Possible two-band superconductivity in PuRhGa$_5$ and CeRhIn$_5$

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1. Introduction

A class of lanthanide- or actinide bearing intermetallic compounds in which the f-electrons are close to a valence instability and hybridize strongly with conduction electrons of neighboring ligands, giving rise to an enhanced effective mass often 1000 times larger than simple metals such as copper, have generated considerable interest for the last 25 years\[1,2\]. In many cases, to coexist with long-range magnetic order, providing a rich playground to investigate the interplay of magnetism and superconductivity. The CeMIn$_5$ family of heavy-fermion superconductors are the most recent prototypical example of this playground\[3\]. In these compounds, the coexistence of antiferromagnetism and superconductivity under pressure in CeRhIn$_5$\[4,5\] or with chemical substitution (e.g., CeRh$_{1-x}$Ir$_x$In$_5$[6], CeRh$_{1-x}$Co$_x$In$_5$[7]), and a new magnetic phase deep within the superconducting state near the upper critical field in CeCoIn$_5$\[8,9\], are just some of the emergent phenomena indicating that the superconductivity is unconventional, involving electrons paired in finite angular momentum and spin states through magnetic fluctuations, and is strongly coupled to the magnetism. While no direct evidence for the coexistence of magnetic order and superconductivity has been found in the isostructural PuMGa$_5$ (M = Co, Rh) materials, which have transition temperatures of $T_c = 18.5$ [10] and $T_c = 8.7$ K [11], respectively, a number of similarities with their CeMIn$_5$ counterparts, including the powerlaw behavior of the upper critical field, the linear $|H_c|^2$ dependence of $H_c$ at $T_c$, indicate that the superconductivity may also be unconventional in these PuMGa$_5$ materials. In the following, we report the similarity of the upper critical field in PuRhGa$_5$ and CeRhIn$_5$ at a pressure where CeRhIn$_5$ exhibits the coexistence of antiferromagnetic order and superconductivity [5], suggesting proximity to magnetism in PuRhGa$_5$.

2. Experimental details

Single crystals of PuRhGa$_5$ and CeRhIn$_5$ were grown by Ga and In flux, respectively. Magnetization measurements were performed on PuRhGa$_5$ using a capacitive torque magnetometer in magnetic fields up to 17 T and temperatures below 0.5 K and 300 K in a cryostat equipped with a HEPA-filtered gas handling system. The magnetometer was immersed in $^3$He liquid/gas to ensure good thermalization of the sample and minimize the effects of self-heating due to the radioactive decay of $^{239}$Pu. The orientation of the magnetic field along the a- and c-crystallographic directions was achieved by alignment perpendicular to the natural faces of a rectangular PuRhGa$_5$ single crystal and confirmed by the observed anisotropy of the upper critical field. The superconducting upper critical field was determined as the inflection of the step in torque, and was unambiguously identified as being associated with superconductivity by performing set of zero-field cooled sweeps, which exhibited strong hysteresis associated with the trapping of flux as the field is reduced below $H_c2$, and subsequent thermal cycling in low field ($< 0.1$ T), where the magnetic hysteresis was observed to collapse at temperatures exceeding $T_c$, as the trapped flux was released. The maximum relative deflection of the magnetometer, and hence the sample alignment, as a function of magnetic field and temperature is less than 1°. The actual angle of the magnetic field with respect to the crystallographic axes was determined to be $\sim 20°$ from the c-axis by scaling the observed upper critical field.
$H_{c2}$ to that of Haga et al. [14], and is consistent with previous measurements of the angular dependence of $H_{c2}$ [14].

### 3. Results

Fig. 1 shows the torque ($\propto$ magnetization), plotted as the difference in capacitance $\Delta C$, vs. magnetic field up to 17 T of PuRhGa$_5$ at temperatures of 0.62 K and 1.6 K for $H||c$. At 1.6 K, a clear signature of the upper critical field, denoted by the large change in $\Delta C$ and subsequent hysteresis between the zero-field-cooled (ZFC) and field-cooled curves. The lack of a similar anomaly and hysteresis in the ZFC/FC curves up to 17 T indicates that the upper critical field is above 17 T at 0.62 K.

The upper critical field $H_{c2}(T)$ of PuRhGa$_5$ is displayed in Fig. 2 (scaled to account for the misalignment) for $H||c$ and $H||ab$, along with the results of Haga et al. [14]. For both field directions, $H_{c2}$ is linear in temperature, and extends down to $\sim T_c/10$ for $H||c$. Extrapolating the upper critical field linearly to zero temperature yields the values $H_{c2}^{0,0} = 31.1$ T and $H_{c2}^{0,1} = 17.3$ T. This anisotropy of $H_{c2}^{0,0} / H_{c2}^{0,1} = 1.8$ in PuRhGa$_5$ is comparable to the anisotropy of the upper critical field of the pressure-induced superconductor CeRhIn$_5$ at 14.7 kbar ($\beta = 1.7$) and of CeCoIn$_5$ ($\beta = 2.4$) and presumably reflects cylindrical (two-dimensional) nature of the Fermi surface in these “115” materials [15–17]. It should be noted that the anisotropy of $H_{c2}$ of PuCoGa$_5$ near $T_c$ is considerably smaller (\sim 10%) ; further measurements at higher magnetic fields are required to determine whether this smaller $H_{c2}$ anisotropy persists to lower temperature.

### 4. Discussion

A magnetic field may suppress superconductivity in two main ways. The orbital supercurrents surrounding the flux lines produced by the external field in the mixed state will break the Cooper pairs when the kinetic energy of the orbital currents exceeds the superconducting condensation energy, leading to a suppression of superconductivity and the upper critical field. The applied field will also interact with the spins of the superconducting electron pair via the Zeeman effect. The upper critical field data of PuRhGa$_5$ are compared to the limiting case of these two effects within a single-band model, to coupled superconducting order parameters in a two-band model, and also to those of the pressure-induced antiferromagnetic superconductor CeRhIn$_5$ in the following sections.

#### 4.1. Single-band superconductivity

In the presence of a Zeeman term it is well known that for the case of a single conduction band with isotropic gapped superconductivity, the normal state is favored over the superconducting state when the magnetic energy exceeds the condensation energy $(1/2)|\chi_H|^2 > (1/2)|\Delta_0|^2$, where $|\chi_H|$ is the Pauli susceptibility, $\Delta_0$ is the zero-temperature superconducting gap, and $N_f$ is the density of states per spin. The Pauli limiting critical field is then given by $H_p = -\Delta_0 / \sqrt{2}$, assuming $|\chi_H| = \mu_B$ the Bohr magneton. In this fully gapped case with $T_c = 8.7$ K, the Pauli limiting field is $H_p = 1.84 T_c = 16.0$ T for PuRhGa$_5$. This value is slightly modified for an order parameter with nodes (i.e., $d$-wave), as indicated by NMR measurements on PuRhGa$_5$ [19], the Pauli field is $H_p = 1.77 T_c = 15.2$ T [20]. As illustrated in Fig. 2, $H_p$ agrees with $H_{c2}^0(0)$ for PuRhGa$_5$, but $H_{c2}^0(0)$ exceeds this value and cannot be reconciled with a purely Pauli limiting field without an unlikely $g$-factor anisotropy. This fact, along with the linear $T$-dependence of $H_{c2}$, would suggest that PuRhGa$_5$ is not Pauli limited. In addition, $H_{c2}^0(0)$ is larger than expected for pure orbital contribution to the upper critical field [18], $H_{c2}^0(0) = -0.7 T_c dH/dT |_{T_c} = 12.4$ T, which is predicted to have a nearly quadratic temperature dependence close to zero temperature, as shown in Fig. 2. Spin-orbit coupling could, in principle, lead to better agreement with the experimental $H_{c2}^0(0)$, but would not yield the $T$-linear dependence of the upper
4.2. Two-band superconductivity

The observed anomalous linear temperature dependence of \( H_{c2} \) in Fig. 2 may be understood in terms of a two-band model of superconductivity. Assuming that the superconducting order parameter of a heavy \( f \)-electron band is coupled to an order parameter of a light \( d \)-electron band, the response to a magnetic field may be gradually tuned between the heavy and light band to obtain a nearly linear temperature dependence. Near \( T_c \) and \( H \approx 0 \) the heavy band will dominate, while at low temperatures and high fields the light band will dominate the magnetic response. To this end, a Ginzburg–Landau (GL) two-band model is constructed to capture the essential behavior of the upper critical field yielding a qualitative explanation of the \( H-T \) phase diagram of superconductivity.

Let the two bands be described by two GL free energies \( \bar{F}_{i} = \frac{1}{2} \sum_{\alpha} (\bar{a}_{i}^{(\alpha)} \bar{\psi}_{i}^{\dagger} \bar{\psi}_{i}^{(\alpha)} + \bar{a}_{i}^{(\alpha)} \bar{\psi}_{i}^{\dagger} \bar{\psi}_{i}^{(\alpha)} + \bar{a}_{i}^{(\alpha)} \bar{\psi}_{i}^{\dagger} \bar{\psi}_{i}^{(\alpha)} + \bar{a}_{i}^{(\alpha)} \bar{\psi}_{i}^{\dagger} \bar{\psi}_{i}^{(\alpha)}) + \frac{1}{2} \sum_{\alpha} \bar{m}_{i}^{(\alpha)} \bar{\psi}_{i}^{\dagger} \bar{\psi}_{i}^{(\alpha)} \) for \( i = 1, 2 \).

The GL parameters are \( a_{i}^{(\alpha)} = \alpha_{i} \bar{T}_{i} (1 - \frac{1}{2} \bar{V}_{i}) \) and \( h_{i} = h_{i}^{(0)} \). Here \( T_{i} \) is the transition temperature in the absence of coupling, \( v = 0 \), while \( H = \nabla \times A \) is the magnetic field inside the superconductor. The factor \( (1 - v H_{c2}) \) accounts for the Zeeman effect and the spin-split conduction bands. The factor \( \frac{T_{c}^{2}}{2} \) is the critical transition temperature in the absence of coupling, \( v = 0 \), while \( H = \nabla \times A \) is the magnetic field inside the superconductor. The factor \( \frac{T_{c}^{2}}{2} \) is the critical transition temperature in the absence of coupling, \( v = 0 \), while \( H = \nabla \times A \) is the magnetic field inside the superconductor.

Minimization of the free energy results in the coupled GL equations. The transition temperature is obtained by solving the linearized GL equations at \( H = 0 \) for a uniform superconductor, which gives \( T_{c} = (T_{1} + T_{2} + \gamma_{1} T_{c}^{2} + \gamma_{2} T_{c}^{2})^{2}/2 \), with \( \gamma_{1} = \eta/\alpha_{1} \) and \( \gamma_{2} = (\eta/\alpha_{2})^{2} \).

Next, in order to calculate the upper critical magnetic field, we solve the coupled linearized GL equations in a magnetic field assuming the Abrikosov vortex lattice solution. After some lengthy calculations, and neglecting the Zeeman term, the upper critical field is found to be

\[
H_{c2}(T) = \frac{h_{1} h_{2}}{2} \left[ t_{1} + t_{2} \pm \sqrt{(t_{1} - t_{2})^{2} + 4 \eta_{1} \gamma_{2} h_{2}/(h_{1} h_{2})} \right]
\]

where \( t_{1} = (T_{1} + \gamma_{1} T_{c}/h_{2} + \gamma_{2} T_{c}/h_{1}, \) and \( h_{i} \) are the slopes of the upper critical fields. When including the Zeeman term, it is easier to solve for \( T_{c}(H) \), which is given by

\[
T_{c}(H) = \frac{1}{2} \left[ T_{c}(H) + \sqrt{T_{c}(H)^{2} + 4D(H)} \right]
\]

where \( D(H) = \gamma_{1} \gamma_{2} + \bar{T}_{1} \bar{T}_{2} + H(\bar{T}_{1}/h_{2}) + (\bar{T}_{2}/h_{1}) \) and \( T_{c}(H) = \bar{T}_{1} \bar{T}_{2} + (H/h_{1}) + (H/h_{2}), \) and \( \bar{T}_{1} = (1 - \gamma_{2} H^{2})/\gamma_{1}. \) Fig. 3 shows the upper critical fields for a specific choice of GL parameters with a small coupling term \( \eta = 0.06 K \) and \( \alpha_{1} = 10 \) \( \alpha_{2} = 10 \) (density of states \( N_{l}^{0} \propto \alpha_{i} \)). By including the Zeeman term, which leads to significantly Pauli-limited upper critical fields, good agreement to the observed measurements is obtained (Fig. 3). However, the results at temperatures below \( T_{c}/2 \) should be considered with care, because at such low temperatures and high magnetic fields (i) the GL expansion is no longer valid and (ii) the coefficients \( \beta_{1} \) and \( \kappa_{1} \) may change sign leading to a first-order phase transition or new phases, such as a Fulde–Ferrell–Larkin–Ovchinnikov state, observed in the isostructural compound CeCoIn\( _{5} \) [21].

4.3. Isostructural compound CeRhIn\( _{5} \)

The linear temperature dependence of the upper critical field has also been observed in the isostructural compound CeRhIn\( _{5} \) at \( P = 14.7 \) kbar where unconventional superconductivity coexists with long-range antiferromagnetic order. As shown in the inset of Fig. 4, the upper critical field of CeRhIn\( _{5} \) is linear in temperature down to the lowest temperature measured for both field directions, with an anisotropy similar to that of PuRhGa\( _{5} \). As we have shown [22], \( H_{c2} \) can qualitatively be described within a two-band model of superconductivity in CeRhIn\( _{5} \) at \( P = 14.7 \) kbar, in which a band of light, weakly hybridized electrons that dominate the antiferromagnetic state is (weakly) coupled to one composed of heavy, strongly hybridized superconducting electrons. The remarkable similarity between \( H_{c2} \) of both compounds is more clearly illustrated in Fig. 4 where it is scaled by the zero-temperature value \( H_{c2}(0) \) vs. \( T/T_{0} \). While PuRhGa\( _{5} \) is not in an ordered state at low temperature, it does appear to be closer to magnetic order than PuCoGa\( _{5} \) [13], for which

**Fig. 3.** Upper critical field within two-band Ginzburg-Landau model with bare transition temperatures \( T_{1} = 8.55 K, T_{2} = 7.3 K \) and slopes \( h_{1} = -1.8 T/K, h_{2} = -4.5 T/K \) for \( H/c \) and \( h_{1} = -4.0 T/K, h_{2} = -70 T/K \) for \( H/ab \). The solid (dashed) lines are with (without) the Zeeman effect. The dotted curves are bare phase transition lines in the two-band model. The symbols are measurements of PuRhGa\( _{5} \) for \( H/c \) (blue circles) and \( H/ab \) (filled red squares). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.).

**Fig. 4.** Upper critical field, scaled by the zero-temperature value \( H_{c2}(0) \) vs. \( T/T_{0} \) of PuRhGa\( _{5} \) for \( H/c \) (blue circles) and \( H/ab \) (red squares) [data (closed symbols) from Ref. [14]] and CeRhIn\( _{5} \) at \( P = 14.7 \) kbar for \( H/c \) (blue triangles) and \( H/ab \) (red triangles). Inset: Anisotropic upper critical field of CeRhIn\( _{5} \) at \( P = 14.7 \) kbar for \( H/c \) (blue triangles) and \( H/ab \) (red triangles). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.).
there is evidence for an itinerant and localized “dual-nature” of the 5f-electrons [23]; perhaps these more localized f-electrons in PuRhGa5 comprise the heavy band, while the itinerant f-electrons contribute to the light within this two-band model of superconductivity; further work is underway to test this two-band scenario in these PuMGa5 superconductors and to fully characterize the nature of their 5f-electrons.

5. Conclusions

The upper critical field $H_{c2}$ of PuRhGa5 has been determined by means of torque magnetometer measurements at temperatures down to 0.6 K and magnetic fields up to 17 T. A linear temperature dependence of $H_{c2}$ are inconsistent with both purely Pauli limiting or orbital effects dominating the upper critical field within a single-band model, but is well-described by a two-band model of superconductivity involving coupled bands of heavy and light electrons. A close similarity between PuRhGa5 and the pressure-induced superconductor CeRhIn5 suggests that PuRhGa5 may be more localized than its isostructural counterpart PuCoGa5.

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References

[22] M. J. Graf et al., unpublished.