Pauli-limited upper critical field in dirty d-wave superconductors

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We calculate the Pauli-limited upper critical field and the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) instability for dirty d-wave superconductors within the quasiclassical theory using the self-consistent t-matrix approximation for impurities. We find that the phase diagram depends sensitively on the scattering rate and phase shift of nonmagnetic impurities. The transition into the superconducting state is always second order for weak (Born) scattering, while in the unitarity (strong) scattering limit a first-order transition into both uniform and spatially modulated superconducting states is stabilized. Contrary to general belief, we find that the FFLO phase is robust against disorder and survives impurity scattering equivalent to a Tc suppression of roughly 40%. Our results bear on the search of FFLO states in heavy-fermion and layered organic superconductors.

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Introduction. In type-II singlet superconductors a magnetic field suppresses superconductivity for two reasons: (1) the phase of the Cooper pair wave function couples to the vector potential resulting in the appearance of vortices; (2) Zeeman coupling of the magnetic field to the electron spins polarizes and splits the conduction band, which destroys superconductivity when the loss in magnetic energy equals the energy gain from pair condensation. This latter mechanism is referred to as Pauli limiting and leads to a first- or second-order transition from the normal (N) to superconducting (SC) state depending on the value of the magnetic field. It has been predicted that a clean system at high fields can remain superconducting beyond the Pauli limit by forming the nonuniform Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state with a spatially modulated order parameter. This state, however, is suppressed by disorder.

In contrast to conventional (isotropic s-wave) superconductors, unconventional (d-wave) superconductors are affected by nonmagnetic impurities even at zero field; scattering averages the gap over the Fermi surface and suppresses Tc. The different rates of suppression of the uniform and FFLO states determine the phase diagram in the field-temperature (B-T) plane. Agterberg and Yang found that in two-dimensional (2D) d-wave superconductors with purely Zeeman coupling, the N-SC transition is of second order at all Tc, with the Larkin-Ovchinnikov (LO) modulation, \(\Delta_{LO} \sim \cos q \cdot R\), and the uniform (USC) state, \(\Delta_{USC} = \text{const}\), favored at low and high temperatures, respectively, and a narrow intermediate T region, where the nodeless Fulde-Ferrell (FF) state, \(\Delta_{FF} \sim e^{i q \cdot R}\), is stabilized. Ref. reported that under combined orbital and Zeeman coupling in impure d-wave superconductors the first-order transition into the vortex state appears at intermediate temperatures. Very recently, Houzet and Mineev studied orbital and impurity effects in s-wave and d-wave Pauli-limited superconductors and concluded that orbital effects are necessary for a first-order transition to occur in 2D d-wave superconductors. In contrast, for s-wave in 3D the transition to the FFLO state is first-order.

Remarkably, our understanding of impurity effects in nonuniform states is still incomplete. In Refs., the discussion was limited to weak (Born) impurity scattering and focused only on the Ginzburg-Landau (GL) regime close to the onset of the FFLO instability, using an expansion in the modulation wave vector q. However, q increases rapidly to values comparable to the inverse superconducting coherence length, \(q \xi_0 \sim 1\), so this expansion quickly becomes invalid away from the critical point.

In this Letter, we present a microscopic treatment of impurity effects on the superconducting states in purely Pauli-limited quasi-2D d-wave superconductors. Impurities are treated in the self-consistent t-matrix approximation (SCTA) covering the weak (Born) and strong (unitarity) scattering limits. The latter limit, never considered previously, is especially important because of a search for FFLO-like states in heavy-fermion and layered organic superconductors, where impurity scattering is strong. Our approach is not limited to an expansion in q, and hence is valid for any temperature and impurity concentration along the second-order upper critical field \(T_{c2}\). We show that the phase diagram of a Pauli-limited dirty d-wave superconductor is very different from nonmagnetic impurities in the Born and unitarity limits. The differences originate from the dependence on scattering strength of quartic and higher order coefficients in the GL functional. The first order N-SC transition, absent for Born scattering, is stabilized by strong impurities, and is therefore expected in heavy fermion systems.

Quasiclassical equations. We follow Refs. and solve the quasiclassical equations for the 4 \times 4-matrix Green’s functions in particle-hole and spin space, which...
satisfy the normalization condition, \( \hat{g}^2 = -\pi^2 \hat{1} \), and the transport equation,

\[
[i\varepsilon_{\tau} \hat{s}_{\tau} - \mu B \cdot \hat{S} - \hat{\Delta}(\hat{R}, \hat{p}) - \hat{\sigma}_{imp}(\hat{R}; \varepsilon_m), \quad (1) \\
\hat{g}(\hat{R}, \hat{p}; \varepsilon_m) + i\hbar v_F(\hat{p}) \cdot \nabla_R \hat{g}(\hat{R}, \hat{p}; \varepsilon_m) = 0.
\]

Here \( \mu \) is the magnetic moment, \( \varepsilon_m = \pi k_B T (2n + 1) \) \( \alpha_{imp} \) are the Matsubara frequencies, \( \hat{S} \) is the mean-field superconducting order parameter depending on the coordinate, \( \hat{R} \), and momentum direction, \( \hat{p} \), at the Fermi surface with velocity \( v_F \). The electron spin operator is \( \hat{S} = \alpha \tau/2(1 + \hat{\gamma}_3) + \sigma^* \tau(1 - \hat{\gamma}_3) \). The Pauli matrices \( \sigma \) and \( \tau \) operate in spin and particle-hole space, respectively.

\[\text{Eq. (1) is complemented by self-consistency equations for } \hat{\Delta} \text{ and the impurity self-energy } \hat{\sigma}_{imp}. \text{ We use } h = k_B = 1.\]

In the SCTA \( \hat{\sigma}_{imp} = n_{imp} \hat{f} \), with impurity concentration \( n_{imp} \). For isotropic scattering the \( t \)-matrix satisfies \( \hat{t}(\hat{R}; \varepsilon_m) = u_0 \hat{1} + u_0 N_f(\hat{g}(\hat{R}, \varepsilon_m), \hat{p}) \hat{t}(\hat{R}; \varepsilon_m) \), where angular brackets \( \langle \ldots \rangle \) denote a normalized Fermi surface average. The strength of the nonmagnetic impurity potential, \( u_0 \), is expressed via the isotropic scattering phase shift, \( \delta_0 = \arctan(\pi u_0 N_f) / N_f \) is the density of states per spin at the Fermi surface. For Born (unitarity) scattering \( \delta_0 = 0 \) (\( \hat{\sigma}_{imp} \)) and the normal-state scattering rate \( \Gamma = 1/2 T N_f = \Gamma_0 \sin^2 \delta_0 \), with \( \Gamma_0 = n_{imp} / \pi N_f \).

If we choose the direction of the spin quantization along \( B = B_\parallel \) (which is allowed if the hamiltonian has spin-rotation symmetry in the absence of the field), both \( \hat{g} \) and \( \hat{\sigma}_{imp} \) have block-diagonal structure corresponding to the two spin projections. Hence, the quasiclassical equations for the spin-up and spin-down sectors decouple, and we solve separately for the diagonal, \( g_s \), and off-diagonal, \( f_s, f_d \), components of \( \hat{g} \), with \( s = \pm 1 \{1, 1 \} \), with the constraint \( g^2_s - f_s f_d = -\pi^2 \). However, both spin projections enter the self-consistency equation for \( \hat{\Delta} \). We assume a separable pairing interaction \( \hat{Y}(\hat{p}) \hat{Y}(\hat{p}') \), where \( \hat{Y}(\hat{p}) \) gives the angular dependence of the gap function with the normalization \( \langle \hat{Y}^2(\hat{p}) \rangle = 1 \). For \( \hat{\Delta}(\hat{R}, \hat{p}) = \Delta(\hat{R}) \hat{Y}(\hat{p}) \), we find

\[
\Delta(\hat{R}) \ln \frac{T}{T_c} = T \sum_{\varepsilon_m} \left( \langle \hat{F}(\hat{R}, \hat{p}; \varepsilon_m) \rangle - \frac{\pi \Delta(\hat{R})}{\varepsilon_m} \right), \quad (2)
\]

\[
\hat{\sigma}_{imp} = S_\alpha \left( \cot \delta_0 + \langle \hat{g}_s \rangle / \pi \right) \left( \frac{(f_s)}{\pi} \cot \delta_0 - \langle \hat{g}_s \rangle / \pi \right). \quad (3)
\]

Here \( \hat{F}(\hat{R}, \hat{p}; \varepsilon_m) = \frac{1}{2} \hat{Y}(\hat{p})[f_s(\hat{R}, \varepsilon_m) + f_d(\hat{R}, \varepsilon_m)] \) and \( S_\alpha = \Gamma/[1 - \pi^2 \delta_0^2 (\langle \hat{g}_s \rangle^2 - (f_s)^2 + \pi^2)] \). To calculate the \( B-T \) phase diagram, we derive the Ginzburg-Landau functional (expansion in \( \Delta \) for arbitrary \( q \)) by taking \( \Delta(\hat{R}) = \sum \Delta_q \exp(i q \hat{R}) \) and solving Eqs. (1)-(3) together with the normalization condition for \( \hat{g} \) to third order in \( \Delta \). We substitute the \( n \)-th order solutions \( f_s^{(n)} \), \( f_d^{(n)} \) into Eq. (2) to obtain the GL free energy difference between the SC and N states,
plicitly depends on the scattering phase shift, Eqs. (1c)-(1d). For example, it controls the location of the first-order transition to the USC state, $T_P$, which competes with the FFLO instability. In unconventional superconductors $\langle \gamma \rangle = \langle \gamma_0 \rangle = 0$ and the critical point $T_P$ is determined by a sign change of the GL coefficient $\beta$ at $q = 0$,

$$\beta_0 = \pi T \text{ Re} \sum_{\epsilon_m > 0} \left( \frac{\langle \gamma \rangle}{D_0^b} - \frac{\Gamma(1 - 2 \sin^2 \delta_0)}{D_0^l} \right). \quad (6)$$

For $\Gamma = 0$, both $\kappa$ and $\beta_0$ become negative at exactly the same temperature, $T_{q\neq0} = T_P \approx 0.5615 T_{c0}$. Since the transition into the FFLO state has a higher critical field at any temperature $T < T_P$, the first-order transition is superseded by the onset of the FFLO state.

A comparison of $\kappa$ and $\beta_0$ shows that in dirty unconventional superconductors $T_{q\neq0} = T_P$ only for $\delta_0 = \pi/4$. For Born (B) and unitarity (u) scattering $\beta_0$ depends on $\delta_0$, such that $T_P^B$ and $T_P^u$ shift in opposite directions relative to $T_{q\neq0}$, hence $T_P^B < T_{q\neq0} < T_P^u$ as shown in Fig. 1. The latter inequality is especially important since it shows that for strong scatterers Pauli limiting leads to a first-order transition into the USC state at high fields/low temperatures in the $B$-$T$ phase diagram. As the system becomes dirtier, i.e., the lifetime $\tau_N$ decreases, these characteristic temperatures are suppressed to zero in the following order, $T_P^B \to 0$ at $\Gamma/\pi T_{c0} \gtrsim 0.18$, $T_{q\neq0} \to 0$ at $\Gamma/\pi T_{c0} \gtrsim 0.20$, and $T_P^u \to 0$ for $\Gamma/\pi T_{c0} \gtrsim 0.22$. Note that for larger $\Gamma$ the N-USC transition line is of second order at all $T$.

Fig. 1 gives the upper critical field lines for different states. Second-order transition lines are found by the largest spatial modulation vector $q \equiv Q$ that maximize $B_{c2}$. In clean $d$-wave SC [12, 13, 14, 19], the modulation is along a gap maximum (antinode) at low $T/T_{c0} < 0.06$, and along a gap node for $0.06 < T/T_{c0} < 0.56$; see Fig. 1(a). However, already for small impurity scattering, $\Gamma/\pi T_{c0} > 0.02$, the critical field for $q||\text{antinode}$ is lowered below $B_{c2}^{u||\text{node}}$, and the stable configuration is with $q||\text{node}$ over the entire range of existence of the FFLO state, see Fig. 1(b).

Determining the first-order transition lines of $B_{c2}$ requires a self-consistent calculation of the full free energy functional, the details of which will be given elsewhere [20]. We find that in the Born limit the first-order transition always is below $B_{c2}^{FFLO}$, in agreement with [5]. In contrast, in the unitarity limit $T_{q\neq0} < T_P^u$ and $B_{c2}^{FFLO}$ is below the first-order transition to the USC state, see Figs. 1(b-d).

For intermediate impurity scattering, the phase diagram is given in Fig. 2. To determine the structure of the SC state near $B_{c2}$, we analyze the GL free energy, Eq. (4), for four possible phases: USC [$\Delta(R)$ = $\Delta_{USC}$], FF with a single Fourier component $Q_1$ = $(Q, 0)$ [$\Delta(R)$ = $\Delta_{FF} \exp(iQx)$], LO with $\{Q_1, Q_2\} = \{\pm 0, 0\}$ [$\Delta(R)$ = $\Delta_{LO} \cos Qx$], and square lattice (SQ) with $\{Q_1, Q_3, Q_2, Q_4\} = \{0, 0, 0\}$ [$\Delta(R)$ = $\Delta_{SQ} \sqrt{\cos Qx + \cos Qy}$]. The $x$-$y$ axes are along the gap nodes. For each phase, we calculate $\Delta Q^GL = -\alpha^2/\beta_i$, with $\beta_{FF} = 2/1111$, $\beta_{LO} = 2/111 + 2/1313$ and $\beta_{SQ} = 0.5(1111 + 2/1212 + 2/313 + 2/1414 + 2/3214)$, where $\beta_{ijkl} = \beta(T, B; Q_i, Q_j; Q_k, Q_l)$. Along the second-order transition line the phase with the lowest positive value of $\beta$ has the lowest energy.

For Born impurities (Fig. 2 right), $\beta_i > 0$ for all nonuniform states, and the LO state is favored in most of the phase diagram except a small region below $T_{q\neq0}$, where the FF phase is stabilized for the impure case [5]. Analysis of $\Delta Q^GL$ indicates that this phase is separated by a second-order transition from the USC and by first-order from the LO state.

The situation is very different for strong impurities (Fig. 2 left). Following the $B_{c2}$ line from $T_c(B = 0)$ to
the Γ-T transition is of first order. At T < Tc, the LO state just below Tc is suppressed below the onset of the nonuniform state, Tc(Γ) < Tc, and the transition is always of second order. Impurities stabilize a narrow region of the Fulde-Ferrell state just below Tc. In contrast in the unitarity limit (relevant to recent experiments) Tc < Tc, and the first-order transition into the uniform state preempts a modulated state. Below T < Tc, the transition into the Larkin-Ovchinnikov state begins as a first-order line and becomes second-order at lower T. Importantly, in this limit the interplay of Zeeman splitting and disorder, even without orbital effects, drives the transition between the normal and superconducting state first order.

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