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Complex state induced by impurities in multiband superconductors

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We study the role of impurities in a two-band superconductor, and elucidate the nature of the recently predicted transition from the $s_\pm$ state to the s_{++} state induced by interband impurity scattering. Using a Ginzburg-Landau theory, derived from microscopic equations, we demonstrate that close to T_c this transition is necessarily a direct one, but deeper in the superconducting state an intermediate complex state appears. This state has a distinct order parameter, which breaks the time-reversal symmetry, and is separated from the $s_\pm$ and s_{++} states by continuous phase transitions. Based on our results, we suggest a phase diagram for systems with weak repulsive interband pairing, and discuss its relevance to iron-based superconductors.

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It has been long recognized that nonmagnetic impurities strongly influence the properties of multiband superconductors [1–6], especially in the case of an order parameter with a sign change between different bands (the $s_\pm$ state) [2,7–9]. Recently, it has been pointed out that impurity-induced interband scattering can continuously change the order parameter of a two-band superconductor from the $s_\pm$ to s_{++} state [10–12]. This is particularly relevant for iron-based superconductors [13,14], most of which are believed to be in some form of $s_\pm$ state (see recent reviews [15,16]).

As we demonstrate in this Rapid Communication, the $s_\pm$-to-s_{++} transformation may follow a nontrivial scenario, and occur via an intermediate complex state at which a finite phase shift develops between the gap parameters in the two bands. We derive the simplest possible two-band Ginzburg-Landau (GL) free energy of the system from microscopic theory, and show that the presence of interband impurity scattering has important consequences for the different possible order parameters the theory can support. In the case of repulsive interband pairing we indeed observe the $s_\pm$-to-s_{++} transition [17] with increasing the degree of disorder. We demonstrate that the transition is necessarily a direct one only close to the critical line; deeper in the superconducting state the $s_\pm$ state gives way to an intrinsically complex order parameter (which can be thought as an $s_\pm + is_{++}$ state), and only then to a pure s_{++} state. This complex state breaks time-reversal symmetry and is separated from the other two superconducting states by continuous phase transitions. We discuss the reason and conditions for the appearance of this state. Based on our results, we propose the phase diagram shown in Fig. 1 for two-band superconductors with weak repulsive interband coupling.

We consider a system of two parabolic bands, with partial and total densities of states (DOS) N_1, N_2, and $N = N_1 + N_2$, respectively. The pairing interactions are described by a 2×2 coupling matrix $\hat{\lambda}$, with $\det[\hat{\lambda}] \equiv w = \lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21}$. In the superconducting state there are two gap parameters Δ_1 and Δ_2, which are assumed to be complex constants for each band $\Delta_m = |\Delta_m|e^{i\phi_m}$. The relative phase $\varphi = \phi_1 - \phi_2$ is a gauge-invariant quantity, and it is 0 or π in the s_{++} or $s_\pm$ states, respectively. The presence of impurities introduces scattering rates parametrized by γ_{mn}, where $m,n = (1,2)$ are the band indices. For the interband terms ($m \neq n$) we can write $\gamma_{mn} = N_n\Gamma$, with $\Gamma = n_{\rm imp}\pi u^2$, where $n_{\rm imp}$ and u are the impurities' concentration and potential, respectively. On general grounds, point defects, such as atomic substitutions or vacancies, can scatter carriers with a large momentum change and therefore are expected to give comparable intraband and interband scattering rates. In the case of the iron-based superconductors this was indeed confirmed by the first-principles calculations [18].

Close to the critical temperature the free energy can be expanded in powers of $|\Delta_1|$ and $|\Delta_2|$. (Although GL theory has been generalized to the case of multicomponent order parameters without impurities [19,20], the proper justification for this multiband extension is a matter of ongoing debate [21–25].) In the presence of impurities this can be done systematically, starting from the Usadel equations [6,21]. The resulting GL free energy up to quartic in Δ terms can be written as

$$\mathcal{F}_{\rm GL} = \mathcal{F}_{11} + \mathcal{F}_{22} + \mathcal{F}_{12} + \mathcal{F}_{\rm EM}. \tag{1}$$

We present the derivation of $\mathcal{F}_{\rm GL}$ from the microscopic theory, and give exact expressions for its coefficients in the Supplemental Material [26]. If the gap parameters are uniform in space and constant within each band, the intraband impurity scattering rate γ_{mm} drops out of the theory completely, as a direct consequence of the Anderson theorem [27]. In contrast, the interband terms play an important role. The first two terms look similar to the standard GL theory

$$\mathcal{F}_{mm}(\Delta_i) = a_{mm}|\Delta_m|^2 + \frac{b_{mm}}{2}|\Delta_m|^4, \tag{2}$$

but with a_{mm} and b_{mm} modified by the presence of impurities [26]. $\mathcal{F}_{\rm EM}$ combines the electromagnetic field contribution, and the derivative terms that couple Δ_1 and Δ_2 to the electromagnetic vector potential. For the rest of this Rapid Communication we assume no field and uniform order parameter, so $\mathcal{F}_{\rm EM} = 0$. The third term in $\mathcal{F}_{\rm GL}$ couples Δ_1 and Δ_2, and without impurities it is $2a_{12}|\Delta_1||\Delta_2|\cos\varphi$. In the presence of interband scattering processes, however, $\mathcal{F}_{12}$ becomes more complicated:

$$\begin{aligned}\mathcal{F}_{12} = {} & 2a_{12}|\Delta_1||\Delta_2|\cos\varphi + b_{12}|\Delta_1|^2|\Delta_2|^2 \\ & + 2(c_{11}|\Delta_1|^3|\Delta_2| + c_{22}|\Delta_1||\Delta_2|^3)\cos\varphi \\ & + c_{12}|\Delta_1|^2|\Delta_2|^2\cos 2\varphi.\end{aligned} \tag{3}$$

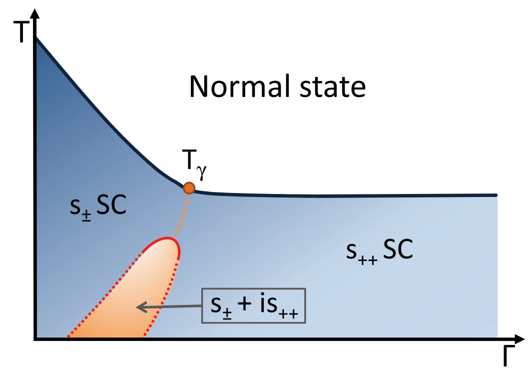

FIG. 1. (Color online) Phase diagram of systems with weak repulsive interband pairing. The x axis represents the interband impurity scattering rate. The orange dashed line denotes the direct $s_\pm$-to-s_{++} transition, and the orange region represents the complex $s_\pm + is_{++}$ state. The phase transition lines between the complex state and the other states are shown as red, and the dashed red indicates the conjectured extension of the complex state at low temperatures.

We can see that the presence of impurities introduces several new quartic interband terms in the GL theory [28]. In the limit $\Gamma \to 0$, a_{12} becomes proportional to λ_{12} and all other coefficients in Eq. (3) vanish. As a consequence, for a clean system the only possible solutions for φ are 0 and π, and which one minimizes $\mathcal{F}_{\rm GL}$ is determined by the sign of λ_{12}. When impurities are present, this is no longer necessarily true, and other solutions are possible, due to the $\cos 2\varphi$ term—it can destabilize the $s_\pm$ and s_{++} states, provided c_{12} is positive [29]. Thus, the dirty two-band superconductor can have quite a rich phase diagram.

The critical temperature at a given disorder strength is determined by the quadratic terms in Eq. (1). The equation for T_c derived in the Supplemental Material [26] takes the form $\det[\mathbb{M} - \mathbb{I}] = 0$, with $\mathbb{I}$ being the 2×2 identity matrix, and

$$\mathbb{M} \equiv \begin{bmatrix} \lambda_{11} I_2 + \lambda_1 n_1 (I_1 - I_2) & \lambda_{12} I_2 + \lambda_1 n_2 (I_1 - I_2) \\ \lambda_{21} I_2 + \lambda_2 n_1 (I_1 - I_2) & \lambda_{22} I_2 + \lambda_2 n_2 (I_1 - I_2) \end{bmatrix}.$$

We have defined $n_m = N_m/N$, $\lambda_m = \lambda_{mm} + \lambda_{mn}$, and

$$I_1 = 2\pi T \sum_{\omega_n > 0}^{\omega_0} \frac{1}{|\omega_n|}, \quad I_2 = 2\pi T \sum_{\omega_n > 0}^{\omega_0} \frac{1}{|\omega_n| + \gamma_{12} + \gamma_{21}},$$

where ω_0 is a high-energy cutoff (e.g., the Debye frequency). In the clean limit, $\Gamma = 0$, this equation gives a transition temperature $T_{c0} \approx 1.13\omega_0 \exp(-1/\lambda)$, where λ is the largest eigenvalue of the $\hat{\lambda}$ matrix. Note that the interband impurity scattering processes are always pair breaking (unless $\Delta_1 = \Delta_2$), and suppress T_c, in contrast with the intraband scattering, which has disappeared.

In general, the dependence $T_c(\gamma_{mn})$ has to be found numerically but the extreme dirty limit can be analyzed analytically. Depending on $\hat{\lambda}$, there are two qualitatively different regimes. If interband pairing is attractive, or negative but weak (i.e., when w is positive), no amount of disorder can completely suppress the superconductivity. In this case the critical temperature in the extreme dirty limit can be obtained [26]:

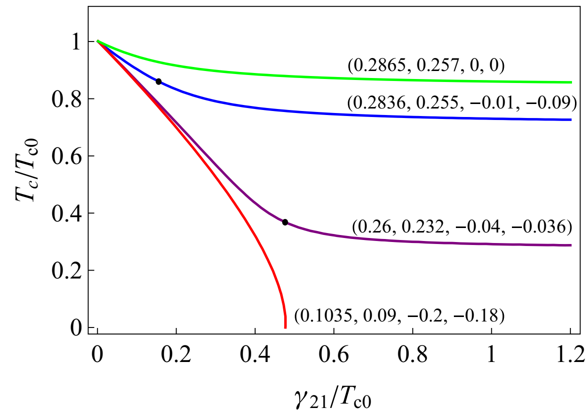

FIG. 2. (Color online) The T_c lines for systems with different $\hat{\lambda}$, as functions of γ_{21}. The coupling constants are shown inside the figure, in $(\lambda_{11}, \lambda_{22}, \lambda_{12}, \lambda_{21})$ format. In the cases of weak interband pairing (green, blue, and purple lines), T_c is initially suppressed, but eventually saturates. For repulsive *and* strong interband pairing (red line), superconductivity is completely suppressed by impurities. The dots indicate the positions of the T_γ points for the blue and the purple curves.

$$T_{c\infty} \approx 1.13\omega_0 \exp\left(-\frac{n_1(\lambda_{22} - \lambda_{12}) + n_2(\lambda_{11} - \lambda_{21})}{w}\right). \quad (4)$$

However, if the interband pairing is repulsive *and* strong, such that w is negative, there is a critical amount of disorder which brings T_c down to zero, in analogy with the Abrikosov-Gor'kov theory [30]. Numerical calculations of T_c for the different regimes are shown in Fig. 2. We see that for some systems, after the initial drop in T_c from its clean limit T_{c0}, the critical temperature saturates and stays finite in the limit $\Gamma \to \infty$. The reason is that the impurity scattering gradually averages the two gaps, and the closer they get to each other, the less effective is the pair breaking from the impurities; thus the superconductivity can survive even in the extremely dirty regime (in that limit, $\Delta_1 = \Delta_2$). The second regime is also easy to understand—if the sign change between the gaps is necessary for the existence of superconductivity (i.e., if the repulsive interband pairing interactions dominate), then the averaging produced by impurities completely suppresses the order parameter. Note that although our results are broadly consistent with the ones obtained in Ref. [11], our Eq. (4) somewhat disagrees with the dirty limit T_c derived there, since in our expression the effective coupling constant is $\langle \lambda^{-1} \rangle^{-1}$ rather than $\langle \lambda \rangle$.

For the rest of this Rapid Communication we concentrate on systems with positive w and repulsive interband pairing—as we will see, these are the systems with the most interesting phase diagram. We turn to the coefficient a_{12} of the Josephson-like term $|\Delta_1||\Delta_2| \cos\varphi$, and its evolution with Γ. The role of a_{12} is to couple the gaps, guaranteeing that they appear simultaneously, and close to T_c its sign fixes the relative phase of Δ_1 and Δ_2. In the presence of impurity scattering it is

$$a_{12} = -g - n_1 n_2 N (I_1 - I_2), \quad (5)$$

with $g = \lambda_{12}N_1/w = \lambda_{21}N_2/w$. In the clean limit $I_2 \to I_1$, $a_{12} \to -g$, and, as a result, φ is temperature independent, and can only be 0 or π. For finite Γ, however, a_{12} becomes a function of both disorder strength *and* temperature, and can even change its sign. This has important consequences for the order parameter. Negative g leads to the $s_\pm$ state in the clean limit. However, the second term in Eq. (5) is negative, and for strong disorder it can overcome the $-g$ term. If T_c is not completely suppressed (i.e., if the intraband pairing dominates), this sign change of a_{12} means a transition from the $s_\pm$ to s_{++} state at the $T_c(\Gamma)$ line [11]. This happens at temperature $T_\gamma \approx 1.13\omega_0 \exp[-(\lambda_{22} - \lambda_{12})/w]$ [26]. At this point the bands are effectively decoupled, and one of them stays normal. At a smaller disorder strength the system condenses in the $s_\pm$ state, while at a larger disorder strength it goes into the s_{++} state.

Below the critical line the quartic terms in the theory become important. Let us consider a system with T_c slightly higher than T_γ (meaning that immediately below T_c it is in the $s_\pm$ state). If $a_{22}(T)$ is positive, then Δ_2 is nonzero, solely because of its coupling to Δ_1 through a_{12}. In the vicinity of T_γ we can keep only the linear in Δ_2 terms in the equation $\partial\mathcal{F}_{\rm GL}/\partial|\Delta_2| = 0$ (while keeping the cubic in Δ_1 terms), and on the $s_\pm$ side we get

$$|\Delta_2| = -\frac{a_{12} + c_{11}|\Delta_1|^2}{a_{22} + c_{12}|\Delta_1|^2 + b_{12}|\Delta_1|^2}|\Delta_1|. \quad (6)$$

It is clear that the equation $a_{12} + c_{11}|\Delta_1|^2 = 0$ defines a line in the (Γ,T) space, originating from T_γ, and separating the $s_\pm$ from the s_{++} regions. On this line the bands are decoupled and Δ_2 is zero. If, for a fixed Γ, a given system has T_c slightly higher than T_γ, with decreasing the temperature it will cross the line, and Δ_2 will change its sign. We demonstrate this in Fig. 3. At this $s_\pm$-s_{++} transition point the second band becomes normal again [remember that we are assuming that $a_{22}(T)$ is still positive]. Note, however, that neither of the gap parameters have any singularity at this point; in a thermodynamic sense this is a crossover, rather than a real phase transition.

What happens if, with decreasing the temperature, the system gets close to the $a_{22}(T) = 0$ point *before* the $s_\pm$-s_{++} transition occurs? It can be easily shown that on the $a_{12} + c_{11}|\Delta_1|^2 = 0$ line the $|\Delta_2| = 0$ solution becomes unstable, and nonzero and purely imaginary Δ_2 appears when $a_{22} - c_{12}|\Delta_1|^2 + b_{12}|\Delta_1|^2$ turns negative. Since Δ_2 is now a superconducting gap in its own right, we have to keep all cubic terms in the equations. More generally, apart from the always present 0 and π solutions, φ can now take nontrivial values. From the condition $\partial\mathcal{F}_{\rm GL}/\partial\varphi = 0$ we obtain for φ the equation

$$\cos\varphi = -\frac{a_{12} + c_{11}|\Delta_1|^2 + c_{22}|\Delta_2|^2}{2c_{12}|\Delta_1||\Delta_2|}. \quad (7)$$

This solution represents a distinct, intrinsically complex superconducting state. The physical picture behind it is simple; instead of changing the relative sign of the gaps by taking one of them through zero, there is alternative, more elegant way—continuous evolution of φ from π to 0. This intermediate superconducting state can be understood as a linear combination (with complex coefficients) of the two "real" order parameters $s_\pm$ and s_{++}. More physically, this means that the fluctuations in the densities of the two condensates (which are induced by fixing the phases) are not in phase, as in s_{++}, and not in antiphase, as in the $s_\pm$, but have some nontrivial time shift. One of the modes is lagging the other, and as a consequence the time-reversal symmetry is spontaneously broken (as it should in such an intrinsically complex state). It is also easy to understand why such a state appears at finite temperatures below T_c; close to the critical line only the $s_\pm$ state exists. For the s_{++} state to condense within the $s_\pm$ state, $a_{22}(T)$ has to turn negative, and only then the complex admixture of $s_\pm$ and s_{++} becomes possible. This strongly suggests the necessary condition for the existence of such a complex state—the presence of *two* attractive superconducting channels at the same temperature (which means that w has to be positive).

By minimizing the GL free energy, we demonstrate that this solution is indeed realized, as illustrated in Fig. 4. The order parameter starts as $s_\pm$ ($\varphi = \pi$) at the critical temperature. However, at some finite temperature below T_c, φ deviates from the π solution, and the superconducting state is no longer pure $s_\pm$, but an intrinsically complex state. According to our model, the time-reversal symmetry breaking state is separated from

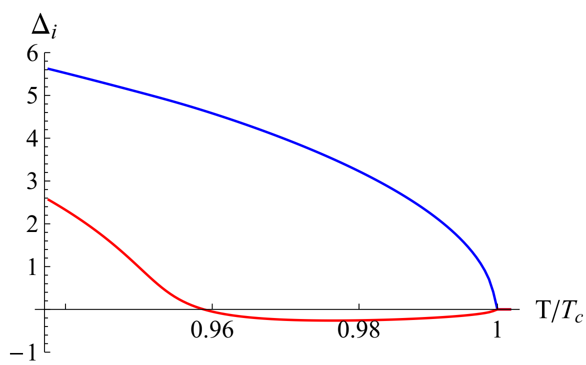

FIG. 3. (Color online) The behavior of Δ_1 (blue) and Δ_2 (red) with temperature, demonstrating the $s_\pm$-s_{++} transition; Δ_2 is negative close to T_c, but goes through zero and changes its sign. The coupling constants are $\lambda_{11} = 0.3$, $\lambda_{22} = 0.297$, $\lambda_{12} = -0.011$, $\lambda_{21} = -0.011$, and $\Gamma = 1.63$ (at the T_γ, $\Gamma \approx 1.67$).

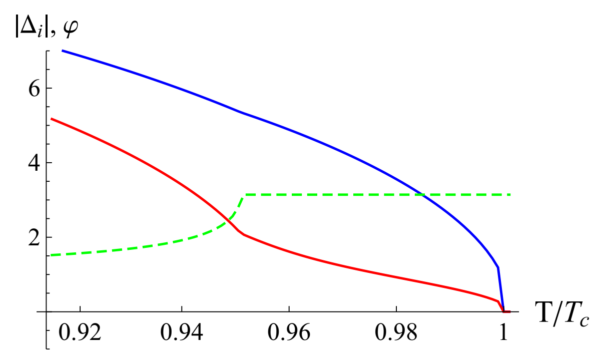

FIG. 4. (Color online) The behavior of $|\Delta_1|$ (blue), $|\Delta_2|$ (red), and φ (green, dashed), for the same $\hat{\lambda}$ as in Fig. 3, but for $\Gamma = 1.57$. Close to T_c the relative phase is π (the system is in the $s_\pm$ state), but around $0.95T_c$ it starts decreasing continuously. Both gaps stay finite.

both "real" order parameters (which preserve the symmetry) by lines of continuous phase transitions.

Similar complex states in one-band systems ($s + id$ states) [31–34] and in three-band systems ($s + is$ states) [35–42] recently have attracted a lot of attention. There are some similarities in the underlying physics between these states and the $s_{\pm} + is_{++}$ state discussed here. As in the $s + id$ case, in our model the complex state appears as a way of avoiding the appearance of the nonsuperconducting parts of the Fermi surface (either the nodes of the d-wave state, or an entire band in our model). The similarity with the three-band model is that in both cases the complex order parameter admixes two superconducting states in the trivial A_{1g} representation. Our impurity-induced complex state is also somewhat similar to the surface complex state predicted in the case of strong interband reflection at the boundary [43].

We summarize our findings in the phase diagram presented in Fig. 1. Strictly speaking, our results are valid only in the region of applicability of the extended GL theory. To observe the complex state in this region, we had to keep λ_{11} and λ_{22} quite close. In case they are not close, the complex state is realized at temperatures significantly lower than T_c and it has to be treated within the full microscopic theory. Nevertheless, using an analogy with the physics and the phase diagrams discussed in Refs. [36,41], we make two conjectures: (i) The $s_{\pm} + is_{++}$ state is present if the system has an $s_{\pm}$ to s_{++} crossover, even if it is not observable in the GL region; (ii) this state extends down to $T = 0$, without any significant modifications. Confirming or rejecting these conjectures is an important direction for future work.

What do our results imply for the iron-based superconductors? Recently a roughly universal complete suppression of T_c was reported for several FeAs-122 compounds [44]. This suggests that these materials are in the $s_{\pm}$ state with strong interband pairing, and thus no complex state is expected there. On the other hand, substantial variations in the effects of different impurities in similar 122 systems were observed in Ref. [45]. Also, a very recent study of T_c suppression in iron chalcogenides [46] showed a nonuniversal behavior, with some of the curves showing T_c, which initially decreases, but eventually saturates, as expected for the $s_{\pm}$-to-s_{++} transition. Although more studies are needed, it is already clear that these materials are surprisingly diverse in their normal and superconducting state properties, so it is entirely possible that the $s_{\pm} + is_{++}$ state can be induced by impurities (for example, by systematically irradiating a sample) in some of them.

In conclusion, we have studied the role of impurities in a two-band superconductor. We derived a Ginzburg-Landau theory to describe the system, and we showed that the interband impurity scattering has a significant impact on the theory. Due to the impurity-induced $\cos 2\varphi$ term in the theory, a complex order parameter may appear between the $s_{\pm}$ and s_{++} states.

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[26] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.89.100505 for derivation of the GL coefficients from the microscopic Usadel equations, by expanding them in powers of the gap parameters. We also derive

approximate formula for the critical temperature in the extreme dirty limit.

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