Identification of Odd-Parity Superconductivity in UPt_3 from Paramagnetic Effects on the Upper Critical Field

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We present a theoretical explanation of the unusual temperature-dependent anisotropy of H_{c2} reported by Shivaram *et al.* for single crystals of UPt_3, which we argue provides strong support for unconventional *odd-parity* pairing. We show that the observed low-temperature crossover in the anisotropy ratio, $H_{c2}^{\perp}/H_{c2}^{\parallel}$, can be explained by paramagnetic limiting for **H** parallel to the sixfold axis within BCS pairing theory if the order parameter has *odd parity* and there is strong spin-orbit coupling of the pair spin to the crystal axes.

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The properties of the heavy-fermion superconductors (e.g., $CeCu_2Si_2$, UBe_{13}, UPt_3) have led to a broad effort to understand superconductors with an *unconventional* order parameter (we use this term to mean an order parameter which has a smaller rotational symmetry group than that of the normal metallic crystal).[1,2] One important area involves studies of the upper critical field H_{c2} that separates the normal metallic state from the Abrikosov state of a type-II superconductor, and which can play a key role in determining the nature of the underlying order parameter.[3,4] In this paper we present a theoretical explanation of the unusual temperature-dependent anisotropy of H_{c2} reported by Shivaram, Rosenbaum, and Hinks[5] for single crystals of UPt_3, which we argue provides strong support for unconventional *odd-parity* pairing.

Theoretical arguments have been put forth by many authors (see, e.g., Ref. 1) that the order parameter for UPt_3 transforms according to one of the nonidentity representations of the hexagonal point group D_{6h}. These arguments are based in part on interpretations of the observed power-law temperature dependences of the ultrasonic attenuation, and also on RPA-like models of the pairing interaction. Authors[6-8] have argued that exchange of antiferromagnetic (AFM) spin fluctuations suppresses pairing in the odd-parity *and* the even-parity identity representations, but favors an even-parity nonidentity representation for the order parameter. More recent variants of the AFM exchange model[9] yield odd-parity superconductivity. However, these arguments are based on models of the pairing interaction that are not grounded in a first-principles theory for the pairing instability.[2] Likewise, the interpretation of the power-law behavior of the ultrasound attenuation in terms of an unconventional order parameter is based on an assumed identification with the excitation gap, not a fundamental relationship. Therefore, considerable theoretical effort has been devoted to identifying strong tests of the symmetry of the order parameter (see, e.g., Ref. 2), including the anisotropy of $dH_{c2}/dT|_{T_c}$ for tetragonal and cubic symmetry,[3] as well as tests of broken time-reversal symmetry.[10,11] As yet there are no reports of positive identifications of unconventional superconductivity based on strong tests of broken symmetry.

We have considerable confidence in the predictions of the BCS theory, even for unconventional pairing (e.g., ^{3}He), provided we know the relevant material parameters like T_c, the Fermi velocity, etc., and most importantly the symmetry class of the pairing state. Thus, predictions of the BCS theory that are *qualitatively* different for order parameters belonging to different symmetry classes are reasonably strong tests of unconventional pairing. The loophole here is whether or not one has identified all the relevant input material parameters.

Nevertheless, there is increasing evidence that UPt_3 has an order parameter belonging to a nonidentity representation. The strongest evidence so far comes from observations of multiple superconducting phases in UPt_3 (see Ref. 12 and references therein), which are naturally interpreted in terms of an order parameter belonging to a fundamental representation with dimensionality >1. The double transition in zero field,[13] as well as the observed kinks in $H_{c2}^{\perp}$ (we use $\perp$ and $\parallel$ to refer to **H** perpendicular to and **H** parallel to the sixfold axis) and H_{c1},[14] has been interpreted in terms of a weak breaking of the hexagonal symmetry which splits the transition into a 2D representation into two nearly degenerate 1D representations.[15-18] The existence of multiple superconducting states suggests that the order parameter belongs to one of the 2D representations: even-parity E_{1g} or E_{2g}, or odd-parity E_{1u} or E_{2u}.

Although attention has recently been focused on the Ginzburg-Landau region, where anomalies in $H_{c2}^{\perp}(T)$ have been observed, the low-temperature measurements of $H_{c2}^{\perp}$ and $H_{c2}^{\parallel}$ contain an important clue to the identification of the order parameter. While measurements of the parallel critical field are considerably larger than those measured in the basal plane for $T>200$ mK, Shivaram, Rosenbaum, and Hinks[5] found that $H_{c2}^{\parallel}$ was strongly *suppressed* relative to $H_{c2}^{\perp}$ below ~ 200 mK. A plot of the anisotropy ratio from Ref. 5, $R(T)=H_{c2}^{\perp}/H_{c2}^{\parallel}$ (inset of Fig. 1) crosses unity at $T \simeq 200$ mK from

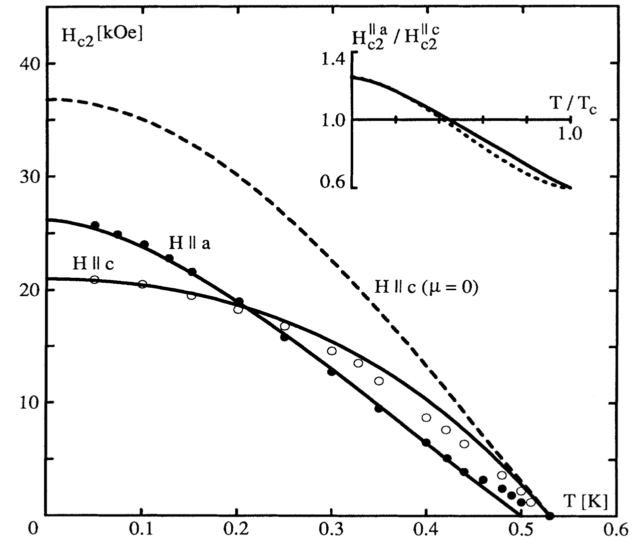

FIG. 1. $H_{c2}^{\perp}$ and $H_{c2}^{\parallel}$ vs T for the E_{1u} representation. The anisotropy ratio is $\eta=3.37$, the effective moment is $\bar{\mu}=0.68$ (except where noted), and $H_0=4.8$ T. Note that $H_{c2}^{\parallel}$ is suppressed by paramagnetism at low temperatures, while $H_{c2}^{\perp}$ is independent of $\bar{\mu}$. For comparison, the data of Ref. 5 are shown. The discrepancy between theory and experiment near T_c is due to the splitting of the transition, which is not accounted for in the calculation except for different T_c's for the two orientations of the field. Inset: The ratio $H_{c2}^{\perp}/H_{c2}^{\parallel}$ vs T/T_c. The solid curve is the result for the E_{1u} representation, while the dotted curve is obtained from Ref. 5.

$R(T_c)\simeq 0.6$ to $R(T\to 0)\simeq 1.25$. This crossover in the anisotropy ratio is a unique feature of UPt_3 that has so far not been explained by BCS theory, even with an unconventional order parameter, but is qualitatively and quantitatively accounted for within BCS theory if (i) the uniaxial anisotropy of the Fermi surface is included, (ii) the paramagnetic coupling to the field is included, and (iii) the order parameter belongs to one of the following odd-parity ($S=1$) representations: E_{1u}, A_{1u}, A_{2u}, B_{1u}, or B_{2u}, appropriate for strong spin-orbit coupling of the Cooper-pair spin to the lattice.

The Fermi-surface anisotropy determines the anisotropy ratio near T_c, where the paramagnetic coupling is unimportant, while the interplay between the paramagnetic effect, strong spin-orbit coupling, and the spin of the Cooper pairs for odd-parity pairing is crucial for explaining the crossover in the anisotropy ratio at low temperatures. For even-parity states, the upper critical field is bounded by the paramagnetic effect for all directions of field. But for odd-parity states, there is no paramagnetic suppression of superconductivity if the external field is perpendicular to the direction $\mathbf{\Delta}$ along which the Cooper pairs have zero total spin, i.e., $\mathbf{\Delta}\cdot\mathbf{S}=0$; whereas other orientations of the magnetic field relative to $\mathbf{\Delta}$ will be pair breaking.[19] Thus, the paramagnetic limit can have a dramatic effect on the anisotropy of H_{c2} for odd-parity states with strong spin-orbit coupling, which serves to lock the Cooper-pair spin to the crystal lattice.

The results presented here are based on the quasiclassical theory of superconductivity (we use the notation of Alexander *et al.*[20]), which allows us to incorporate an anisotropic Fermi surface, paramagnetic coupling, spin-orbit coupling, impurity scattering, and an unconventional order parameter into our calculations. The formalism and details of the calculations presented here are contained in a forthcoming article.[21]

We present results for the clean limit, $1/\tau_{\rm imp}\ll 2\pi T_c$, appropriate for high-quality single crystals of UPt_3. The central equation is the linearized weak-coupling gap equation; for odd-parity ($S=1$) pairing,

$$\mathbf{\Delta}(\vec{k}_f,\mathbf{R})=2\pi T\sum_{\epsilon}\int_0^{\infty}d\tau\int d^2k'_f\, n(\vec{k}'_f)\hat{V}(\vec{k}_f,\vec{k}'_f)\exp\{-2\tau|\epsilon|-\mathrm{sgn}(\epsilon)\tau\mathbf{v}'_f\cdot\boldsymbol{\partial}\}[1+\{\cos(2\tau\mu H)-1\}\hat{\mathbf{h}}\otimes\hat{\mathbf{h}}]\mathbf{\Delta}(\vec{k}'_f,\mathbf{R})\,, \qquad (1)$$

where the sum is over Matsubara frequencies and $\boldsymbol{\partial}=\boldsymbol{\nabla}_{\mathbf{R}}+i(2e/c)\mathbf{A}$ includes the coupling to the field through the vector potential $\mathbf{A}(\mathbf{R})$. The density of states $n(\vec{k}_f)$, the Fermi velocity $\mathbf{v}_f$, and the pairing interaction $\hat{V}(\vec{k}_f,\vec{k}'_f)$, are functions of position $\vec{k}_f$ on the Fermi surface, as is the effective magnetic moment μ that determines the paramagnetic coupling to the field. The uniaxial tensor, $\hat{\mathbf{h}}\otimes\hat{\mathbf{h}}$, is defined by the direction of magnetic field, $\hat{\mathbf{h}}$. For even-parity states the scalar ($S=0$) order parameter satisfies a similar equation to (1) with $\hat{\mathbf{h}}\otimes\hat{\mathbf{h}}\to 1$. The upper critical field is then computed as the largest value of H for which Eq. (1) has a nontrivial solution. Note that the paramagnetic term is unimportant close to T_c for both odd- and even-parity states; however, it has an effect on H_{c2} at lower temperatures, except for odd-parity states with $\mathbf{\Delta}\perp\mathbf{H}$.

Our calculations of H_{c2} for UPt_3 depend on the following material parameters: (i) Principal Fermi velocities, $v_f^{\perp}$ and $v_f^{\parallel}$, to parametrize our uniaxial model for the Fermi surface. (ii) An effective moment for quasiparticles, μ. (The effective moment is in general a crystal tensor; however, our conclusions are insensitive to this structure, so μ is taken to be a single parameter here. A more detailed discussion of this point is given in Ref. 21.) (iii) A transition temperature T_c (in zero field) determined by the most attractive pairing channel, which will be one of the irreducible representations of the point group D_{6h}. We assume that only one pairing channel is significant, thus excluding an accidental degeneracy of two representations. We also omit all symmetry-breaking fields that might reduce the 2D representations to two nearly degenerate 1D representations. Thus, the calculations we present do not show the kink in $H_{c2}^{\perp}$ near T_c, but since we are interested in the effect of paramagnetism on the intermediate- and low-temperature behavior of H_{c2} this is not a serious limitation.

It is useful to introduce the coherence lengths $\xi_\perp = v_f^\perp/2\pi T_c$ and $\xi_\parallel = v_f^\parallel/2\pi T_c$, and a magnetic-field scale $H_0=(hc/2e)/\pi\xi_\perp^2$, as well as the dimensionless parameters

$$\eta=(\xi_\parallel/\xi_\perp)^2,\quad h=H/H_0,\quad \bar{\mu}=\mu H_0/\pi T_c\,. \tag{2}$$

We have solved Eq. (1) for the upper critical field of the odd-parity representations (and a similar equation for the even-parity representations) of the hexagonal group with **H** along $\hat{\mathbf{z}}$, the axis of sixfold symmetry, and also for **H** in the basal plane.

We first consider solutions for the odd-parity 2D representations, E_{1u} and E_{2u}, and also the B_{1u} representation, which for our purposes is representative of all the 1D odd-parity states. Consider the E_{1u} order parameter (see, e.g., Ref. 22),

$$\Delta(\vec{k}_f,\mathbf{R})=\hat{\mathbf{z}}[\eta_+(\mathbf{R})f^*(\vec{k}_f)+\eta_-(\mathbf{R})f(\vec{k}_f)]\,, \tag{3}$$

where $f(\vec{k}_f)$ is given in Table I. Note that Δ is along $\hat{\mathbf{z}}$, so that the Cooper-pair spin is in the basal plane. The amplitudes $\eta_\pm(\mathbf{R})$ satisfy a homogeneous equation that depends on H and is simply obtained from Eqs. (1) and (3). The order parameter and upper critical field are obtained for all temperatures by a standard method of introducing raising and lowering operators and a set of eigenfunctions $\{\phi_n(\mathbf{R})\}$ of the harmonic-oscillator problem.[23]

When the magnetic field is along $\hat{\mathbf{z}}$, the maximum value of H occurs for $(\eta_+,\eta_-)\sim(\phi_2,c_0\phi_0)$.[24] Without paramagnetism, $h_{c2}^\parallel$ has the limiting values shown in Table I. Since the pairs have $S_z=0$, the paramagnetic term reduces $h_{c2}^\parallel$. Figure 1 shows the reduction in $h_{c2}^\parallel(T)$ at low temperatures due to paramagnetic limiting as well as the absence of a paramagnetic effect near T_c.

TABLE I. Limiting values for the upper critical field of hexagonal crystals with unconventional order parameters. Column 1 lists the representations of the unconventional order parameter discussed in the text. The form of the order parameter at H_{c2} is given in column 2 along with the (unnormalized) basis functions used to evaluate the Fermi-surface integrals. The upper (lower) row corresponds to $\mathbf{H}\parallel\hat{\mathbf{z}}$ ($\mathbf{H}\perp\hat{\mathbf{z}}$). Variational calculations are denoted by a †. Columns 3 and 4 give the limiting values of the upper critical field in units of H_0 for $\bar{\mu}=0$, $T=0$ and $T=T_c$, respectively.

Rep.	Order parameter at H_{c2}	$h_{c2}(0)$	$-T_c\,dh_{c2}/dT$
E_{1u}	$\phi_2\hat{\mathbf{z}}(k_x-ik_y)+c_0\phi_0\hat{\mathbf{z}}(k_x+ik_y)$	0.77	1.08
	$\phi_0\hat{\mathbf{z}}k_x$	$1.01/\sqrt{\eta}$	$1.19/\sqrt{\eta}$
E_{2u}	$\phi_0(\hat{\mathbf{x}}+i\hat{\mathbf{y}})(k_x+ik_y)$	0.37	0.59
	$\phi_0(\hat{\mathbf{x}}k_y+\hat{\mathbf{y}}k_x)$ †	$0.65/\sqrt{\eta}$	$0.84/\sqrt{\eta}$
B_{1u}	$\phi_0\hat{\mathbf{z}}(k_x^3-3k_xk_y^2)$ †	0.32	0.53
	$\phi_0\hat{\mathbf{z}}(k_x^3-3k_xk_y^2)$ †	$0.92/\sqrt{\eta}$	$1.07/\sqrt{\eta}$
E_{1g}	$\phi_2k_z(k_x-ik_y)+c_0'\phi_0k_z(k_x+ik_y)$	1.15	1.51
	$\phi_0k_zk_x$ †	$0.70/\sqrt{\eta}$	$0.96/\sqrt{\eta}$

By contrast, $h_{c2}^\perp(T)$ is independent of the paramagnetic effect because the pairs have no amplitude with zero spin projection for directions in the basal plane.

In Table I and Fig. 1 we summarize the results for the E_{1u} representation as a function of T/T_c for relevant choices of the effective-mass ratio η and scaled effective moment $\bar{\mu}$. Both $h_{c2}^\parallel$ and $h_{c2}^\perp$ are scaled in units of H_0; $h_{c2}^\parallel$ is then independent of η, while $h_{c2}^\perp$ scales as $1/\sqrt{\eta}$. As noted earlier the paramagnetic effect can signficantly reduce the value of $h_{c2}^\parallel$ at low tempertures, whereas it has no effect on $h_{c2}^\perp$. Thus, by adjusting η and $\bar{\mu}$, we obtain $h_{c2}^\parallel$ and $h_{c2}^\perp$ that cross over at an intermediate temperature. We have a crossover around $0.4T_c$ for $\eta=3.37$ and $\bar{\mu}=0.68$; the inset of Fig. 1 shows the anisotropy ratio $h_{c2}^\perp/h_{c2}^\parallel$ as a function of T/T_c for this choice of parameters, compared with the experimental values from Ref. 5. Figure 1 also shows the measured values of $H_{c2}^\perp$ and $H_{c2}^\parallel$ compared with the theoretical calculations. We have purposely chosen slightly different values for T_c of $H_{c2}^\perp$ and $H_{c2}^\parallel$, which emphasizes the discrepancy in the narrow temperature interval of the double transition, but provides a better fit to the data away from T_c. An essentially perfect fit to the full temperature range, including the region of the double transition, can be obtained by including the splitting of the E_{1u} representation by a weak symmetry-breaking field.[15]

For the E_{2u} representation, the order parameter,

$$\Delta(\vec{k}_f,\mathbf{R})=\eta_+(\mathbf{R})\frac{\hat{\mathbf{x}}+i\hat{\mathbf{y}}}{\sqrt{2}}f(\vec{k}_f)+\eta_-(\mathbf{R})\frac{\hat{\mathbf{x}}-i\hat{\mathbf{y}}}{\sqrt{2}}f^*(\vec{k}_f)\,, \tag{4}$$

differs significantly in its magnetic structure from that of E_{1u}. When $\mathbf{H}\parallel\hat{\mathbf{z}}$ the upper critical field is *independent* of the paramagnetic term and has a maximum eigenvalue for $(\eta_+,\eta_-)\sim(\phi_0,0)$, and for fields in the basal plane H_{c2} is quite sensitive to $\bar{\mu}$. We can obtain a very weak crossover in the anisotropy ratio even without the paramagnetic term for the limited range $2<\eta<3$; however, this crossover is much too weak to account for that observed in UPt_3, and becomes more isotropic as $\bar{\mu}$ is increased (see Ref. 21 for details).

There are four odd-parity, 1D representations in the limit of strong spin-orbit coupling, all of which have a similar spin structure to that of the E_{1u} representation and, therefore, exhibit similar anisotropic paramagnetic effects. We choose B_{1u} to illustrate the 1D representations with $\Delta(\vec{k}_f,\mathbf{R})=\varphi(\mathbf{R})\hat{\mathbf{z}}f(\vec{k}_f)$. A numerical calculation of the anisotropy ratio $h_{c2}^\perp/h_{c2}^\parallel$ for $\eta=11.0$ and $\bar{\mu}=1.2$ gives an almost identical fit to the experimental data of UPt_3 as that shown in Fig. 1 for the E_{1u} representation. Similar results can be obtained for all of the 1D odd-parity representations with suitable choices of the anisotropy and paramagnetic parameters.

The important distinction of the even-parity states is that the paramagnetic term is important for any direction of the magnetic field. For the E_{1g} representation,

$\Delta(\vec{k}_f,\mathbf{R})=\eta_+(\mathbf{R})f^*(\vec{k}_f)+\eta_-(\mathbf{R})f(\vec{k}_f)$ has a similar orbital form to that of E_{1u}. And not surprisingly, the solutions for (η_+,η_-) are of the same form as for the odd-parity E_{1u} representation; for $\mathbf{H}\parallel\hat{\mathbf{z}}$ the upper critical field occurs for $(\eta_+,\eta_-)\sim(\phi_2,c_0\phi_0)$. For $\mathbf{H}\parallel\hat{\mathbf{x}}$ we performed a variational calculation for h_{c2}, the precision of which is $\simeq 1\%$. The important point is that we do not find a crossover in the anisotropy ratio $h_{c2}^{\perp}/h_{c2}^{\parallel}$, with $h_{c2}^{\perp}>h_{c2}^{\parallel}$ at $T=0$, for any values of η and $\bar{\mu}$. Similar results are obtained for the other even-parity representations, and are discussed in Ref. 21.

We have calculated the upper critical field for order parameters belonging to the even- and odd-parity representations of the hexagonal point group with strong spin-orbit coupling. The effects of paramagnetism and anisotropy of Fermi surface on the anisotropy of the upper critical field, in particular the crossover behavior of $h_{c2}^{\perp}/h_{c2}^{\parallel}$, were examined in detail. For odd-parity states E_{1u} and B_{1u}, with $\mathbf{\Delta}$ parallel to $\hat{\mathbf{z}}$, the anisotropy ratio $h_{c2}^{\perp}/h_{c2}^{\parallel}$ fits the data on UPt_3 well with two parameters that measure the Fermi-surface anisotropy and the paramagnetic effect. Although our model for the Fermi-surface anisotropy is simple, we note for comparison that the values, $\eta=3.37$ (11.0), $H_0=4.8$ T (9.4 T), and $\bar{\mu}=0.7$ (1.2), which we obtain from our fit for the E_{1u} (B_{1u}) representation imply an effective moment of $\mu=0.34\mu_B$ ($\mu=0.30\mu_B$), in reasonable agreement with several other estimates.[25] The important point is that the Fermi-surface anisotropy and paramagnetic coupling give rise to a crossover in the anisotropy ratio of the right magnitude and temperature dependence for realistic values of the material parameters only for odd-parity states with $\mathbf{\Delta}$ along the $\hat{\mathbf{z}}$ axis. In particular, for the E_{2u} representation ($\mathbf{\Delta}\perp\hat{\mathbf{z}}$), the paramagnetic term affects only $h_{c2}^{\perp}$, and not $h_{c2}^{\parallel}$, and thus cannot account for the observed crossover. For even-parity states, the paramagnetic effect suppresses both $h_{c2}^{\perp}$ and $h_{c2}^{\parallel}$, and even tends to reduce the anisotropy of the upper critical field at low temperatures. These characteristics lead us to conclude that the experimental data of Ref. 5 provide strong support for odd-parity pairing in UPt_3.

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