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On the phase diagrams of the ferromagnetic superconductors UGe_2 and $ZrZn_2$

Michael G. Cottam [a], Diana V. Shopova [b], Dimo I. Uzunov [a,b,*]

[a] Department of Physics and Astronomy, University of Western Ontario, London, Ontario, N6A 3K7, Canada
[b] CP Laboratory, Institute of Solid State Physics, Bulgarian Academy of Sciences, BG-1784 Sofia, Bulgaria

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ABSTRACT

A general phenomenological theory is presented for the phase behavior of ferromagnetic superconductors with spin-triplet electron Cooper pairing. The theory accounts in detail for the temperature-pressure phase diagram of $ZrZn_2$, while the main features of the diagram for UGe_2 are also described. Quantitative criteria are deduced for the U-type (type I) and Zr-type (type II) unconventional ferromagnetic superconductors with spin-triplet Cooper electron pairing. Some basic properties of quantum phase transitions are also elucidated.

The remarkable coexistence of itinerant ferromagnetism and unconventional (spin-triplet) superconductivity at low temperatures ($T < 1$ K) was discovered experimentally in the intermetallic compounds UGe_2 [1–3], $ZrZn_2$ [4], and URhGe [5]. Other metallic compounds, such as UCoGe [6,7] and UIr [8,9], were also found to be spin-triplet ferromagnetic superconductors. In $ZrZn_2$, URhGe, and UCoGe, the mixed phase of coexistence of ferromagnetism and unconventional superconductivity (labeled the FS phase) occurs over a wide range of pressure (i.e., from ambient pressure $P \sim 1$ bar up to 10 kbar). By contrast, in other compounds (e.g., UGe_2 and UIr) this FS phase is found only in the high-pressure part ($P \sim 10$ kbar) of the T–P phase diagram.

Another feature of the above compounds is that the FS phase occurs only in the ferromagnetic phase domain of the T–P diagram. Specifically, at equilibrium and a given P, the temperature $T_F(P)$ of the normal-to-ferromagnetic phase (or N–FM) transition is never lower than the temperature $T_{FS}(P)$ of the ferromagnetic-to-FS phase (or FM–FS) transition. This is consistent with the point of view that the superconductivity in these compounds is triggered by the spontaneous magnetization $\boldsymbol{M}$, by analogy with the well-known triggering of the superfluid phase A_1 in ^{3}He at mK temperatures by the external magnetic field $\boldsymbol{H}$. This helium-analogy has been used in some theoretical studies (see, e.g., [10–13]), where Ginzburg–Landau (GL) free energy terms to describe the FS phase were derived by symmetry arguments.

For the spin-triplet ferromagnetic superconductors the trigger mechanism was recently examined in detail [13]. The main system properties are affected by a term in the GL expansion of the form $\boldsymbol{M}|\boldsymbol{\psi}|^2$, which represents the interaction of $\boldsymbol{M} = \{M_j;\ j = 1, 2, 3\}$ with the complex superconducting vector field $\boldsymbol{\psi} = \{\psi_j\}$. Specifically, this term triggers $\boldsymbol{\psi} \neq 0$ for certain T and P values. An analogous trigger mechanism is familiar in the context of improper ferroelectrics [14].

A crucial consideration in this work is the nonzero magnetic moment of the spin-triplet Cooper pairs of the electrons. While the spin-singlet Cooper pairs have net spin zero and are quite sensitive to the magnitude of the magnetic induction $\boldsymbol{B}$, the spin-triplet pairs are known to be robust with respect to relatively large $\boldsymbol{B}$. The phenomena of spin-triplet superconductivity and itinerant ferromagnetism are both due to the same electron bands of the compounds: the f-band electrons in uranium-based compounds and the d-band electrons in $ZrZn_2$. However, the microscopic band theory of magnetism and superconductivity in non-Fermi liquids of strongly interacting heavy electrons is either too complex or insufficiently developed to describe the complicated behavior in itiner-

* Corresponding author at: Department of Physics and Astronomy, University of Western Ontario, London, Ontario, N6A 3K7, Canada.
E-mail address: d.i.uzunov@gmail.com (D.I. Uzunov).

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ant ferromagnetic compounds. Consequently, several authors (see [10–13,15]) have explored a phenomenological description within self-consistent mean field theory, and we build on a similar approach here.

In this Letter, by focusing on $ZrZn_2$ and UGe_2 with their contrasting types of behavior, we show that the T–P phase diagrams of spin-triplet ferromagnetic superconductors can be successfully described starting from the general GL free energy $F(\boldsymbol{\psi}, \boldsymbol{M})$ established in Refs. [10–13]. The present phenomenological approach includes both mean-field and spin-fluctuation theory (SFT), as in [16], considerations. We propose a simple, yet comprehensive, modeling of the P dependence of the free energy parameters, from which it is shown that the phase diagram of $ZrZn_2$ is obtained in good quantitative agreement with the experimental data [4]. Further, the main features [1] of the T–P diagram of UGe_2 are also well-described within our approach. The theory is capable of outlining several different possible topologies for the T–P phase diagram, depending on the GL parameters of the material which can be chosen in accordance with experiment. Quantitative criteria emerge for two distinct types of behavior for unconventional ferromagnetic superconductors, which we label Zr-type and U-type. Further possible applications are to URhGe, UCoGe, and UIr. Our results address questions regarding the order of the quantum phase transitions at ultra-low and zero temperatures. They also pose intriguing questions pointing to further experimental investigations of (e.g.) the detailed structure of the phase diagrams in the high-P/low-T region.

Following Ref. [13] the free energy per unit volume, $F/V = f(\boldsymbol{\psi}, \boldsymbol{M})$, can be written in the form

$$f(\boldsymbol{\psi}, \boldsymbol{M}) = a_s|\boldsymbol{\psi}|^2 + \frac{b_s}{2}|\boldsymbol{\psi}|^4 + \frac{u_s}{2}\left|\boldsymbol{\psi}^2\right|^2 + \frac{v_s}{2}\sum_{j=1}^{3}|\psi_j|^4 + a_f\boldsymbol{M}^2 + \frac{b_f}{2}\boldsymbol{M}^4 + i\gamma_0\boldsymbol{M}\cdot(\boldsymbol{\psi}\times\boldsymbol{\psi}^*) + \delta\boldsymbol{M}^2|\boldsymbol{\psi}|^2. \quad (1)$$

The material parameters satisfy, as in [13], $b_s > 0$, $b_f > 0$, $a_s = \alpha_s(T - T_s)$, and $a_f = \alpha_f[T^n - T_f^n(P)]$, where $n = 1$ gives the standard form of a_f, and $n = 2$ applies for SFT [16]. The terms proportional to u_s and v_s describe, respectively, the anisotropy of the spin-triplet electron Cooper pairs and the crystal anisotropy. Next, $\gamma_0 \sim J$ (with $J > 0$ the ferromagnetic exchange constant) and $\delta > 0$ are parameters of the $\boldsymbol{\psi}$–$\boldsymbol{M}$ interaction terms. Previous mean-field studies have shown that the anisotropies represented by the u_s and v_s terms in Eq. (1) slightly perturb the size and shape of the stability domains of the phases, while similar effects can be achieved by varying the b_s factor in the $b_s|\boldsymbol{\psi}|^4$ term. For these reasons, in the present analysis we ignore the anisotropy terms, setting $u_s = v_s = 0$, and consider $b_s \equiv b > 0$ as an effective parameter. Then, without loss of generality, we are free to choose the magnetization vector to have the form $\boldsymbol{M} = (0, 0, M)$.

A convenient dimensionless free energy can now be defined by $\tilde{f} = f/(b_f M_0^4)$, where $M_0 = [\alpha_f T_{f0}^n/b_f]^{1/2} > 0$ is the value of M corresponding to the pure magnetic subsystem ($\boldsymbol{\psi} \equiv 0$) at $T = P = 0$ and $T_{f0} = T_f(0)$. On scaling the order parameters as $m = M/M_0$ and $\boldsymbol{\varphi} = \boldsymbol{\psi}/[(b_f/b)^{1/4}M_0]$ we obtain

$$\tilde{f} = r\phi^2 + \frac{\phi^4}{2} + tm^2 + \frac{m^4}{2} + 2\gamma m\phi_1\phi_2\sin\theta + \gamma_1 m^2\phi^2, \quad (2)$$

where $\phi_j = |\varphi_j|$, $\phi = |\boldsymbol{\varphi}|$, and θ is the phase angle between the complex φ_2 and φ_1. The dimensionless constants are $t = [\tilde{T}^n - \tilde{T}_f^n(P)]$, $r = \kappa(\tilde{T} - \tilde{T}_s)$ with $\kappa = \alpha_s b_f^{1/2}/\alpha_f b^{1/2} T_{f0}^{n-1}$, $\gamma = \gamma_0/[\alpha_f T_{f0}^n b]^{1/2}$, and $\gamma_1 = \delta/(bb_f)^{1/2}$. The reduced temperatures are $\tilde{T} = T/T_{f0}$, $\tilde{T}_f(P) = T_f(P)/T_{f0}$, $\tilde{T}_s(P) = T_s(P)/T_{f0}$.

The analysis involves making simple assumptions for the P dependence of the t, r, γ, and γ_1 parameters in Eq. (2). Specifically, we assume that only T_f has a significant P dependence, described by $\tilde{T}_f(P) = (1 - \tilde{P})^{1/n}$, where $\tilde{P} = P/P_0$ and P_0 is a characteristic pressure deduced later. In $ZrZn_2$ and UGe_2 the P_0 values are very close to the critical pressure P_c at which both the ferromagnetic and superconducting orders vanish, but in other systems this is not necessarily the case. As we will discuss, the nonlinearity ($n = 2$) of $T_f(P)$ in $ZrZn_2$ and UGe_2 is relevant at relatively high P, at which the N–FM transition temperature $T_F(P)$ may not coincide with $T_f(P)$.

The simplified model in Eq. (2) is capable of describing the main thermodynamic properties of spin-triplet ferromagnetic superconductors. There are three stable phases: (i) the normal (N) phase, given by $\phi = m = 0$; (ii) the pure ferromagnetic (FM) phase, given by $m = (-t)^{1/2} > 0$, $\phi = 0$; and (iii) the FS phase, given by $\phi_1^2 = \phi_2^2 = (\gamma m - r - \gamma_1 m^2)/2$, $\phi_3 = 0$, where $\sin\theta = -1$ and m satisfies

$$\left(1 - \gamma_1^2\right)m^3 + \frac{3}{2}\gamma\gamma_1 m^2 + \left(t - \frac{\gamma^2}{2} - \gamma_1 r\right)m + \frac{\gamma r}{2} = 0. \quad (3)$$

We note that FS is a two-domain phase as discussed in Refs. [12, 13]. Although Eq. (3) is complicated, some analytical results follow, e.g., we find that the second order phase transition line $\tilde{T}_{FS}(P)$ separating the FM and FS phases is the solution of

$$\tilde{T}_{FS}(P) = \tilde{T}_s + \frac{\gamma_1}{\kappa}t(T_{FS}) + \frac{\gamma}{\kappa}\left[-t(T_{FS})\right]^{1/2}. \quad (4)$$

Under certain conditions, the $T_{FS}(P)$ curve has a maximum at $\tilde{T}_m = \tilde{T}_s + (\gamma^2/4\kappa\gamma_1)$ with pressure P_m found by solving $t(T_m, P_m) = -(\gamma^2/4\gamma_1^2)$. Examples will be given later, but generally this curve extends from ambient P up to a tri-critical point labeled B, with coordinates (P_B, T_B), where the FM–FS phase transition occurs at a straight line of first order transition up to a critical end-point C. The lines of all three phase transitions (N–FM, N–FS, and FM–FS) terminate at C. For $P > P_C$ the FM–FS phase transition occurs on a rather flat, smooth line of equilibrium transition of first order up to a second tri-critical point A with $P_A \sim P_0$ and $T_A \sim 0$. Finally, the third transition line terminating at C describes the second order phase transition N–FM. The temperatures at the three multi-critical points correspond to $\tilde{T}_A = \tilde{T}_s$, $\tilde{T}_B = \tilde{T}_s + \gamma^2(2 + \gamma_1)/4\kappa(1 + \gamma_1)^2$, and $\tilde{T}_C = \tilde{T}_s + \gamma^2/4\kappa(1 + \gamma_1)$, while the P values can be deduced from the previous equations. These results are valid whenever $T_f(P) > T_s(P)$, which excludes any pure superconducting phase ($\boldsymbol{\psi} \neq 0$, $m = 0$) in accord with the available experimental data. Note that, for any set of material parameters, $T_A < T_C < T_B < T_m$ and $P_m < P_B < P_C$.

A calculation of the T–P diagram from Eq. (2) for any material requires some knowledge of P_0, T_{f0}, T_s, κ γ, and γ_1. The temperature T_{f0} can be obtained directly from the experimental phase diagrams. The model pressure P_0 is either identical to or very close to the critical pressure P_c at which the N–FM phase transition line terminates at $T \sim 0$. The characteristic temperature T_s of the generic superconducting transition is not available from the experiments and thus has to be estimated using general consistency arguments. For $T_f(P) > T_s(P)$ we must have $T_s(P) = 0$ at $P \geqslant P_c$, where $T_f(P) \leqslant 0$. For $0 \leqslant P \leqslant P_0$, $T_s < T_C$ and therefore for cases where T_C is too small to be observed experimentally, T_s can be ignored. For systems where T_C is measurable this argument does not hold. This is likely to happen for $T_s > 0$ (for $T_s < 0$, T_C is very small). However, in such cases, pure superconducting phase should be observable. To date there are no experimental results reported for such a feature in $ZrZn_2$ or UGe_2, and thus we can put $T_s = 0$. We remark that negative values of T_s are possible, and they describe a phase diagram topology in which the FM–FS transition line terminates at $T = 0$ for $P < P_c$. This might be of relevance for other compounds, e.g., URhGe.