Generalized spin fluctuation feedback in heavy fermion superconductors

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Experiments reveal that the superconductors UPt_3, $PrOs_4Sb_{12}$, and $U_{1-x}Th_xBe_{13}$ undergo two superconducting transitions in the absence of an applied magnetic field. The prevalence of these multiple transitions suggests a common underlying mechanism. A natural candidate theory which accounts for these two transitions is the existence of a small symmetry-breaking field; however, such a field has not been observed in $PrOs_4Sb_{12}$ or $U_{1-x}Th_xBe_{13}$ and has been called into question for UPt_3. Motivated by arguments originally developed for superfluid ^{3}He, we propose that a generalized spin fluctuation feedback effect is responsible for these two transitions. We first develop a phenomenological theory for ^{3}He that couples spin fluctuations to superfluidity, which correctly predicts that a high-temperature broken time-reversal superfluid ^{3}He phase can emerge as a consequence. The transition at lower temperatures into a time-reversal invariant superfluid phase must then be first order by symmetry arguments. We then apply this phenomenological approach to the three superconductors UPt_3, $PrOs_4Sb_{12}$, and $U_{1-x}Th_xBe_{13}$, revealing that this naturally leads to a high-temperature time-reversal invariant nematic superconducting phase, which can be followed by a second-order phase transition into a broken time-reversal symmetry phase, as observed.

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I. INTRODUCTION

There has been renewed interest in unconventional superconductors, as they provide a natural platform for topological states [1–4]. Correlated fermion superconductors such as UPt_3, $PrOs_4Sb_{12}$, $U_{1-x}Th_xBe_{13}$, and URu_2Si_2 have been intensely studied as they show time-reversal symmetry breaking [5–9] and may host Majorana modes as well as Bogoliubov Fermi surfaces [10–13]. Of these, UPt_3, $PrOs_4Sb_{12}$, and $U_{1-x}Th_xBe_{13}$ show a rich phase diagram, with two superconducting phases under zero field. The high-temperature A phase is time-reversal symmetric and the low-temperature B phase is a broken time-reversal symmetry state [9,14–19].

The presence of two transitions in three different materials raises a question about the underlying mechanism. UPt_3 has been the most studied of these materials and has a phase diagram as shown in Fig. 1 [15]. The most common explanation for this phase diagram relies on coupling the superconducting order parameter to a weak symmetry-breaking field, which splits the degeneracy between the different order parameter components [20,21]. The symmetry-breaking field is associated with an antiferromagnetic (AFM) order seen in older neutron scattering measurements [22]. However, recent experiments show that there is no static order near T_c, though antiferromagnetic fluctuations are present [23,24], which casts serious doubts on the use of a symmetry-breaking field to generate two transitions. Meanwhile, there is no accepted model which accounts for the double transition in $U_{1-x}Th_xBe_{13}$ or $PrOs_4Sb_{12}$, though there are signatures of antiferroquadrupolar (AFQ) fluctuations in $PrOs_4Sb_{12}$ and antiferromagnetic fluctuations in $U_{1-x}Th_xBe_{13}$ as seen in inelastic neutron scattering [25,26]. It is natural to ask if these fluctuations can account for the generic observation of two transitions.

To gain insight into this question, it is reasonable to consider superfluid ^{3}He, which also exhibits multiple phases. In this case, conversely, there is a high-temperature high-pressure time-reversal symmetry-breaking A phase and a low-temperature low-pressure time-reversal symmetric B phase as shown in Fig. 2 [27,28]. Originally, the stability of the A phase was a puzzle, as weak-coupling theory predicted that the B state was stable for all temperatures [29]. This paradox was resolved by Anderson and Brinkman, who showed that coupling superfluidity to paramagnetic fluctuations can stabilize the A state, through a mechanism called the spin fluctuation feedback effect [30–32].

In this paper, we propose a mechanism for multiple transitions in correlated fermion materials by coupling superconductivity to fluctuations (both antiferromagnetic and antiferroquadrupolar), analogous to superfluid ^{3}He. We initially formulate a simple phenomenological method to capture the essential physics of superfluid ^{3}He and show that it reproduces the microscopic spin fluctuation feedback effect developed by Anderson and Brinkman. We then apply this to UPt_3, $U_{1-x}Th_xBe_{13}$, and $PrOs_4Sb_{12}$ and show that these fluctuations change the coefficients of the Ginzburg-Landau theory and allow for the possible stabilization of a time-reversal symmetric A phase. We then consider a transition into the broken time-reversal symmetric state, implementing the symmetry constraints associated with observing a polar Kerr

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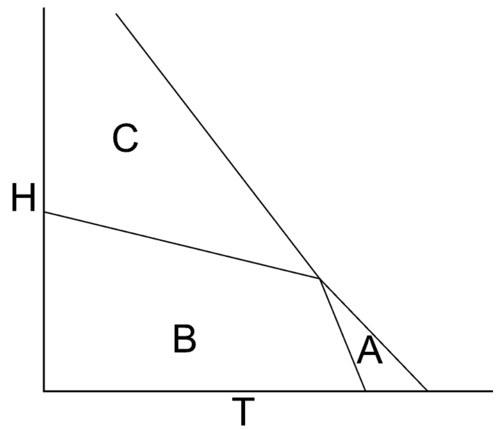

FIG. 1. Magnetic-field–temperature H-T phase diagram of UPt_3 with the B field perpendicular to the z axis.

effect, when applicable. These considerations constrain the possible order parameters. We obtain the following results.

(i) We show that, except for the three-dimensional (3D) $T_{g/u}$ irreducible representations of $PrOs_4Sb_{12}$, the only possible way to undergo two successive transitions is for the B state to be a time-reversal broken state.

(ii) We also find that the Kerr effect measurement rules out the two dimensional (2D) $E_{g/u}$ irreducible representation scenario for $PrOs_4Sb_{12}$. We suggest that subsequent Kerr measurements with different training fields directions may further constrain the order parameters for the 3D $T_{g/u}$ irreducible representation case.

(iii) For $U_{1-x}Th_xBe_{13}$ in the case of its 3D $T_{g/u}$ irreducible representation, the form of the spin fluctuation feedback effect allows for only one A state symmetry out of two possible states.

(iv) We also suggest that a polar Kerr measurement be conducted on $U_{1-x}Th_xBe_{13}$, as the presence of a polar Kerr signal would rule out the 2D ($E_{g/u}$) scenario for $U_{1-x}Th_xBe_{13}$ and would constrain the pairing channels for the 3D ($T_{g/u}$) irreducible representation scenario of $U_{1-x}Th_xBe_{13}$.

These results are tabulated in Table I. We have only written one representative state when there are degeneracies; the other degenerate states appear in the main text and the Appendix.

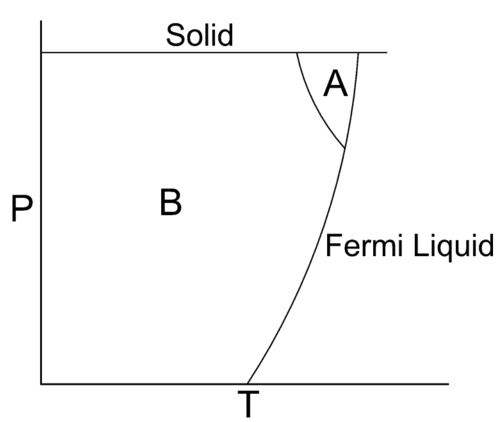

FIG. 2. Pressure-temperature P-T phase diagram of ^{3}He.

II. ^{3}He

^{3}He is a strongly correlated Landau-Fermi liquid, whose quasiparticle excitations pair to form a spin-triplet p-wave superfluid [27,28]. The gap function is $\Delta(\mathbf{k}) = i[d_i(\mathbf{k})\sigma_i]\sigma_y$, with $d_i = d_{i\alpha}\hat{\mathbf{k}}_\alpha$; in this paper we use the Einstein summation convention. The order parameter $d_{i\alpha}$ is a 3×3 matrix with complex entries, where i is the spin index and α is the orbital index and both run over x, y, and z. By comparing to experiments, the ^{3}He–A phase was identified with the Anderson-Brinkman-Morel state with $d_{xx} = \frac{\Delta}{\sqrt{2}}$, $d_{xy} = i\frac{\Delta}{\sqrt{2}}$, and all other $d_{ij} = 0$, while the B state was associated with the Balian-Wethamer state, which has $d_{ij} = \frac{\Delta}{\sqrt{3}}\delta_{i,j}$ [27,28]. Weak-coupling theory showed that the BW state is stable for all temperatures [29], implying that a strong-coupling approach was needed to explain the existence of the high-temperature high-pressure A phase. Anderson and Brinkman [30] used spin fluctuation feedback effect to stabilize the A phase, which relied on the pairing glue in ^{3}He being paramagnetic fluctuations. This implies that the formation of the superfluid alters the pairing interaction, where the type of modification depends on which state is formed [27]. Thus the A state can be stabilized despite being unstable under weak-coupling theory. The A–B transition in this case is first order as the B state is not a subgroup of the A state.

Here spin fluctuation feedback effect will be recaptured in a phenomenological manner, by coupling the superfluid order parameter to paramagnetic fluctuations and calculating the resulting change to the bare free energy. The bare free-energy density of superfluid ^{3}He is given as

$$\beta f_{\text{sf}} = \alpha d_{i\alpha}d^*_{i\alpha} + \beta_1 d_{i\alpha}d_{i\alpha}d^*_{j\beta}d^*_{j\beta} + \beta_2 d_{i\alpha}d_{j\alpha}d^*_{i\beta}d^*_{j\beta} + \beta_3 d_{j\alpha}d_{j\beta}d^*_{i\alpha}d^*_{i\beta} + \beta_4 d_{i\alpha}d^*_{i\alpha}d_{j\beta}d^*_{j\beta} + \beta_5 d^*_{i\alpha}d_{j\alpha}d_{i\beta}d^*_{j\beta}. \tag{1}$$

To this we add the coupling of superfluidity and magnetic fluctuations, which is constructed to be invariant under independent rotations in orbital and spin space. The free-energy density is given as

$$\beta f_{\text{sf-}m} = A_1 m_i m_i + K_1 m_i m_i d_{j\alpha}d^*_{j\alpha} + K_2 m_i m_j d_{i\alpha}d^*_{j\alpha} + K_3 i m_i(\epsilon_{ijk}d_{j\alpha}d^*_{k\alpha}) + B(\vec{\nabla}_i m_j)^2, \tag{2}$$

where m_i is the magnetic order parameter associated with spin fluctuations. The K's are the couplings between the spin fluctuations and the superfluid order parameter, and the B term is the spatial variation, i.e., q dependence, of the spin order. We assume that A_1 is parametrically small and positive, i.e., $A_1 \to 0$, to indicate that we have large fluctuations. We have the Hamiltonian

$$\begin{aligned}\beta\mathcal{H} &= \beta\mathcal{H}_0 + \mathcal{K}\\ &= \int d^3x\big[A_1 m_j^2 + B(\vec{\nabla}_i m_j)^2\big] + \int d^3x\big[K_1\big(m_i^2\big)(d_{j\alpha}d^*_{j\alpha})\\ &\quad + K_2 m_i m_j d_{i\alpha}d^*_{j\alpha} + K_3 i m_i(\epsilon_{ijk}d_{j\alpha}d^*_{k\alpha})\big],\end{aligned} \tag{3}$$

where $\beta\mathcal{H}_0$ is the Gaussian theory for spin fluctuations and $\mathcal{K}$ is the coupling between superfluid order and the spin order

TABLE I. Summary of our results. Column 2 shows the fluctuations responsible for the spin fluctuation feedback effect, whose Q vector dependence can be found in the text. Column 3 shows the irreducible representation of the order parameter. Column 4 gives a representative basis function of the superconducting order parameter and displays the symmetry properties of its components. Column 5 is the time-reversal symmetric state A, where each state is multiplied by an overall η_0 magnitude. Column 6 is the time-reversal symmetry-breaking state B grown out of the A state, where the "or" separates different B irreducible representations and we only write one state for each irreducible representation; the other degenerate states are in the text and the Appendix. Here δ represents a small number, which grows positively from 0 at the second transition. The $( )^*$ is for A–B transitions which are Kerr inactive. The $\times$ represents that this channel is ruled out.

Material	Fluctuation	Irreducible representation	Order parameter	A	B
UPt_3	AFM	E	k_xk_z, k_yk_z	(1,0)	$(1,i\delta)$
$PrOs_4Sb_{12}$	AFQ	E	$2k_z^2-k_x^2-k_y, k_x^2-k_y^2$	$\times$	$\times$
$PrOs_4Sb_{12}$	AFQ	T	k_yk_z, k_xk_z, k_xk_y	(1,0,0)	$(1, i\delta, 0)$ or $(1, 0, i\delta)$
$PrOs_4Sb_{12}$	AFQ	T	k_yk_z, k_xk_z, k_xk_y	(1,1,1)	$(1, 1+i\delta\cos\theta, 1+i\delta\sin\theta)$
$U_{1-x}Th_xBe_{13}$	AFM	E	$2k_z^2-k_x^2-k_y, k_x^2-k_y^2$	(1,0)	$(1+i\delta, 0)^*$ or $(1, i\delta)^*$
$U_{1-x}Th_xBe_{13}$	AFM	E	$2k_z^2-k_x^2-k_y, k_x^2-k_y^2$	(0,1)	$(0, 1+i\delta)^*$ or $(i\delta, 1)^*$
$U_{1-x}Th_xBe_{13}$	AFM	T	k_yk_z, k_xk_z, k_xk_y	(1,0,0)	$(1+i\delta, 0, 0)^*$ or $(0, 1+i\delta, 0)$

fluctuations. After Fourier transforming we get

$$\beta\mathcal{H} = \int \frac{d^3\mathbf{q}}{(2\pi)^3}(A_1 + Bq^2)|\tilde{m}(q)|^2 + \mathcal{K}. \tag{4}$$

The coupling between superfluidity and magnetic fluctuations in Fourier space is

$$\begin{aligned}\mathcal{K} = \int &\frac{d^3\mathbf{q}_1 d^3\mathbf{q}_2 d^3\mathbf{q}_3 d^3\mathbf{q}_4}{(2\pi^3)^4}(2\pi)^3\delta^3(\mathbf{q}_1+\mathbf{q}_2+\mathbf{q}_3+\mathbf{q}_4)\\ &\times [K_1\tilde{m}_i(\mathbf{q}_1)\tilde{m}_i(\mathbf{q}_2)\tilde{d}_{j\alpha}(\mathbf{q}_3)\tilde{d}^*_{j\alpha}(\mathbf{q}_4)\\ &+ K_2\tilde{m}_i(\mathbf{q}_1)\tilde{m}_j(\mathbf{q}_2)\tilde{d}_{i\alpha}(\mathbf{q}_3)\tilde{d}^*_{j\alpha}(\mathbf{q}_4)]\\ &+ K_3\int \frac{d^3\mathbf{q}_1 d^3\mathbf{q}_2 d^3\mathbf{q}_3 d^3\mathbf{q}_4}{(2\pi^3)^3}(2\pi)^3\delta^3(\mathbf{q}_1+\mathbf{q}_2+\mathbf{q}_3)i\tilde{m}_i\\ &\times(\mathbf{q}_1)\epsilon_{ijk}\tilde{d}_{j\alpha}(\mathbf{q}_2)\tilde{d}^*_{k\alpha}(\mathbf{q}_3).\end{aligned} \tag{5}$$

Near the superfluid transitions, the coupling $\mathcal{K}$ can be treated perturbatively around the Gaussian theory of the magnetic fluctuations, i.e., $\beta\mathcal{H}_0$. This quadratic theory for spin fluctuations is valid as long as we are more than a Ginzburg temperature away from the critical temperature associated with magnetic ordering. Thus the coupling can be evaluated as

$$\beta\mathcal{H} = \beta\mathcal{H}_0 - \ln\langle\exp^{-\mathcal{K}}\rangle_m, \tag{6}$$

where $\langle\,\rangle_m$ implies that we are calculating the expectation values with respect to the Gaussian $\beta\mathcal{H}_0$ theory. The expression (6) can be expanded perturbatively around the Gaussian theory (see, e.g., [33]) as

$$\ln\langle\exp^{-\mathcal{K}}\rangle_m = -\langle\mathcal{K}\rangle_m + \tfrac{1}{2}(\langle\mathcal{K}^2\rangle_m - \langle\mathcal{K}\rangle_m^2) + \cdots. \tag{7}$$

We keep up to only the second order in $\mathcal{K}$, as this will introduce corrections of $O(d^4)$ to the superfluid free-energy density, which will be responsible for stabilizing the A state. The first term $\langle\mathcal{K}\rangle_m$ can be evaluated and will introduce a correction to quadratic term in the superfluid Hamiltonian

$$\begin{aligned}-\langle\mathcal{K}\rangle_m &= -\frac{3K_1+K_2}{2}\int\frac{d^3\mathbf{q}_1 d^3\mathbf{q}_2 d^3\mathbf{q}_3 d^3\mathbf{q}_4}{(2\pi)^{12}}(2\pi)^3\delta^3(\mathbf{q}_1\\ &\quad+\mathbf{q}_2+\mathbf{q}_3+\mathbf{q}_4)\frac{(2\pi)^3\delta^3(\mathbf{q}_1+\mathbf{q}_2)}{A_1+Bq_1^2}\tilde{d}_{i\alpha}(\mathbf{q}_3)\tilde{d}^*_{i\alpha}(\mathbf{q}_4)\\ &= -\frac{3K_1+K_2}{2}\int\frac{d^3\mathbf{q}_3}{(2\pi)^3}\tilde{d}_{i\alpha}(\mathbf{q}_3)\tilde{d}^*_{i\alpha}(-\mathbf{q}_3)\\ &\quad\times\int\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{A_1+Bk^2}.\end{aligned} \tag{8}$$

Performing an inverse Fourier transformation, we obtain the correction to the bare superfluid free-energy density

$$\begin{aligned}\beta f_{\text{eff}} = &\left(\alpha + \frac{(3K_1+K_2)(2\pi)^3}{2\Lambda^3}\int^{\Lambda^3}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{A_1+Bk^2}\right)d_{i\alpha}d^*_{i\alpha}\\ &+\beta_1 d_{i\alpha}d_{i\alpha}d^*_{j\beta}d^*_{j\beta} + \beta_2 d_{i\alpha}d_{j\alpha}d^*_{i\beta}d^*_{j\beta} + \beta_3 d_{j\alpha}d_{j\beta}d^*_{i\alpha}d^*_{i\beta}\\ &+\beta_4 d_{i\alpha}d^*_{i\alpha}d_{j\beta}d^*_{j\beta} + \beta_5 d^*_{i\alpha}d_{j\alpha}d_{i\beta}d^*_{j\beta},\end{aligned} \tag{9}$$

where Λ^3 is a cutoff wavelength.

We see that the first-order correction from the spin fluctuation feedback effect has changed the bare T_c. The coefficient of the quadratic term is the inverse susceptibility, i.e., $\chi^{-1} = A_1 + Bq^2 + \frac{(3K_1+K_2)(2\pi)^3}{\Lambda^3}\int^{\Lambda^3}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{A_1+Bk^2}$. Next we calculate the crucial second-order $\frac{1}{2}(\langle\mathcal{K}^2\rangle_m - \langle\mathcal{K}\rangle_m^2)$ correction which will change the coefficient of the quartic terms in the bare free-energy density. We follow the exact same process as outlined above and after inverse Fourier transforming we get the free-energy density

$$\begin{aligned}\beta f_{\text{eff}} = &\left(\alpha + \frac{(3K_1+K_2)(2\pi)^3}{2\Lambda^3}\int^{\Lambda^3}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{A_1+Bk^2}\right)d_{i\alpha}d^*_{i\alpha} + \beta_1 d_{i\alpha}d_{i\alpha}d^*_{j\beta}d^*_{j\beta} + \beta_2 d_{i\alpha}d_{j\alpha}d^*_{i\beta}d^*_{j\beta}\\ &+\left(\beta_3 - \frac{K_2^2(2\pi)^3}{4\Lambda^3}\int^{\Lambda^3}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{(A_1+Bk^2)^2}\right)d_{j\alpha}d_{j\beta}d^*_{i\alpha}d^*_{i\beta} + \left(\beta_5 - \frac{K_2^2(2\pi)^3}{4\Lambda^3}\int^{\Lambda^3}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{(A_1+Bk^2)^2}\right)d^*_{i\alpha}d_{j\alpha}d_{i\beta}d^*_{j\beta}\end{aligned}$$

$$+\left(\beta_4-\frac{\left(K_2^2+6K_1^2+4K_1K_2\right)(2\pi)^3}{2\Lambda^3}\int^{\Lambda^3}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{(A_1+Bk^2)^2}\right)d_{i\alpha}d^*_{i\alpha}d_{j\beta}d^*_{j\beta}$$
$$-\frac{K_3^2(2\pi)^3}{2\Lambda^3}\int^{\Lambda^3}\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{(A_1+Bk^2)}(i\vec{d}\times\vec{d}^*)^2. \tag{10}$$

Let us now consider a simpler situation, where we ignore the cost of spatial variation in the spin fluctuation order parameter, i.e., $B=0$. Thus we have the coupling

$$\beta f_{\text{sf-}m}=A_1m_im_i+K_1m_im_id_{j\alpha}d^*_{j\alpha}+K_2m_im_jd_{i\alpha}d^*_{j\alpha}+K_3im_i(\epsilon_{ijk}d_{j\alpha}d^*_{k\alpha}). \tag{11}$$

The magnetic partition function is given as

$$\mathcal{Z}_m=\int Dm_ie^{-\beta\mathcal{H}}=\int Dm_i\exp\left(-\int d^3xf_{\text{sf-}m}\right)=\int Dm_i\exp\left(-\int d^3xA_{ij}m_im_j\right), \tag{12}$$

where A_{ij} contains couplings between magnetic and superconducting orders and gives corrections for the bare superconducting free-energy density. Integrating out the quadratic (Gaussian) magnetic fluctuations gives an effective free-energy density

$$\beta f_{\text{eff}}=\left(\alpha+\frac{3K_1}{2A_1}+\frac{K_2}{2A_1}\right)d_{i\alpha}d^*_{i\alpha}+\beta_1d_{i\alpha}d_{i\alpha}d^*_{j\beta}d^*_{j\beta}+\beta_2d_{i\alpha}d_{j\alpha}d^*_{i\beta}d^*_{j\beta}+\left(\beta_3-\frac{K_2^2}{4A_1^2}\right)d_{j\alpha}d_{j\beta}d^*_{i\alpha}d^*_{i\beta}$$
$$+\left(\beta_4-\frac{K_2^2+6K_1^2+4K_2K_1}{2A_1^2}\right)d_{i\alpha}d^*_{i\alpha}d_{j\beta}d^*_{j\beta}+\left(\beta_5-\frac{K_2^2}{4A_1^2}\right)d^*_{i\alpha}d_{j\alpha}d_{i\beta}d^*_{j\beta}-\frac{K_3^2}{2A_1}(i\vec{d}\times\vec{d}^*)^2. \tag{13}$$

Now if we compare Eqs. (10) and (13), we note that even though the corrections introduced to the free-energy density have different forms in both cases, the sign of the correction introduced is the same, i.e., in both cases the spin fluctuation feedback effect leads to corrections which can stabilize a different state than the state preferred by the weak-coupling theory. Subsequently, for clarity, we use the simplified result since it yields qualitatively the same results.

The β_i quartic terms in Eq. (13) are associated with the free-energy density without coupling to fluctuations, here assumed to be derived from weak-coupling theory and the quartic terms with the K's originating from spin fluctuation feedback effect. For large paramagnetic fluctuations, i.e., a small but positive A_1, the terms that dominate are those that are proportional to A_1^{-2} and we ignore terms of order A_1^{-1}, e.g., couplings of the form m^2d^4. The K_3 term in Eq. (13) shows that paramagnetic fluctuations can favor nonunitary states [34–37], but will be neglected here due to its weaker A_1^{-1} dependence.

Weak-coupling theory gives $\beta_2=\beta_3=\beta_4=-\beta_5=2\beta_1=\frac{6}{5}s$, where s is a positive-valued constant [27,28]. When the spin fluctuation feedback effect is turned off, i.e., $K_i=0$, the BW state is energetically favorable with $f_{\text{eff}}=\frac{5}{3}s$, while the A state has a slightly larger free-energy density of $f_{\text{eff}}=2s$. The spin fluctuation feedback effect coupling lowers the energy of the A state by $K_2^2/3A_1^2$ compared to the B state and thus for large fluctuations, i.e., $K_2^2/3A_1^2>\frac{1}{3}s$, can stabilize the A state [32].

The A–B transition stems from the different temperature dependence of the weak-coupling quartic terms versus the spin fluctuation feedback effect corrections to the quartic terms. Quartic, i.e., β, terms originating from weak-coupling theory generically have a $\frac{1}{T^2}$ dependence [35], while microscopic calculations show that quartic terms originating from the spin fluctuation feedback effect have a $\frac{1}{T}$ dependence [27,31,32]. These calculations assume quadratic Gaussian spin fluctuations and imply that at high temperatures, strong fluctuations may stabilize the A phase, while at lower temperature the weak-coupling terms will dominate and the system will undergo a first-order transition into the state preferred by weak-coupling theory, i.e., the B phase. Recent calculations done for twisted bilayer graphene [38] show that the same temperature dependence of the quartic terms is seen for both spin density and charge density wave fluctuations, again considering Gaussian fluctuations. This suggests that the weak-coupling quartic terms and spin fluctuation corrections to these terms generically have a $1/T$ and a $1/T^2$ temperature dependence, respectively. We will assume this to be the case in the correlated fermion materials considered below.

III. UPt_3

UPt_3 is a hexagonal crystal with D_{6h} point group symmetry and has two distinct phases under zero field: a high-temperature A phase and a low-temperature B phase [15,39]. However, unlike ^{3}He, the A phase is time-reversal symmetric while the B phase is time-reversal symmetry breaking as seen in muon spin relaxation (μSR) and polar Kerr measurements [5,14]. UPt_3 has four 2D irreducible representations labeled $E_{1u/g}$ and $E_{2u/g}$, where the order parameter transforms like $\eta_1\sim k_xk_z$ and $\eta_2\sim k_yk_z$ and like $\eta_1\sim k_x^2-k_y^2$ and $\eta_2\sim 2k_xk_y$, respectively. The free-energy density is the same for all the E irreducible representations and is given as [40]

$$\beta f_{\text{sc}}=\alpha(|\eta_1|^2+|\eta_2|^2)+\beta_1(|\eta_1|^2+|\eta_1|^2)^2+\beta_2\left|\eta_1^2+\eta_2^2\right|^2. \tag{14}$$

The coefficient of $|\eta_i^2|^2$ determines the behavior below T_c, with $\beta_2>0$ favoring the time-reversal symmetry-breaking $(1,i)$ state and $\beta_2<0$ stabilizing the time-reversal symmetric (1,0) state. To explain the existence of multiple phases, the currently accepted model involves coupling superconductivity to antiferromagnetic order [20,21]. This splits the T_c between η_1 and η_2 and allows for two transitions.

However, recent experiments raise questions over the existence of true antiferromagnetic order near the two closely spaced superconducting transitions. The Bragg peaks in inelastic neutron scattering are not resolution limited near the superconducting transitions of $T_{cA} = 530$ mK and $T_{cB} = 480$ mK [15,23,41], in addition to the absence of signatures of a magnetic transition in specific heat, magnetization, and NMR Knight shift experiments around this temperature. The peaks start narrowing only below 50 mK and become resolution limited at 20 mK [23,41], which seems consistent with anomalies seen in specific heat [42,43], thermal expansion [44], and magnetization measurements [45] seen near 20 mK. Interestingly, NMR experiments show an anomaly at 50 mK which is associated with the fluctuations slowing down, though there is no sign of static order down until 15 mK [46,47]. This has led to the current interpretation that these experiments imply the presence of antiferromagnetic fluctuations [24], instead of antiferromagnetic order, and we suggest that a generalized spin fluctuation feedback effect then stabilizes a time-reversal symmetric A state.

Our theory is a Ginzburg-Landau theory and is valid near the A–B transition and cannot be extended deep into the B state. However, the use of spin fluctuation feedback effect fluctuations to generate two closely spaced transitions should be valid near the superconducting transitions, i.e., $T_c \sim 500$ mK. We note that the Ginzburg temperature for magnetic transitions is generically of the order 1–10^{-2} mK of the magnetic transition temperature (which for UPt_3 is $T_{cm} \sim 50$ mK). Consequently, the superconducting A–B transitions will be insensitive to critical phenomena stemming from the possible ultralow-temperature magnetic ordering. Additionally, there are no signatures of quantum critical effects in this material and the other materials considered in this paper. Hence arguments similar to those developed for superfluid ^{3}He can be used to explain two transitions in UPt_3 and it suffices to consider a Gaussian theory of spin fluctuations.

We will proceed analogously to ^{3}He and assume that the B time-reversal symmetry-breaking state is favored by the weak-coupling theory, while strong fluctuations can stabilize a time-reversal symmetric A phase. The fluctuations are characterized by wave vectors $\mathbf{Q}_1 = \frac{1}{2}\mathbf{a}^*$, $\mathbf{Q}_2 = \frac{1}{2}(\mathbf{b}^* - \mathbf{a}^*)$, and $\mathbf{Q}_3 = -\frac{1}{2}\mathbf{b}^*$ [22,23] which are associated with the magnetic order parameters m_1, m_2, and m_3, respectively. The coupling of superconductivity to the magnetic fluctuations is constructed to be invariant under $D_{6h} \times \mathrm{U}(1) \times \mathscr{T}$, where $\mathscr{T}$ is time-reversal symmetry and is expressed as [21]

$$\begin{aligned}\beta f_{\text{sc-}m} = {} & A_1 m_i^2 + K_1\big(m_i^2\big)(\eta_j\eta_j^*) \\ & + K_2\big[\big(2m_1^2 - m_2^2 - m_3^2\big)(|\eta_1|^2 - |\eta_2|^2) \\ & + \sqrt{3}\big(m_3^2 - m_2^2\big)(\eta_1\eta_2^* + \eta_2\eta_1^*)\big]. \end{aligned} \tag{15}$$

Integrating out the fluctuations as before gives the free-energy density

$$\begin{aligned}& \left(\alpha + \frac{3}{2}\frac{K_1}{A_1}\right)(|\eta_1|^2 + |\eta_2|^2) + \left(\beta_1 - \frac{3}{4}\frac{K_1^2}{A_1^2}\right)(|\eta_1|^2 + |\eta_1|^2)^2 \\ & + \left(\beta_2 - \frac{6}{4}\frac{K_2^2}{A_1^2}\right)|\eta_1^2 + \eta_2^2|^2. \end{aligned} \tag{16}$$

We see that the terms originating from the generalized spin fluctuation feedback effect have introduced negative-value corrections to the quartic terms, which most importantly changes the $|\eta_i^2|^2$ coefficient. Thus, for large fluctuations, these spin fluctuation feedback effect terms can stabilize the A state, instead of the time-reversal symmetry breaking B by making this coefficient negative.

This also interestingly implies that two transitions will occur only if the B state is a broken time-reversal symmetry state. In particular, if the B state were time-reversal symmetric then the spin fluctuation feedback effect terms would simply further stabilize the time-reversal symmetric nematic state.

As argued earlier, the quartic terms that stem from weak-coupling theory should increase more strongly as temperature is decreased than the quartic terms which arise from the generalized spin fluctuation feedback effect. This allows the coefficient of the $|\eta_1^2 + \eta_2^2|^2$ to change sign as temperature is decreased so that a transition into a broken time-reversal symmetry state is possible. We discuss this in more detail below.

A. Effective theory for A–B transitions

A complete phenomenological description of a second phase transition within a single multidimensional irreducible representation requires a free-energy density that is at least eighth order in the order parameter [48,49]. For this reason, we consider a simpler approach and model the A–B transition as an effective phenomenological theory in which we start with the (1,0) state for UPt_3 and allow the B state to continuously grow out of this, i.e., $(1 + \tilde{\eta}_{1i}, 0 + \tilde{\eta}_{2i})$, where $\tilde{\eta}_i$ is small near the transition. Time-reversal symmetry allows us to classify the order parameter $\tilde{\eta}_i$ for the A–B transition into a real part $\tilde{\eta}_R$, which is invariant under $\mathscr{T}$, and an imaginary part $\tilde{\eta}_I$, which changes sign under $\mathscr{T}$, with the transformation properties

$$\tilde{\eta}_R \xrightarrow{\mathcal{T}} \tilde{\eta}_R, \quad \tilde{\eta}_I \xrightarrow{\mathcal{T}} -\tilde{\eta}_I. \tag{17}$$

The condition that the second transition is observed to break time-reversal symmetry allows us to consider only the imaginary order parameter. The (1,0) state has $D_2(C_2) \times \mathscr{T}$ and $D_2 \times \mathscr{T}$ [34] symmetries for the $E_{1u/g}$ and $E_{2u/g}$ irreducible representations, respectively. The order parameter $\tilde{\eta}_{1I}$ belongs to the A_1 irreducible representation of D_2, while $\tilde{\eta}_{2I}$ belongs to the B_1 irreducible representation. Hence our mechanism only allows these two possible symmetries for the B phase.

B. Constraints from the polar Kerr effect

The observation of a polar Kerr signal for the A–B transition [5] further constrains the possible order parameters. In particular, polar Kerr experiments shows that the signal can be trained with an applied magnetic field [5]. This implies that the only viable order parameters are those which belong to the same representation as a component of the magnetic field (H_x, H_y, and H_z). This follows because a Kerr signal that can be trained by a magnetic field is only possible if the superconducting order parameter couples linearly to the applied field. These order parameters will be referred to as

Kerr active (in the literature, this is sometimes labeled as belonging to a ferromagnetic class [50]). This rules out the A_1 order parameter since it is Kerr inactive. The $\tilde{\eta}_{2I}$ order parameter has H_z symmetry, which is consistent with the direction of the training field applied along the c axis in the experiment. Thus the A–B transition can be modeled by the effective order parameter $\tilde{\eta}_{2I}$ with the free energy $\beta f_{A\to B} = \alpha_{1I}\tilde{\eta}_{2I}^2 + \beta_{1I}\tilde{\eta}_{2I}^4$. We will use this approach subsequently to constrain the possible symmetries of the order parameters.

IV. $PrOs_4Sb_{12}$

$PrOs_4Sb_{12}$ is a Pr-based tetrahedral correlated fermion skutterudite superconductor with a T_h point group that, like UPt_3, has two distinct phases [51]. Polar Kerr and μSR measurements show a time-reversal symmetry-breaking B phase [6,18], while the A phase is time-reversal symmetric. $PrOs_4Sb_{12}$ has been studied by phenomenological methods [52]; however, there is no satisfactory mechanism for the double transition. Inelastic neutron scattering experiments indicate the presence of antiferroquadrupolar fluctuations with a $\mathbf{Q} = (1, 0, 0)$ [25,53], which is a single $\mathbf{Q}$ order, invariant under the point group operations. The order parameter of these antiferroquadrupolar fluctuations is three dimensional with components that transform as $m_1 \sim k_yk_z$, $m_2 \sim k_xk_z$, and $m_3 \sim k_xk_y$ [54,55]. The antiferroquadrupolar fluctuations can stabilize a time-reversal symmetric A phase for both the E and T irreducible representations as shown below.

For the E irreducible representation, where the order parameter transforms as $\eta_1 \sim 2k_z^2 - k_x^2 - k_y^2$ and $\eta_2 \sim k_x^2 - k_y^2$, the coupling is

$$\begin{aligned}\beta f_{\text{sc-}m} = {} & A_1\big(m_i^2\big) + K_1\big(m_j^2\big)(\eta_j\eta_j^*)\\ & + K_2\big[\big(2m_3^2 - m_1^2 - m_2^2\big)(|\eta_1|^2 - |\eta_2|^2)\\ & - \sqrt{3}\big(m_1^2 - m_2^2\big)(\eta_1\eta_2^* + \eta_2\eta_1^*)\big]\\ & + K_3\big[\sqrt{3}\big(m_1^2 - m_2^2\big)(|\eta_1|^2 - |\eta_2|^2)\\ & + (\eta_1\eta_2^* + \eta_2\eta_1^*)\big(2m_3^2 - m_1^2 - m_2^2\big)\big]. \end{aligned} \tag{18}$$

As noted in UPt_3, we will consider the simple case of $B(\vec{\nabla}\vec{m})^2 = 0$. Integrating out the antiferroquadrupolar fluctuations, we obtain the effective free-energy density

$$\begin{aligned}\beta f_{\text{eff}} = {} & \left(\alpha + \frac{3K_1}{2A_1}\right)(\eta_j\eta_j^*)\\ & + \left(\beta_1 - \frac{3K_1^2}{4A_1^2} - \frac{3K_2^2}{2A_1^2} - \frac{3K_3^2}{2A_1^2}\right)(\eta_j\eta_j^*)^2\\ & + \left(\beta_2 - \frac{3K_2^2}{2A_1^2} - \frac{3K_3^2}{2A_1^2}\right)(\eta_1\eta_2^* - \eta_2\eta_1^*)^2. \end{aligned} \tag{19}$$

This generalized spin fluctuation feedback effect may again stabilize a time-reversal symmetric A state (ϕ_1, ϕ_2) with $D_2 \times \mathscr{T}$ symmetry [52,56], instead of the time-reversal symmetry-breaking B phase $(1, i)$ with $T(D_2)$ symmetry by changing the sign of the $(\eta_1\eta_2^* - \eta_2\eta_1^*)^2$ term from positive to negative. The A phase has the two components with the same magnitude but an arbitrary phase [52]. Similar to UPt_3, two transitions are possible only when the B state is time-reversal symmetry breaking. The A–B transition is modeled similarly to UPt_3; however, both $\eta_{1I/2I}$ have A_1 Kerr inactive symmetry and are ruled out due to the presence of the Kerr effect, thereby eliminating the 2D E irreducible representation scenario for $PrOs_4Sb_{12}$. Thus we can see that, just using these general symmetry considerations, we are able to rule out a pairing scenario with this mechanism.

For the 3D $T_{g/u}$ irreducible representation, with an order parameter which transforms, for example, as $\eta_1 \sim k_yk_z$, $\eta_2 \sim k_xk_z$, and $\eta_3 \sim k_xk_y$, the coupling is

$$\begin{aligned}\beta f_{\text{sc-}m} = {} & A_1\big(m_i^2\big) + K_1(\eta_j\eta_j^*)\big(m_j^2\big)\\ & + K_2\big[3(|\eta_2|^2 - |\eta_3|^2)\big(m_2^2 - m_3^2\big)\\ & + (2|\eta_1|^2 - |\eta_2|^2 - |\eta_3|^2) \times \big(2m_1^2 - m_2^2 - m_3^2\big)\big]\\ & + K_3\big[(|\eta_2|^2 - |\eta_1|^2)\big(2m_1^2 - m_2^2 - m_3^2\big)\\ & - (2|\eta_1|^2 - |\eta_2|^2 - |\eta_3|^2)\big(m_2^2 - m_3^2\big)\big]\\ & + K_4[(\eta_2\eta_3^* + \eta_2^*\eta_3)m_2m_3 + (\eta_3\eta_1^* + \eta_3^*\eta_1)m_3m_1\\ & + (\eta_1\eta_2^* + \eta_1^*\eta_2)m_1m_2]. \end{aligned} \tag{20}$$

Integrating out the Gaussian antiferroquadrupolar fluctuations gives the effective free-energy density

$$\begin{aligned}\beta f_{\text{eff}} = {} & \left(\alpha + 3\frac{K_1}{A_1}\right)(\eta_j\eta_j^*)\\ & + \left(\beta_1 - \frac{6K_1^2}{A_1^2} + \frac{6K_2^2}{A_1^2} + \frac{2K_3^2}{A_1^2} - \frac{K_4^2}{4A_1^2}\right)(\eta_j\eta_j^*)^2\\ & + \left(\beta_2 - \frac{K_4^2}{4A_1^2}\right)|\eta_i^2|^2 + \left(\beta_3 - \frac{18K_2^2}{A_1^2} - \frac{6K_3^2}{A_1^2} + \frac{K_4^2}{2A_1^2}\right)\\ & \times(|\eta_1|^4 + |\eta_2|^4 + |\eta_3|^4). \end{aligned} \tag{21}$$

Again the generalized spin fluctuation feedback effect has changed the coefficient of the bare free-energy density and hence allows for the possibility of a time-reversal symmetric A state. Interestingly, here we may have two transitions even if the A state is time-reversal symmetry breaking, due the indeterminant sign of the correction to the β_3 coefficient. However, since this does not agree with the experimental identification of the B state having broken time-reversal symmetry, we do not consider this possibility. This irreducible representation has two states which are time-reversal symmetric: the (1,0,0) state with $D_2(C_2) \times \mathscr{T}$ symmetry and the (1,1,1) state with $C_3 \times \mathscr{T}$ symmetry. Both of these allow for a transition to a time-reversal symmetry-breaking B state which is Kerr active and hence provide two viable channels for the transition. The physics of this is similar to the 2D irreducible representation case for UPt_3 and is worked out in the Appendix, the results of which are collected in Table I. It would be of interest to carry out Kerr measurements [6] under different directions of the training field to further constrain the possible pairing channels.

V. $U_{1-x}Th_xBe_{13}$

$U_{1-x}Th_xBe_{13}$ is a cubic material with an O_h point group, which also has two transitions [9,17], but only for a doping range of $2\% < x < 4\%$. The B phase is again a time-reversal symmetry-breaking state [9]. Antiferromagnetic fluctuations are seen in inelastic neutron scattering with a wave vector of $\mathbf{Q}_3 = (1/2, 1/2, 0)$ [26]. We consider both the E and T irreducible representations and model the system with O_h symmetry, having antiferromagnetic fluctuations with wave vector $\mathbf{Q}_3$. The star of $\mathbf{Q}_3$ gives two additional wave vectors $\mathbf{Q}_2 = (1/2, 0, 1/2)$ and $\mathbf{Q}_1$, each of which corresponds to a 1D order parameter $m_1 = (0, 1/2, 1/2)$, m_2, and m_3. Here η_1 and η_2 transform exactly as the E irreducible representation of $PrOs_4Sb_{12}$. The coupling is

$$\beta f_{\text{sc-}m} = A_1\left(m_i^2\right) + K_1\left(m_i^2\right)(\eta_j\eta_j^*) + K_2\left[\left(2m_3^2 - m_1^2 - m_2^2\right)(|\eta_1|^2 - |\eta_2|^2) - \sqrt{3}\left(m_1^2 - m_2^2\right)(\eta_1\eta_2^* + \eta_2\eta_1^*)\right]. \quad (22)$$

The K_3 term present in Eq. (18) is absent above, due to additional symmetry elements present in the O_h as compared to T_h point group. The correction to the free-energy density from these fluctuations is

$$\beta f_{\text{eff}} = \left(\alpha + \frac{3K_1}{2A_1}\right)(\eta_j\eta_j^*) + \left(\beta_1 - \frac{3K_1^2}{4A_1^2} - \frac{3K_2^2}{2A_1^2}\right)(\eta_j\eta_j^*)^2 + \left(\beta_2 - \frac{3K_2^2}{2A_1^2}\right)(\eta_1\eta_2^* - \eta_2\eta_1^*)^2. \quad (23)$$

There are two possible time-reversal symmetric A states. The details of the A–B effective theory follow that of UPt_3 and are in the Appendix, with the possible A state being (1,0) with $D_4 \times \mathscr{T}$ symmetry and (0,1) with $D_4^{(1)}(D_2) \times \mathscr{T}$ symmetry [50]. These states are Kerr inactive, and we suggest that a polar Kerr measurement be performed on this material. If a field-trainable polar Kerr signal is seen, then the 2D order parameter can be ruled out.

For the 3D order parameter case, we note that $U_{1-x}Th_xBe_{13}$ has four T irreducible representations: one $T_{2g/u}$, which transforms exactly like T irreducible representations of $PrOs_4Sb_{12}$, and the other $T_{1g/u}$, which transforms as $\eta_1 \sim k_yk_z(k_y^2 - k_z^2)$, $\eta_1 \sim k_zk_x(k_z^2 - k_x^2)$, and $\eta_3 \sim k_xk_y(k_x^2 - k_y^2)$. The coupling for these T irreducible representations is

$$f_{\text{sc-}m} = A_1\left(m_i^2\right) + K_1\left(m_i^2\right)(\eta_j\eta_j^*) + K_2\left[3(|\eta_2|^2 - |\eta_3|^2)\left(m_2^2 - m_3^2\right) + (2|\eta_1|^2 - |\eta_2|^2 - |\eta_3|^2)\left(2m_1^2 - m_2^2 - m_3^2\right)\right]. \quad (24)$$

The K_3 term found in Eq. (20) is absent because of higher O_h symmetry compared to the T_h symmetry, while the K_4 term is forbidden as m_im_j is not translationally invariant for this $\mathbf{Q}$ vector. The effective free-energy density obtained is

$$\beta f_{\text{eff}} = \left(\alpha + 3\frac{K_1}{A_1}\right)(\eta_j\eta_j^*) + \left(\beta_1 - \frac{3K_1^2}{4A_1^2} + \frac{3K_2^2}{A_1^2}\right)(\eta_j\eta_j^*)^2 + \beta_2\left|\eta_j^2\right|^2 + \left(\beta_3 - \frac{9K_2^2}{A_1^2}\right)(|\eta_1|^4 + |\eta_2|^4 + |\eta_3|^4). \quad (25)$$

Interestingly, here, unlike $PrOs_4Sb_{12}$, there is no change to β_2, which is a result of the K_4 coupling being absent for Eq. (25) in contrast to Eq. (20). There are four possible time-reversal symmetric A states, two for each T irreducible representation, i.e., (1,0,0) and (1,1,1). However, due to there being no change to the coefficient of the $|\eta_i^2|^2$ term, the (1,0,0) is the only time-reversal symmetric A state which allows for a viable transition to a time-reversal symmetry-breaking B state [34]. This state has $D_4(C_4) \times \mathscr{T}$ and $D_4^{(2)}(D_2) \times \mathscr{T}$ symmetry for the $T_{1g/u}$ and $T_{2g/u}$ irreducible representations, respectively [50]. The A–B transition follows similarly to UPt_3 and is modeled in the Appendix.

Polar Kerr measurements, especially if trainable by the field, may be useful as they can rule out the $E_{g/u}$ irreducible representation scenario and can eliminate other 3D $T_{g/u}$ order parameters.

VI. CONCLUSION

In analogy to superfluid ^{3}He, we have argued that a generalized spin fluctuation feedback effect can account for multiple transitions seen in correlated fermion materials. We have provided a simple phenomenological framework to capture this and show that this naturally provides a unifying mechanism for two superconducting transitions observed in UPt_3, $PrOs_4Sb_{12}$, and $U_{1-x}Th_xBe_{13}$. In addition, the use of this generalized spin fluctuation feedback effect has allowed us to constrain various pairing symmetries. The results of this analysis were tabulated in Table I. In particular, we were able to rule out the 2D ($E_{g/u}$) scenario for $PrOs_4Sb_{12}$, while for the 3D ($T_{g/u}$) irreducible representation of $U_{1-x}Th_xBe_{13}$ only one of two possible A states was allowed. Additionally, if a polar Kerr signal were observed for $U_{1-x}Th_xBe_{13}$, we would be able to rule out the 2D ($E_{g/u}$) scenario for $U_{1-x}Th_xBe_{13}$ and place further constraints on other pairing channels.

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APPENDIX: EFFECTIVE THEORY FOR THE A–B TRANSITION

1. $PrOs_4Sb_{12}$

For the 3D $T_{1g/u}$ irreducible representation, we have two possible time-reversal symmetric A states: the (1,0,0) state with $D_2(C_2) \times \mathscr{T}$ symmetry and the (1,1,1) state with $C_3 \times \mathscr{T}$ symmetry. Both of these allow for a transition to a time-reversal symmetry-breaking B state which is Kerr active and hence provides two viable channels for the transition. In the case of $(1 + \tilde{\eta}_{1I}, 0 + \tilde{\eta}_{2I}, 0 + \tilde{\eta}_{3I})$, $\tilde{\eta}_{1I}$ belongs to the A_1 Kerr inactive irreducible representation and hence is not considered, while $\tilde{\eta}_{2I}$ and $\tilde{\eta}_{3I}$ belong to the Kerr active B_1 and B_2 irreducible representations, respectively. The form of the free energy will look similar to the 1D A–B transition in UPt_3 and the exact transition would depend on which irreducible representation has a higher T_c. The free energy will be

$$\beta f_{A\to B} = \alpha_I\tilde{\eta}_{2/3I}^2 + \beta_I\tilde{\eta}_{2/3I}^4. \quad (A1)$$

For the $(1 + \tilde{\eta}_{1I}, 1 + \tilde{\eta}_{2I}, 1 + \tilde{\eta}_{3I})$ state we have a 1D A_1 Kerr inactive order parameter, defined as $\eta_A = \tilde{\eta}_1 + \tilde{\eta}_2 + \tilde{\eta}_3$ and which is ignored, and a 2D Kerr active E order parameter, defined as

$$\eta_x = \frac{1}{\sqrt{6}}(2\tilde{\eta}_3 - \tilde{\eta}_2 - \tilde{\eta}_3), \quad \eta_y = \frac{1}{\sqrt{2}}(\tilde{\eta}_1 - \tilde{\eta}_2), \tag{A2}$$

where η_x and η_y belong to the 2D Kerr active E irreducible representation of C_3. The free energy for this E irreducible representation is

$$\beta f^E_{A\to B} = \alpha_I \eta_i^2 + \beta_{1I}\left(\eta_i^2\right)^2 + \gamma\left(\eta_i^2\right)^3 + \gamma_1\eta_{xI}^2\left(\eta_{xI}^2 - 3\eta_{yI}^2\right)^2 + \gamma_2\eta_{yI}^2\left(3\eta_{xI}^2 - \eta_{yI}^2\right)^2 + \gamma_3\eta_{xI}\eta_{yI}\left(\eta_{xI}^2 - 3\eta_{yI}^2\right) \times\left(3\eta_{xI}^2 - \eta_{yI}^2\right). \tag{A3}$$

Once α_I changes sign the ground state will be $\eta = \eta_I(\cos\theta, \sin\theta)$, where the value of θ will depend on the value of the coefficients (γ_i) of the sixth-order terms and hence provides a viable mechanism for a transitions to a time-reversal symmetry-breaking B phase.

2. $U_{1-x}Th_xBe_{13}$

For the 2D $E_{g/u}$ irreducible representation there are two possible time-reversal symmetric A states. We model the A–B transition as done before, with the A state being (1,0) possessing $D_4 \times \mathscr{T}$ symmetry and (0,1) with $D_4^{(1)}(D_2) \times \mathscr{T}$ symmetry [50]. The B state grows as $(1 + \tilde{\eta}_{1I}, 0 + \tilde{\eta}_{2I})$, where $\tilde{\eta}_{1I}$ belongs to the A_1 and $\tilde{\eta}_{2I}$ belongs to the B_1 irreducible representation of D_4, while for the (0,1) state, the situation is reversed. In that case the B state grows as $(0 + \tilde{\eta}_{1I}, 1 + \tilde{\eta}_{2I})$, with $\tilde{\eta}_{1I}$ having B_1 symmetry and $\tilde{\eta}_{2I}$ belonging to the A_1 irreducible representation of D_4. The free energy will be the same as for the 1D order parameters

$$\beta f_{A\to B} = \alpha_I \tilde{\eta}^2_{1/2I} + \beta_I \tilde{\eta}^4_{1/2I}. \tag{A4}$$

Interestingly due the absence of a Kerr measurement experiment, the A_1 irreducible representation is viable and the same physics applies here as discussed for the $s + is$ states in the iron-based superconductors [57–60]. These states are Kerr inactive and can be ruled out depending on the results of a trainable polar Kerr measurement in $U_{1-x}Th_xBe_{13}$.

For the 3D irreducible representations $T_{1g/u}$ and $T_{2u/g}$ there are four possible time-reversal symmetric A states, two for each T irreducible representation, i.e., (1,0,0) and (1,1,1). However, as explained in the main text, the (1,0,0) is the only viable time-reversal symmetric A state which allows for a transition to a time-reversal symmetry-breaking B state, due to the form of the coupling between superconductivity and antiferromagnetic fluctuations. This state has $D_4(C_4) \times \mathscr{T}$ and $D_4^{(2)}(D_2) \times \mathscr{T}$ symmetries for the $T_{1g/u}$ and $T_{2g/u}$ irreducible representations, respectively [50]. The A–B transition is modeled similarly to $PrOs_4Sb_{12}$, i.e., $(1 + \tilde{\eta}_{1I}, 0 + \tilde{\eta}_{2I}, 0 + \tilde{\eta}_{3I})$, except here $\tilde{\eta}_{1I}$ belongs to the A_1 irreducible representation of D_4 and would again have the $s + is$ physics with the standard free energy for 1D irreducible representations

$$\beta f_{A\to B} = \alpha_I \tilde{\eta}^2_{1I} + \beta_I \tilde{\eta}^4_{2I}, \tag{A5}$$

while $(\tilde{\eta}_{2I}, \tilde{\eta}_{3I})$ belong to the 2D Kerr active E irreducible representation of D_4. The free energy for the E irreducible representation is

$$\beta f^E_{A\to B} = \alpha_I \tilde{\eta}_i\tilde{\eta}_i + \beta_{1I}(\tilde{\eta}_i\tilde{\eta}_i)^2 + \beta_{2I}\tilde{\eta}^2_{2I}\tilde{\eta}^2_{3I}. \tag{A6}$$

For $\beta_2 > 0$ we pick the $\eta_I(1, 0)$ state and for $\beta_2 < 0$ the $\eta_I(1, 1)$ state, both of which break time-reversal symmetry.

[1] X.-L. Qi and S.-C. Zhang, Topological insulators and superconductors, Rev. Mod. Phys. **83**, 1057 (2011).

[2] M. Sato and Y. Ando, Topological superconductors: A review, Rep. Prog. Phys. **80**, 076501 (2017).

[3] A. P. Schnyder, S. Ryu, A. Furusaki, and A. W. W. Ludwig, Classification of topological insulators and superconductors in three spatial dimensions, Phys. Rev. B **78**, 195125 (2008).

[4] N. Read and D. Green, Paired states of fermions in two dimensions with breaking of parity and time-reversal symmetries and the fractional quantum hall effect, Phys. Rev. B **61**, 10267 (2000).

[5] E. R. Schemm, W. J. Gannon, C. M. Wishne, W. P. Halperin, and A. Kapitulnik, Observation of broken time-reversal symmetry in the heavy-fermion superconductor UPt_3, Science **345**, 190 (2014).

[6] E. M. Levenson-Falk, E. R. Schemm, Y. Aoki, M. B. Maple, and A. Kapitulnik, Polar Kerr Effect from Time-Reversal Symmetry Breaking in the Heavy-Fermion Superconductor $PrOs_4Sb_{12}$, Phys. Rev. Lett. **120**, 187004 (2018).

[7] E. R. Schemm, R. E. Baumbach, P. H. Tobash, F. Ronning, E. D. Bauer, and A. Kapitulnik, Evidence for broken time-reversal symmetry in the superconducting phase of URu_2Si_2, Phys. Rev. B **91**, 140506(R) (2015).

[8] Y. Kasahara, T. Iwasawa, H. Shishido, T. Shibauchi, K. Behnia, Y. Haga, T. D. Matsuda, Y. Onuki, M. Sigrist, and Y. Matsuda, Exotic Superconducting Properties in the Electron-Hole-Compensated Heavy-Fermion "Semimetal" URu_2Si_2, Phys. Rev. Lett. **99**, 116402 (2007).

[9] R. H. Heffner, J. L. Smith, J. O. Willis, P. Birrer, C. Baines, F. N. Gygax, B. Hitti, E. Lippelt, H. R. Ott, A. Schenck *et al.*, New Phase Diagram for $(U,Th)Be_{13}$: A Muon-Spin-Resonance and HC1 Study, Phys. Rev. Lett. **65**, 2816 (1990).

[10] Y. Yanase and K. Shiozaki, Möbius topological superconductivity in UPt_3, Phys. Rev. B **95**, 224514 (2017).

[11] V. Kozii, J. W. F. Venderbos, and L. Fu, Three-dimensional Majorana fermions in chiral superconductors, Sci. Adv. **2**, e1601835 (2016).

[12] P. Goswami and L. Balicas, Topological properties of possible Weyl superconducting states of URu_2Si_2, arXiv:1312.3632.

[13] D. F. Agterberg, P. M. R. Brydon, and C. Timm, Bogoliubov Fermi Surfaces in Superconductors with Broken Time-Reversal Symmetry, Phys. Rev. Lett. **118**, 127001 (2017).

[14] G. M. Luke, A. Keren, L. P. Le, W. D. Wu, Y. J. Uemura, D. A. Bonn, L. Taillefer, and J. D. Garrett, Muon Spin Relaxation in UPt_3, Phys. Rev. Lett. **71**, 1466 (1993).

[15] R. Joynt and L. Taillefer, The superconducting phases of UPt_3, Rev. Mod. Phys. **74**, 235 (2002).
[16] H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phase transition in the superconducting state of $U_{1-x}Th_xBe_{13}$ ($x = 0$–0.06), Phys. Rev. B **31**, 1651 (1985).
[17] R. J. Zieve, R. Duke, and J. L. Smith, Pressure and linear heat capacity in the superconducting state of thoriated UBe_{13}, Phys. Rev. B **69**, 144503 (2004).
[18] Y. Aoki, A. Tsuchiya, T. Kanayama, S. R. Saha, H. Sugawara, H. Sato, W. Higemoto, A. Koda, K. Ohishi, K. Nishiyama *et al.*, Time-Reversal Symmetry-Breaking Superconductivity in Heavy-Fermion $PrOs_4Sb_{12}$ Detected by Muon-Spin Relaxation, Phys. Rev. Lett. **91**, 067003 (2003).
[19] S. Adenwalla, S. W. Lin, Q. Z. Ran, Z. Zhao, J. B. Ketterson, J. A. Sauls, L. Taillefer, D. G. Hinks, M. Levy, and B. K. Sarma, Phase Diagram of UPt_3 from Ultrasonic Velocity Measurements, Phys. Rev. Lett. **65**, 2298 (1990).
[20] D. W. Hess, T. A. Tokuyasu, and J. A. Sauls, Broken symmetry in an unconventional superconductor: A model for the double transition in UPt_3, J. Phys.: Condens. Matter **1**, 8135 (1989).
[21] K. Machida, M.-a. Ozaki, and T. Ohmi, Unconventional superconducting class in a heavy Fermion system UPt_3, J. Phys. Soc. Jpn. **58**, 4116 (1989).
[22] G. Aeppli, E. Bucher, C. Broholm, J. K. Kjems, J. Baumann, and J. Hufnagl, Magnetic Order and Fluctuations in Superconducting UPt_3, Phys. Rev. Lett. **60**, 615 (1988).
[23] Y. Koike, N. Metoki, N. Kimura, E. Yamamoto, Y. Haga, Y. Onuki, and K. Maezawa, Long range antiferromagnetic ordering observed below 20 mK in the heavy fermion superconductor UPt_3, J. Phys. Soc. Jpn. **67**, 1142 (1998).
[24] W. J. Gannon, W. P. Halperin, M. R. Eskildsen, P. Dai, U. B. Hansen, K. Lefmann, and A. Stunault, Spin susceptibility of the topological superconductor UPt_3 from polarized neutron diffraction, Phys. Rev. B **96**, 041111(R) (2017).
[25] K. Kuwahara, K. Iwasa, M. Kohgi, K. Kaneko, N. Metoki, S. Raymond, M.-A. Méasson, J. Flouquet, H. Sugawara, Y. Aoki *et al.*, Direct Observation of Quadrupolar Excitons in the Heavy-Fermion Superconductor $PrOs_4Sb_{12}$, Phys. Rev. Lett. **95**, 107003 (2005).
[26] A. Hiess, R. H. Heffner, J. E. Sonier, G. H. Lander, J. L. Smith, and J. C. Cooley, Neutron elastic and inelastic scattering investigations of $U_{0.965}Th_{0.035}Be_{13}$, Phys. Rev. B **66**, 064531 (2002).
[27] A. J. Leggett, A theoretical description of the new phases of liquid ^{3}He, Rev. Mod. Phys. **47**, 331 (1975).
[28] D. Vollhardt and P. Wölfle, *The Superfluid Phases of Helium 3* (Courier, North Chelmsford, 2013).
[29] R. Balian and N. R. Werthamer, Superconductivity with pairs in a relative p wave, Phys. Rev. **131**, 1553 (1963).
[30] P. W. Anderson and W. F. Brinkman, Anisotropic Superfluidity in ^{3}He: A Possible Interpretation of Its Stability as A Spin-Fluctuation Effect, Phys. Rev. Lett. **30**, 1108 (1973).
[31] W. F. Brinkman, J. W. Serene, and P. W. Anderson, Spin-fluctuation stabilization of anisotropic superfluid states, Phys. Rev. A **10**, 2386 (1974).
[32] Y. Kuroda, Paramagnon and the BCS state in He^3. II, Prog. Theor. Phys. **53**, 349 (1975).
[33] M. Kardar, *Statistical Physics of Fields* (Cambridge University Press, Cambridge, 2007).
[34] L. P. Gor'kov, Superconductivity in heavy fermion systems, Sov. Sci. Rev. A Phys. **9**, 1 (1987).
[35] V. P. Mineev and K. Samokhin, *Introduction to Unconventional Superconductivity* (CRC, Boca Raton, 1999).
[36] C. H. Choi and P. Muzikar, Gradient free energy and intrinsic angular momentum in unconventional superconductors, Phys. Rev. B **40**, 5144 (1989).
[37] M. B. Walker and K. V. Samokhin, Model for Superconductivity in Ferromagnetic $ZrZn_2$, Phys. Rev. Lett. **88**, 207001 (2002).
[38] V. Kozii, H. Isobe, J. W. F. Venderbos, and L. Fu, Nematic superconductivity stabilized by density wave fluctuations: Possible application to twisted bilayer graphene, Phys. Rev. B **99**, 144507 (2019).
[39] G. R. Stewart, Z. Fisk, J. O. Willis, and J. L. Smith, Possibility of Coexistence of Bulk Superconductivity and Spin Fluctuations in UPt_3, Phys. Rev. Lett. **52**, 679 (1984).
[40] M. Sigrist and K. Ueda, Phenomenological theory of unconventional superconductivity, Rev. Mod. Phys. **63**, 239 (1991).
[41] Y. Koike, N. Metoki, N. Kimura, E. Yamamoto, Y. Haga, Y. Ōnuki, and K. Maezawa, Neutron scattering study of the antiferromagnetic ordering in UPt_3 at mK-temperatures, Physica B **259**, 662 (1999).
[42] E. A. Schuberth, B. Strickler, and K. Andres, Specific-Heat Anomaly in Superconducting UPt_3 at 18 mK, Phys. Rev. Lett. **68**, 117 (1992).
[43] E. A. Schuberth, Some heavy fermion systems at very low temperatures, Int. J. Mod. Phys. B **10**, 357 (1996).
[44] A. Sawada, T. Kubo, Y. Fujii, T. Komatsubara, Y. Ònuki, N. Kimura, E. Yamamoto, and Y. Haga, Thermal expansion anomaly of UPt_3 below 70mK, Czech. J. Phys. **46**, 803 (1996).
[45] S. Schöttl, E. A. Schuberth, K. Flachbart, J. B. Kycia, J. I. Hong, D. N. Seidman, W. P. Halperin, J. Hufnagl, and E. Bucher, Anisotropic Dc Magnetization of Superconducting UPt_3 and Antiferromagnetic Ordering Below 20 mK, Phys. Rev. Lett. **82**, 2378 (1999).
[46] Y. Kitaoka, H. Tou, K. Ishida, N. Kimura, Y. Ōnuki, E. Yamamoto, Y. Haga, and K. Maezawa, NMR studies on UPt_3, Physica B **281**, 878 (2000).
[47] N. H. Van Dijk, P. Rodière, B. Fåk, A. Huxley, and J. Flouquet, Weak antiferromagnetic order and superconductivity in UPt_3 studied by neutron scattering, Physica B **319**, 220 (2002).
[48] Y. M. Gufan, Superconducting classes generated by a single irreducible multidimensional representation, Zh. Éksp. Teor. Fiz. **107**, 855 (1995) [Sov. J. Exp. Theor. Phys. **80**, 485 (1995)].
[49] J.-C. Toledano and P. Toledano, *The Landau Theory of Phase Transitions: Application to Structural, Incommensurate, Magnetic and Liquid Crystal Systems* (World Scientific, Singapore, 1987), Vol. 3.
[50] G. E. Volovik and L. P. Gor'kov, Superconducting classes in heavy-fermion systems, Sov. Phys. JETP **61**, 843 (1985).
[51] R. Vollmer, A. Faißt, C. Pfleiderer, H. v. Löhneysen, E. D. Bauer, P.-C. Ho, V. Zapf, and M. B. Maple, Low-Temperature Specific Heat of the Heavy-Fermion Superconductor $PrOs_4Sb_{12}$, Phys. Rev. Lett. **90**, 057001 (2003).
[52] I. A. Sergienko and S. H. Curnoe, Superconducting states in the tetrahedral compound $PrOs_4Sb_{12}$, Phys. Rev. B **70**, 144522 (2004).
[53] K. Kaneko, N. Metoki, R. Shiina, T. D. Matsuda, M. Kohgi, K. Kuwahara, and N. Bernhoeft, Neutron scattering study on

the field-induced O_{xy}-type antiferroquadrupolar ordering of the heavy-fermion superconductor $PrOs_4Sb_{12}$, Phys. Rev. B **75**, 094408 (2007).

[54] R. Shiina, Multipolar moments in Pr-based filled-skutterudite compounds with singlet-triplet crystal-field levels, J. Phys. Soc. Jpn. **73**, 2257 (2004).

[55] R. Shiina and Y. Aoki, Theory of field-induced phase transition in $PrOs_4Sb_{12}$, J. Phys. Soc. Jpn. **73**, 541 (2004).

[56] S. Mukherjee and D. F. Agterberg, Microscopic theories for cubic and tetrahedral superconductors: Application to $PrOs_4Sb_{12}$, Phys. Rev. B **74**, 174505 (2006).

[57] B. Mettout, P. Tolédano, and V. Lorman, Unconventional *s*-Pairing Wave Superconductivity, Phys. Rev. Lett. **77**, 2284 (1996).

[58] J. Carlström, J. Garaud, and E. Babaev, Length scales, collective modes, and type-1.5 regimes in three-band superconductors, Phys. Rev. B **84**, 134518 (2011).

[59] S. Maiti and A. V. Chubukov, $s + is$ state with broken time-reversal symmetry in fe-based superconductors, Phys. Rev. B **87**, 144511 (2013).

[60] J. Garaud, A. Corticelli, M. Silaev, and E. Babaev, Properties of dirty two-band superconductors with repulsive interband interaction: Normal modes, length scales, vortices, and magnetic response, Phys. Rev. B **98**, 014520 (2018).