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Phenomenological theory in reentrant uranium-based superconductors

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We develop a phenomenological theory for a family of uranium-based heavy fermion superconductors (URhGe, UCoGe, and UTe_2). The theory unifies the understanding of both superconductivity (SC) with a weak magnetic field and reentrant superconductivity (RSC) that appears near the first-order transition line with a high magnetic field. It is shown that the magnetizations along the easy and hard axes have opposite effects on SC. The RSC is induced by the fluctuation parallel to the direction of the magnetic field. The theory makes specific predictions about the variation of triplet SC order parameters $\vec{d}$ with applied external magnetic fields and the existence of a metastable state for the appearance of the RSC.

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Heavy-fermion superconductors UCoGe and URhGe are promising spin-triplet superconductors. The spin-triplet pairing is supported by their highly anisotropic upper critical fields which greatly exceed the Pauli limit along all three crystallographic directions [1–6], and the coexistence of ferromagnetism (FM) and superconductivity (SC) [7–10].

Very recently, another uranium-based superconductor (UBS) UTe_2 has been found. Considerable research has been conducted, such as on large residual Sommerfeld coefficients [11,12], the coexistence of ferromagnetic fluctuations and superconductivity [13,14], field-boosted superconductivity [15,16], chiral superconducting states [17], quasi-two-dimensional Fermi surfaces [18], and so on. The new superconductor shares many common features with previous counterparts, such as highly anisotropic upper critical fields and reentrant superconductivity (RSC) under high magnetic fields. However, unlike the previous ones, there is no sign of FM order in UTe_2 down to 25 mK [13,14]. In all these superconductors, the SC transition temperature T_c is first suppressed by the magnetic field (h_y) perpendicular to both the hardest (x) and easy axes (z). But when the magnetic field is strong enough, the T_c arises again [3,15,16,19–22].

The difference between these superconductors brings new challenges and calls for a unified understanding. On the basis of Landau phenomenological theory and weak-coupling theory for URhGe given by Mineev [23,24], the jump of the magnetic moment m_{z0} enhances the fluctuations along the easy axis to induce the RSC. This mechanism cannot be applied to understand the RSC in UTe_2 [15,16] because UTe_2 has no magnetic order along the easy axis [6,14–16]. The increase of the fluctuation along the easy axis cannot be the only cause of the RSC. Experimentally, it has also been found that both longitudinal (along the easy axis) and transverse (along the magnetic field) fluctuations exist near the RSC region in $URh_{0.9}Co_{0.1}Ge$ by ^{59}Co nuclear magnetic resonance (NMR) measurements [25].

Herein, we generalize the phenomenological theory of the spin fluctuation feedback effect (SFFE) proposed by Amin *et al.* [26] to explain the physics in the family of UBS. We show that the decrease of T_c in a weak magnetic field and the appearance (disappearance) of the RSC near the first-order transition in URhGe, UCoGe, and UTe_2 can be understood in a unified manner. In the weak magnetic field region, T_c decreases with the decrease of static magnetic order along the easy axis and the increase of magnetic moment along the field directions. In the strong-field region, the RSC is caused by fluctuations along magnetic field directions. However, RSC can be killed by destroying the metastable state near a first-order transition and a sudden increase of magnetic moment along the field directions. Our theory further predicts the $\vec{d}$ vector of the RSC in these superconductors and a metastable RSC state during the magnetic hysteresis loop, providing a sound theoretical basis for further investigation of the RSC in microscopic theory.

Ferromagnetic SC. We first focus on the SC and RSC in FM UBS, and take URhGe as an example. The phase diagram is sketched in Fig. 1. With weak magnetic fields, the SC coexists with FM, and as the spin-orbital coupling is strong, the symmetry is described by the magnetic group $D_{2h}(E, C_{2z}, I, \sigma_{xy})$ [27]. The spin-triplet SC order parameter, the $\vec{d}$ vector, is expanded in the basis of the A_u or B_u antisymmetric corepresentations of this magnetic group. For both A_u and B_u, the free energy of the magnetic ($\vec{m}$) and magnetism-SC coupling parts are the invariants [28,29] of the magnetic group [see Supplemental Material (SM) Sec. I [30]],

$$f_{\text{sc-}m} = A_{1i}(m_i)^2 + B_{ij}(m_i)^2(m_j)^2 - h_y m_y + K_{1ij}(m_i)^2|d_j|^2 + K_{2z}m_z(i\vec{d} \times \vec{d}^*)_z, \quad (1)$$

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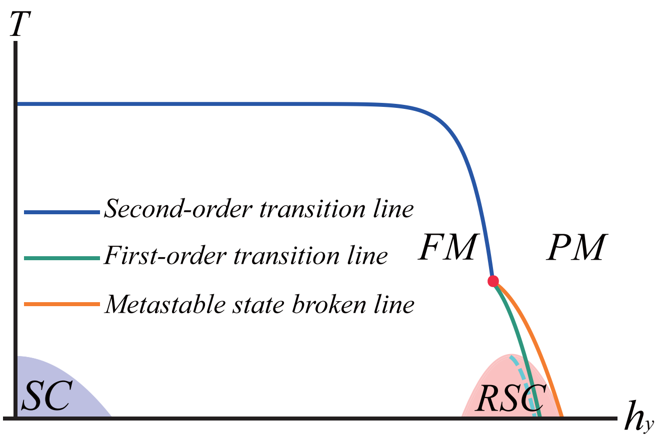

FIG. 1. The sketched phase diagram of URhGe. The dashed cyan line indicates the proposed magnetic-hysteresis-loop type of behavior for the upper critical magnetic field for RSC.

where $i, j = x, y, z$ and the repeated subscripts indicate a summation throughout this Rapid Communication. Except for $A_{1z} < 0$, other A_{1i}, B_{ij}, K_{1ij}'s are positive or positive definite to ensure the FM ground state. Positive K_{2i}'s are the amplitudes of the couplings between the FM and SC order parameters [26].

In the weak magnetic field region, the relevant magnetic part of the free energy can be simplified as $f_{m,1} \approx A_{1z}m_z^2 + B_z m_z^4 + A_{1y}m_y^2 - h_y m_y$. The minimization gives the magnetic moment $\vec{m}_0 = (0, \frac{h_y}{2A_{1y}}, \sqrt{\frac{-A_{1z}}{2B_z}})$. We integrate out the magnetic fluctuation $\delta\vec{m}$ ($\vec{m} = \vec{m}_0 + \delta\vec{m}$) in $f_{\text{sc-}m}$ to get the effective SC free energy,

$$f_{\text{sc}} = \alpha_i'|d_i|^2 + K_{2z}m_{z0}p_z + \beta_{1ij}|d_i|^2|d_j|^2 + \beta_{2z}p_z^2 + \beta_{3iz} \cdot p_z|d_i|^2, \tag{2}$$

where $\vec{p} = i\vec{d} \times (\vec{d})^*$ and $\alpha_i' = \alpha_i + K_{1zi}m_{z0}^2 + \frac{K_{1zi}}{8B_z m_{z0}^2} + \frac{K_{1yi}}{2A_{1y}} + K_{1yi}m_{y0}^2$, with α_i being the bare quadratic coefficient of SC without SFFE. The positive or positive-definite quartic coefficients β's are renormalized from the SFFE as listed in Sec. II of the SM [30]. Here, different components d_i are not degenerate.

To track the evolution of T_c under varying magnetic field, we rescale the $\vec{d}$ vector with $\alpha_i'|d_i|^2 = \alpha'|d^I|^2$ (see SM Sec. II [30]), where α' is the α_i' corresponding to the highest T_c. By minimizing the free energy, we obtain the nonunitary SC with a rescaled order parameter $\vec{d}^I = \frac{d_0}{\sqrt{2}}(\vec{r})(1, -i, 0)$ (see SM Sec. III [30]). This SC state has an intrinsic z-polarized magnetic moment proportional to $\vec{p}^I = -d_0^2(\vec{r})\hat{z}$ [28] and $T_c = \Delta T_c + T_{c0}$, with

$$\Delta T_c = -\frac{K_{1yi}m_{y0}^2 + K_{1zi}m_{z0}^2 - K_{2z}'m_{z0}}{\alpha_0} - \frac{K_{1zi}}{8B_z m_{z0}^2\alpha_0}, \tag{3}$$

where $\alpha_i = \alpha_0(T - T_{c0})$ with $\alpha_i' = \alpha'$ and $K_{2z}' = \frac{\sqrt{\alpha_x'\alpha_y'}}{\alpha'}K_{2z}$ (here, the weak temperature dependence of the K_{2z}' can be ignored, as discussed in SM Sec. II [30]). For a weak FM superconductor [24,31], we can assume that $m_{z0}^2 < \frac{1}{2\sqrt{2B_z}}$. In this case, it can be seen from Eq. (3) that either a decrease of m_{z0} or an increase of m_{y0} results in the decrease of T_c. Namely, T_c decreases with increasing magnetic field h_y, corresponding to the SC phase of URhGe as shown in Fig. 1. By the way, from Eq. (3), we can see that the K_{1yi} and K_{1zi} coupling terms dominate when the magnetic field along the y axis is weak, at least in URhGe, since the $K_{2z}'m_{z0}$ terms could not lead to the disappearance of the T_c.

Now we consider the strong magnetic field region to discuss the rotation of the $\vec{d}$ vector and appearance of the RSC close to the magnetic first-order transition. When the magnetic field is strong enough, the symmetry of URhGe and UCoGe is described by the magnetic group $D_{2h}(E, C_{2y}, I, \sigma_{xz})$. We obtain (see SM Sec. I [30])

$$f_{\text{sc-}m} = A_{1i}(m_i)^2 + B_{ij}(m_i)^2(m_j)^2 - h_y m_y + K_{1ij}(m_i)^2|d_j|^2 + K_{2y}m_y(i\vec{d} \times \vec{d}^*)_y - \lambda h_y p_y. \tag{4}$$

On the low-field side of the first-order transition, the y component of the magnetic moment enters the free energy as $f_{m,2} \approx f_{m,1} + B_{yz}m_y^2 m_z^2$, giving the magnetic moments as $m_{z0}^2 = -\frac{A_{1z}+B_{yz}m_{y0}^2}{2B_z}$ and $m_{y0} = \frac{h_y}{2(A_{1y}+B_{yz}m_{z0}^2)}$. Following the same procedure, integrating out magnetic fluctuations and rescaling the $\vec{d}$ vector, we obtain the effective free energy,

$$f_{\text{sc}}' \approx \alpha'\left|\vec{d}^I\right|^2 + K_{2y}'m_{y0}p_y^I - \lambda' h_y p_y^I + \text{HO}, \tag{5}$$

where again $\alpha' = \alpha_i'$ and the high-order (HO) terms are not specified. The coupling K_{2y}' makes the SC with $\vec{d}^I = \frac{d_0(\vec{r})}{\sqrt{2}}(1, 0, \pm i)$ and $\vec{p}^I = \mp d_0(\vec{r})^2\hat{y}$ at the highest T_c. Close to the first-order transition critical magnetic field h_m, before the sudden jump of m_{z0}, the variation of m_{z0} is small, so ΔT_c tuned by the magnetic field can be approximated as

$$\Delta T_c = -\frac{1}{\alpha_0}\left(\frac{K_{1yi}}{4A_{1y}'^2}h_y^2 - \left|\frac{K_{2y}'}{2A_{1y}'} - \lambda'\right|h_y\right), \tag{6}$$

where $A_{1y}' = A_{1y} + B_{yz}m_{z0}^2$, $K_{2y}' = \frac{\sqrt{\alpha_x'\alpha_z'}}{\alpha'}K_{2y}$, and $\lambda' = \frac{\sqrt{\alpha_x'\alpha_z'}}{\alpha'}\lambda$. The phase diagram in the strong magnetic field region can be explained if we assume $h_m < h_q \equiv \frac{|K_{2y}'A_{1y}' - 2\lambda' A_{1y}'^2|}{K_{1yi}}$. In this case, from Eq. (6), the T_c increases with increasing magnetic field ($h_y > 0$) at first and then decreases when the magnetic field $h_y > h_q$. However, it is noteworthy that Eq. (6) is valid only for the region which is on the left-hand side of the first-order transition line and close to it. From Eq. (6), we know that the key coupling terms which lead to the appearance of the reentrant superconductivity under a strong magnetic field are K_{2y} and λ terms. When the magnetic field continues to increase and exceeds the critical value h_m, as will be analyzed next, the RSC disappears with increasing magnetic field.

The first-order transition and the disappearance of RSC close to it can be further understood within our theory. With a strong magnetic field and a small m_{z0} in the FM phase, the free energy to describe the first-order transition can be derived

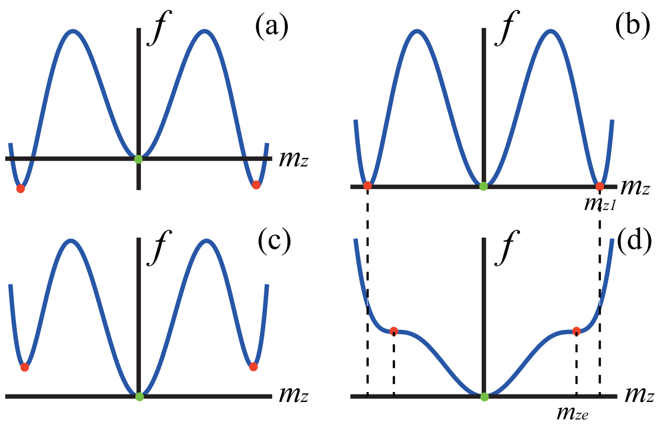

FIG. 2. The f_m-m_z relations from Eq. (7) with $B'_z < 0$, $A'_{1z} > 0$, and $C'_z > 0$ for gradually increasing h_y: (a) FM state with $h_y = 0$. (b) First-order transition point $m_{z0} = m_{z1}$. (c) A metastable state $m_{ze} < m_{z0} < m_{z1}$. (d) The local minima broken state $m_{z0} = m_{ze}$.

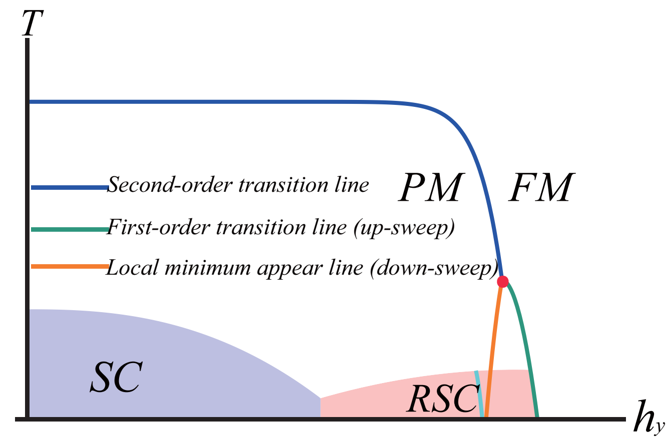

FIG. 3. The sketched phase diagram of UTe_2. The solid cyan line indicates the magnetic-hysteresis-loop type of behavior of the upper critical field for RSC.

[23,24],

$$f_m = -\frac{h_y^2}{4A_{1y}} + A'_{1z}m_z^2 + B'_z m_z^4 + C'_z m_z^6, \tag{7}$$

where $A'_{1z} = A_{1z} + \frac{B_{yz}h_y}{4A_{1y}^2}$, $B'_z = B_z - \frac{B_{yz}^2 h_y^2}{4A_{1y}^3}$, and $C'_z = C_z + \frac{B_{yz}^3 h_y^2}{4A_{1y}^4}$. So one can learn from Eq. (7) that the magnetic field h_y modifies the coefficients of the free energy f_m. Thus the magnetic moment dependence of the free energy changes with increasing magnetic field as shown in Fig. 2. From Eq. (7), we can derive (see SM Sec. IV [30]) the condition for the first-order transition, $m_{z1}^2 = -\frac{B'_z}{2C'_z}$, as well as the condition when the local minima are broken, $m_{ze}^2 = -\frac{B'_z}{3C'_z}$. Here, as $m_{z1} > m_{ze}$, there is a metastable state as displayed in Fig. 2(c), corresponding to the state between the green and orange lines in Fig. 1. During the upsweep of magnetic field, the system can cross the first-order transition line, and the magnetic moment m_{z0} does not collapse abruptly to zero but decreases continuously before the local minima are broken.

The existence of the metastable state is important to the RSC. By substituting $\frac{\partial f_m}{\partial m_z}\big|_{m_{z0}} = 0$ and $m_z = m_{z0} + \delta m_z$ into the free energy Eq. (7), one can get the magnetic part of the free energy f_m. Using this f_m and the new $\vec{p}$ is parallel to the direction of the magnetic field which can be derived from $\vec{d}^I = \frac{d_0(\vec{r})}{\sqrt{2}}(1, 0, -i)$, we obtain ΔT_c from $f_{\text{sc-}m}$ with the same method as before,

$$\Delta T_c = -\frac{K_{1zi}}{2\left(6C'_z m_{z0}^4 - 2A'_{1z}\right)\alpha_0} - \frac{K_{1zi}m_{z0}^2}{\alpha_0} + \frac{|\lambda'|h_y}{\alpha_0}. \tag{8}$$

The second term in Eq. (8) shows that if the metastable state is broken, namely, $m_{z0} = m_{ze}$ [$m_{ze}^4 = \frac{A'_{1z}}{3C'_z}$, as shown in Fig. 2(d)], T_c reaches $-\infty$, indicating the truncation of RSC right before the metastable state's broken line. Moreover, Eq. (8) also shows the K_{1zi} coupling terms are the key couplings which are responsible for the truncation of the RSC.

Paramagnetic SC. There are several known experimental facts for the paramagnetic UBS, UTe_2 [6,14]. The SC, as the weak-field region, is initially suppressed by the increasing magnetic field h_y. However, when the magnetic field is sufficiently strong, RSC appears. Finally, when the magnetic field arrives at 34.9 T [32], a first-order transition occurs with the increasing jump of the magnetic moment m_{y0}, and RSC disappears simultaneously. The phase diagram of the UTe_2 is summarized in Fig. 3.

Due to the absence of FM order, the absence of the K_{2i} coupling terms in our free energy hardly supports the nonunitary triplet SC states in the weak magnetic field region, consistent with the measurements of heat capacity and thermal conductivity in UTe_2, which indicates a point-node gap structure [33]. Similar to the method in the ferromagnetic UBS, ΔT_c can be derived,

$$\Delta T_c = -\left(\frac{K_{1zi}}{2A_{1z}} + \frac{K_{1yi}}{2A_{1y}}\right) - K_{1yi}m_{y0}^2, \tag{9}$$

where T_{c0} is the superconducting critical temperature without the SFFE. Since m_{y0} increases with increasing h_y, Eq. (9) implies T_c decreases as shown in Fig. 3. In addition, from Eq. (9), we can learn that, for UTe_2, the key coupling terms in the weak magnetic field region are K_{1yi} coupling terms.

However, when the magnetic field is strong enough, from our theory, the symmetry of UTe_2 can be described by a magnetic group $D_{2h}(E, C_{2y}, I, \sigma_{xz})$, thus the nonunitary SC can appear because of the K_{2i} coupling term in Eq. (4) as discussed before in FM UBS. For both corepresentations A_u and B_u, the free energy can be expressed as (see SM Sec. I [30])

$$f_{\text{sc-}m} = A_{1z}m_z^2 + A_{1y}m_y^2 + K_{1zi}m_z^2|d_i|^2 + K_{1yj}m_y^2|d_j|^2 + K_{2y}m_y p_y - \lambda h_y p_y, \tag{10}$$

and ΔT_c can be derived as

$$\Delta T_c = -\frac{K_{1yi}m_{y0}^2}{\alpha_0} + \frac{|K'_{2y} - 2\lambda' A_{1y}|}{\alpha_0} m_{y0}. \tag{11}$$

This parabolic function on m_{y0} explains the RSC close to the first-order transition line in UTe_2 and shows the key coupling terms which lead to the appearance of the RSC are K_{2y} and λ coupling terms.

Similar to the FM case, we can also describe the first-order transition and metastable state in UTe_2, which have been detected in experiment [32]. In this case, the magnetic part of the free energy in a strong magnetic field can be written as

$$f_m = a_y m_y^2 - c_y m_y^3 + \tfrac{1}{2} b_y m_y^4 - \mu_1 h_y m_y + a_z m_z^2. \quad (12)$$

We can derive (see SM Sec. V [30]) the condition for the first-order transition, $h_{yc1} = \frac{c_y}{\mu_1}(\frac{a_y}{b_y} - \frac{c_y^2}{2b_y^2})$ as well as the condition where the local minimum appears, $h_{yc2} = \frac{\Delta_y(8a_y b_y - c_y\Delta_y)}{36 b_y^2 \mu_1}$, where $\Delta_y = 3c_y + \sqrt{9c_y^2 - 12a_y b_y}$. Here, $h_{yc2} < h_{yc1}$, so the metastable state is located in the paramagnetic region as shown in Fig. 3. Then, considering the fluctuation $\vec{m} = (0, m_{y0} + \delta m_y, \delta m_z)$, we obtain the total free energy and ΔT_c as follows,

$$f_{\text{sc-}m} = a_z \delta m_z^2 + \left(-a_y + \mu_1 \frac{h_y}{m_{y0}}\right)\delta m_y^2 + K_{1zi} m_z^2 |d_i|^2 + K_{1yj} m_y^2 |d_j|^2 + K_{2y} m_y p_y - \lambda h_y p_y, \quad (13)$$

$$\Delta T_c = -\frac{1}{\alpha_0}\left(\frac{K_{1yi}}{2\left(\mu_1 \frac{h_y}{m_{y0}} - a_y\right)} - K_{1yi} m_{y0}^2 + |K'_{2y} m_{y0} - \lambda' h_y|\right). \quad (14)$$

When the first-order transition occurs, the magnetic moment m_{y0} increases abruptly. The second term in Eq. (14) increases suddenly. As for the last two terms of Eq. (14), the jump of the magnetic moment m_{y0} can lead to $m'_{y0} \gg \frac{|K'_{2y} - 2\lambda' A_{1y}|}{2K_{1yi}}$, which belongs to the right-hand side of the first-order transition line. In this case, the T_c decreases abruptly as shown in Fig. 3. This explains the experimental observation of the sudden truncation of the RSC in UTe_2 upon the first-order transition [15,16] and shows the key coupling terms leading to this sudden truncation are K_{1yi} coupling terms.

Summary. We develop a phenomenological theory with respect to the full magnetic groups to describe the SC and RSC in UBS in a unified way. The theory explains the global phase diagram of this family of superconductors. In our theory, the SC at the weak magnetic region is suppressed with the increasing transverse magnetic field h_y, due to the energy cost from the mismatch of the induced transverse magnetic moment m_{y0} with the z-polarized nonunitary SC order p_z and the unitary SC order, for ferromagnetic and paramagnetic superconductors, respectively. However, the RSC in both ferromagnetic and paramagnetic superconductors is induced by a fluctuation parallel to the magnetic fields, rather than the sudden jump of the magnetic moment upon the first-order transition. Instead, the sudden jump of the magnetic moment indeed truncates the RSC and there should be a shift of the RSC dome upon a magnetic-hysteresis-loop type of measurement.

Moreover, due to the nondegenerate nature of the triplet SC $\vec{d}$ vector under the magnetic group, another interesting phenomenon of a multijump of the specific heat at different temperatures corresponding to the transition of each component might be observed. The nonunitary coupling K_{2z} term can further cause the splitting of the transition temperature of d_x and d_y from that of d_z, as derived with simplification in Sec. VI of SM [30]. Assuming a small difference among the bare quadratic coefficients, their renormalization would only tune T_c, leaving the jumps of specific heat at the transition temperature $\frac{\Delta C}{T_c}$ intact to the varying magnetic field.

Our theory makes a few explicit predictions. First, we predict the rotation of the spin-triplet pairing $\vec{d}$ vector in different magnetic field regions. In the ferromagnetic superconductors UCoGe and URhGe, with increasing magnetic field, the rescaled $\vec{d}$ vector rotates from $\frac{d_0(\vec{r})}{\sqrt{2}}(1, -i, 0)$ to $\frac{d_0(\vec{r})}{\sqrt{2}}(1, 0, \pm i)$. In UTe_2, the SC is unitary at first. However with a high enough magnetic field, it becomes a nonunitary SC with a rescaled $\vec{d}$ vector, $\frac{d_0(\vec{r})}{\sqrt{2}}(1, 0, \pm i)$. The rotation of the $\vec{d}$ vector by a magnetic field was studied in Sr_2RuO_4 [34], whose spin-triplet pairing symmetry has been questioned [35]. In principle, this prediction can be examined experimentally in superconducting junctions made by these materials. The $\vec{d}$ vector can be visualized from quasiparticle interference technique in scanning tunneling microscopy (STM) experiments [36].

Second, we predict that it is the metastable state that ensures the extension of the RSC over the right-hand side of the first-order transition line in URhGe and UCoGe. This prediction can be checked by performing a magnetic-hysteresis-loop type of measurement around the first-order transition line. We can apply a strong magnetic field to destroy the metastable state at first and then reduce it to induce the RSC. The maximum of the upper critical magnetic field is predicted to have a magnetic-hysteresis-loop type of behavior. Namely, it is much smaller than the one with a normal procedure where the field crosses the first-order transition line from its left-hand side. The RSC dome upon a downswept magnetic field would shift to the left of the first-order transition line, as depicted by the dashed cyan line in Fig. 1.

Finally, we predict that a metastable state also exists in UTe_2 and affects the behavior of the RSC in UTe_2 because of the magnetic hysteresis [32]. The metastable state indicates the remaining large magnetic moment m_{y0} during the downsweep process. Since the RSC is truncated by the sudden increase of m_{y0}, during the upsweep, the RSC would exist until the first-order transition line. However, during the downsweep, the magnetic moment does not decrease abruptly when the system crosses the first-order transition line so that the RSC will not appear until the system crosses the metastable state's broken line (the cyan downsweep line in Fig. 3). (Recently, we noticed that a magnetic-hysteresis-loop type of behavior near the first-order transition line in UTe_2 was detected [37], which is strong support for our theory.) By the way, we also notice that in UTe_2, a new reentrant superconductivity which exists only in the FM region, has been detected in Ref. [16]. In the frame of our theory, the reason why this reentrant superconductivity only exists in the FM region is highly related to the downsweep path in URhGe and UCoGe, in which the superconductivity does not appear until the system cross the first-order transition line (as mentioned in our second prediction about URhGe and UCoGe). In addition, since the direction of the magnetic field is in a specific region between the b axis and c axis, it may also be related to both of the field-induced fluctuations along the c axis and b axis, which needs further investigation.

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