

Accidental Near Degeneracy of the Order Parameter for Superconducting UPt_3

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UPt_3 appears to have a tetracritical point in the H - T plane for all orientations $\hat{\mathbf{H}}$ of \mathbf{H} . Such a point can exist only if the Ginzburg-Landau (GL) "Hamiltonian" has a conserved quantity Q . It is shown that the model of a two-component vector order parameter admits a Q for $\mathbf{H}\parallel\mathbf{c}$, but has no tetracritical point. Hence, accidentally nearly degenerate order parameters are examined, particularly the model where one behaves as an A and the other as a B representation of D_{6h} . This model can yield a tetracritical point for all $\hat{\mathbf{H}}$. Its physical properties are calculated and GL parameters are fitted to experiment.

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The finding of multiple superconducting phases in the heavy-electron material UPt_3 [1-4] makes it almost certain that it is an infrasympmetric superconductor [5]. The field-temperature (H - T) phase diagram is particularly interesting; for all orientations of \mathbf{H} studied so far, three vortex phases appear to coexist at a tetracritical point, and the inner phase boundaries $H_{ci}(T)$ lie below the extensions of the outer, i.e., $H_{c2}(T)$, lines [2]. These features are clearest for $\mathbf{H}\perp\mathbf{c}$, and less so for $\mathbf{H}\parallel\mathbf{c}$ and for \mathbf{H} at 45° to \mathbf{c} . Thus, although it can be argued that the phase boundaries merely approach each other very closely without actually meeting for the latter two cases, the natural assumption at this stage of experimental development is that there is a tetracritical point for *all* field orientations.

In this Letter we shall argue that the existence of a tetracritical point puts very strong constraints on the order parameter. We shall work within the framework of Ginzburg-Landau (GL) theory. In a previous paper by one of us [6], it was shown that since H_{c2} is determined by the smallest eigenvalue of the operator (which we will call a GL "Hamiltonian," \mathcal{H}_{GL}) in the linearized GL equations, one should think of the tetracritical point as a point of intersection of two smooth energy curves of \mathcal{H}_{GL} that vary with some parameter. One knows from quantum mechanics that such an intersection cannot occur unless there is some conserved quantity Q that commutes with \mathcal{H}_{GL} , and unless the two intersecting curves belong to different Q eigenvalues. The first task in understanding the tetracritical point is thus the identification of a Q .

The GL theory studied in Ref. [6] is based on a two-component order parameter $\boldsymbol{\eta}=(\eta_x,\eta_y)$, which covers any of the four two-dimensional representations of D_{6h} , the point group of UPt_3 . There is also a weak symmetry breaking field to account for the split transition [4] when $H=0$. This has been a popular model for UPt_3 [7-9]. It was shown in Ref. [6] that the corresponding GL Hamiltonian \mathcal{H}_{GL} does indeed admit conserved quantities Q_{\parallel} and Q_{\perp} , for both $\mathbf{H}\parallel\mathbf{c}$ and $\mathbf{H}\perp\mathbf{c}$ [10], but the tetracritical point itself was only analyzed for $\mathbf{H}\perp\mathbf{c}$. There is no Q for other orientations $\hat{\mathbf{H}}$ of \mathbf{H} , and it was pointed out that careful studies of the phase diagram for nonspecial

$\hat{\mathbf{H}}$ would provide a discriminating test of this model. See also Ref. [11].

In this Letter we point out a serious difficulty with this model. We find that although there is a Q_{\parallel} , the lowest energy curve (which yields H_{c2}) never intersects with any other [12]. (There are plenty of intersections among the higher energy curves.) Thus a strict tetracritical point is impossible. We argue below that it is difficult to amend the model so as to lead to a "weakly" avoided crossing. The only other way to get multiple superconducting phases is to have two accidentally nearly degenerate order parameters η_a and η_b , transforming as different representations of D_{6h} . We therefore search for two representations that can yield a crossing for all $\hat{\mathbf{H}}$. One possibility is for η_a and η_b to have different parity, in which case they can have any rotational symmetry. The other is for η_a and η_b to have the same parity, in which case we find that one of them must belong to an A representation (A_1 or A_2), and the other to a B representation (B_1 or B_2). We study the second case (which we call the "AB" model) in detail, and obtain formulas for specific heat jumps when $H=0$, H_{c1} , H_{c2} , and the slopes of the inner transition lines [13].

Note that although both order parameters in the AB model are one dimensional, at least one must not transform as the identity, A_{1g} , so in our terminology it still represents infrasympmetric superconductivity. The combined order parameter can still have nodes that may be detectable either by power-law behavior of thermodynamic and transport quantities, or by the anisotropy of the nonlinear Meissner effect [14]. If the A representation is A_1 , i.e., either A_{1g} or A_{1u} , there are no nodes required by symmetry. If it is A_{2g} (and the other is therefore B_{1g} or B_{2g}), then there are line nodes along three meridional "great circles" and six additional point nodes along the "equator." (The Fermi surface is not implied to be a sphere.) If it is A_{2u} (and the other is therefore B_{1u} or B_{2u}), then there are only two point nodes along the c axis.

It is worth noting here that recent work [15] appears to directly tie the zero field splitting to the existence of weak basal plane antiferromagnetism in UPt_3 , in that both

disappear under hydrostatic pressures of about 4 kbar. These data seem at first to disfavor the AB model and to support the two-component model, with the antiferromagnetism providing the symmetry breaking field [7,8]. The implied p - T plane phase boundaries pose severe thermodynamic problems, however; the Néel line $T_N(p)$ ends in midplane, which is impermissible, as is the junction of three second-order lines [the superconducting $T_c(p)$'s] with nonzero specific heat jumps [16], so that in our view, these data cannot be regarded as conclusive. We shall, therefore, focus only on the better understood H - T phase diagram in this Letter.

We first consider the two-component model. To fourth order in the η 's the GL free energy density is given by

$$f_{GL} = \alpha_+ |\eta_y|^2 + \alpha_- |\eta_x|^2 + \beta_1 (\boldsymbol{\eta} \cdot \boldsymbol{\eta}^*)^2 + \beta_2 |\boldsymbol{\eta} \cdot \boldsymbol{\eta}|^2 + \kappa_1 p_i^* \eta_j^* p_i \eta_j + \kappa_2 p_i^* \eta_i^* p_j \eta_j + \kappa_3 p_i^* \eta_j^* p_j \eta_i + \kappa_4 p_z^* \eta_i^* p_z \eta_i + h^2/8\pi, \quad (1)$$

where $i, j = x, y$, $p_i = -i\partial_i - 2eA_i/\hbar c$, \mathbf{A} and \mathbf{h} are the local vector potential and magnetic field, and $\alpha_{\pm} = \alpha_0(T - T_{c\pm})$, $T_{c\pm} = T_{c0} \pm \epsilon/\alpha_0$, with $T_{c0} \gg \epsilon/\alpha_0$, the symmetry breaking perturbation. We require $\beta_1 > 0$ for stability, and $\beta_2 > 0$ to get two transitions in zero field [8(a)].

To cast the eigenvalue equation for H_{c2} into dimensionless form, we scale all lengths by the magnetic length $l = (\hbar c/2eH)^{1/2}$, and define

$$\kappa_{23} = \kappa_2 + \kappa_3, \quad \kappa_b = \kappa_1 + (\kappa_{23}/2), \quad u = \kappa_{23}/2\kappa_b,$$

$$v = (\kappa_2 - \kappa_3)/2\kappa_b, \quad \tilde{\epsilon} = \epsilon l^2/\kappa_b, \quad E = \alpha_0(T_{c0} - T)l^2/\kappa_b.$$

For the case $\mathbf{H}\parallel c$, writing $\eta_{\pm} = (\eta_x \mp i\eta_y)/\sqrt{2}$ and $a = (p_x + ip_y)/\sqrt{2}$, we get

$$\begin{pmatrix} 2a^\dagger a + 1 - v & 2ua^\dagger a + \tilde{\epsilon} \\ 2ua^2 + \tilde{\epsilon} & 2a^\dagger a + 1 + v \end{pmatrix} \begin{pmatrix} \eta_+ \\ \eta_- \end{pmatrix} = E \begin{pmatrix} \eta_+ \\ \eta_- \end{pmatrix}. \quad (2)$$

We have also set $p_z \boldsymbol{\eta} = 0$ as the energy is raised by taking $p_z \boldsymbol{\eta} \neq 0$. The conserved quantity is $Q_{\parallel} = e^{ina^\dagger a}$, the parity of the Landau level number. If (as is not unlikely [17]) the ground state is homogeneous when $H = 0$, the gradient terms in f_{GL} must be separately positive definite, which implies that $\kappa_b > 0$, $\kappa_4 > 0$, and that u and v lie in the triangular region [18]

$$1 - v > 0, \quad 1 + v > 2|u|. \quad (3)$$

Our goal is to find the eigenvalue spectrum $\{E(\tilde{\epsilon})\}$ when $\epsilon \neq 0$ for fixed u and v constrained by Eqs. (3), and in particular to see if the lowest energy solution has different parities for low and high $\tilde{\epsilon} \sim 1/H$. In the infinite field limit $\tilde{\epsilon} = 0$, it is easy to solve Eq. (2) exactly, and one can show that the lowest two energy states have even parity [19]. In the low field limit $\tilde{\epsilon} \rightarrow \infty$, it is better to use the η_x, η_y basis, and do perturbation theory in $1/\tilde{\epsilon}$. To lowest order the η_x and η_y subspaces separate, and the lowest state is the zeroth Landau level for η_y , which also has even parity. An odd-parity curve must thus intersect the lowest even-parity curve either never or at least twice.

The latter is unlikely, but to rule it out, we have solved for the energies $\{E(\tilde{\epsilon})\}$ numerically for various u and v satisfying Eq. (3), with a grid $\Delta u = \Delta v = 0.1$. We do not find an intersection with the lowest energy curve for any u and v . This is illustrated in Fig. 1, where we replot the first four energies $E(\tilde{\epsilon})$ in the H - T plane; $h_{c2} = 1/\tilde{\epsilon}$ and $t = \alpha_0(T - T_{c0})/\epsilon$. The same general structure is obtained for all u and v .

We therefore conclude that the simple two-component model of Eq. (1) cannot explain the observed phase diagram of UPT_3 for $\mathbf{H}\parallel c$. We now ask if it is possible to obtain an intersection and thus save this model by assuming that the symmetry breaking term ϵ decreases with H_z , vanishing and changing sign at some value of H_z . Such an assumption is not implausible, since the symmetry breaking perturbation is usually ascribed to a small antiferromagnetic moment in the basal plane, which can be expected to respond differently to fields parallel and perpendicular to the c axis. The eigenvalue problem is still given by Eq. (2), with $\tilde{\epsilon} \sim \epsilon(H)/H$. Note, however, that the spectrum $\{E(\tilde{\epsilon})\}$ is even in $\tilde{\epsilon}$, since the change $\tilde{\epsilon} \rightarrow -\tilde{\epsilon}$ in Eq. (2) is equivalent to the unitary transformations $a \rightarrow ia$, $a^\dagger \rightarrow -ia^\dagger$, and $\eta_- \rightarrow -\eta_-$. Since even when $\tilde{\epsilon} = 0$, the lowest energy is separated from the next, there is no intersection. Further, as stated above, the lowest two energies have even parity when $\tilde{\epsilon} = 0$, so the lowest odd-parity curve does not approach the lowest even-parity one near the corresponding value of H anyway.

To explain a split transition in zero field, one is therefore compelled to use a sum of two order parameters unrelated by symmetry, which happen to have nearby transition temperatures by accident [20]. Depending on the representations of D_{6h} to which the two order parameters (denoted η_a and η_b) belong, the GL free energy may or

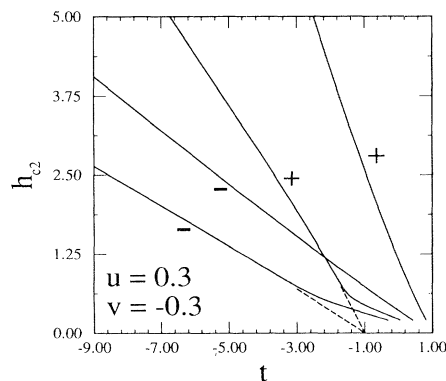


FIG. 1. The four lowest energy curves for the two-component model with $\mathbf{H}\parallel c$, shown in the H - T plane. The \pm signs are the Landau level parities. Note that there is no intersection with the lowest energy (highest h_{c2}) curve. The dashed lines show how the high field behavior of two of the curves extrapolates to $t = -1.0$ for $h = 0$, and thus illustrate avoided crossings. The same behavior is seen for all other values of u and v .

may not contain terms such as [21]

$$p_{\perp}^* \eta_a^* p_{\perp} \eta_b, \quad p_{\perp}^* \eta_a^* p_z \eta_b. \quad (4)$$

We wish to look for pairs of representations in which such terms are *not allowed* by symmetry, for otherwise the η_a and η_b subspaces will be coupled together in the eigenvalue problem for H_{c2} , and it is extremely unlikely that a conserved quantity (and therefore a tetracritical point) will exist for all \mathbf{H} . There are now two cases. The first is that η_a and η_b have different parities. Gradient coupling terms are then obviously excluded, but η_a and η_b can have any rotational symmetry, and it is hard to reduce the number of possibilities further. The second case, and the one we shall study in more detail, is that η_a and η_b have the same parity. In this case one can show that one of the types of terms in Eq. (4) is always allowed, except when one representation is A_1 or A_2 , and the other is B_1 or B_2 . We dub the resulting model the "AB" model.

We now proceed to analyze the AB model. The GL free energy density is the same for all four cases and can be taken to quartic order as

$$f_{\text{GL}} = \sum_{i=a,b} \alpha_i |\eta_i|^2 + \beta_i |\eta_i|^4 + \kappa_i |p_{\perp} \eta_i|^2 + \kappa_i' |p_z \eta_i|^2 + \beta_1 |\eta_a|^2 |\eta_b|^2 + \beta_2 [(\eta_a \eta_b^*)^2 + \text{c.c.}] + h^2/8\pi. \quad (5)$$

Here, $\alpha_i = \alpha_0(T - T_i)$, with $T_a = T_0 + \epsilon$, $T_b = T_0 - \epsilon$, with $T_0 \gg \epsilon > 0$. We note immediately that there is no symmetry breaking term, and Eq. (5) is actually invariant under *all* rotations about the c axis. Quantities such as H_{c1} and H_{c2} calculated on the basis of Eq. (5) will thus be isotropic in the basal plane, as is observed.

We first consider solutions in zero external field. We can clearly take η_a to be real and positive. The energy is minimized by taking η_b to be real if $\beta_2 < 0$, and pure imaginary if $\beta_2 > 0$. Writing $\beta' = (\beta_1 - 2|\beta_2|)/2$, the quartic coupling terms in Eq. (5) can be combined into $2\beta' \eta_a^2 \eta_b^2$, where the phase of η_b (if any) has been removed. The stability constraints are then

$$\beta_a > 0, \quad \beta_b > 0, \quad \beta' > -(\beta_a \beta_b)^{1/2}. \quad (6)$$

There are three solutions for which f_{GL} is stationary: (i) $\eta_a \neq 0$, $\eta_b = 0$, (ii) $\eta_a = 0$, $\eta_b \neq 0$, and (iii) $\eta_a \neq 0$, $\eta_b \neq 0$. Clearly, (i) nucleates immediately below T_a , which is therefore the observed upper transition temperature T_{c+} . To obtain a continuous transition to solution (iii) at a lower temperature, we require $\beta' < \beta_a$ and $\beta' < \beta_b$ [22]. The lower transition temperature, T_{c-} , and the specific heat jumps (both measured from the normal state) are then given by

$$T_{c-} = T_{c0} - \epsilon(\beta_a + \beta')/(\beta_a - \beta'), \quad (7)$$

$$\frac{\Delta C_+}{T_{c+}} = \frac{\alpha_0^2}{2\beta_a}, \quad \frac{\Delta C_-}{T_{c-}} = \frac{\alpha_0^2(\beta_a + \beta_b - 2\beta')}{2(\beta_a \beta_b - \beta'^2)}. \quad (8)$$

The lower transition is thus enhanced or suppressed depending on the sign of β' : $T_{c-} \leq T_b$ if $\beta' \geq 0$. The ratio of

specific heat jumps, r_{sh} is given by

$$r_{\text{sh}} = \frac{\Delta C_-/T_{c-}}{\Delta C_+/T_{c+}} = 1 + \frac{(\beta_a - \beta')^2}{\beta_a \beta_b - \beta'^2}. \quad (9)$$

The zero-field solutions also enable one to calculate H_{c1} in the London approximation. We have done this, and find a kink at T_{c-} for all θ [13].

We next consider the problem of H_{c2} and the inner transition lines. The calculation of the outer lines reduces to finding H_{c2} for a one-component order parameter with a uniaxially anisotropic mass tensor. For a field applied at an angle θ to the c axis, we get [23]

$$H_{c2}^{a,b}(T) = \alpha_0(T_{a,b} - T)\phi_0/2\pi g_{a,b}, \quad (10)$$

where $\phi_0 = hc/2e$ is the flux quantum, and

$$g_i(\theta) = [\kappa_i(\kappa_i \cos^2 \theta + \kappa_i' \sin^2 \theta)]^{1/2}. \quad (11)$$

The condition for an intersection to exist for all θ is $g_a(\theta) > g_b(\theta)$ (assuming $\epsilon \ll T_0$), or equivalently, $\kappa_a > \kappa_b$, $\kappa_a \kappa_a' > \kappa_b \kappa_b'$. (All κ 's must be positive if the zero-field equilibrium solution is to be homogeneous.) The tetracritical point (T^* , H^*) is located at

$$T^* = T_0 - \epsilon(g_a + g_b)/(g_a - g_b), \quad (12)$$

$$H^* = \alpha_0 \phi_0 \epsilon / \pi (g_a - g_b).$$

We now sketch the inner line (H_{ci}) calculation. (See also Refs. [6] and [9].) Suppose η_a has nucleated and is given by the usual centered rectangular Abrikosov flux lattice. The linearized instability condition for η_b is

$$[\kappa_b p_y^2 + g_b^2(\theta) p_x^2 / \kappa_b] \eta_b + \beta_1 |\eta_a|^2 \eta_b + 2\beta_2 \eta_a^2 \eta_b^* = -\alpha_b \eta_b. \quad (13)$$

This is the problem of a charge in a magnetic field [24] plus a periodic potential given by the η_a lattice. Writing $\bar{t} = (T - T_{c2})$, we have $\eta_a \propto \bar{t}^{1/2}$, and we can get H_{ci} to order \bar{t} by first-order perturbation theory in the potential. The η_b solution is a vortex lattice with the same periodicity as η_a , but shifted by a vector \mathbf{r} , and has an overall relative phase. The potential energy can be evaluated in terms of two sums $S_1(\mathbf{r})$ and $S_2(\mathbf{r})$, given by

$$[S_1, S_2] = \frac{\langle |\eta_a \eta_b|^2 \rangle, \langle (\eta_a \eta_b^*)^2 \rangle}{\langle |\eta_a|^2 \rangle \langle |\eta_b|^2 \rangle}, \quad (14)$$

where the angular brackets denote spatial averages. Then H_{ci}^b is found to be

$$H_{ci}^b(T, \theta) = (H_{c2}^b g_b - \Gamma H_{c2}^a g_a) / (g_b - \Gamma g_a), \quad (15)$$

where

$$\Gamma \approx (\beta_1 S_1 - 2|\beta_2 S_2|) / 2\beta_a \beta_b. \quad (16)$$

Here, $\beta_{\Delta} = \langle |\eta_a|^4 \rangle / \langle |\eta_a|^2 \rangle^2 = 1.1596$ is the usual Abrikosov lattice parameter, and we have set the effective GL ratio ξ/λ to zero, since it is of order 0.03 for UPT₃. It is

to be understood that the displacement \mathbf{r} is chosen so as to minimize Γ , which maximizes H_{ci} in Eq. (16). S_1 and S_2 are functions only of \mathbf{r} and the ratio ξ_b/ξ_a , where $\xi_i = l(\kappa_i/g_i)^{1/2}$. S_2 vanishes unless the two lattices coincide or are displaced by half of either of the two primitive lattice vectors, \mathbf{u}_1 and \mathbf{u}_2 . Note that since $|S_2| < S_1$, Eqs. (15) and (16) imply that the inner (H_{c1}) lines will lie below the extensions of the outer (H_{c2}) lines if $\beta' > 0$. The latter does appear to be so for UPt_3 (see below).

A comparison of our results with existing data is useful even though it leaves a large leeway in fixing the GL parameters. As the experimental H_{c2} curves for $\mathbf{H}\parallel\mathbf{c}$ [2] show substantial curvature, a quantitative fit to the theory for H_{ci} is not possible, and the only useful information that can be extracted from them is that $\kappa_b/\kappa_a \approx 1$. We can use the data for $\mathbf{H}\perp\mathbf{c}$, however, along with the measured $T_{c\pm}$ values to obtain relative values for the four β parameters. Using the H_{c2} slopes, we estimate that $g_b/g_a \approx 0.8$, implying that $\xi_b/\xi_a \approx 1.1$. We estimate T_{c+} , T_{c-} , T^* , and ϵ to be 500, 438, 400, and 10 mK, respectively, and $H^* \approx 0.47$ T. This gives $\beta'/\beta_a \approx 0.68$. Using this value of β'/β_a , we can reproduce the measured H_{c1}^b slope by taking $\mathbf{r} = \mathbf{u}_1/2$ [25]. We obtain $\beta_1/\beta_a \approx 1.38$, and $|\beta_2|/\beta_a \approx 0.015$. With these values of β_1 and β_2 , the lowest Γ at the H_{c1}^a transition is obtained by taking $\mathbf{r} = \mathbf{u}_1/2$, and the slope is fitted by taking $\beta_b/\beta_a \approx 0.74$, and a specific heat jump ratio $r_{sh} \approx 1.37$. (Note though that putting the η_a vortices at the centroid of an η_b vortex triangle is almost as good. We get $\beta_b/\beta_a \approx 0.75$, and $r_{sh} \approx 1.36$.) This is reasonably close to the value of 1.18 for r_{sh} that one would deduce from Ref. [4]. It thus appears that the vortex lattices are displaced by $\mathbf{r} = \mathbf{u}_1/2$ at both inner transitions, but this cannot be regarded as certain.

In summary, we believe we have shown that the AB model can explain all global features of the phase diagram and is therefore a strong candidate for UPt_3 . The most useful test of the model, in our view, would come from careful investigation of the phase diagram, especially for nonspecial $\hat{\mathbf{H}}$.

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