Theory of spin-polarized superconductors – an analogue of superfluid $^3$He A-phase—

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It is shown theoretically that ferromagnetic superconductors, UGe$_2$, URhGe, and UCoGe can be described in terms of the A-phase like triplet pairing similar to superfluid $^3$He in a unified way, including peculiar reentrant, S-shape, or L-shape $H_c2$ curves. The associated double transition inevitable between the A$_1$ and A$_2$-phases in the H-T plane is predicted, both of which are characterized by non-unitary state with broken time reversal symmetry and the half-gap. UTe$_2$, which has been discovered quite recently to be a spin-polarized superconductor, is analyzed successively in the same view point, pointing out that the expected A$_1$-A$_2$ transition is indeed emerging experimentally. Thus the four heavy Fermion compounds all together are entitled to be topologically rich solid state materials worth further investigating together with superfluid $^3$He A-phase.

Much attention has been focused on ferromagnetic superconductors, such as UGe$_2$, URhGe, and UCoGe. Recently a new member of a superconductor UTe$_2$ with $T_c$=1.6K is discovered, which is almost ferromagnetic, and attracts much excitement. Many researchers start devoting to UTe$_2$ experimentally and theoretically and show renewed interest on the former three compounds too. Those heavy Fermion materials belong to a strongly correlated system where 5f electrons responsible for it is believed to form a coherent narrow band with the large mass enhancement below a Kondo temperature. Since $H_c2$ in those compounds exceeds the Pauli paramagnetic limitation, it is thought that a triplet or odd parity pairing state is realized. However, the detailed studies of the pairing symmetry remains lacking in spite of the long history of the first three compounds over two decades. Some of those features are, interesting enough, common for spin-polarized flat band superconductivity found in double bilayer magic angle graphene, where the in-plane $H_c2$ shows a similar “reentrance” behavior, as will see shortly, due to linear Zeeman effect in the superconducting (SC) state, appearing next to a spin-polarized insulating state.

To understand those four spin-polarized superconductors in a unified way, here we develop a phenomenological theory based on the assumption that the four compounds are commonly described in terms of the triplet pairing symmetry analogous to the superfluid $^3$He-A phase under Zeeman effect. Namely the A$_1$-A$_2$ phase transition is induced by an applied field, which is observed as the clear double specific heat jumps.

There exist prominent SC properties observed commonly in those superconductors:

(1) Above $T_c$ the ferromagnetic transition (FM) occurs in the first three compounds. Thus the SC state survives under a strong internal field coming through an exchange interaction. However, in UTe$_2$ “static” FM is not detected so far although FM fluctuations are probed above $T_c$, i.e. a diverging static susceptibility along the $a$-axis, and $1/T_c$. When $H$ is applied parallel to the magnetic hard axis $b$ in orthorhomic crystals, $H_c2$ exhibits the reentrant behavior for URhGe where the SC state once disappeared reappears again at higher fields, or an S-shape $H_c2$ in UCoGe and a L-shape in UTe$_2$ in the H-T plane.

(2) When $H$ is applied parallel to the magnetic hard axis $b$ in orthorhomic crystals, $H_c2$ exhibits the reentrant behavior for URhGe where the SC state once disappeared reappears again at higher fields, or an S-shape $H_c2$ in UCoGe and a L-shape in UTe$_2$ in the H-T plane.

(3) The gap structure is unconventional characterized by either point in UTe$_2$ or line nodes in the other three. A similar indication for the double transitions in the ambient pressure is found in UCoGe at $T_c$~0.2K where $1/T_cT$ exhibits a plateau corresponding to the “half residual DOS” value at the intermediate $T$ below $T_c$=0.5K. Upon further lowering $T$ it starts decreasing again at 0.2K. In UTe$_2$ under ambient pressure a quite similar $1/T_cT$ is also observed in NMR experiment. The saturated $1/T_cT = 0.25$ normalized by the normal value, which corresponds to the “half residual DOS”, restarts decreasing to zero at the second transition at $T_{c2}$=0.2K. This value nicely matches with the missing second phase transition in the $P$-$T$ plane, namely, a smooth extrapolation of $T_{c1}$ and $T_{c2}$ toward $P$→0.

(4) Recent specific heat $C/T$ data for several high quality samples of UTe$_2$ commonly show the residual DOS amounting to 0.5N(0), a half of the normal DOS N(0) while some claims zero. Thus this “residual” half DOS issue is controversial at this moment. We will propose to resolve it later in this paper.

In order to address those seemingly “complicated and mutually conflicting”, but quite intriguing experimental facts, we postulate the A phase like triplet pair symmetry which responds to the spontaneous FM moment under perpendicular external fields to yield the A$_1$-A$_2$ double transitions. This scenario coherently explains the observed reentrant $H_c2$ in URhGe, an S-shape in UCoGe, and L-shape in UTe$_2$ in a unified way.

As mentioned, the A$_1$-A$_2$ phase transition in $^3$He A-phase is controlled by the linear Zeeman effect due to applied field, which acts to split $T_{c2}$. Thus in the FM superconductors $T_c$ is controlled by spontaneous magnetic moment of ferromagnetism, which is linearly coupled to the non-unitary order parameter. We apply a Ginzburg-Landau theory to describe those characteristic $H_c2$ curves. We also identify the pairing symmetry
based on group theoretic classification\cite{Note1}. The pairing symmetry is non-unitary triplet\cite{Note1,Note2}, where the \textit{d}-vector is a complex function and points to the perpendicular direction to the magnetic easy axis and the gap function in the normal state \( F \) symmetry is non-unitary triplet written as \( \langle \eta^+ | \eta^- \rangle = 0 \). This pairing, namely invariance under the \( \sqrt{2} \times \sqrt{2} \) vector. \( \sqrt{2} \times \sqrt{2} \) vectors). We assume the weak spin-orbit coupling scheme whose strength depends on the compounds and will be appropriately tuned relative to the experimental situations. There exists \((1) \times 2\) symmetry in this pairing, namely invariance under \( \eta \rightarrow -\eta \) and the gauge transformations.

Under \( D_{2h} \) symmetry GL free energy functional is given by

\[
F = \alpha_0 (T - T_c) \eta^+ \cdot \eta^- + \frac{\beta_1}{2} (\eta^+ \cdot \eta^-)^2 + \frac{\beta_2}{2} |\eta|^4 + i \kappa \vec{M} \cdot \eta^+ \times \eta^-.
\]

The last invariant \( (\kappa > 0) \) comes from the non-unitarity of the pairing function in the presence of the spontaneous moment \( \vec{M}(H) \), which responds to external field directions differently. Since the fourth order term are written as \( F^{(4)} = \frac{\beta_1}{2} (\eta^+ \cdot \eta^- + \eta^+ \cdot \eta^+)^2 + \frac{\beta_2}{2} |(\eta^+ \cdot \eta^- - \eta^+ \cdot \eta^-)|^2 \), for \( \beta_1, \beta_2 > 0 \), we can find the minimum when \( |\eta^+| = |\eta^-| \) and \( \eta^+ \mp \eta^- \). Note that the weak coupling estimate leads to \( \frac{\partial F^{(4)}}{\partial \eta} = -2 \), thus we assume the strong coupling effects in the following arguments. The quasi-particle spectra are given by \( E_{k,\sigma} = \sqrt{\epsilon(k)^2 + (|\eta|^2 + |\eta|^2)} \phi(k)^2 \). If we choose \( \eta^+ = \eta_0 \beta \) and \( \eta^- = \eta_0 \beta \), those are rewritten as \( E_{k,\sigma} = \sqrt{\epsilon(k)^2 + \Delta_\sigma(k)^2} \) where the gap functions for two branches are \( \Delta_+ (k) = |\eta_0 + \eta_0 \phi(k)| \) and \( \Delta_-(k) = |\eta_0 - \eta_0 \phi(k)| \). Note that if \( |\eta_0| = 0 \), \( \Delta_+ (k) = \Delta_-(k) \), which is the A phase. \( T_c \) (2) When \( |\eta_0| = |\eta_0| \), \( \Delta_+(k) \neq 0 \) and \( \Delta_-(k) = 0 \) which is the non-unitary A 1 phase. The gap of one of the two branches identically vanishes and remains normal. Therefore, if we naively assume that in the normal state \( N_T(0) = N_T(0) \) which is consistent with the NMR data in \( UTe_2 \), the A 1 phase is characterized by having the ungapped DOS \( N_T(0) = N_T(0)/2 \) with \( N(0) = N_T(0) + N_T(0) \). In the non-unitary state with the complex \( d - \)vector, the time reversal symmetry is broken.

It is convenient to consider \( \eta^+ = 0, \eta_0, \eta_\sigma \) or \( \eta_\sigma = \frac{1}{\sqrt{2}} (\eta_\pm + i \eta_\mp) \) for \( M = \langle M, 0, 0 \rangle \). Then we see from eq. (2), the quadratic term \( F^{(2)} \) becomes

\[
F^{(2)} = \alpha_0 (T - T_{c+}) |\eta|^2 + \alpha_0 (T - T_{c-}) |\eta^-|^2
\] with \( T_{c+} = T_c \pm \frac{\beta_1}{\beta_2} M_a \). The actual second transition temperature is modified to \( T_{c+} = T_c - \frac{\beta_1}{\beta_2} M_a \), which could be larger or smaller than the original \( T_{c+} \equiv T_c - \frac{\beta_1}{\beta_2} M_a \) due to the fourth order terms. For \( 1 \leq \beta_1/\beta_2 \leq 3, T_{c+} > T_{c-} \). This remark becomes important to understand the asymmetric L-shape \( H_c^b \) observed in \( UTe_2 \) as see later. It is easy to see the ratio of the specific heat jumps \( \frac{T_{c+}^{(0)} - T_{c-}^{(0)}}{T_{c-}^{(0)} - T_{c+}^{(0)}} \). The jump at \( T_{c+} \) can be quite small for \( T_{c+} \gg T_{c-} \) which is the case for \( UTe_2 \) since \( T_{c+} = 1.6K, T_{c+} = 0.2K \), and the second factor is an order of one, anticipating the difficulty to observe the second transition thermodynamically.

The FM moment \( M_a \) acts to shift the original transition temperature \( T_{c0} \) and split it into \( T_{c+} \) and \( T_{c-} \). The external field \( H \) comes in through \( M_a(H) \) in addition to usual vector potential which gives rise to orbital depairing. The magnetic coupling is estimated\cite{Note1} by \( \kappa = T_{c+}^{(0)}(0)/\sqrt{N(0)} \) \( N(0) \) is energy derivative of the normal DOS and \( \omega \) energy cutoff. This term arises from the electron-hole asymmetry near the Fermi level. \( \kappa \) indicates the degree of this asymmetry, which can be substantial for a narrow band, thus the Kondo coherent band in heavy Fermion material of our cases is expected to be important. We can estimate \( N(0)/\sqrt{N(0)} \sim 1/EF \) with the Fermi energy \( E_F \). Since \( T_{c+} = 2mK \) and \( E_F = 1K \) in \( ^3\text{He} \), \( \kappa \sim 10^{-3} \), while the present compounds \( T_{c+} \sim 1K \) and \( E_F \sim T_K \) with the \( T_K \) Kondo temperature typically \( 10-50K \), thus \( \kappa \) is an order of \( 10^{-1} \).

Let us now consider the action of external field \( H_B \) applied to the hard axis \( b \) on the FM moment \( M_a \), pointing parallel to the \( a \)-axis. The \( a \)-axis component of the moment \( M_a(H_B) \) generally decreases by rotating it toward the \( b \)-axis as shown in Fig. 1(b). In fact it is actually observed in \( URhGe \) by neutron experiment.\cite{Note3} Here we display the generic and typical magnetization curves of \( M_a \) and \( M_b \) in Fig. 1(c) where \( H_R \) denotes a characteristic field for \( M_b(H_B) = M_a \). Experimentally it is realized by a meta-magnetic transition via a first order transition in \( UTe_2 \) or gradual change in \( UCoGe \).

Thus as displayed in Fig. 1(a), \( T_{c+} (T_{c-}) \) by increasing \( H_B \) decreases (increases) according to eq. (2). The two transitions \( T_{c+} = T_{c-} \) meet at \( H_R \). Upon further increasing \( H_B, T_{c-} \) could keep increasing further by rotating the \( d \)-vector direction so that \( d \) is now perpendicular to \( M_b \), which maximally gains the magnetic coupling energy \( i \kappa \vec{M} \cdot \eta \times \eta^\prime \). This process occurs gradually or suddenly, depending on the situations of the magnetic subsystem and also on the spin-orbit coupling which locks \( d \) to underlying lattices. Therefore \( H_R \) may indicate simultaneously the \( d \)-vector rotation. It should be noted, however, that if the spin-orbit coupling is so strong, the \( d \)-vector rotation is prevented. In this case \( H_B^l \) exhibits a Pauli limited behavior as observed in \( UTe_2 \) under pressure\cite{Note1}.

Within the GL scheme it is easy to estimate \( H_B^l \) as follows. We start with the \( H_B^l \) expression: \( H_B^l(T) = A_0 (H_c(T_c - T)) \) with \( A_0 = \frac{\phi_0}{2\pi \kappa M_a \alpha_0}, m \) effective mass,
and $\Phi_0$ quantum unit flux. Here $T_c$ depends on $H$ though $M_a(H)$ as described above. Thus the initial slope of $H'_{c2}$ at $T_c$ is simply given by $H'_{c2}(T) = A_0 \frac{dT_c}{dH_{c2}} - A_b$. It is seen that if $\frac{dT_c}{dH_{c2}} = 0$ for the ordinary superconductors, $H'_{c2}(T) = -A_0 < 0$. The slope $H'_{c2}(T)$ is always negative. However, it is easily written as

$$\frac{1}{|H'_{c2}|} = \frac{1}{|H'_{c2}^0|} + \frac{dT_c}{dH_{c2}}.$$

The condition for attaining the positive slope, $H'_{c2}(T) > 0$ implies $|H'_{c2}^0| > |(dH_{c2}/dT_c)|$. This is a necessary condition to achieve an S-shaped or L-shaped $H_{c2}$ curves. This is fulfilled when $|H'_{c2}^0|$ is large enough, that is, the orbital depairing is small or $\frac{dT_c}{dH_{c2}}$ at $H_{c2}$ is large, or the $T_c$ rise is strong enough. It is to be noted that when $1 - A_0(\frac{dT_c}{dH_{c2}}) = 0$, the $H_{c2}(T)$ curve has a divergent part, which is indeed observed in UCoGe as a part of the S-shape. It is clear from the above that when $dT_c/dH < 0$, $|H'_{c2}(T)| < |H'_{c2}^0|$. Namely, in this case the slope $|H'_{c2}|$ is always smaller than the original $|H'_{c2}^0|$ as expected.

When the magnetic field is applied to the magnetic easy $a$-axis, the spontaneous moment $M_a(H_a)$ increases monotonically as shown in Fig.1(c). According to eq. (2), $T_{c\parallel}$ ($T_{c\perp}$) increases (decreases) as $H_a$ increases. Thus $H'_{c2}$ can have a positive slope at $T_{c\parallel}$ in principle. However, the existing data in UCoGe[22] show that it is negative and there is no report for it in other compounds. This is simply because the orbital depairing $H'_{c2}^0$ overcomes the positive rise of $T_{c\parallel}$. Moreover, $H'_{c2}$ is strongly suppressed by both $T_{c\perp}$ and the orbital effect $H'_{c2}^0$, resulting in quite a low $H'_{c2}$, compared with $H'_{c2}^0$. This large $H_{c2}$ anisotropy is common in those compounds[3]. From the above calculation, it is apparent that the large $H'_{c2}$ comes because the higher field part of $H_{c2}$ belongs to $H_{c2}^{\text{upper}}$, which has a positive slope.

There could be several types of the $H'_{c2}$ curves depending on the magnitude of the spontaneous moment $M_a$, and its growth rate against $H_b$. The coupling constant $\kappa$, $T_{c0}$, and etc. Possible representative $H'_{c2}$ curves are displayed in Fig. 2.

When the hypothetical $T_{c0}$ is situated in the negative temperature side, the realized phase is the $A_{12}$-phase only at $H_b = 0$. In the high field regions, $T_{c\parallel}$ ($H_b$) increases by increasing $M_a(H_b)$ as the $A_{2}$-phase, which is shown in Fig. 2(a). This is the situation of URhGe where the reentrant phase appears around the magnetic rotation field $H_{c2}$. We also note that under uniaxial stresses along the $b$-axis $H'_{c2}$ curve continuously deforms and two separated SC regions merge into an S-shaped $H_{c2}^{\text{upper}}$ in URhGe (see Fig. 2(a) in Ref. [33]). This can be understood because under stresses $H_R$ is known to decrease, thus the detached high field part approaches and eventually touches the lower part to form an S-shape $H_{c2}^{\text{upper}}$, resulting in a positive slope $(dH_{c2}^{\text{upper}}/dT_c) > 0$.

The second example shown in Fig. 2(b) is the case where $M_a$ and $\kappa$ are relatively small, thus the splitting $T_{c\parallel}$ and $T_{c\perp}$ at $H_b = 0$ is small. The small moment $M_a = 0.09\mu_B$ and a rapid growth of $M_b$ in UCoGe[33] are consistent with this picture. The resulting $H_{c2}^{\text{upper}}$ curve possess an S shape, similar to that observed in UCoGe. Note that there must always exist the “absolute” upper limit of $H_{c2}$ for any superconductors. This is set by the orbital depairing where $H_{c2}^{\text{upper}} = \phi_0/2\pi\xi^2$ at $T=0$ with $\xi$ coherent length. This absolute upper limit $H_{c2}^{\text{upper}}$ has a triangle region in the $H$ and $T$ plane which is denoted by a dotted line in Fig. 2. The SC state is allowed only inside this $H_{c2}^{\text{upper}}$ curve. If the rising $T_{c\parallel}(H_b)$ hits this line, the $H_{c2}$ curve follows this limiting curve.

In Fig. 2 (c), we show the $H_{c2}^{\text{upper}}$ curve similar to UTe$_2$ with an L shaped[4]. It occurs when $H_R$ situates at lower field and $T_{c0}$ is positive and low. All three phases $A_1$, $A_2$ and $A$ are realized for $H \parallel b$-axis. The observed $H_{c2}^{b}$ curve in UTe$_2$ is different from Fig.2 (c) in that the upper part of the A$_2$-phase is horizontally cut out at $H_{c2}^{b} \sim 40$
T because beyond that field the first order meta-magnetic transition completely alters the background electronic state, so that the SC state is abruptly wiped out. However, note that the field direction is appropriately chosen away from the b-axis, $H_{c2}$ is greatly enhanced by avoiding the meta-magnetic transition at $H \parallel b$, reaching $\sim 60T$
. This means that the intrinsic absolute upper limit of the orbital $H_{c2}^{\text{upper}}$ can reach remarkably $\sim 60T$ in this system. Thus it was quite reasonable to ignore the orbital depairing effect in the above $H_{c2}$ arguments. Therefore, it is possible to reproduce the essential features associated with $H_{c2}$ for all compounds in terms of the A$_1$-A$_2$ transitions. To confirm our scenario, we now examine possible signatures of the double transition. The recent NMR experiment\textsuperscript{12} on UT$_2$ clearly demonstrates it at $T_{c1} \sim 0.2K$. Namely, upon lowering $T$, $1/T_1T$ which is proportional to square of DOS exhibits a sudden drop at $\sim 0.2K$ after a prolonged $T$-constant plateau at 0.25 normalized by its normal value corresponding to $N(0)/2$. This behavior is backed up by the simultaneous Knight shift measurement\textsuperscript{12}. $T_{c1} = 0.2K$ moves up from $T_c(0)$ which is expected to situate at a negative $T$ as shown in Fig. 2(c) because of the fourth order terms in eq. (1) mentioned before.

Under $P$ in UT$_2$ the successive double transitions are indeed discovered\textsuperscript{13} and vary systematically as explained in the $P$-$T$ phase diagram. The missing second transition at ambient $P$ is now found mentioned above.

As for UCoGe, thermal conductivity experiment\textsuperscript{14} indicates an anomaly at $T \sim 0.2K$, which coincides roughly with our prediction shown in Fig. 2(b). As a function of $H \parallel b$ the thermal conductivity anomaly is detected as a sudden increase at $H \sim 0.6H_{c2}$ (see Fig. 5 in Ref. \textsuperscript{38}). Moreover, under $H$ parallel to the easy axis $H_{c2}$ curve shows low $T$ enhancement indicative of the underlaying phase transition (see Fig. 2(b) in Ref. \textsuperscript{35}). According to NMR by Manago et al\textsuperscript{22}, $1/TT_1$ shows very similar behaviors: plateau at $N(0)/2$ and sudden drop mentioned above. We expect further careful experiments to detect the A$_1$-A$_2$ transitions in all four compounds.

While for UGe$_2$, URhGe, and UCoGe “static” FM transitions are established, in UT$_2$ slow FM fluctuations are found\textsuperscript{26,10,11}, which could be the origin of the symmetry breaking of $T_{c\uparrow} \neq T_{c\downarrow}$ under the assumption that FM fluctuations are slow enough compared with the conduction electron motion. We recall a similar circumstance in UT$_3$: The fluctuating antiferromagnetism at $T_N = 5K$ is detected only by the fast probe: neutron diffraction\textsuperscript{37,38} and undetected by other “static” probes, such as specific heat, $\mu$SR, and NMR. Yet, this is believed to be the origin of the double transition in UT$_3$\textsuperscript{39,40}.

In summary, we have discussed the SC properties of UGe$_2$, URhGe, UCoGe, and UT$_2$ in depth in terms of a non-unitary triplet pairing state in a unified way. The FM moment governs and produces the various types of the $H_{c2}$ curves observed. The possible pairing function is described by the complex $\mathbf{d}$-vector whose direction is perpendicular to the magnetic easy axis. The orbital part $\phi(k)$ of the order parameter could be line nodes allowed group-theoretically\textsuperscript{47} in the present orthorhombic symmetry, which is experimentally suggested in UCoGe\textsuperscript{43,44}.

As for UT$_2$, the specific heat experiment\textsuperscript{25,38,45} exhibit $C/T \sim T^2$, suggesting the gap structure with point nodes, which is also consistent with microwave measurement\textsuperscript{13}. Thus if this is true, the pairing function of UT$_2$ is symbolically given by $(b + ic)(k_b + i{k_c})$, a solid state analogous literally to superfluid $^3$He A$_1$-phases\textsuperscript{41}, which is double chiral both in the spin and orbital parts with points nodes. This chiral $p$-wave form in a simplest, but somewhat “ad hoc” possibility is consistent with the observed chirality by STX\textsuperscript{42}. The orbital angular moment $\mathbf{L}$ spontaneously induced by this chiral state can gain the extra energy through the coupling $\mathbf{M}$-$\mathbf{L}$ with the spontaneous magnetic moment. We can explore a variety of interesting topological properties, such as Weyl nodes associated with point nodes, known in $^3$He A-phase\textsuperscript{41}, which was difficult to access experimentally.

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