Coexistence of ferromagnetism and superconductivity near quantum phase transition: The Heisenberg- to Ising-type crossover

Andriy H. Nevidomskyy

Theory of Condensed Matter, Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, UK

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A microscopic mean-field theory of the phase coexistence between ferromagnetism and superconductivity in the weakly ferromagnetic itinerant electron system is constructed, while incorporating a realistic mechanism for superconducting pairing due to the exchange of critical spin fluctuations. The self-consistent solution of the resulting equations determines the superconducting transition temperature which is shown to depend strongly on the exchange splitting. The effect of phase crossover from isotropic (Heisenberg-like) to uniaxial (Ising-like) spin fluctuations near the quantum phase transition is analysed and the generic phase diagram is obtained. This scenario is then applied to the case of itinerant ferromagnet ZrZn$_2$, which sheds light on the proposed phase diagram of this compound. Possible explanation of superconductivity in UGe$_2$ is also discussed.

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There has been extensive experimental research done recently on the possible coexistence of superconductivity (SC) and ferromagnetism (FM) in strongly correlated electron materials. First, superconductivity was discovered in the ferromagnetic metal UGe$_2$ at high pressure [1]. Later a low-temperature SC phase was found in another f-electron compound URhGe$_2$ [2] and in the d-electron ferromagnet ZrZn$_2$ [3]. In the best studied cases of UGe$_2$ and URhGe the experiment strongly suggests that superconductivity coexists with itinerant ferromagnetism in these compounds. Notably, it is the same electrons that are involved in both SC and FM, which leads to inter-dependence of the corresponding order parameters.

The aim of this paper is to construct a mean-field theory of the phase coexistence between FM and SC on the border of magnetism, while adopting a realistic mechanism for superconducting pairing. We show that interplay between FM and SC order parameters has crucial effect on the resulting phase diagram. We then propose a mechanism that would explain the enhancement of SC transition temperature in the FM phase and discuss the application of this mechanism to ZrZn$_2$ and UGe$_2$.

The fact that SC is observed inside the FM region imposes strict limitations on the nature of the SC state. The very large internal molecular field due to the exchange interaction (measured [4] to be $\sim 240$ T in UGe$_2$) excludes, due to the Pauli limitation, not only any singlet-pairing SC but also any unitary triplet states [5]. In this paper we analyse consequences of the so-called non-unitary triplet SC state [6] on the resulting phase diagram, and then develop a microscopic theory based on spin-fluctuation mediated pairing to proceed beyond the phenomenological level of treatment reported in Ref. [5].

A nonunitary triplet state is described [7] by the order parameter $\Delta_{\alpha\beta}(k) \equiv \langle \hat{c}_{k,\alpha}c_{k,\beta} \rangle = \langle i(d(k) \cdot \sigma)\gamma_y \rangle_{\alpha\beta}$, where $\sigma = \hat{x}\sigma_x + \hat{y}\sigma_y + \hat{z}\sigma_z$ denote the usual Pauli matrices and the basis of symmetric matrices $i\sigma\gamma_y$ was used to represent odd angular momentum pairing. The three-dimensional complex vector $d(k)$ fully characterizes the triplet pairing state. In what follows we assume for simplicity an easy axis of magnetization in the $z$-direction. Because of the pair-breaking effect of strong exchange field $M$, only the Cooper pairs with parallel spins will survive. In this case of equal-spin pairing we can write vector $d$ in the form $d = (d_x, d_y, 0)$. Denoting $\Delta_{\perp} \equiv d_x + id_y$, the SC order parameter becomes

$$\Delta(k)_{\alpha\beta} = \begin{pmatrix} -\Delta_{-}(k) & 0 \\ 0 & \Delta_{+}(k) \end{pmatrix}. \quad (1)$$

We shall start from the effective Heisenberg model for itinerant electrons with spin $s(r) = \sum_{\alpha\beta} \psi_\alpha(r)\sigma_{\alpha\beta}\psi_\beta(r)$, where $\psi_\alpha(r)$, $\psi_\beta(r)$ are electron field operators. Some attractive pair-forming interaction $V$ is also assumed:

$$H_{FM+SC} = \sum_{k,\alpha} \epsilon_k \hat{c}_{k,\alpha}^\dagger \hat{c}_{k,\alpha} - I \int dr \, s(r) \cdot s(r) + \frac{1}{2} \sum_{k,\alpha,\beta,\lambda\mu} V_{\alpha\beta,\lambda\mu}(k,k') \hat{c}_{-k,\alpha}^\dagger \hat{c}_{k,\beta} \hat{c}_{k',\lambda} \hat{c}_{-k',\mu}. \quad (2)$$

Making use of the Hubbard–Stratonovich transformation and integrating out fermionic degrees of freedom in order to arrive at the effective action in terms of the bosonic field operators $\Delta_{\pm}(k)$ and $M(r) \equiv s_z(r)$, we then deduce the mean-field equations for the order parameters in the saddle-point approximation. The resulting equations for the SC order parameter have usual BCS-like form:

$$\begin{cases}
\Delta_{-}(k) = -\frac{1}{2} \sum_{k'} V(k,k') \frac{1-2f(E_{-}(k'))}{E_{-}(k')} \Delta_{-}(k') \\
\Delta_{+}(k) = -\frac{1}{2} \sum_{k'} V(k,k') \frac{1-2f(E_{+}(k'))}{E_{+}(k')} \Delta_{+}(k'),
\end{cases} \quad (3)$$

where $f(E)$ is the Fermi-Dirac distribution function.

The magnetic order parameter $M$ enters above equations via the quasiparticle spectrum $E_{\pm}(k) = \epsilon_k \mp i\Delta_{\pm}(k)$.
\[ \sqrt{(\epsilon_k + M)^2 + |\Delta_\pm(k)|^2} \]. The equation for \( M \) looks as
\[ \frac{2M}{I} = \frac{1}{V} \sum_k \left\{ \epsilon_k^+ \left[ 1 - 2f(E_-) \right] - \epsilon_k^- \left[ 1 - 2f(E_+) \right] \right\} \]
where \( \epsilon_k^\pm \equiv \epsilon_k \pm M \). In the limit of pure magnetism (i.e. \( \Delta_\pm \to 0 \) when \( T \to 0 \)), this equation reduces to the Stoner criterion for itinerant ferromagnet \( 1/I = 1/V \sum_k \left( \frac{\partial \tilde{E}_k}{\sigma_k} \right) \approx N(0) \).

In order to illustrate the interplay between the FM and SC order parameters that follows from this model, we solved Eqs. \( 3,4 \) self-consistently for the simple case of spherical Fermi surface at half filling, while assuming that SC pairing strength in the \( p \)-channel \( V(k, k') \equiv V_{l=1}(k, k') \sum_{m=-1}^1 Y_{1m}(k) Y_{1m}^*(k') \) has BCS-like form (i.e. \( V_1(k, k') \) vanishes everywhere except the narrow region near the Fermi surface) and does not depend on exchange interaction \( I \). The resulting SC transition temperature was calculated in the weak-coupling BCS approximation and is shown in Fig. 1 as a function of dimensionless interaction constant \( I = N(0)I \). It is apparent that exchange splitting has large effect on superconductivity, enhancing \( T_{SC} \) in the FM phase for the majority spin channel and suppressing it for the minority spin. This is not surprising since exchange splitting enhances (suppresses) the density of states (DOS) \( N_\sigma \) in the majority (minority) spin channel, which enters the expression for the dimensionless pairing strength \( \lambda_\sigma^\prime = N_\sigma(0)|V_1| \). We note that at this stage the symmetry of the magnetic state has no effect on \( T_{SC} \) since the mass renormalization effects have not been taken into account when calculating \( T_{SC} \). These effects will prove to be very important in what follows.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{The calculated SC transition temperature as a function of interaction strength \( I \) obtained as a result of solving self-consistent Eqs. \( 3,4 \) in the weak-coupling BCS approximation \( T_{SC} \approx 1.14 \omega_c \exp(-1/\lambda_\sigma^\prime) \), where frequency cutoff \( \omega_c \) was chosen arbitrarily \( \omega_c = 0.01 E_F \). The solid (dashed) line shows \( T_{SC} \) for majority (minority) spin in units of \( E_F \). Exchange splitting \( M \) is plotted with dotted line (right scale).}
\end{figure}

We shall now address the issue of the mechanism of the superconducting pairing that arises in the vicinity of the magnetic quantum phase transition. It has been shown that exchange of spin fluctuations, called paramagnons, can lead to an attractive pairing interaction. On the paramagnetic side the strength of this interaction was derived in the context of superfluidity in \(^3\)He \( \lambda \). On the FM side of the transition the corresponding formulae were obtained in Ref. \( 10,11 \). Following their approach, the (attractive) equal-spin pairing interaction is given by
\[ V^{ss}(k, q + k) = - \left( \frac{I^2 \chi_0^{ss}}{1 - I^2 \chi_0^{ss}} \right) \]
where \( \chi_0^{ss}(q) \equiv \chi_0^{ss}(i\omega_n, q) \) is the Lindhard function of the non-interacting system in the given spin channel. The usual BCS pairing parameter \( \lambda \) in the triplet channel is given by \( \lambda_{ss}^\prime = N_\sigma(0)|V_{ss}^\prime| \), where \( N_\sigma(0) \) is the DOS at Fermi level and \( V_{ss}^\prime \) is the strength of the interaction in the \( l \)-orbital channel, which for spherical Fermi surface is
\[ V_{ss}^\prime = \int_0^{2\pi} dq \frac{dq}{2q^2} P_l \left( 1 - \left( q^2/2k_F^2 \right) \right) \]
where \( P_l(z) \) denotes the Legendre polynomial of order \( l \). The dominant contribution to the integral comes from the small-\( q \) region since \( V^{ss}(q) \) is strongly peaked for \( q \to 0 \). This allows us to employ the small-\( q \) approximation for the Lindhard function \( \chi_0^{ss}(q) \approx N_\sigma(0)[1 - \frac{1}{12}(q/k_F)^2 + O(q^4)] \), which enters Eq. 5.

The mass enhancement near magnetic instability renormalizes the BCS pairing parameter \( \lambda_0 \) to the new value \( \lambda^* = \lambda_0^2/Z_\sigma(0) \equiv \lambda_0^2/(1 + \lambda_0^2) \), where \( \lambda_0 \) is the s-wave pairing interaction parameter and \( Z(0) \) is the mass enhancement factor \( 29 \) at the Fermi surface given by
\[ \frac{m^*}{m} \approx Z(0) = \left( 1 - \frac{\partial \Sigma(\omega, |k| = k_F)}{\partial \omega} \right) \]
where \( \Sigma \) is the single-particle self-energy.

It is remarkable that experimentally no SC is observed in the paramagnetic (PM) phase of UGe\(_2\) and ZrZn\(_2\). Admittedly, this is different from the result of Fay and Appel \( 11 \) who obtained comparable values of \( T_{SC} \) on both sides of the magnetic transition. Here we propose a tentative generic explanation for the strong suppression of SC on the PM side of the transition in the nearly ferromagnetic metal, which is due to the different nature of spin fluctuations on the two sides of the magnetic quantum phase transition. We consider a nearly ferromagnetic metal which has no preferred magnetization axis in the PM phase and thus is characterized by spin fluctuations that are of Heisenberg type. In this case both longitudinal and transverse spin fluctuations contribute to the effective mass enhancement, so that
\[ Z(0) = 1 + \lambda_0^2 + \lambda_0^2 \]
Consequently, the renormalized pairing strength \( \lambda^* = \lambda_0/Z(0) \) and the SC transition temperature are both small on the PM side. Remarkably, higher
values of $T_{SC}$ on the FM side could be achieved if spin fluctuations there were of Ising-type, so that only longitudinal spin fluctuations contribute to the effective mass enhancement $Z(0) = 1 + \lambda^L Q_0$. This would lead to $Z(0)$ being about three times larger in the PM phase than it is in the FM phase, as illustrated in Fig. 2a).

The SC transition temperature is notoriously difficult to calculate. For the purpose of comparison of $T_{SC}$ on both sides of the FM transition a simple McMillan-type formula [12] should suffice:

$$T_{SC}^{1,\sigma} \approx 1.14 \omega_c \exp[-1/\lambda^\sigma Q_0]$$

(8)

where cutoff $\omega_c$ simulates in a crude way the fact that in reality $\lambda^\sigma(\omega, q)$ is strongly frequency-dependent, being sharply peaked at small energy transfers. It turns out [12] that $\omega_c$ depends strongly on exchange interaction, as shown in the inset of Fig. 2a). The resulting dependence

$T_{SC}(T)$ is plotted in Fig. 2c), which indicates clearly that the SC transition temperature is an order of magnitude higher in the FM phase than it is in the PM phase.

Our calculations suggest that $T_{SC}$ goes through a maximum and then approaches zero at the quantum critical point, in accordance with Ref. [11, 12]. A recent strong-coupling calculation by Roussev and Millis [13] suggests however that $T_{SC} > 0$ generically at the magnetic critical point, contrary to our result. We note that though interesting from the fundamental point of view, the behaviour of $T_{SC}$ directly at the magnetic phase transition is not so important in practice, since experimentally magnetic transition proves to be first order [1, 2], thereby eliminating the low values of $(T - T_c)$ from consideration.

For outlined scenario to take place, two crucial conditions are necessary. Firstly, the contribution of soft transverse spin fluctuations to $\lambda_0$ must be quenched on the FM side. This is achieved due to spin waves taking over the available phase space as the magnetization $M$ increases. Indeed, the fraction of the momentum space available to gapless spin fluctuations is $q > q_\perp \equiv k_{F\perp} - k_{F\perp}^*$, the rest being taken by spin waves at $0 \leq q \leq q_\perp$. Long-wavelength spin waves themselves do not contribute to the mass enhancement in the leading order of the perturbation theory [15]. Thus as the exchange splitting increases, the soft spin fluctuations shift to larger $q$-values, thereby decreasing their contribution to $\lambda_0$.

However, this suppression also affects the pairing strength $\lambda_L \approx \chi_{LL}$ due to longitudinal spin fluctuations which become quenched as well. The situation can be cured by the second condition: the existence of the quantum meta-magnetic transition (MMT) [16] somewhere in the FM phase. Indeed, the longitudinal susceptibility $\chi_{LL}$ is peaked near the jump in magnetization accompanying such a transition, as seen experimentally in UGe$_2$ [17] and in Sr$_3$Ru$_2$O$_7$ [18]. As a result, longitudinal spin fluctuations will be enhanced and the material will appear effectively Ising-like near the MMT, justifying the assumption made above. However unlikely the “coincidental” presence of meta-magnetic transition near the quantum transition to the FM phase may appear at first sight, the experiment suggests that this is not uncommon in the ferromagnetic strongly correlated electron materials. Indeed, the MMT has been observed in Sr$_3$Ru$_2$O$_7$ [19], UGe$_2$ [17, 20], and recently in ZrZn$_2$ [21].

We now turn to the application of the above model to the experimentally studied materials. Figure 3 shows the generic phase diagram of an itinerant ferromagnet that arises from studies of ZrZn$_2$ [3, 21] and UGe$_2$ [1, 20, 22, 23]. The Curie temperature $T_c$ is suppressed to zero at pressure $p_c$, where the transition appears to be first order [1, 8] in both compounds. Another feature, the crossover line $T_c$ between the two ferromagnetic phases, FM1 and FM2, is also shown. This crossover exhibits itself as an anomaly in the measurements of resistivity [1, 22, 26] and specific heat [26, 27] in UGe$_2$, where it
indicates a crossover, rather than a sharp phase transition.

The situation is more intricate in UGe$_2$ where evidence of strong uniaxial anisotropy exists on both sides of magnetic transition at $p_c$. However in the light of recent measurements of specific heat $^{27}$ it becomes evident that the very narrow SC region is centred around the MMT at $p_x$ rather than $p_c$. This can be easily understood given that the transition at $p_c$ is strongly first order $^1$ and therefore has a strong pair-breaking effect on SC. It is hence not surprising that no SC is seen on both sides of $p_c$. The presence of SC in UGe$_2$ is instead due to critical spin fluctuations at $p_x$, which is only a weakly first order transition. We note that this view has been expressed already in earlier works on UGe$_2$ $^{22,27}$. Our proposed theoretical model has thus an indirect application to UGe$_2$ in a sense that the observed uniaxial (Ising) symmetry enhances $T_{SC}$, which would have been much more suppressed if the Heisenberg-like spin fluctuations had prevailed in this compound.

The above argument already suggests that SC phase must be suppressed both in FM1 and PM phases of UGe$_2$ close to $p_c$. It should be noted that the qualitative change in the Fermi surface observed at the magnetic transition by de Haas–van Alphen experiment $^{28}$ may be another factor that suppresses SC near $p_c$. In this context, the existence of a double peak structure in the electronic density of states very close to the Fermi level has been proposed $^{24}$ as a possible microscopic explanation. We also note that absence of SC in the PM phase of UGe$_2$ may be partly due to the spin degeneracy of the Fermi surface as it can, in principle, enhance spin-flip processes of the electrons forming a Cooper pair, which would have detrimental consequence on spin-triplet SC state.

In conclusion, we have formulated a mean-field theory of coexisting FM and SC in terms of the equations for the corresponding order parameters that have to be solved self-consistently. We have also incorporated a microscopic mechanism of the SC pairing due to the exchange of spin fluctuations in our model. A scenario based on Heisenberg- to Ising-type crossover has been proposed, which provides a natural explanation of the enhancement of SC on the FM side of magnetic transition, observed experimentally in ZrZn$_2$. The apparent suppression of SC in the PM phase of UGe$_2$ is explained by the detrimental effect that the strongly first-order phase transition at $p_c$ has on pair-forming spin fluctuations. The proposed theoretical model supports the evidence of SC in UGe$_2$, as superconductivity is predicted to be enhanced by Ising-like spin fluctuations near $p_c$.

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$^{*}$ahl229@phy.cam.ac.uk

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