Critical superconductors

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Bogomolnyi critical point, originally introduced in string theories, is fundamental to superconductivity. At the critical temperature $T_c$ it marks the sharp border between ideally diamagnetic bulk type-I superconductors and type-II ones that can host vortices, while itself it harbors infinitely degenerate distributions of magnetic flux in the superconducting state. Here we show that below $T_c$ the physics of the Bogomolnyi critical point projects onto a wider range of microscopic parameters, even extremely wide for multiband superconductors. In this critical range, the degeneracy of the superconducting state at $T_c$ is lifted into a sequence of novel topological equilibria and the customary understanding of superconducting phenomena does not suffice. As a radical departure from traditional views, we introduce the paradigm of critical superconductors, discuss their distinct magnetic properties, advocate their subdivision in terms of possible intermediate states, and demonstrate direct relevance of this paradigm to superconducting materials of prime interest today.

Following textbooks [1–3], superconductors are classified by the relation between the magnetic penetration depth $\lambda$ and the coherence length $\xi$. According to the Ginzburg-Landau (GL) theory [4] types I and II of superconductivity interchange when the GL parameter $\kappa = \lambda/\xi$ crosses the critical value $\kappa_0 = 1/\sqrt{2}$. However, detailed experimental studies as well as the microscopic theory revealed that the interchange actually occurs in a rather narrow but finite interval around $\kappa_0$ [5–8]. A long-range attraction of vortices was observed in this interval, leading to different patterns of intercalated Meissner and Abrikosov-lattice domains [5], often referred to as the type-II/1 state. Recently, similar vortex configurations have been reported for MgB$_2$ [9], where several carrier bands contribute to the superconducting condensate. This led to a controversial idea of a special superconducting type for multiband compounds, named “type-1.5”, where multiband materials are considered to comprise several coupled condensates, one for each band, and a new type arises for at least one band-condensate being type I and others type II [9,10]. It was however immediately argued that unusual features of the “type 1.5” are not specific to multiband materials and are actually similar to the earlier discussed type-II/1 [11]. Still, no explanation has been given how to reconcile the large GL parameter of MgB$_2$ ($\kappa > 5$, see e.g. Ref. [12]) with the narrow critical range around $\kappa_0$ where the type-II/1 behavior is expected. To date, the applicability of the standard classification of superconductors to multiband materials remained an unresolved and highly debated issue.

We here demonstrate that the appearance of the critical interval near $\kappa_0$ actually defines a new class of superconducting materials, which generalizes types II/1 and “1.5” and by far exceeds them in complexity. The new properties of this class are directly related to the fact that all states of a superconductor, with arbitrary vortex configurations, have the same energy at the Bogomolnyi point $(\kappa, T) = (\kappa_0, T_c)$ when the applied magnetic field is equal to the thermodynamic critical field $H_c$. [13–15]. This topological degeneracy, lifted at $T < T_c$, is then the source of a wide diversity of unconventional phenomena in the critical interval of $\kappa$ around $\kappa_0$, where the long-range attraction of vortices mentioned above is only a particular example. Since this distinct behavior stems from the critical character of the Bogomolnyi point we refer to it as critical type of superconductivity. Interestingly, even some seemingly understood superconductors (such as Pb) may turn out to belong to the critical type, so that parts of the long established knowledge in the field must be revisited. More importantly, we find that coexistence of multiple bands in a single material gives rise to a large, even giant enlargement of the critical domain near $\kappa_0$. This appreciably widens the class of critical superconductors, arguably including most of the recently discussed materials (such as metal-borides and iron-based ones [16]).

Our work is based on the analysis of the critical GL parameter $\kappa^*$ at which superconductivity should change its type. There are several standard ways to calculate $\kappa^*$: (i) using the condition of zero surface energy for the superconductor-normal (S-N) interface (the corresponding critical GL parameter is further denoted as $\kappa^*_s$); (ii) from conditions $H_c = H_{c1} (\kappa_1^*)$ and $H_c = H_{c2} (\kappa_2^*)$, where $H_{c1}$ and $H_{c2}$ are the lower and upper critical fields, respectively; and (iii) using the condition of the sign change for the long-range interaction between vortices [7,15] ($\kappa^*_a$). The GL theory yields $\kappa^* = \kappa_0$ for all above definitions, resulting in just two possibilities: type I for $\kappa < \kappa_0$ and type II when $\kappa > \kappa_0$. However, beyond the standard
GL theory one obtains different and temperature dependent values for $\kappa^* \mathbb{Z} [14]$. Consequences of this fact were never fully understood and, as a result, are completely ignored in standard textbooks.

We obtain $\kappa^*$ from the Extended GL (EGL) approach, derived by the perturbation expansion of the microscopic BCS theory in powers of $\tau = 1 - T/T_\lambda$ to one order beyond the standard GL theory [17]. The EGL theory therefore improves the standard GL model but still allows for simple analytical solutions in many important cases. In order to study the critical domain around $\kappa_0$, we combine EGL with the expansion over $\delta \kappa = \kappa - \kappa_0$, in the similar way as was done in Ref. [15]. Then, the density of the Gibbs free energy of the superconducting state (measured from that of the normal metal) is obtained at thermodynamic critical field $H = H_c$ as

\[ g = g^{(0)} + (\delta g^{(0)}/\delta \kappa) \delta \kappa + g^{(1)} \tau, \quad (1) \]

where the coefficients, calculated at $\kappa = \kappa_0$, are

\[ g^{(0)} = \frac{1}{2} \left( \frac{B}{\sqrt{2}\kappa} - 1 \right)^2 |\psi|^2 + \frac{1}{2} |\psi|^4 + \frac{1}{2\kappa^2} |\mathbf{D}\psi|^2, \quad (2a) \]

\[ g^{(1)} = \left( \frac{B}{\kappa \sqrt{2}} - 1 \right) \left[ \frac{1}{2} \bar{\psi} + \bar{G}(\bar{\alpha} - \bar{\beta}) \right]^2 + \frac{1}{\kappa^2} |\mathbf{D}\psi|^2 \]

\[ - \frac{1}{2} |\psi|^2 + |\psi|^4 + \bar{\psi} |\psi|^6 + \bar{G} |\psi|^2 (\bar{\alpha} - \bar{\beta}) |\psi|^2 \]

\[ + \frac{\bar{\mathbf{Q}}}{4\kappa^2} [\mathbf{D}^2 |\psi|^2 + \frac{1}{3} (\text{rot} \mathbf{B})^2 + \mathbf{B}^2 |\psi|^2 ] \]

\[ + \frac{\bar{\mathbf{L}}}{4\kappa^2} [8 |\psi|^2 |\mathbf{D}\psi|^2 + (\psi^*)^2 (\mathbf{D}^* \psi^2) + |\psi|^2 (\mathbf{D}^* \psi^2)] \quad (2b) \]

Here $\mathbf{D} = \nabla + i\mathbf{A}$, the magnetic field is assumed to be in the $z$-direction, $\mathbf{B} = (0,0, B)$, and we used the dimensionless quantities $r/\lambda \sqrt{2}$, $\kappa \mathbf{A}/\lambda H_c$, $\kappa \sqrt{2} \mathbf{B}/H_c$, $\sqrt{b}/\sqrt{a\tau}$ and $4\pi g/H_c^2$, with $H_c = \tau \sqrt{4\pi a^2/b}$ (the lowest order contribution to the thermodynamic critical field). The order parameter $\psi$ and the magnetic field $B$ (both in the lowest order in $\tau$) obey the standard GL equations

\[ \psi(1 - |\psi|^2) + \frac{1}{2\kappa^2} \mathbf{D}^2 \psi = 0, \quad \text{rot} \mathbf{B} = \mathbf{j}. \quad (3) \]

where $\mathbf{j} = -i(\psi \mathbf{D}^* \psi^* - \psi^* \mathbf{D} \psi)$. Eqs. [1] - [3] are valid for a system with an arbitrary number of carrier bands (see Ref. [17]), where $\psi$ is a single Landau order parameter [19]. Differences between the band condensates are accounted for in the coefficients of the equations. In particular, for the two-band case we obtain

\[ a = \frac{a_1}{S} + S a_2, \quad b = \frac{b_1}{S^2} + S^2 b_2, \quad \kappa = \frac{K_1}{S} + S K_2, \]

\[ \alpha = \frac{a_1}{S} - S a_2, \quad \beta = \frac{b_1}{S^2} - S^2 b_2, \quad \Gamma = \frac{K_1}{S} - S K_2, \]

\[ c = \frac{c_1}{S^3} + S^3 c_2, \quad \mathcal{Q} = \frac{Q_1}{S} + S Q_2, \quad \mathcal{L} = \frac{L_1}{S^2} + S^2 L_2, \]

\[ \bar{\gamma} = \frac{ca}{3b^2}, \quad \bar{\mathcal{Q}} = \frac{Q_0 a}{K_2^2}, \quad \bar{\mathcal{L}} = \frac{L a}{b K}, \quad \bar{G} = \frac{Ga}{4g_{12}}, \quad \bar{\alpha} = \frac{\alpha}{a} - \frac{\kappa}{K}, \quad \bar{\beta} = \frac{\beta}{b} - \frac{\kappa}{K}. \quad (4) \]

where indices label the bands, and $G = g_{11} g_{22} - g_{12}^2$, with $g_{ij}$ denoting the interaction matrix. The relative weights of the band contributions to Eqs. [4] are controlled by the quantity

\[ S = \frac{1}{2\lambda_{12}} \left[ \lambda_{22} - \frac{\lambda_{11}}{\chi} + \sqrt{\left( \lambda_{22} - \frac{\lambda_{11}}{\chi} \right)^2 + \frac{4\lambda_2^2}{\chi}} \right], \quad (5) \]

where $\chi = N_2(0)/N_1(0)$ is the ratio of the band dependent densities of states (DOS), and $\lambda_{ij} = g_{ij} N(0)$ is the dimensionless coupling constant, with $N(0) = \sum_j N_0(j)$ the total DOS. Parameters $a_i, b_i, c_i, K_i, Q_i, L_i$ are calculated separately for each band with a chosen model for the carrier states (for procedure, see [17]). Notice that in the limit $\chi \to \infty$ or $\chi \to 0$ one recovers the single band result.

Derivative $\delta g^{(0)}/\delta \kappa$ in Eq. [1] accounts for both explicit $\kappa$-dependence of the functional and for the implicit one through the solution $\psi$. Since $\psi$ satisfies Eq. [3], i.e. $\delta g^{(0)}/\delta \psi = 0$, the implicit dependence does not contribute to the derivative and one obtains

\[ \frac{\delta g^{(0)}}{\delta \kappa} = \frac{\partial g^{(0)}}{\partial \kappa} = - \frac{B}{\kappa^2 \sqrt{2}} \left( \frac{B}{\kappa \sqrt{2}} - 1 \right) - \frac{1}{\kappa^3} |\mathbf{D}\psi|^2. \quad (6) \]

Special character of the Bogomolnyi point (BP) follows formally from the fact that $\psi$ satisfies the Sarma equation $\Pi - \psi = 0$ where $\Pi^\pm = D_x \pm iD_y$, which leads to relation $|\psi|^2 = 1 - B \mathbb{Z} [2]$. In string theories these relations are often referred to as the self-duality Bogomolnyi equations [20]. Substituting them into Eq. [1] and integrating the result, we obtain the total energy difference as ($\mathcal{G} = \int g \, d^3r$)

\[ \mathcal{G} = - \sqrt{2} \mathcal{L} \delta \kappa + \mathcal{J} \left[ 1 - \bar{\psi} + \bar{G}(\bar{\alpha} - \bar{\beta}) \right]^2 + J \left[ \bar{\mathcal{L}} - \bar{\psi} - \frac{5}{3} \bar{\mathcal{Q}} - \bar{G}\bar{\beta}^2 \right], \quad (7) \]

where $I = \int d^3r |\psi|^2(1 - |\psi|^2)$ and $J = \int d^3r |\psi|^4(1 - |\psi|^2)$, and $g^{(0)} = 0$ manifests the BP degeneracy. In the single-band case this expression is similar to the perturbation functional used in Ref. [15]. The contribution with $\mathcal{G}$ in Eq. [7] appears due to the difference in the spatial
profiles (and, respectively, length-scales) of different band condensates: $\tilde{G} = 0$ for single-band systems.

Equation (7) enables calculating $\kappa^*$ according to the definitions given above. This yields the general expression

$$\kappa^* = \kappa_0 \left\{ 1 + \tau \left[ 1 - \tilde{c} + 2 \tilde{Q} + \tilde{G} \tilde{\beta} (2 \tilde{\delta} - \tilde{\beta}) \right] + \frac{J}{I} \left( 2 \tilde{\mathcal{L}} - \tilde{c} - \frac{5}{3} \tilde{Q} - \tilde{G} \tilde{\beta}^2 \right) \right\}, \quad (8)$$

For $\kappa_{li}^*$, at which $\tilde{G}$ calculated for the S-N interface is zero, the numerical solution yields $J/I = 0.559$. For $\kappa_{li}^*$, obtained from the condition $H_c = H_{c1}$ (which marks the onset of the stability of an Abrikosov vortex), one obtains $J/I = 0.735$. For $\kappa_{li}^*$, at which the long-range asymptote of the vortex-vortex interaction changes sign, one finds the exact result $J/I = 2$. For the remaining critical parameter $\kappa_{li}^*$ calculated from $H_c = H_{c2}$ one obtains $J/I = 0$, as $|\psi| \to 0$.

For a single-band system in the clean limit and with a spherical Fermi surface one then obtains the following universal expressions, independent of the particular material parameters:

$$\kappa_1^* = \kappa_0 (1 + 0.951 \tau), \kappa_1^* = \kappa_0 (1 + 0.093 \tau),$$

$$\kappa_2^* = \kappa_0 (1 - 0.027 \tau), \kappa_2^* = \kappa_0 (1 - 0.407 \tau). \quad (9)$$

We note that similar linear dependencies were obtained earlier in Ref. [11] (within the approximate Neumann-Twordt model) and recently in Ref. [15] (although with different numerical coefficients). At $T < T_c$ ($\tau > 0$) we obtain $\kappa_2^* < \kappa_1^* < \kappa_0^*$. The upper and lower limits of this inequality define the critical interval $[\kappa_{li}^*, \kappa_{li}^*]$, where superconductivity types I and II interchange. At $\kappa < \kappa_{li}^*$ only the Meissner state is possible (type I), at $\kappa > \kappa_{li}^*$ the system can develop a mixed state with repulsive Abrikosov vortices (type II). Experimentally, $\kappa_{li}^*$ and $\kappa_{li}^*$ can be obtained from the field dependence of the magnetization $M(H)$ [6, 21], since inside the critical domain the magnetization has a discontinuity at $H_{c2}^*$ ($< H_c^*$) [22], accompanied by a nonzero tail at $H_{c2}^*$ due to the existence of a mixed state. Fig. 1 shows $\kappa_{li}^*$ and $\kappa_{li}^*$ from Eq. (9) alongside experimental data obtained for nearly clean single-band materials [6, 21]. The experimental boundaries of the critical interval are indeed material-independent and follow straight lines down to temperature $T \approx 0.4T_c$, in remarkable agreement with our theoretical prediction.

Although differences between $\kappa^*$ obtained using different definitions have been noted earlier [11], their relation to the Bogomolny point has been noted only recently [15]. However, the nontrivial internal structure of the critical domain, related to the topological degeneracy of the Bogomolny point, has not been considered. When this degeneracy is lifted at $T < T_c$, different $\kappa^*$ delimit subdomains of very different magnetic behavior [24]. Here we describe the critical-domain structure by addressing crucial changes in the vortex behavior at $\kappa_{li}^*$ and $\kappa_{li}^*$.

When $\kappa_{li}^* < \kappa < \kappa_{li}^*$, the system hosts single-quantum vortices for $H > H_{c1}$, in an Abrikosov lattice. However, unlike vortices in conventional type-II superconductors, these vortices are attractive at long distances [17, 22], so the lattice constant has a finite maximal value. One consequence of this is a first-order transition with a magnetization jump at $H_{c2}^* = H_{c1}$ (see Fig. 1). This behavior is
known in literature and often referred to as type II/1 [6–8]}. However, so far it was associated with the entire critical domain, i.e., for $\kappa_2 < \kappa < \kappa_{s1}$, and this is not correct.

Namely, at $\kappa < \kappa_{s1}$, an isolated single-quantum vortex becomes energetically unfavorable at $H^*_c$, since $H^*_c < H_c$ and $H_{c3} > H_c$. However, we still have the mixed state, since $H_c < H_{c2}$. As the closest alternative, we have put into consideration the $N$-quanta vortices, even though they are supposed to be unstable in the conventional type-II picture. Quite surprisingly, Fig. 2(a) shows that when $\kappa$ decreases in the interval $[\kappa_{s1}, \kappa_1^*]$, the multi-quanta vortices proliferate, i.e., vortices of progressively larger $N$ become more favorable than $N$ isolated single-quantum vortices. As a consequence, the Meissner state becomes unstable at the field $H^*_c = H_{c1,N} < H_c < H_{c3}$, where $H_{c1,N}$ labels $H_{c1}$ for the $N$-quanta vortex.

Using the condition $H_c = H_{c1,N}$, we define the critical parameter $\kappa_{s1,N}^*$ at which $N$-quanta vortices first appear in the system. We also consider $\kappa_{s1,N}^*$ and $\kappa_{s1,N}^*$ at which the short-range and, respectively, the long-range asymptotes of the vortex-vortex interaction change their sign. In Fig. 2(b), we show the calculated $\kappa_{s1,N}^*$ and $\kappa_{s1,N}^*$, which also satisfy Eq. (3) but for $N$-dependent integrals $J$ and $I$. $\kappa_{s1,N}^*$ decreases monotonically for increasing $N$, leading to a conclusion that only vortices with progressively larger $N$ remain stable against fusion when decreasing $\kappa$. On the other hand, $\kappa_{s1,N}^* = \kappa_{s1}^*$ is independent of $N$.

When $\kappa$ descends below $\kappa_{s1}^*$, the S-N surface energy becomes positive and fragmentation of S-N interfaces into vortices is no longer energetically advantageous. Isolated vortices are, therefore, unstable for any $N$ and $H^*_c \neq H_{c1,N}$. Nevertheless, as $H_c < H_{c2}$, the mixed state must exist and should be densely inhomogeneous. An educated guess into its shape (e.g., a dense distribution of strongly overlapping multiple-quanta vortices, stripe structures, or something similar to the Fulde-Ferrel-Larkin-Ovchinnikov pattern [20]) is necessary to determine the corresponding $H^*_c$. For the present analysis, it is most important that the single-vortex picture becomes inadequate here. We label this subdomain critical type I (type Ic). In contrast, when $\kappa > \kappa_{s1}^*$, isolated vortices are more energetically favorable than the Meissner state at $H^*_c < H < H_{c2}$. However, unlike the conventional type II, vortices are attractive at long distances and each can carry multiple flux-quanta. We refer to this behavior as critical type II (type IIc). The summary of this analysis is illustrated in Fig. 3 with delimiting lines obtained from Eq. (6). It must be noted at this point that the complete analysis should include the field dependence of the most favorable configurations, since our present consideration of possible intermediate states is restricted to fields in vicinity of $H_c$.

As best seen from Fig. 3 in single-band superconductors the critical domain is limited to GL parameters relatively close to $\kappa_0$, thus critical superconduc-
tivity is of relevance to a restricted number of materials. This changes drastically for multiband superconductors, where the interband coupling enhances non-local effects [27], known to be responsible for the long-range vortex attraction [11]. An amplification of the critical type of behavior can thus be anticipated. Our calculations for the two-band system fully confirm this expectation. Fig. 4 demonstrates a very significant widening of the critical region for two-band superconductors as a function of the ratio of the band Fermi velocities $v_2/v_1$. The conclusion that critical superconductivity is enhanced by disparate length-scales of two or more involved band-condensates is rather general and holds for a wide selection of the material parameters. In particular the results in Fig. 4 are calculated for the parameters of: (a) MgB$_2$ [28], $\lambda_{11} = 1.88$, $\lambda_{22} = 0.5$, $\lambda_{12} = 0.21$, and $N_2(0)/N_1(0) = 1.33$; (b) OsB$_2$ [29], $\lambda_{11} = 0.387$, $\lambda_{22} = 0.291$, $\lambda_{12} = 0.0084$, and $N_2(0)/N_1(0) = 1.22$; (c) FeSe$_{0.94}$ [30], $\lambda_{11} = 0.48$, $\lambda_{22} = 0.39$, $\lambda_{12} = 0.005$, and $N_2(0)/N_1(0) = 1$; and (d) LiFeAs [31] $\lambda_{11} = 0.63$, $\lambda_{22} = 0.64$, $\lambda_{12} = 0.008$, and $N_2(0)/N_1(0) = 1$. As a main conclusion of Fig. 4, the critical domain in two band materials is always larger than in single-band ones. One should however notice its extraordinary sensitivity to $v_2/v_1$: for $v_2 > 2v_1$ in Figs. 4(a)-(c) and for $2v_2 < v_1$ in Fig. 4(d), tangents of $\kappa^*_1$ and $\kappa^*_2$ change by an order of magnitude or even more. As a consequence, critical type of superconductivity becomes very likely at low temperatures and should always be reckoned with in multiband materials.

In summary, we have shown analytic distinction of the class of critical superconductors, which draw their unique properties from the physics of the Bogomolnyi critical point. Critical type of superconductivity is intrinsic to some long known superconductors, such as e.g. Nb, or Ta with impurities, but could also be relevant to Pb which was always assumed type-I [32]. Our analysis suggests that critical superconductivity should always be taken into account in the studies of novel superconducting compounds, particularly multiband ones with notably different Fermi surfaces (such as e.g. FeSe$_x$Te$_{1-x}$ [16]). In critical superconductors, the rich structure of the critical domain will undoubtedly reflect itself in a wealth of unconventional superconducting states, with unique sensitivity to the applied field, current, temperature but also to different geometric parameters of a specimen. Detailed description of those states warrants further theoretical as well as experimental investigations. Finally, it is important to note that in string models mathematically similar to the superconducting theory (such as the Abelian Higgs model [13]), by analogy to our findings a similar critical domain of unusual equilibria could also be observed.

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smaller than the BCS coherence length.

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