observe no qualitative difference in the phase diagrams compared to the one obtained previously when the two order parameters lock in the same phase. In fact, it has been suggested that enhanced nematic height may favor this type of $T$ violating mixed states.

Finally, from the point of view of BCS theory, unconventional particle-hole condensates like spin singlet or triplet Density Waves (DW) behave similarly to SC condensates when the two bands are perfectly nested. Qualitatively, our present findings could be extrapolated to the DW condensates suggesting that the nematicity that accompanies the antiferromagnetic transition in underdoped and underdoped iron-pnictides may well indicate the presence of a nematic mixed $S_{\pm} + d_{x^2-y^2}$ spin DW state. Note that in particle-hole asymmetric systems a spin DW and charge DW exhibiting the same momentum state having the higher critical temperature as well. On conclusion, we claim that nematicity in iron-based superconductors indicates the presence of mixed condensates in which $d_{x^2-y^2}$ and $S_k$ order parameters coexist. We suggest the experimental search for an in-plane field-induced melting of nematicity in films of FeSe by an abrupt first order transition at low temperatures. Mixed states that break time reversal invariance are also shown to be accessible and should be taken into consideration in the analysis of experiments. If our findings for nematic mixed SC states are proven to be relevant in FeSe films, then it is probable that nematicity in non-superconducting phases of underdoped pnictides originates from analogous mixed nematic density wave condensates.

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