Discontinuous Superconducting Transitions in the Paramagnetic limit: a Non-Perturbative approach

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Abstract. The unusual superconducting (SC) phase transitions occurring under competing orbital and spin pair breaking, which characterize clean strongly type-II superconductors at low temperatures, are investigated within a non-perturbative approach, which avoids the difficulties encountered in various perturbative approaches and enables comparison with recent experimental data. It is shown that in a 3D system with strong spin-splitting, a spatial (FFLO) modulation of the order parameter along the magnetic-field direction preserves the continuous nature of the SC transition. However, at a magnetic field slightly below $H_{c2}$ the FFLO state becomes unstable, transforming discontinuously into a uniform SC state via a first-order phase transition. Our calculation shows that the entropy jump at the first-order phase transition is significantly larger than its total variation in the continuous region between the two transitions, in agreement with recent thermal-conductivity measurements performed on the heavy-fermion compound URu$_2$Si$_2$.

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The competition between orbital and spin pair-breaking in strongly type-II superconductors in the Pauli paramagnetic limit is known to control the occurrence of discontinuous SC transitions at sufficiently low temperatures. Qualitative descriptions of these first-order phase transitions [1] usually exploit perturbative Ginzburg-Landau (GL) approaches (see, e.g. [2, 3]). However, a quantitative (and sometimes even qualitative) description of discontinuous transitions, where the SC order parameter changes abruptly by a finite value, usually requires a non-perturbative approach. This is particularly important in the presence of a spatially modulated order parameter along the magnetic-field direction [4, 5], due to the oscillatory dependence of the quartic and higher-order terms in the expansion on the modulation wave number $q$, which makes the utilization of a uniquely defined expression for the SC free energy impossible within perturbation theory.

In this paper, we present our results for some thermodynamic properties of clean 3D strongly type-II superconductors in the Pauli paramagnetic limit, obtained by use of calculations based on a non-perturbative method, which removes the difficulties associated with the various perturbative approaches and leads to a clear physical picture of the corresponding SC phase transitions. Application to the heavy-fermion superconductor URu$_2$Si$_2$, which is characterized by a 3D Fermi surface (FS) [6] with parameters favoring strong spin pair breaking, is in good agreement with the step-like structure observed experimentally in thermal-conductivity data at the SC transition of URu$_2$Si$_2$ [7].

Technically, this is achieved by considering the general term in the order-parameter expansion
of the Grand thermodynamic potential (see e.g. [8, 9, 10]) and then performing analytical summation of the corresponding infinite series to all orders of the perturbation expansion. This approach becomes tractable due to factorization of the general term, provided that a special representation for the electron Green’s functions is exploited [11]. The details of the derivation will be given elsewhere. We restrict our calculation here to conventional (s-wave) electron pairing interaction, since as shown in Ref. [11], the main results in the clean limit are found to be independent of the type of electron pairing.

The resulting thermodynamic potential (TP) for an isotropic 3D electron gas under an external stationary magnetic field \( \mathbf{H} = H \hat{z} \) is written as:

\[
\frac{\Omega}{V} = \frac{a_x}{\sqrt{2\pi}} \left[ \frac{\Delta_{\text{max}}^2}{g \nu} - \frac{k_B T}{\alpha_H} \Re \sum_{\nu > 0} \frac{2}{\sqrt{\pi}} \int_0^\infty du \int \frac{d^2 k d k_z}{(2\pi)^3} \ln \left( 1 + e^{-u^2 X_{\nu}} \right) \right] \tag{1}
\]

\[
X_{\nu} = \frac{\Delta_{\text{max}}^2}{g \nu} \Phi_{\nu}(k, k_z, g, q) \Phi^*_\nu(k, k_z, -g, -q) \tag{2}
\]

\[
\Phi_{\nu}(k, k_z, g, q) \approx \int_0^\infty d \tau e^{-\tau^2 |\varphi_{\nu, k, k_z} - |k|^2 r^2|} \tag{3}
\]

where \( \xi_{k, k_z} = \frac{1}{\nu} |k|^2 + \frac{1}{2} (k_z - q)^2 - q - n_F \), \( k = \hat{x}k_x + \hat{y}k_y \), \( n_F = \mu/\hbar \omega_c \), \( \mu \) is the chemical potential, \( \omega_c = \nu_0/\hbar \omega_c \) is the reduced Matsubara frequency, \( g = m^*/m_0 \) (with \( m^* \) and \( m_0 \) the effective and free electron mass, respectively), and \( \omega_c = eH/m^* c \). The dimensionless momentum \( k \) is measured in units of the inverse magnetic length \( a_H^{-1} = \sqrt{eH/c\hbar} \). The above expression for \( \Omega \) is similar to the well-known expression obtained for a superconductor at zero magnetic field except for the following modifications: The effective electron Green’s function in the quasi-classical approximation, \( \Phi_{\nu}(k, k_z, g, q) \), includes a Gaussian factor, \( e^{-\frac{1}{2} |k|^2 r^2} \), which takes into account the combined diamagnetic effect of the SC order parameter \( \Delta(r, z) \), \( r = x \hat{x} + y \hat{y} \) (in the plane perpendicular to \( \mathbf{H} \)), and the gauge factors of the single-electron Green’s functions. The other modification involves, of course, the kinetic energy cost of the Fulde-Ferrell (FF)-type spatial modulation of the order parameter \( \Delta(r, z) = \Delta_{\text{max}} \varphi_0(x, y) e^{i h z} \). Here, \( \varphi_0(x, y) \) describes (in symmetric gauge) a lowest Landau level (LLL) hexagonal vortex-lattice wavefunction with inter-vortex distance \( a_x \) and \( \Delta_{\text{max}} = \Delta_{\text{max}}^2/\hbar \omega_c \), with \( \Delta_{\text{max}} \) being the maximal value of the SC order parameter in the vortex-lattice unit cell. The latter is related to the spatial average \( \Delta_0^2 = V^{-1} \int d^2 r dz |\Delta(r, z)|^2 \) through \( \Delta_{\text{max}}^2 = \left( \frac{2\pi}{a_x^2} \right)^{1/2} \Delta_0^2 \). Finally, the LLL constraint, appearing in Eq. (1) as an additional integration over \( u \), is another feature of the high-magnetic-field situation investigated.

For saving heavy numerical calculations the effective Green’s function is approximated as \( \Phi_{\nu}(k, k_z, g, q) \approx \frac{1}{\omega_{\nu} + \frac{1}{\alpha} k^2 - i \xi_{k, k_z, (g, q)} \omega} \), which amounts to replacing the Gaussian \( e^{-\frac{1}{2} |k|^2 r^2} \) in Eq. (3) with the simple exponential \( e^{-\frac{1}{2} |k|^2 r^2} \). This simplification does not change the results qualitatively in the high-magnetic-field region relevant to the discontinuous phase transitions of interest here.

The thermodynamic functions, in the mean-field approximation can be obtained from \( \Omega \) by minimization with respect to both \( \Delta_{\text{max}}^2 \) and \( g \). Below we present results for the SC magnetization, \( M = -\frac{\partial \Omega}{\partial H} \), and entropy, \( S = -\frac{\partial \Omega}{\partial T} \), which are calculated from the following expressions:

\[
\mathcal{M} = \frac{1}{2\pi} \frac{M}{M_0} = -\sum_{\nu > 0} \frac{1}{\pi \sqrt{\pi}} \int k^2 dk \int \sin \alpha d \alpha \int_0^\infty du \int \frac{k^2 dk}{(2\pi)^3} \ln \left( 1 + e^{-u^2 X_{\nu}} \right) \tag{4}
\]

\[
\times \Re \left\{ \left[ 1 - \left( 1 + e^{-u^2 X_{\nu} (\kappa, \alpha)} \right)^{-1} \left( \frac{1}{\varepsilon_{\nu} (\kappa, \alpha)} + \frac{1}{\varepsilon^*_\nu (\kappa, \alpha)} \right) \right] \left( \frac{\eta_0}{2\pi^{1/2}} k b^{-1/2} \sin \alpha + \varphi \right) \right\}
\]
\[ S = \frac{1}{\pi} \int \frac{S}{S_0} = - \frac{1}{2 \pi \sqrt{E}} \int \kappa^2 d\kappa \int \sin \alpha d\alpha \frac{1}{\sqrt{\pi}} \int_0^{\infty} du Re \left[ \log \left( 1 + e^{-u^2} X_{\nu=0} (\kappa, \alpha) \right) \right], \quad (5) \]

where \( X_{\nu} (\kappa, \alpha) = \frac{\Delta_{\text{max}}}{\nu} \frac{\tau}{\pi k_B T_0}, \) \( \tau = \frac{\mu}{\pi k_B T_0}, \) \( \mu \equiv \sqrt{\frac{\hbar \omega_{c0}/2}{\pi k_B T_0}} \approx 0.365, \) and \( \omega_{c0} \equiv e H_{c20}/m^* c. \) Here, \( H_{c20} \) is the theoretical upper critical field (at \( T = 0 \) and zero spin splitting), and \( T_0 \) is the transition temperature at \( H = 0. \) The normalization factors are given by \( M_0 = \frac{\Omega_0}{H_{c20}} \) and \( S_0 = \frac{\Omega_{00}}{k_B T_0}. \)

\[ \Omega_{00} = \frac{4 \pi k_B T_0}{(2\pi)^3} \left( \frac{2m^* \pi k_B T_0}{\hbar^2} \right)^{3/2}. \]

**Figure 1.** The TP and the corresponding self-consistent SC order parameter as function of \( b \equiv H/H_{c20} \) at \( T/T_0 = 0.1. \) Note the restricted range of magnetic fields around the transitions in the plot of the TP. The following parameters have been used: \( \Omega = 100, \) \( \Omega_{00} = 3. \)

Using the above procedure it is found that for a large spin-splitting parameter, \( \tilde{\gamma} \), characterizing, e.g. the heavy-fermion superconductor URu$_2$Si$_2$, the general structure of the function \( \Omega \left( \tilde{\Delta}^2_{\text{max}} \right) \) can account for the nature of the SC phase transitions observed in this material. The usual (diamagnetic limiting, \( \tilde{\gamma} = 0 \)) situation corresponds to real positive values of \( X_{\nu} (\kappa, \alpha) \) for which \( \Omega \left( \tilde{\Delta}^2_{\text{max}} \right) \) possesses the single minimum structure characterizing the usual GL theory. For \( \tilde{\gamma} \gtrsim 1.8 \), \( X_{\nu} (\kappa, \alpha) \) can become negative and complex (features that can be "healed" by the presence of the FF modulation wavenumber \( q \) ), so that \( \Omega \left( \tilde{\Delta}^2_{\text{max}} \right) \) shows a maximum at small \( \tilde{\Delta}^2_{\text{max}} \) which is followed by a minimum at larger \( \tilde{\Delta}^2_{\text{max}} \). Thus, considering SC states with \( q = 0 \) one expects the formation of a maximum at small \( \tilde{\Delta}_{\text{max}} \) and a local minimum at larger \( \tilde{\Delta}_{\text{max}} \) by the strong spin splitting effect as the magnetic field is reduced. Upon further field decrease the minimum becomes global and so should drive a first-order normal-to-SC phase transition. Allowing for states with \( q \neq 0 \), however, the unusual features (i.e., \( Re X_{\nu} (\kappa, \alpha) < 0 \), \( Im X_{\nu} (\kappa, \alpha) \neq 0 \)) associated with the strong spin-splitting effect are "healed", and the usual single-minimum picture is recovered.

Thus, instead of the expected first-order transition to a uniform SC state one finds a second-order phase transition to a nonuniform (FF) SC state. However, following the compensation effect of the FF modulation, the field range of stability of the modulated phase is quite small. Hence, by slightly reducing the field below the second-order normal-to-SC transition, the \( q = 0 \) state becomes energetically more favorable and the system transforms from the nonuniform to a uniform SC state via a first-order phase transition. Our calculated TP, minimized with respect to both \( q \) and \( \tilde{\Delta}^2_{\text{max}} \), is shown in Fig. 1 as a function of the magnetic field. The two stages of the
SC transition are apparent. The TP for the two SC phases has different field derivatives (i.e. different magnetizations) at the low-field transition point. Fig. 1 also shows the corresponding self-consistent SC order parameter as a function of magnetic field.

The SC magnetization and entropy, estimated by use of Eqs. (4) and (5), respectively, for parameters characteristic for URu$_2$Si$_2$, are shown in Fig. 2. It is interesting to note that the relative size of the entropy (as well as of the magnetization) jump, with respect to its value just above the first-order transition, and the width of its high-field precursor (i.e., the region between the continuous and the discontinuous transition points), are very similar to those characterizing the magnetic-field dependence of the thermal conductivity observed in URu$_2$Si$_2$ [7]. These remarkable features are not sensitive to the values of the material parameters (compare, e.g., Fig. 1 to Fig. 2, which were calculated for different sets of parameters), provided that the temperature is well below the tricritical point.

In conclusion, a non-perturbative approach to the thermodynamics of strongly type-II superconductors in the Pauli paramagnetic limit, which is crucially important for determining correctly the nature of their low-temperatures SC phase transitions, is presented and applied to 3D systems, such as the heavy-fermion superconductor URu$_2$Si$_2$. We predict a sharp, double-stage transition to the SC state, and provide reliable estimates of the relative size of the jump in the SC order parameter, and in related thermodynamical properties, as well as of the overall width of the two-stage transition, in good agreement with experiment.

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