Phenomenological Theory of Ferromagnetic Superconductivity

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It is argued that the pairing symmetry realized in a ferromagnetic superconductor UGe_2 must be a nonunitary triplet pairing. This particular state is free from the Pauli limitation and can survive under a huge internal molecular field. To check our identification we examine its basic properties and several experiments are proposed. In particular, the external field is used to raise T_c by controlling the internal spontaneous dipole field.

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Ferromagnetism (FM) and superconductivity (SC) are thought to be basically mutually repulsive. Since Ginzburg [1] points out a possibility of its coexistence under the condition that the magnetization is less than the thermodynamic critical field, there are many experimental investigations performed, starting by Matthias *et al.* [2] who consider impurity ferromagnetism in a superconductor. Although the coexistence between antiferromagnetism and superconductivity is rather easy to realize and actually observed in several compounds [3] because the antiferromagnetic moments spatially averaged over the SC coherent length vanish, the ferromagnetic case is difficult to realize. Rare exceptions of this case are rare-earth ternary compounds $HoMo_6S_8$ and $ErRh_4B_4$ where in a narrow temperature region just below the Curie temperature T_{FM} the coexistence of FM and SC is attained [4]. When the rare-earth 4*f* moments completely align at lower T, SC is wiped out by a strong internal field. So far there is no known SC which can fully sustain such a large molecular field.

As for the theoretical developments, Anderson and Suhl [5] discuss the cryptoferromagnetism as a possible coexistence phase where the FM is modified to a long-period modulated spin structure. This modified RKKY interaction is mediated by SC pairs. Similarly to this idea, Blount and Varma [6] propose a magnetic spiral phase coexisting with SC to gain electromagnetic dipole interaction between localized moments. The coexistence of weak itinerant FM with s-wave SC is also discussed [7,8].

The recent discovered material UGe_2 [9] seems to be difficult to understand in terms of these theories so far proposed and seems to require a novel concept to interpret the data, because previous theories assume that two groups of electrons are distinguishable and clearly separated spatially. FM comes from the well localized 4*f* electrons while SC pairs are formed by conduction electrons. Here the situation is much more intricate; we cannot separate two groups in a well defined way from the outset, where 5*f* electrons from U atoms play double roles both for FM and SC. It is quite an interesting problem how to describe its double roles played by 5*f* electrons microscopically; here we confine our discussions to a phenomenological level without going into pairing mechanism, which is closely related to it.

At ambient pressure UGe_2 is an itinerant metallic ferromagnet whose Curie temperature $T_{FM} = 52$ K and the spontaneous moment $\sim 1.4\mu_B$/U atom. The easy axis is the a axis in orthorhombic crystal [10]. Upon increasing pressure P, both T_{FM} and the spontaneous moment moderately decrease gradually and at $P \sim 1$ GPa the SC starts to appear and the SC transition temperature T_c increases, taking a maximum $T_c \sim 0.8$ K above which FM already sets in at a high T of $T_{FM} \sim 30$ K whose moment $\sim 1\mu_B$/U atom. Further increasing P, both T_{FM} and T_c drop down to disappear almost simultaneously around $P \sim 1.7$ GPa. Thus the SC region is entirely covered by FM. The SC is confirmed to coexist with FM by neutron experiment [11]. This phenomenon is difficult to understand in terms of the previous framework based on a conventional singlet pairing state. The internal ferromagnetic molecular field coming from the ordered magnetic system through the exchange interaction amounts to an order of a few hundred tesla (T) in view of the exchange splitting $\sim$70 meV for the up-spin and down-spin Fermi surfaces [12]. According to Tsutsui *et al.* [13] the hyperfine field probed by Mössbauer spectroscopy on uranium is $\sim$240 T. This huge exchange field apparently excludes not only any singlet pairing category, but also certain forms of the triplet pairing category, namely, unitary triplet states. These are all limited by the Pauli paramagnetic field $H_p \sim 1.3T_c$(T). The theories based on s-wave SC [5–8] are not applicable here, and the Fulde and Ferrell state only slightly enhances H_{c2} by at most 10% [14], but definitely not possible for $>$100 T.

In fact, the only possible pairing symmetry under such a strong internal field is the nonunitary triplet state, which is free from the Pauli limit [15]. Thus, we investigate this possibility in light of the present material UGe_2, examine the available data, and predict some of the interesting phenomena associated with this nonunitarity. A spin-fluctuation mediated pairing mechanism of a triplet SC coexisting with weak itinerant FM is proposed previously by Fay and Appel [16] who base it on RPA. We note

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in general that the strong coupling effects may be needed to stabilize a nonunitary state [17].

A nonunitary triplet state is described by the order parameter $\hat{\Delta}(\mathbf{k}) = i[\mathbf{d}(\mathbf{k}) \cdot \vec{\sigma}]\sigma_y$ in a 2×2 matrix form. The three-dimensional complex vector $\mathbf{d}(\mathbf{k})$ fully characterizes the triplet pairing state. If $\mathbf{d}(\mathbf{k})$ is a complex number, the product $\hat{\Delta}(\mathbf{k})\hat{\Delta}(\mathbf{k})^\dagger = |\mathbf{d}(\mathbf{k})|^2\sigma_0 + i[\mathbf{d}(\mathbf{k}) \times \mathbf{d}^*(\mathbf{k})] \cdot \vec{\sigma}$ is not a multiple of the unit matrix and $\hat{\Delta}(\mathbf{k})$ becomes nonunitary. Thus in a nonunitary state time reversal symmetry necessarily is broken spontaneously and spontaneous moment $\mathbf{m}(\mathbf{k}) \propto i\mathbf{d}(\mathbf{k}) \times \mathbf{d}^*(\mathbf{k})$ appears at each $\mathbf{k}$ point, yielding the macroscopic averaged moment $\langle \mathbf{m}(\mathbf{k}) \rangle$ provided that its Fermi surface average nonvanishes. There are two kinds of nonunitary states among several possible forms, depending on its orbital structure: bipolar and nonbipolar. In the former (latter) the real and imaginary parts of the complex $\mathbf{d}(\mathbf{k})$ vector are ascribed by different (same) orbital function(s). The average spontaneous moment in the latter (former) is nonvanishing (vanishing) in general by symmetry. Since the additional Zeeman magnetic energy is gained in the nonbipolar, in the following we consider only the nonbipolar state.

We derive an appropriate Ginzburg-Landau (GL) free energy density functional allowed in the presence of the FM described in terms of the complex order parameter $\vec{\eta}$ of the coefficients of the d vector. The macroscopic ferromagnetic order parameter $\mathbf{M}$ is proportional to the magnetization which is estimated as ~ 0.2 T in the ambient pressure [9]. The complex order parameter $\vec{\eta}$ is coupled linearly and quadratically with $\mathbf{M}$, namely, $i\gamma \mathbf{M} \cdot \vec{\eta} \times \vec{\eta}^*$ and $M^2 \vec{\eta} \cdot \vec{\eta}^*$, respectively. Thus it is convenient to write $\vec{\eta} = (\eta_x, \eta_y, 0)$ or $\eta_\pm = \frac{1}{\sqrt{2}}(\eta_x \pm i\eta_y)$ by choosing $\mathbf{M} = (0, 0, M)$, implying that the Cooper pair spin orientation points to the $\mathbf{M}$ direction. We choose a coordinate system: $x \parallel b, y \parallel c, z \parallel a$, where the magnetic easy axis is the a axis. In view of the fact that the lattice constant a and c are almost the same, only a 2% difference, we regard the crystal structure as tetragonal where the (a, c) forms the basal plane.

So far we have discussed the possible coupling terms with FM allowed by symmetry in order to explicitly see the effects of the FM formation. If we further consider the microscopic origin of SC, which appears under the Zeeman split Fermi surfaces (FS), leading to the distinct transition temperatures and the different fourth order terms for $\eta_\pm$ from the outset. Thus we finally arrive at the following generic GL form appropriate to UGe_2:

$$f_{\mathrm{bulk}} = \alpha_+|\eta_+|^2 + \alpha_-|\eta_-|^2 + \frac{1}{2}\beta_+|\eta_+|^4 + \beta|\eta_+|^2|\eta_-|^2 + \frac{1}{2}\beta_-|\eta_-|^4, \tag{1}$$

where we introduce $\alpha_\pm = \alpha_0^\pm(T - T_c^\pm)$ and $\beta_\pm > 0$ for the stability condition of the system. In the following, T_c^+ is identified to the observed T_c below which η_+ becomes nonvanishing.

It is easy to derive the condition for the second SC transition to occur: $\alpha_0^- T_c^- / \alpha_0^+ T_c^+ > \beta/\beta_+$ at $T_{c2} = (\beta_+ \alpha_0^- T_c^- - \beta \alpha_0^+ T_c^+)/(\beta_+ \alpha_0^- - \beta \alpha_0^+)$ at which the remaining order parameter η_- starts to appear. Thus there is a chance to find the double transition near the critical pressure 1.7 GPa where FM is suppressed and T_c^- will approach T_c^+. We note that this transition is analogous to the A_1-A phase transition in superfluid ^{3}He described by Ambegaokar and Mermin [18]. Near the critical pressure where M is small, we can estimate relative changes of the two transitions by expanding the density of states $N(\epsilon_F)$ at ϵ_F, giving rise to $T_{c1,2} = T_{c0}(1 \pm M \frac{N'(\epsilon_F)}{N(\epsilon_F)} \ln \frac{\omega}{T_{c0}})$, the derivative of the densities of states $N'(\epsilon_F)$ and the energy cutoff ω.

The quasiparticle excitation spectrum for a nonbipolar type $\mathbf{d}(\mathbf{k}) = \vec{\eta}\phi(\mathbf{k})$, where the spin $\vec{\eta}$ and the orbital $\phi(\mathbf{k})$ parts are separable, has two branches: $E_\sigma(\mathbf{k}) = \sqrt{\epsilon_\sigma^2(\mathbf{k}) + \Delta_\sigma^2(\mathbf{k})}$ $(\sigma = \pm)$. The gap functions are given by $\Delta_\pm(\mathbf{k}) = |\eta_x \pm \eta_y|\phi(\mathbf{k}) = |\eta_\pm|\phi(\mathbf{k})$. Thus one of the branches, say, $\Delta_-(\mathbf{k})$, vanishes identically. That means that on the spin-down Fermi surface there is no superconducting gap formed, leaving it normal.

The nodal structure associated with nonunitarity leads to several observable predictions without specifying a particular orbital function $\phi(\mathbf{k})$. The total density of states is given by $N(E) = N_+(E) + N_-(E)$, where $N_\sigma(E) = \Sigma_{\mathbf{k}}\delta[E - E_\sigma(\mathbf{k})]$. At the SC transition temperature, only the spin-up FS opens the gap, thus the jump of the specific heat is substantially reduced from the BCS value (1.43). In the lowest T the specific heat becomes $C(T) = \gamma_- T$, where γ_- is the densities of states at the FS for the spin-down band. According to an estimate by the spin polarized band structure calculation [12] γ_- is substantial compared with the total γ $(= \gamma_+ + \gamma_-)$. Thus, it is quite observable. The existence of the γ_- term governs the lowest-T thermodynamics, such as a T-linear term in thermal conductivity and the I-V characteristics of quasiparticle tunneling between a normal metal and UGe_2, etc.

We notice that if the second transition T_{c2} really takes place at lower T, there is no residual γ_- contribution to the thermodynamic quantities mentioned above since the remaining spin-down FS is also gapped by η_-.

As for the spin susceptibility χ_i $(i = a, b$ and $c)$ probed by the Knight shift experiment, we naively expect that χ_b and χ_c (χ_a) must decrease (be unchanged) below T_c because the $\mathbf{d}(\mathbf{k})$ vector lies in the (b, c) plane, but remain a finite value, corresponding to γ_-. Note that χ_b and χ_c may not decrease appreciably below T_c because these transverse susceptibilities in FM are known to be determined by the whole bands and insensitive to the gap formation near FS. The direction of the $\mathbf{d}(\mathbf{k})$ vector is strongly locked to $\mathbf{M} \parallel a$ where UGe_2 is known to have a strong easy axis type of the magnetic anisotropy and the magnetization is Ising-like [9].

Having analyzed the basic thermodynamic properties of the nonunitary state for UGe_2, we now proceed to examine electromagnetic properties when the external field is applied, regarding UGe_2 a conventional type II superconductor. We note, however, that without H vortices with each having unit quantum flux are spontaneously created because the internal field ~0.2 T far exceeds the usual H_{c1}. In this sense the system lacks the complete Meissner phase in the H vs T plane. Let us introduce the free energy density f_{grad} related to the magnetic field H

$$f_{\mathrm{grad}} = K_1\Sigma_{\sigma=\pm}\Sigma_{j=y,z}|D_j\eta_\sigma|^2 + K_2\Sigma_{\sigma=\pm}|D_x\eta_\sigma|^2, \tag{2}$$

where $D_j = \partial_j - i\frac{2e}{\hbar c}A_j$ and the unit flux $\phi_0 = \frac{\hbar c}{2e}$. We have assumed a simple orbital function $\phi(\mathbf{k})$ which does not break tetragonal symmetry in the basal plane (a, c) or (y, z) for simplicity. We reserve its extension to a future work when the orbital function turns out to be more complex. The essence of the following arguments is not altered. Near T_c when H is applied parallel to the z axis (the a axis) the above is written as

$$f_{\mathrm{grad}} = K_1\left(\frac{d\eta_+}{dx}\right)^2 + \left(\frac{2\pi}{\phi_0}\right)^2 K_2(M + \mu_0H)^2x^2\eta_+^2 \tag{3}$$

because the magnetic induction $\mathbf{B} = (0, 0, \mu_0H + M)$. The minimization of the free energy

$$\frac{2\pi}{\phi_0}\sqrt{K_1K_2}\,|M + \mu_0H_{c2}^a| = \alpha_0(T_c - T) \tag{4}$$

readily yields the upper critical field H_{c2}^a as

$$\mu_0H_{c2}^a = \frac{\phi_0}{2\pi}\frac{\alpha_0}{\sqrt{K_1K_2}}(T_c - T), \tag{5}$$

where the transition temperature is redefined as $T_c - \frac{2\pi}{\phi_0}\frac{\sqrt{K_1K_2}}{\alpha_0}M \to T_c$ and $\alpha_0^+ \to \alpha_0$. When $H \parallel y(\parallel c)$, by a similar way we obtain

$$f_{\mathrm{grad}} = K_2\left(\frac{d\eta_+}{dx}\right)^2 + \left(\frac{2\pi}{\phi_0}\right)^2 K_1\{M^2 + (\mu_0H)^2\}x^2\eta_+^2 \tag{6}$$

which yields

$$\frac{2\pi}{\phi_0}\sqrt{K_1K_2}\sqrt{M^2 + (\mu_0H_{c2}^b)^2} = \alpha_0(T_c - T) \tag{7}$$

or, approximately near T_c,

$$\mu_0H_{c2}^c \sim \sqrt{2M}\sqrt{\mu_0H_{c2}^a}\,. \tag{8}$$

This means that near T_c, H_{c2}^c exhibits a root singularity as a function of T with the slope being infinite. This is also true for $H \parallel x(\parallel b)$, namely,

$$H_{c2}^b \sim \sqrt{\frac{K_2}{K_1}}H_{c2}^c\,, \tag{9}$$

where it shows not only a root singular behavior at T_c, but also is scaled with H_{c2}^c by a constant factor. The large slope in H_{c2}^b is observed in a certain pressure region of UGe_2 by Huxley [19].

In the basal (a, c) plane the angular dependence of $H_{c2}(\theta)$ (θ is the angle from the a axis) is calculated as

$$\left\{1 - \left(\frac{H_{c2}^a}{H_{c2}^c}\right)^2\right\}\cos\theta\,\frac{H_{c2}(\theta)}{H_{c2}^a} + \left(\frac{H_{c2}(\theta)}{H_{c2}^c}\right)^2 = 1 \tag{10}$$

by simple arithmetics. Since the system is polar axis symmetric, namely, the magnetization is of a vectorial nature, $M > 0$ differs from $M < 0$. Thus when the external field is reversed from the $M > 0$ to the $M < 0$ direction, H_{c2}^{-a} behaves differently; namely, provided that the magnetization stays its original orientation under the reversed field which is $H \lesssim 300$ G, judging from the magnetization curve (Fig. 1 in Ref. [9]) H_{c2}^{-a} is given from Eq. (4):

$$\mu_0H_{c2}^{-a} = \frac{\phi_0}{2\pi}\frac{\alpha_0}{\sqrt{K_1K_2}}(T - T_c) \tag{11}$$

for $T > T_c$. In this situation T_c goes up by 30–40 mK. Then on further increasing field as the magnetization is quickly reversed from its original direction and begins decreasing, the H_{c2}^{-a} curve changes into the original H_{c2}^a curve. Thus it is expected that the SC state is reentrant by increasing H under a fixed T just above T_c. This T_c rises and the associated reentrant phenomenon is deeply rooted in the fact that the SC survives under the influence of the ferromagnetic state. Upon controlling FM by external field we can manipulate and raise T_c. Physically the T_c rise occurs because the external field can cancel the spontaneously building internal field due to the ferromagnetic polarization which is to lower the hypothetical transition temperature and the magnetic induction $\mathbf{B}$ can become smaller than the internal field M.

The possible orbital forms in the triplet pairing case allowed in tetragonal symmetry are given by Volovik and Gorkov [20] for the strong spin-orbit coupling case. Since SC appears in the presence of FM, the spin part of the order parameter is limited to the form $\hat{\mathbf{b}} + i\hat{\mathbf{c}}$ ($\hat{\mathbf{a}}$, $\hat{\mathbf{b}}$, and $\hat{\mathbf{c}}$ are unit vectors along the three crystal axes). In the strong spin orbit coupling case all the one-dimensional representations (A_{1u}, A_{2u}, B_{1u}, and B_{2u}) are excluded; only the pairing function $\hat{k}_a(\hat{\mathbf{b}} + i\hat{\mathbf{c}})$ remains as a candidate among the two-dimensional representation E_u in their classification. Since the spin-orbit coupling is not the largest energy scale in the present ferromagnetic superconductor, the weak spin-orbit coupling scheme shown next is a more relevant classification scheme.

In the weak spin-orbit case [21], which was the case for UPt_3 where another exotic superconducting pairing state (either a nonunitary bipolar state similar to the present material or the triplet planar state) is realized [22] there are several possible orbital functions allowed coupled to the spin function $\hat{\mathbf{b}} + i\hat{\mathbf{c}}$, namely, $k_ak_ck_b(k_a^2 - k_c^2)$ (A_{1u}), k_b (A_{2u}), $k_ak_ck_b$ (B_{1u}), and $k_b(k_a^2 - k_c^2)$ (B_{2u}) as the one-dimensional representations with $k_i (i = a, b, c)$ being the unit vector for reciprocal space. As for the two-dimensional representation E_u, the orbital functions of the forms are allowed: $\lambda_1(k)$, $\lambda_1(k) + \lambda_2(k)$, and $\lambda_1(k) + i\lambda_2(k)$ are listed where $\lambda_1(k) = k_a$ and $\lambda_2(k) = k_c$, or $\lambda_1(k) = k_a k_b^2$ and $\lambda_2(k) = k_c k_b^2$. The bipolar-type pairing state $\lambda_1(k)\hat{\mathbf{a}} + i\lambda_2(k)\hat{\mathbf{c}}$ is excluded. If we take literally the orthorhombic crystal symmetry, the strong spin-orbit classification does not give rise to a suitable spin function of $\hat{\mathbf{b}} \pm i\hat{\mathbf{c}}$ among the four one-dimensional representations such as $k_a\hat{\mathbf{a}}$, $k_c\hat{\mathbf{c}}$ (A_{1u}), and $k_a\hat{\mathbf{c}}$, $k_c\hat{\mathbf{a}}$ (B_{1u}) and its linear combinations. On the other hand, in the weak spin-orbit coupling case where there is no two-dimensional representation all one-dimensional presentations have suitable orbital functions attached to the spin part $\hat{\mathbf{b}} + i\hat{\mathbf{c}}$, namely, $k_a k_c k_b$ (A_{1u}), k_b (B_{1u}), k_c (B_{2u}), and k_a (B_{3u}). It is interesting to note that $[\lambda_1(k) + i\lambda_2(k)](\hat{\mathbf{b}} + i\hat{\mathbf{c}})$ has not only the spin angular moment but also the orbital angular moment, both pointing to the ferromagnetic moment direction. The allowed states have line node(s) in general except for $(k_a + ik_c)(\hat{\mathbf{b}} + i\hat{\mathbf{c}})$ in the two-dimensional representation if the up-spin Fermi surface opens along the b axis as predicted by band structure calculation [12].

We can design several interesting experiments associated with the coexistence between FM and SC in the zero-field cooling where FM domains are formed, consisting of $\mathbf{M}$ and $-\mathbf{M}$. Each domain has its own SC order parameters, either $\hat{\mathbf{b}} + i\hat{\mathbf{c}}$ or $\hat{\mathbf{b}} - i\hat{\mathbf{c}}$, in order to fit its spin angular moment with the ferromagnetic moment direction. When one of the two domains, say, $\mathbf{M}$ with $\hat{\mathbf{b}} + i\hat{\mathbf{c}}$, percolates throughout a whole system, the macroscopic superconducting state is established. In contrast, under the field cooling process whose field strength is strong enough to make the whole system the single domain, say, $\mathbf{M}$, the SC takes place immediately right at T_c.

This reentrant phenomenon mentioned before provides direct evidence for the coexistence. The reentrant behavior is indeed observed at 1.35 GPa at a much higher field region ($\sim$ a few T) and may be different in origin from our prediction. However, it should be kept in mind that the superconducting properties in UGe_2 show a strong sensitivity to the measured current density [11]. It might be related to the domain formation associated with FM. Toward the critical pressure $P = 1.7$ GPa the FM is greatly suppressed. There the two long-range orders FM and SC truly compete with each other while around the optimal pressure $P \sim 1.2$ GPa T_{FM} is much higher than T_c. Thus the relationship between FM and SC is changing upon increasing P. As a consequence it could be possible to change the pairing symmetry from the nonunitary to the unitary state. This is one of the possible explanations of the observed reentrant phenomenon at $P = 1.35$ GPa.

We also remark that along the FM domain boundaries or the Bloch walls where the magnetization direction rotates so as to bridge the two oppositely polarized states $\pm\mathbf{M}$, the spontaneously induced spin current flows because when transforming the $\hat{\mathbf{b}} + i\hat{\mathbf{c}}$ state into $\hat{\mathbf{b}} - i\hat{\mathbf{c}}$ the SC phase also continuously changes.

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Note added.—Shick and Pickett study the same problem [23].

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