Staggered Superconductivity in UPt$_3$: A New Phenomenological Approach

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**Abstract**

We present a new Ginzburg-Landau theory for superconductivity in UPt$_3$, based upon a multicomponent order parameter transforming under an irreducible space group representation; the phase is staggered in real space. Our model can explain the $H - T - P$ phase diagram including the tetracritical point for all field directions. We motivate this unconventional superconducting state in terms of odd-in-time-reversal pairing that may arise in one- or two-channel Kondo models, and suggest experimental tests.

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Heavy fermion superconductors have continued to attract attention since their discovery in 1979 [1]. In these materials, fermionic excitations with effective masses hundreds of times the free electron mass undergo a pairing transition. Anomalous power laws in low temperature thermodynamic and transport properties [1] together with complex phase diagrams suggest interpretation in terms of Cooper pairs with exotic symmetries. For example in the magnetic field-temperature $H - T$ plane for ambient pressure $P \approx 1$ bar, the hexagonal material UPt$_3$ apparently possesses three distinct superconducting phases, shown as A, B, C in Fig. 1 [2]. In the $P - T$ plane for zero $H$ there are similarly three observed phases [3]. Intriguingly, the B phase in UPt$_3$ appears to break time reversal symmetry $\mathcal{T}$ in that muon spin rotation spectroscopy reveals the development of tiny additional magnetic
moments (order 0.001$\mu_B$) \[4\]. So far, no comprehensive microscopic theory exists for these materials and so much theoretical effort has been devoted to phenomenological approaches. No such approach to date has successfully explained UPt$_3$ without substantial fine tuning of parameters.

In this Letter, we propose a new model for the phase diagram of UPt$_3$ in which finite center of mass momentum (FCM) pairs give rise to an order parameter which transforms as a triplet under the operations of the full hexagonal space group. Our model produces a plausible description of the $H−P−T$ superconducting phase diagram. In particular, we can explain the tetracritical point found for all field orientations. While the Ginzburg-Landau free energy contains numerous parameters, experiment constrains the zero field values to a narrow region which allows for a $T$-breaking $B$ phase. The crystalline structure of UPt$_3$ plays a crucial role in our considerations. The proposed superconducting order parameters are microscopically motivated in terms of odd-in-$T$ pairing \[5\] such as may arise in the two-channel Kondo model \[6,7\] suggested for heavy fermion materials \[8\]. Such pairs may generically give rise to negative pair hopping energy favoring staggered superconducting states \[9\]. We shall discuss data supporting this odd-in-$T$ pairing and point out crucial tests of our model as well as some of its limitations. We note that our theory is applicable only near $T_c$ and $H_{c2}$, and is macroscopic in character. Explanation of the low temperature power laws awaits the development of a suitable microscopic approach.

To place our work in context, we first discuss the $E$ doublet model \[10\]. In this picture, the Cooper pairs have zero center of mass momentum (ZCM), and are described by basis functions $\phi_a(\vec{k}), \phi_b(\vec{k})$ which form a closed vector space under the symmetry operations of the hexagonal space group. With each basis function is associated an order parameter amplitude $\eta_{a,b}$, defined through the gap function $\Delta(\vec{k}) \sim \eta_a\phi_a(\vec{k}) + \eta_b\phi_b(\vec{k})$. The phenomenological approach consists of writing down and minimizing a Ginzburg-Landau free energy which is an expansion in the $\eta$ amplitudes. A zero field splitting is present which most likely originates in the orthorhombic strain induced by magnetic order. Then the three phases have $\eta_a \neq 0$, $\eta_b = 0$ ($A$-phase), $\eta_a \neq 0$, $\eta_b \neq 0$ ($B$-phase), $\eta_a = 0$, $\eta_b \neq 0$ ($C$-phase). The apparent
time reversal breaking in the $B$-phase is explained by suitable a choice of Ginzburg-Landau parameters which energetically favor a relative phase factor of $i$ between $\eta_a, \eta_b$. The $E$ model has a flaw: while the data show a tetracritical point for all field orientations, a tetracritical point generically arises only for in-plane magnetic fields [11]. This problem is remedied by fine tuning to zero the gradient term in the free energy which mixes the $a, b$ components [10]. As a possible alternative, the authors of Ref. [11] propose a model of accidentally nearly degenerate ZCM singlet states (DS model) which have no such gradient mixing term. While this can explain the tetracritical point for all field orientations, there is no compelling explanation for the accidental near degeneracy.

Our work proceeds via a different phenomenological route: by considering Cooper pairs with FCM, we are free to examine space group representations which merge the desirable properties of the $E$ doublet and DS models by having order parameter degeneracy and the absence of gradient mixing terms enforced by symmetry.

Before turning to the core of the paper, we shall briefly review the motivation for this work in terms of odd-in-$T$ pairing theory. For an odd in $T$ pair wave function it may be shown within a quasiparticle framework [5,9] that (i) for ZCM pairs, the “Meissner stiffness” which goes as $1/\lambda_L(T)^2$, $\lambda_L$ the London penetration depth, is negative; (ii) an immediate corollary is that the pair transfer energy between two odd-in-$T$ slabs, or Josephson coupling, is negative–alternatively, the coefficients of the gradient terms in the free energy are negative. To the extent that these properties are generically true (it is unknown whether a quasiparticle picture is applicable in all cases), we then expect a non-uniform superconducting state in which the pairs have FCM and the phase is modulated in real space. Indeed, the Majorana Fermion treatment of the ordinary Kondo lattice model favors just such a state, with the superconducting order parameter fully staggered, i.e., the pair hopping from site to site is negative [9].

There is so far no numerical evidence supporting this odd-in-$T$ picture for the single channel Kondo lattice model [12]. There are strong reasons to consider this state in the two-channel Kondo lattice model. In this model, two identical “channels” of spin 1/2 conduction
electrons interact antiferromagnetically with a lattice of spin 1/2 local moments. For this model it has been shown that (i) in the impurity limit, the pair field susceptibility for the odd-in-$T$ order parameter which is a singlet in spin and channel indices diverges as $-\ln(T)$, $T \to 0$, and (ii) all heavy fermion superconductors have appropriate symmetry and dynamical conditions to be describable by two-channel Kondo models \[8,13\]. For the uranium based materials a two-channel quadrupolar Kondo model can apply, with spin corresponding to local quadrupolar indices and the channel labels corresponding to conduction magnetic indices. The spin and channel singlet states transform as the $A_2$ representation for each point group relevant to the heavy fermion materials \[14\] ($A_2 \sim (x^3 - 3xy^2)(y^3 - 3yx^2)$ for the hexagonal point group).

Assuming that such local odd-in-$T$ pairs of $A_2$ point symmetry form at each site with negative pair hopping in the heavy fermion superconductors, we can determine the allowed multidimensional space group representations which describe the ordering in each compound. Note that such a description is reasonable only to the extent that the Kondo screening length is strongly reduced in the lattice relative to the impurity, which is in fact expected in the single channel Kondo lattice model \[15\]. In a hexagonal crystal with one 4f/5f atom per unit cell the pair energy minima will be at the two $K$ points of Fig. 2. For UPt$_3$, the crystal structure with two atoms per unit cell and interlayer frustration for antiferromagnetic (phase) couplings pushes the pair hopping minima away from the $K$ points, and for a wide range of parameter values they move to the $M$ points which have a three point star \[16\]. In addition, the presence of two atoms per unit cell gives rise to non-degenerate bonding and non-bonding combinations of pair orbitals within each cell. We assume that the non-bonding combination is sufficiently high in energy to be negligible in the current analysis.

We now turn to the discussion of a specific Ginzburg-Landau model for UPt$_3$ based upon the 3 dimensional $M$-point representation. While motivated by the odd-in-$T$ order parameter discussion above, we are not limited to this as a potential microscopic source for our phenomenology. Free energy invariants are constructed by standard group theoretical methods, and we have checked the completeness of our set using image group techniques \[17\].
We include a linear coupling to an orthorhombic strain field $\epsilon$, which breaks the hexagonal symmetry. The free energy density is given by

$$F = F_0 + F_{ab}^g + F_c^g$$

(1a)

$$F_0 = \alpha_+ |\eta_3|^2 + \alpha_- (|\eta_1|^2 + |\eta_2|^2) + \beta_1 \sum_n |\eta_n|^4$$

$$+ \beta_2 \sum_{n \neq m} |\eta_n|^2 |\eta_m|^2 + \beta_3 \sum_{n \neq m} \eta_n^2 \eta_m^2$$

(1b)

$$F_{ab}^g = \sum_n \{ \mu_n (|p_+ \eta_n|^2 + |p_- \eta_n|^2) + \mu'_n (|p_+ \eta_n|^2 - |p_- \eta_n|^2)$$

$$- |p_\pm \eta_n|^2) + (\nu_n (p_+ \eta_n)(p_- \eta_n)^* + c.c.) \}$$

(1c)

$$F_c^g = \sum_n \chi_n |p_z \eta_n|^2$$

(1d)

Here, $\eta_n$, $n=1,2,3$, are the order parameter components, $p_\pm = (p_x \pm ip_y)/\sqrt{2}$, $p_\alpha = -i \partial_\alpha - 2eA_\alpha/hc$ ($e < 0$), and $\alpha_\pm = a_0(T - T_\pm)$ with $T_+ = T_{c0} + 2\epsilon/a_0$ and $T_- = T_{c0} - \epsilon/a_0$. For the odd-in-$T$ order parameter considered in the frequency domain, the $\eta_i$ are understood to be the leading order coefficients of the expansion about $\omega = 0$ [5]. We will restrict further analysis to the case $\epsilon > 0$ ($T_+ > T_-$), which is a necessary condition to comply with the suggested $T$-breaking in phase B but not in phase A. The coefficients of the gradient terms are expanded to linear order in $\epsilon$ and are defined by a number of phenomenological coupling constants $\kappa_j$ through $\mu_n = \kappa_1 + \kappa_2 \epsilon \text{Re}(\gamma^n)$, $\mu'_n = \kappa_3 \epsilon \text{Im}(\gamma^n)$, $\nu_n = \kappa_4 \gamma^n + \kappa_5 \epsilon + \kappa_6 \epsilon \gamma^{-n}$, and $\chi_n = \kappa_7 + \kappa_8 \epsilon \text{Re}(\gamma^n)$. The phase factor $\gamma = \exp(i2\pi/3)$ reflects the three-fold symmetry of the star of M. Note that the last term in Eq. (1b) favors $T$-breaking solutions for $\beta_3 > 0$.

Model (1) accounts for a rich phase diagram already when $H = 0$. Fig. 3 summarizes the stability region of zero field solutions. Four regions in the parameter space can be distinguished according to their predicted phase sequence, as shown in Table 1 [20]. Most promising with regard to experiment are regions I and II. Both allow for two second order transitions at $T_+$ and $T_*(< T_+)$, where the second phase manifestly breaks $T$-symmetry and can be identified with phase B of Fig. 1b.

For region I, we recover the experimental $P - T$ diagram, when we assume a pressure dependent strain field, which decreases with increasing $P$ and vanishes for $P > P_c$ [21].
the other hand, a third low-temperature phase for finite $\epsilon$ is predicted for region II, which signals an unlocking of the relative phases of the order parameter components. Consequently, a fourth superconducting phase $D$ appears between $B$ and $C$, and all phase boundaries meet at a pentacritical point. Although this scenario seems to be at variance with Fig. 1b, we cannot immediately reject it because the $BD$ and $DC$ boundaries are predicted to be very close together rendering an observation of the fourth phase difficult. Estimates for the model parameters, as derived from experimental data (shaded area inside the region of stability in Fig. 3) do not unambiguously favor one of the two regions.

Let us now discuss the $H − T$ diagram predicted by model (1). Generally, the upper critical field $H_{c2}$ is determined by the smallest eigenvalue of the linearized Ginzburg-Landau equations, which can be seen as a coupled system of Schrödinger-like equations, with the order parameter playing the role of a multicomponent wavefunction. In our case, this leads to three decoupled equations, which makes it possible to obtain all eigenvalues in a closed analytic form for a general magnetic field. For $\mathbf{H} = H(\cos \varphi \sin \theta, \sin \varphi \sin \theta, \cos \theta)$, $H_{c2}$ is given by the largest of the three values

$$H_n(T) = \frac{\hbar c}{2|\epsilon|} \frac{-\alpha_n(T)}{\sqrt{R_n + \mu'_n \cos \theta}}$$

$$R_n = (\mu_n^2 - |\nu_n|^2) \cos^2 \theta + \chi_n(\mu_n - Re(\nu_ne^{i2\varphi})) \sin^2 \theta$$

(2)

where $\alpha_3 \equiv \alpha_+$ and $\alpha_1 = \alpha_2 \equiv \alpha_-.$

A tetracritical point in the $H − T$ plane corresponds to a crossing of the two largest values $H_n$ as a function of temperature. As mentioned by Garg [22], such a crossing necessarily requires the existence of a conserved quantity associated with the Schrödinger equations. For our model, this property follows immediately from the decoupling of the equations dictated by translational invariance, therefore allowing, in principle, a tetracritical point for all directions of the magnetic field. Within a one domain model, the almost complete isotropy of $H_{c2}$ in the basal plane requires that the in-plane anisotropic gradient terms are very small ($|\nu_n| \ll \mu_n$). For the extreme case $\nu_n = 0$, we find kinks in $H_{c2}$ for all field directions provided that $\kappa_1 > 0$, $\kappa_7 > 0$, $\kappa_2 + |\kappa_3|/\sqrt{3} > 0$, and $\kappa_2 \kappa_7 + \kappa_1 \kappa_8 > 0$ [24]. These
constraints do not represent strong limitations. Note, that the gradient terms proportional to $\epsilon$ are necessary for a tetracritical point to appear because for vanishing strain the slopes of the $H_n$ versus $T$ lines would be identical. As a consequence, the slopes of the low-field and high-field $H_{c2}$-lines ($AN$ and $CN$ phase boundaries in Fig. 1a) approach the same value for $\epsilon \to 0$, i.e. for $P \to P_c$. This holds for any field direction, and represents a qualitative difference from the predictions of the $E$ model, where the slope difference approaches a nonzero limit for in-plane fields and vanishes for c-axis fields.

Our model is further consistent with observed anisotropies of the penetration depth and $H_{c1}$, and predicts a kink of $H_{c1}$ at $T_*$ for all field directions because of an onset of an additional order parameter component below $T_*$.

The above given analysis of the phase diagram assumes a homogeneous symmetry breaking field. This one-domain picture is an oversimplification of the situation in UPt$_3$. Any orthorhombic field is very likely to build a six-fold domain structure, as it was observed for the antiferromagnetic order with domain sizes of less than 150 Å. Such an underlying domain structure can significantly alter the superconducting state [25]. This point deserves further investigation to allow a more reliable comparison with experiment.

Our model may resolve some additional phenomenological puzzles. Most notable is the absence of Josephson tunneling from conventional superconductors into heavy fermion materials, in particular for UPt$_3$ [26]. Assuming the superconductivity to arise from FCM pairs with odd-in-$T$ symmetry, no single pair Josephson coupling to a conventional even-in-$T$ superconductor is possible without applied or internal (spontaneous) magnetic field that breaks $T$. Even in this case the staggered phase across an arbitrary sample face will yield zero net pair tunneling unless: (i) one cleaves along planes perpendicular to the pairing $k$-vectors, or (ii) one uses atomic scale tunneling probes (e.g., STM), which can sample a small region of non-zero phase. This suggests a clear experimental test of the odd-in-$T$ hypothesis.

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REFERENCES

[1] P. A. Lee, et al., Comm. Cond. Matt. Phys. 12, 99 (1985); L. Gorkov, Soviet Sci. Reviews 9A, 1 (1987); N. Grewe and F. Steglich, in Handbook of the Physics and Chemistry of the Rare Earths, Vol. 14, eds. K. A. Gschneidner Jr. and L.i L. Eyring (Elsevier, Amsterdam, 1991), p. 343.

[2] G. Bruls et al., Phys. Rev. Lett. 65, 2294 (1990); S. Adenwella et al., Phys. Rev. Lett. 65, 2298 (1990).

[3] M. Boukhny et al. (to be published).

[4] G. M. Luke et al., Phys. Rev. Lett. 71, 1466 (1993).

[5] A. V. Balatsky and E. Abrahams, Phys. Rev. B 45, 13125 (1992); A. V. Balatsky et al., Physica B 199&200, 363 (1994).

[6] V. J. Emery and S. Kivelson, Phys. Rev. B 46, 10812 (1992).

[7] A. W. W. Ludwig, Physica B 199 & 200, 406 (1994); I. Affleck and A. W. W. Ludwig, preprint (1993).

[8] D. L. Cox, unpublished (1990); to be published (1994).

[9] P. Coleman, E. Miranda, and A. Tsevlik, Phys. Rev. B 49, 8955 (1994).

[10] J. A. Sauls, J. Low Temp. Phys. 95, 153 (1994); Advances in Physics (to be published).

[11] D.-C. Chen and A. Garg, Phys. Rev. Lett. 70, 1689 (1993).

[12] M. Jarrell, private communication (1994).

[13] D. L. Cox, Physica B 186-188, 312 (1993).

[14] D. L. Cox and A.W.W. Ludwig (to be published).

[15] A. J. Millis and P. A. Lee, Phys. Rev. B 35, 3394 (1986).
[16] Incommensurate 6- and 12-point star representations are also possible, but the free energies are actually simpler in form than that of the $M$-point.

[17] J.-C. Toledano and P. Toledano, *The Landau Theory of Phase Transitions* (World Scientific, Singapore, 1987), Ch. II, §4.5.

[18] R. A. Fischer *et al.*, Phys. Rev. Lett. **62**, 1411 (1989).

[19] K. Hasselbach, L. Taillefer, and J. Flouquet, Phys. Rev. Lett. **63**, 93 (1989).

[20] Parameter regions are derived without the more physical condition $T_{\pm} > 0$ (or $T_3 > 0$). Adding this constraint leads to a slight ($T_\pm$ and $\epsilon$ dependent) shift of the region boundaries.

[21] In this simple picture, the BC line is horizontal which would be at variance with ultrasound investigations [3]. However, by replacing the static strain field by a thermodynamic variable $M$ describing the AF-magnetic moment ($\epsilon \propto M^2$), the slope of the BC line can be modified significantly.

[22] A. Garg, Phys. Rev. Lett. **69**, 676 (1992).

[23] J. P. Brison *et al.*, J. Low. Temp. Phys. **95**, 145 (1994).

[24] Given conditions only guarantee a crossing of $H_{\epsilon 2}$ lines below $T_\epsilon$. Demanding a positive crossing temperature changes the constraints only slightly.

[25] R. Joynt *et al.*, Phys. Rev. B **42**, 2014 (1990).

[26] An exception is a weak link CeCu$_2$Si$_2$/Al junction; see U. Poppe, J. Mag. Mag. Mat. **52**, 157 (1985).
TABLES

TABLE I. Order parameters for stable phases of model (1) for $H = 0$ depending on the parameter regions I-IV (cf. Fig. 3). Results apply for a constant (static) strain field $\epsilon$. Phase 1 appears at $T_+$, phase 2 at $T_* = T_+ - \beta_1/(\beta_1 - \beta_2 - \beta_3)(T_+ - T_-)$, $T_* < T_+$, and a possible third phase may exist below $T_3 = T_+ - 2(\beta_1 + \beta_2 - \beta_3)/(\beta_1 - \beta_2 - \beta_3)(T_+ - T_-)$, $T_3 < T_*$. $x$ and $\phi$ are temperature dependent real numbers. $\epsilon = 0$ corresponds to the high-pressure phase (Fig. 1b).

| region | $\epsilon > 0$ | $\epsilon = 0$ |
|--------|----------------|----------------|
|        | phase 1 | phase 2 | phase 3 |                  |
| I      | (0 0 1) | (0 ix 1) | -       | (0 1 i)          |
| II     | (0 0 1) | (ix ix 1) | $(e^{i\phi}x e^{-i\phi}x 1)$ | $(e^{i2\pi/3} e^{-i2\pi/3} 1)$ |
| III    | (0 0 1) | -       | -       | (0 0 1)          |
| IV     | (0 0 1) | (x x 1) | -       | (1 1 1)          |
FIGURES

FIG. 1. Schematic phase diagram of UPt$_3$ in the $P = 0$ and $H = 0$ planes as deduced from experiments [2]. Areas supposed to belong to the same phase in the three-dimensional $H$-$P$-$T$ diagram are given the same letter.

FIG. 2. $k_z = 0$ cross-section of the Brillouin zone of UPt$_3$. $k_1$ are wave vectors of the star of the M-point representation used in our model.

FIG. 3. Region of stability for solutions of model (1) for $H = 0$ as a function of Ginzburg-Landau parameters for $\beta_1 > 0$. The free energy has no lower bound in the unstable region, which holds also for $\beta_1 < 0$. The shaded strip marks the region consistent with specific heat measurements [18,19] (dotted lines: the specific heat jump ratio, $r$, at $T_*$ and $T_+$ with respect to the normal state, $1.25 < r < 1.33$), estimates for $T_-$ from $H$_{c2}-measurements [2] (dashed lines; $x = (T_+ - T_-)/(T_+ - T_*), 0.2 < x < 0.45$) and the assumption that only phase B violates $\mathcal{T}$-invariance. Characteristic phase diagrams found in regions (I)-(IV) are described in the text and in Table 1.
Fig. 1

(a) P=0

(b) H=0

NN
Fig. 2
