Generic nodeless Larkin-Ovchinnikov states due to singlet-triplet mixing in organic superconductors

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Larkin-Ovchinnikov (LO) states typically have a singlet gap that vanishes along real-space lines. These real-space nodes lead to Andreev midgap states which can serve as a signature of LO pairing. We show that at these nodes, an odd-parity, spin-triplet component is always induced, leading to a nodeless LO phase. We find the two-dimensional weak coupling, clean limit s wave phase diagram when this spin-triplet part is included. The triplet component is large and increases the stability of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase. We also show that the spin-triplet contribution pushes the midgap states away from zero energy. Finally, we show how our results can be explained phenomenologically though Lifshitz invariants. These invariants provide a simple approach to understand the role of unconventional pairing states, spin-orbit coupling, and inhomogeneous mixed singlet-triplet states that are not due to an FFLO instability. We discuss our results in the context of organic superconductors.

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I. INTRODUCTION

There are strong reasons to suspect that the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) (Refs. 1 and 2) phases appear in the quasi-one-dimensional Bechgaard salts $(TMTSF)_2X$ (Refs. 3 and 4) and in the quasi-two-dimensional (2D) organics $\kappa-(BEDT\text{-}TTF)_2Cu(NCS)_2$ (Ref. 5) and $\lambda\text{-}(BETS)_2GaCl_4$.[6] FFLO phases have also been argued to be of importance in understanding ultracold atomic Fermi gases[7,8] and in the formation of color superconductivity in high-density quark matter.[9] The understanding of these phases has become a relevant and topical pursuit in physics. A central result of theoretical studies is the ubiquitous appearance of the LO phase, a striped superconducting phase in which the spin-singlet order parameter vanishes spatially along lines.[10] Indeed, it has been suggested that the observation of Andreev bound states localized at these nodes would provide strong evidence for LO phase.[11,12]

Here we argue that the spin-singlet LO phase is generically nodeless due to the appearance of a spin-triplet component at the spatial nodes of the spin-singlet component. We further show that the triplet component is stabilized by "removing" the Andreev bound states, that is, by pushing these states away from zero energy.

We begin with a microscopic derivation of our main results. This derivation considers a 2D superconductor with spin-singlet s wave and spin-triplet p wave pairing interactions. This is followed by a phenomenological description that shows how Lifshitz invariants (LI) account for the microscopic results and allow for a significant generalization to include the effects of unconventional pairing states, spin-orbit coupling (SOC), and inhomogeneous singlet-triplet mixed states not due to an FFLO instability. While there have been prior studies of the role of p wave interactions on the FFLO phase[3,13–17] and in a related phase in cold atoms[18] and vortex states,[19] these studies have focused on the high-field region near the normal to superconducting phase transition where the gap is small. Here we examine the low-field transition from a usual superconductor to a LO phase which requires a solution of the nonlinear Eilenberger equations.

II. MICROSCOPIC FORMULATION

We use the Eilenberger equations as presented by Alexander.[11,12,20] The central equation for the quasiclassical Green's function $\hat{g}(\boldsymbol{R},\hat{\boldsymbol{k}};i\epsilon_n)$ is

$$[i\epsilon_n\hat{\tau}_z-\hat{\Delta}-\hat{v},\hat{g}]+i\boldsymbol{v}_f\cdot\boldsymbol{\nabla}\hat{g}=0, \tag{1}$$

where $\hat{\boldsymbol{k}}$ is the direction of the Fermi momentum, $\epsilon_n=\pi T(2n+1)$ are the Matsubara frequencies, and $\boldsymbol{v}_f$ is the Fermi velocity. We denote the three Pauli matrices in particle-hole space by τ_x, τ_y,and τ_z, and in spin space by $(\sigma_x,\sigma_y,\sigma_z)\equiv\boldsymbol{\sigma}$. The Green's function must satisfy Eilenberger's normalization condition $\hat{g}^2=-\pi^2\hat{1}$. The quasiclassical Green's function in Nambu space is

$$\hat{g}=\begin{pmatrix} g+\boldsymbol{g}\cdot\boldsymbol{\sigma} & (f+\boldsymbol{f}\cdot\boldsymbol{\sigma})i\sigma_y \\ i\sigma_y(f'+\boldsymbol{f}'\cdot\boldsymbol{\sigma}) & -g+\boldsymbol{g}\cdot\boldsymbol{\sigma}^* \end{pmatrix}. \tag{2}$$

The Zeeman coupling with the magnetic field is given by

$$\hat{v}=\begin{pmatrix} \mu\boldsymbol{B}\cdot\boldsymbol{\sigma} & 0 \\ 0 & \mu\boldsymbol{B}\cdot\boldsymbol{\sigma}^* \end{pmatrix}, \tag{3}$$

where μ is the magnetic moment of the electron. The order-parameter matrix in Nambu space is

$$\hat{\Delta}(\boldsymbol{R},\hat{\boldsymbol{k}})=\begin{pmatrix} 0 & (\Delta+\boldsymbol{\Delta}\cdot\boldsymbol{\sigma})i\sigma_y \\ i\sigma_y(\Delta^*+\boldsymbol{\Delta}^*\cdot\boldsymbol{\sigma}) & 0 \end{pmatrix}. \tag{4}$$

The self-consistency relations are

$$\Delta(\boldsymbol{R},\hat{\boldsymbol{k}})=N_0\pi T\sum_n\langle V(\hat{\boldsymbol{k}},\hat{\boldsymbol{k}}')f(\boldsymbol{R},\hat{\boldsymbol{k}}';i\epsilon_n)\rangle_{\hat{\boldsymbol{k}}'}, \tag{5}$$

$$\boldsymbol{\Delta}(\boldsymbol{R},\hat{\boldsymbol{k}})=N_0\pi T\sum_n\langle V(\hat{\boldsymbol{k}},\hat{\boldsymbol{k}}')\boldsymbol{f}(\boldsymbol{R},\hat{\boldsymbol{k}}';i\epsilon_n)\rangle_{\hat{\boldsymbol{k}}'}, \tag{6}$$

where $V(\hat{\boldsymbol{k}},\hat{\boldsymbol{k}}')$ is the pairing interaction, N_0 is the density of states at the Fermi level, and $\langle\,\rangle_{\hat{\boldsymbol{k}}'}$ denotes the average over the Fermi surface. To determine which phase is stable, we use the free energy derived from the Luttinger-Ward functional by Vorontsov and Sauls[21]

$$\Delta f(R) = \frac{1}{2}\int_0^1 d\lambda T \sum_n N_0 \int \frac{d^2p}{2\pi} \mathrm{Tr}\hat{\Delta}\left(\hat{g}_\lambda - \frac{1}{2}\hat{g}\right) \quad (7)$$

g_λ is an auxiliary propagator obtained from the solution to the Eilenberger equation with the physical order parameter scaled by the dimensionless coupling parameter $0 \le \lambda \le 1$

$$[i\epsilon_n \hat{\tau}_z - \lambda\hat{\Delta} - \hat{\upsilon}, \hat{g}_\lambda] + i\boldsymbol{v}_f \cdot \boldsymbol{\nabla}\hat{g}_\lambda = 0. \quad (8)$$

We include both singlet s wave interactions and triplet p wave interactions. We assume a 2D cylindrical Fermi surface and a paring interaction $V(\hat{\boldsymbol{k}},\hat{\boldsymbol{k}}') = V_s + V_t \hat{\boldsymbol{k}}\cdot\hat{\boldsymbol{k}}'$. The relative strength of triplet interaction is given by the parameter $T_p = T_t/T_s$ where $T_s(T_t)$ are the T_c for the singlet (triplet) pairing. Due to spin-rotational invariance, we will get equivalent results for the field chosen along any direction. We therefore set the field along $\hat{z}$ direction for convenience. However, we note that the magnetic field should be in the plane to ensure that vortices can be ignored. Similarly, we also assume spatial variations along the $\hat{x}$ direction. The structure of the Eilenberger equations then ensure that there will be a nonzero spin-triplet component of the order parameter of the form $\boldsymbol{d} = \hat{z}\sqrt{2}k_x/k_f \psi_z(x) = \hat{z}\sqrt{2}\cos\theta\psi_z(x)$. More specifically the self-consistency relations become

$$\psi(\boldsymbol{R}) = N_0 \pi T V_s \sum_n \int_0^{2\pi} \frac{d\theta}{2\pi} f(\boldsymbol{R},\theta;i\epsilon_n), \quad (9)$$

$$\psi_z(\boldsymbol{R}) = N_0 \pi T V_t \sum_n \int_0^{2\pi} \sqrt{2}\cos(\theta)\frac{d\theta}{2\pi} f_z(\boldsymbol{R},\theta;i\epsilon_n). \quad (10)$$

This leads to the gap function $\psi + \sigma_z k \psi_z(x)$ that appears in the Eilenberger equations for $\hat{g}$.

III. PHASE DIAGRAM

In the vicinity of the transition from the normal state to the superconducting states, we set $(\psi,\psi_z) = e^{iqx}(\tilde{\psi},\tilde{\psi}_z)$ and find the instability line H_{c2} by solving the linear-gap equation and optimizing H_{c2} with respect to q. The order parameter for a particular q, ψ_q, is a linear combination of the singlet and triplet parts, that is, $(\tilde{\psi},\tilde{\psi}_z) = (\alpha,\beta)\psi_q$. Due to parity symmetry, this solution has the same H_{c2} as ψ_{-q}, which is given by $(\tilde{\psi},\tilde{\psi}_z) = (\alpha,-\beta)\psi_{-q}$. As a consequence, just below H_{c2}, two solutions can appear: a solution for which only one of ψ_q or ψ_{-q} is nonzero (known as the FF phase); or a solution for which both are nonzero and $|\psi_q| = |\psi_{-q}|$ (known as the LO phase). To determine which of these phases appear at H_{c2} requires an analysis beyond the nonlinear gap equation. keeping up to order $|\psi|^4$ in the free energy, we find that both the FF and the LO phases appear. The FF phase takes up only a small portion of the phase diagram. Nevertheless, this has an important physical consequence. In particular, if the FFLO phase is generated created by a magnetic field applied in the plane, then an additional magnetic field applied along

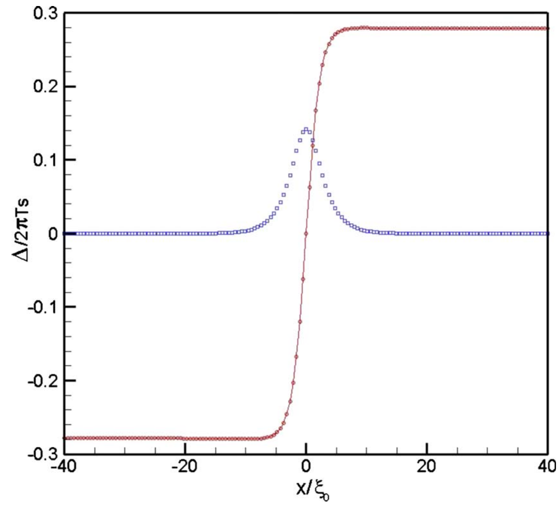

FIG. 1. (Color online) Singlet (circles solid) and triplet (squares) order parameters at $T = 0.2T_s$ for 2D FFLO superconductors with $T_p = 0.5$ and $\xi_0 = v_f/(2\pi T_c)$.

the $\hat{z}$ direction will lead to vortices. For the new order parameter $\boldsymbol{\psi} = (\psi_q, \psi_{-q})$, the general free-energy density can be written as

$$f = \alpha|\psi_q|^2 + \alpha|\psi_{-q}|^2 + \beta_1|\boldsymbol{\psi}|^4 + \beta_2|\psi_q|^2|\psi_{-q}|^2 + \kappa(|\boldsymbol{D}\psi_q|^2 + |\boldsymbol{D}\psi_{-q}|^2). \quad (11)$$

Equation (11) is independent of separate rotations of the phases of ψ_q and ψ_{-q}, revealing a global $U(1)\times U(1)$ gauge invariance. When the vortex core is encircled, the vortices of a $U(1)\times U(1)$ theory lead to a $2n\pi$ phase change in $\psi_q(\boldsymbol{r})$ and a $2m\pi$ phase change in $\psi_{-q}(\boldsymbol{r})$ classified by two integers (n,m), in particular, (1,1), (1,0), and (0,1) vortices. The (1,1) vortex is the usual Abrikosov vortex and it contains the usual flux quantum Φ_0, however, the (1,0) vortex contains a half-quantum flux $\Phi_0/2$. The degeneracy of the FF and LO phases ensures that *there exists a stable vortex lattice of half-quantum vortices*, in which each of the conventional (1,1) vortices decays into a pair of (1,0) and (0,1) vortices, as opposed to the usual Abrikosov lattice of full-quantum vortices.[22] This half-quantum lattice will exist in a region near where these two phases are degenerate.

We also compute the phase boundary from the uniform superconducting phase to the FFLO phase. In general, this requires a numerical solution of the Eilenberger equations. We use an efficient and numerically stable method described by Schopohl and Maki[23] in which the Eilenberger equations are transformed to Riccati equations. The transition from the uniform superconducting phase to the FFLO phases is found by computing the free energy of these two phases. Figure 1 shows the self-consistent order parameter at the transition from the uniform superconducting state to FFLO state for $T_p = 0.5$ and $T/T_s = 0.2$. (Microscopic calculations of Aizawa *et al.*[17] show that a combination of charge fluctuations and spin-fluctuations allows for strongly enhanced spin-triplet interaction so it is possible to have such large values of T_p. Note that a smaller T_p does not remove our predicted effects

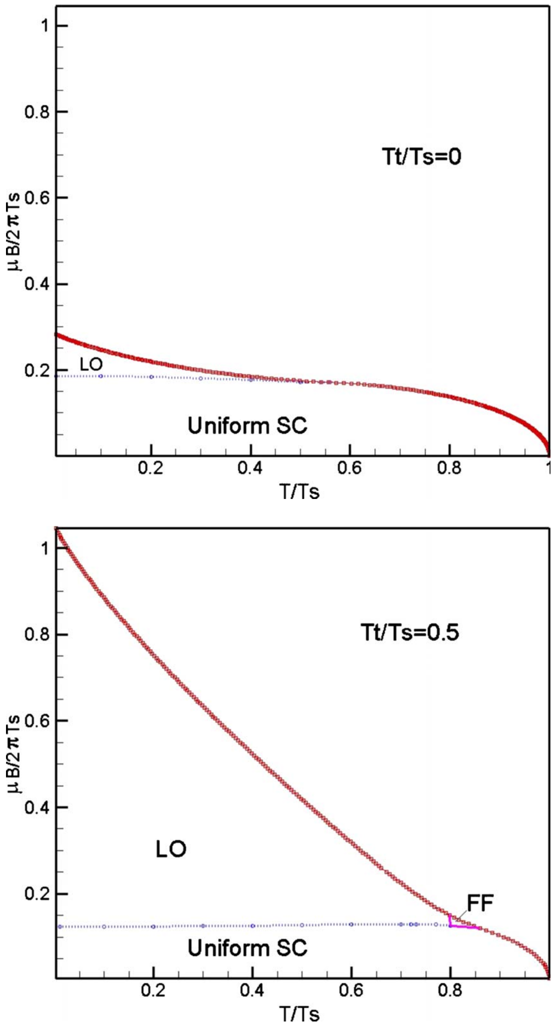

FIG. 2. (Color online) FFLO phase diagrams for $T_p = T_t/T_s = 0$ and $T_p = T_t/T_s = 0.5$. At low fields, the uniform superconducting to LO-phase transition is second order (circles dot). When $T_p = T_t/T_s = 0.5$, a FF phase appears in a small region of the phase diagram (solid lines).

but only makes them smaller). The spin-singlet order parameter is qualitatively similar to previous results on the LO phase.[11] However, the spin-triplet order parameter is maximum where the spin-singlet order parameter vanishes, removing the spatial-line nodes usually predicted in the LO phase. Furthermore, we find that if the spin-singlet order parameter is chosen real, then the spin-triplet order parameter is imaginary. Both the phase and the positions of the maxima of the spin-triplet order parameter are a natural consequence of the phenomenological arguments presented later. The complete H-T phase diagrams are presented in Fig. 2 for $T_p = 0.0$ and $T_p = 0.5$.

IV. QUASIPARTICLE PROPERTIES

Previous studies of the LO phase have found midgap Andreev states associated with sign change of the spin-singlet order parameter.[11,12] Given the removal of the gap through the appearance of a spin-triplet order parameter, we compute

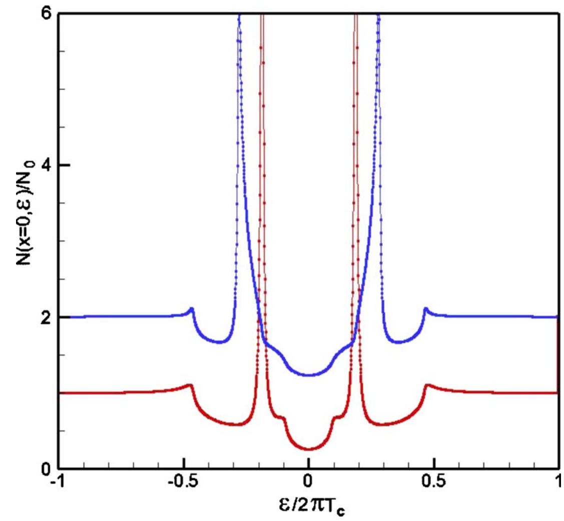

FIG. 3. (Color online) Total LDOS for spin-up/down electrons at nodes of spin-singlet order parameter for $T_p = 0$ (dot-dashed) and $T_P = 0.5$ (solid lines, the y axis has been offset by 1 for clarity).

the single-particle density of states to see what happens to these midgap states. The local quasiparticle density of states (LDOS) at point $\boldsymbol{R}$ with spin direction $\boldsymbol{e}$ can be calculated from

$$N_e(\boldsymbol{R};\epsilon) = -\left\langle \frac{1}{\pi}\mathrm{Im}[g(\boldsymbol{R},\hat{k};\epsilon) + \boldsymbol{e}\cdot\boldsymbol{g}(\boldsymbol{R},\hat{k};\epsilon)] \right\rangle_{\hat{k}}, \quad (12)$$

where $i\epsilon_n \rightarrow \epsilon + i0^+$. In Fig. 3, we show the total LDOS at the nodes of the spin-singlet order parameter including both spin-up and spin-down excitations. These results compare the solutions for $T_p = 0$ and $T_p = 0.5$. The LDOS for spin-down electrons can be found by reflecting LDOS for spin-up electrons through zero energy. When $T_p = 0$, there exist Andreev bound states with energies pinned to the middle of the gap. This agrees with previous studies.[11,12] Once the spin-triplet part becomes nonzero, these states are shifted away from zero energy. This shifting of these provides a microscopic mechanism through which the spin-triplet order parameter is energetically stabilized. We note that a similar Andreev bound-state removal mechanism has been proposed to explain the occasional appearance of spin-density wave (SDW) order at the spin-singlet nodes.[24,25] An important difference with our results is that the spin-triplet order we find is required to appear by symmetry while the SDW order is not.

One physical property associated with the Andreev midgap states is the appearance of an increased ferromagnetic magnetization at the nodes of the spin-singlet order parameter.[11] To investigate the role of the spin-triplet order parameter on this, we calculate the magnetization and find that the spatial peak of magnetization and the total magnetization both decrease due to the shift of the Andreev states to higher energy.

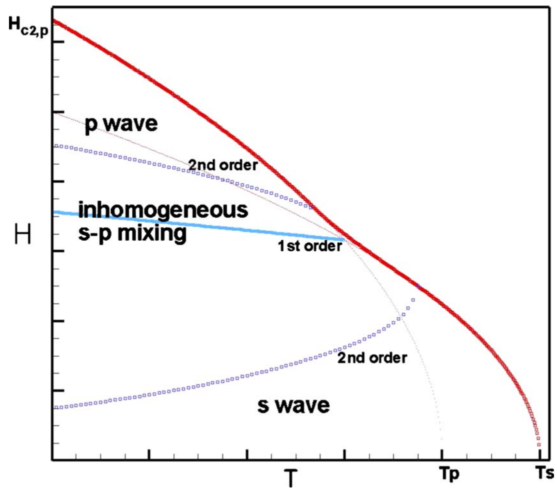

FIG. 4. (Color online) Phase diagram of inhomogeneous singlet-triplet phase with chosen parameters. A first-order transition (solid line) occurs without the LI. Two second-order transitions (squares) from the pure-singlet and pure-triplet phases into an inhomogeneous singlet-triplet mixed state occur when the LI is included.

V. PHENOMENOLOGICAL THEORY

A. Lifshitz invariants

We now turn to a phenomenological description of the above microscopic results. This phenomenological theory shows that the appearance of a spin-triplet component is generic and not specific to the microscopic details. The key point is that the admixture of spin-singlet and spin-triplet order parameters is due to the existence of Lifshitz invariants in the Ginzburg-Landau free energy (such invariants where first discussed by Mineev and Samokhin[26]). In particular, if the spin-triplet order parameter has the form $\boldsymbol{d}(\boldsymbol{k},\boldsymbol{R})=\Sigma_{i,j}A_{i,j}(\boldsymbol{R})\hat{x}_i k_j$ and if $\psi_s(\boldsymbol{R})$ describes the s wave pairing, then symmetry allows the following LI (note that a similar LI has been found in the context of cold atoms[18]):

$$\sum_{i,j} H_i[A_{i,j}(i\nabla_j\psi_s)^* + A^*_{i,j}(i\nabla_j\psi_s)]. \tag{13}$$

If, for example, the spin-singlet order parameter is given by $\psi(\boldsymbol{R})=\psi_0\cos(qR_j)$, then this term implies $A_{l,j}(\boldsymbol{R})=\psi_{t0}iH_l q\sin(qR_j)$ with $\psi_{t0}\neq 0$. This LI ensures that a spin-triplet component is *always induced*. This captures some of the main results found in the microscopic theory: the triplet order parameter is largest where the spin-singlet order parameter vanishes; and the relative phase between the spin-singlet and spin-triplet order parameters is $\pi/2$. The LI can also be generalized to unconventional spin-singlet order parameters and the role of SOC. For example, if the spin-singlet pairing is $d_{x^2-y^2}$, then the following Lifshitz invariant exists:

$$\sum_i H_i[A_{i,x}(i\nabla_x\psi_d)^* + A^*_{i,x}(i\nabla_x\psi_d) - A_{i,y}(i\nabla_y\psi_d)^* - A^*_{i,y}(i\nabla_y\psi_d)]. \tag{14}$$

This implies f wave spin-triplet pairing appears and once again, the magnitude of the f wave component is largest where the d wave component vanishes. This has been argued to be relevant in the organic $(\mathrm{TMTSF})_2X$.[17,27] Furthermore, for example, in a tetragonal material with spin-singlet s wave order ψ_s, spin-orbit interactions allow the following LI:

$$\eta\sum_j [\psi_j(i\nabla_j\psi_s)^* - \psi^*_j(i\nabla_j\psi_s)], \tag{15}$$

where ψ_i is defined through $\boldsymbol{d}(\boldsymbol{k},\boldsymbol{R})=[\psi_y(\boldsymbol{R})k_x-\psi_x(\boldsymbol{R})k_y]\hat{z}$. In this case, the triplet component will have the same phase as the s-wave component (as opposed to the $\pi/2$ phase shift for the field-induced LI).

B. Singlet to triplet-phase transition

The existence of the LI plays another role not tied to the LO phase. In particular, it has been argued that a singlet to triplet-phase transition may occur in $(\mathrm{TMTSF})_2X$ superconductors without the existence of a FFLO phase.[3,17] Such a transition is typically first order. The LI terms can transform this first-order transition into a pair of second-order transitions between which lies an inhomogeneous singlet-triplet mixed phase. To model a singlet to triplet transition, consider the following free-energy density:

$$f=\alpha_s|\psi_s|^2+\alpha_p(|\psi_x|^2+|\psi_y|^2)+\beta_s|\psi_s|^4+\beta_p(|\psi_x|^2+|\psi_y|^2)^2 + \kappa_s|\nabla\psi_s|^2+\kappa_p(|\nabla\psi_x|^2+|\nabla\psi_y|^2), \tag{16}$$

where we take $\alpha_s=\alpha_{s0}(T-T_s+\epsilon_s H^2)$ and $\alpha_p=\alpha_{p0}(T-T_p+\epsilon_p H^2)$. Choosing parameters $T_s>T_p$ and $\epsilon_s>\epsilon_p$ leads to a low-field s-wave state and a high-field spin-triplet state. Without the LI, a first-order transition occurs when $\alpha_s^2/\beta_s=\alpha_p^2/\beta_p$. Now consider the role of the LI, near the normal to superconducting phase boundary, where it is sufficient to consider the quadratic free energy. The singlet to triplet transition will occur at $T=T'$ when $\alpha_s(T')=\alpha_p(T')$. We can expand the s wave and p wave order parameters into the Fourier mode $q\hat{x}$. Close to this point $T=T'$, when the LI is included, the quadratic free energy is always minimized by introducing an inhomogeneous state where $\psi_s\propto e^{iqx}$ and $\psi_x\propto e^{iqx}$ and they are both nonzero. This solution intervenes between the pure singlet and triplet states and the transition into this inhomogeneous phase is second order from both the pure-singlet and pure-triplet phases. A stability condition for this phase is $q^2=\min(va,0)$ where $va=(\kappa_s\alpha_p+\kappa_p\alpha_s-\eta^2)/(2\kappa_s\kappa_p)$. We can also extend the analysis of this phase into the whole phase diagram where the quartic terms in the free energy are also considered. We find that when $\eta^2>\kappa_s(\alpha_s-\alpha_p)^2/(\alpha_p-2\alpha_s)$, there is a second-order transition from pure singlet to the intermediate phase, and when $\eta^2>\kappa_p(\alpha_s-\alpha_p)^2/(\alpha_s-2\alpha_p)$, a second-order transition from pure-triplet to the intermediate phase occurs. A representative phase diagram is given by Fig. 4 with parameters chosen to clearly exhibit the phases ($\alpha_{p0}=0.9$, $T_p=0.8$, $\epsilon_p=0.5$, $\eta=0.2$, and $\alpha_{s0}=T_s=\epsilon_s=\beta_s=\beta_p=\kappa_s=\kappa_p=1$). Consequently, even if there is no FFLO phase in $(\mathrm{TMTSF})_2X$, it would be of interest to look for this inhomogeneous singlet-triplet phase.

VI. CONCLUSION

In conclusion, we present microscopic arguments that show that the spatial-line nodes of spin-singlet LO phases are removed by the appearance of a spin-triplet components. We show that this can be understood phenomenologically through the existence of Lifshitz invariants in the free energy which also ensure that the spin-triplet component always appears in a spin-singlet FFLO phase. This or related inhomogeneous singlet-triplet mixed states are likely to exist in the organic superconductors $(\mathrm{TMTSF})_2X$.

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