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Possible two-band superconductivity in $PuRhGa_5$ and $CeRhIn_5$

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ABSTRACT

Measurements of the upper critical field H_{c2} of the Pu-based heavy-fermion superconductor $PuRhGa_5$ have been made using a torque magnetometer at temperatures down to 0.6 K and magnetic fields up to 17 T along the principal axes of the tetragonal structure. A linear temperature dependence of H_{c2} down to $T_c/10$ for $H||c$ and to $T_c/2$ for $H||ab$ is observed. While the data appear to be inconsistent with both purely Pauli limiting or orbital effects governing the upper critical field, the linear T-dependence of H_{c2} is well described by a two-band model of superconductivity involving coupled bands of heavy and light electrons. A close similarity between $PuRhGa_5$ and the pressure-induced antiferromagnetic isostructural superconductor $CeRhIn_5$ is also found.

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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 a few of these heavy-fermion materials, superconductivity is found to intimately involve a dense array of magnetic *f*-electrons and, 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., $CeRh_{1-x}Ir_xIn_5$ [6], $CeRh_{1-x}Co_xIn_5$ [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 superconducting specific heat and spin-lattice relaxation rate and the linear variation of the ratio of the tetragonal lattice parameters with the superconducting transition temperature T_c [12,13], 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 preformed on $PuRhGa_5$ using a capacitive torque magnetometer in magnetic fields up to 17 T at temperatures between 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

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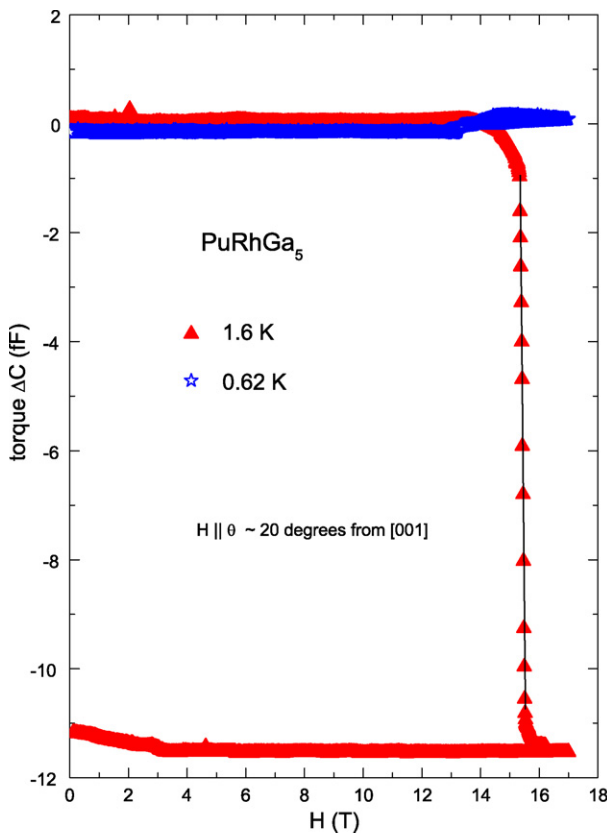

Fig. 1. Change in capacitance ΔC (torque) vs. magnetic field H of $PuRhGa_5$ at 1.6 K (red triangles) and 0.62 K (blue stars), with $H||20^{\circ}$ from the c-axis. The upper critical field H_{c2} is determined by the sharp drop in ΔC at 15.5 T at 1.6 K. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.).

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 ΔC, vs. magnetic field up to 17 T of $PuRhGa_5$ at temperatures of 0.62 K and 1.6 K for $H|| \sim c$. At 1.6 K, a clear signature of the upper critical field, denoted by the large change in ΔC 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 at 0.62 K 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 $H_{c2}^{ab} = 31.1$ T and $H_{c2}^{c} = 17.3$ T. This anisotropy of $H_{c2}^{ab}/H_{c2}^{c} = \beta = 1.8$ 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.

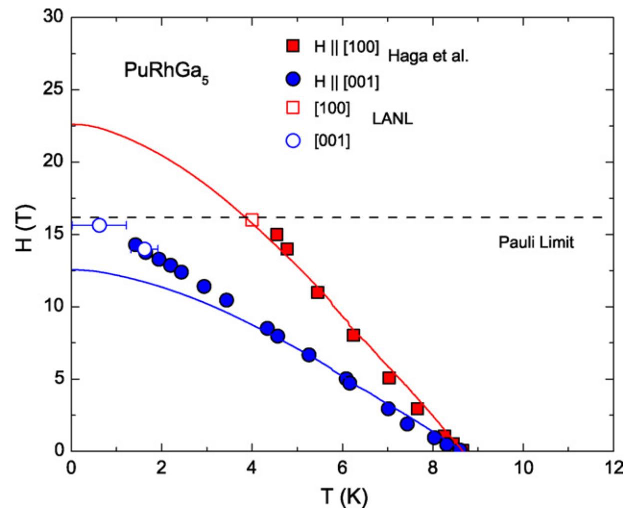

Fig. 2. Upper critical field $H_{c2}(T)$ of $PuRhGa_5$ for $H||c$ (open blue circles) and $H||ab$ (open red squares) in the present study and data from Ref. [14] for $H||c$ (filled blue circles) and $H||ab$ (filled red squares). The solid lines are the theoretical predictions for a purely orbital critical field [18] using the initial slopes of $dH^c/dT|_{T_c} = -2.0$ T/K and $dH^{ab}/dT|_{T_c} = -3.6$ T/K. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.).

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 $(1/2)\chi_P H^2 > (1/2)N_f\Delta_0^2$, where χ_P is the Pauli susceptibility, Δ_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 $H_P = \Delta_0/\sqrt{2}\mu_B$ (assuming a g-factor of 2), where μ_B is the Bohr magneton. In this fully gapped case with $T_c = 8.7$ K, the Pauli limiting field is $H_P = 1.84T_c = 16.0$ 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 $H_P = 1.77T_c = 15.2$ T [20]. As illustrated in Fig. 2, H_P agrees with $H_{c2}^{c}(0)$ for $PuRhGa_5$, but $H_{c2}^{ab}(0)$ 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, $H_{c2}^{c}(0)$ is larger than expected for pure orbital contribution to the upper critical field [18], $H_{c2}^{*}(0) = -0.7\,T_c dH/dT|_{T_c} = 12.4$ 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 $H_{c2}^{c}(0)$, but would not yield the T-linear dependence of the upper critical field down to $T_c/10$ in $PuRhGa_5$. An alternative explanation for the upper critical fields may be a two-band model as discussed below.

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 with a small set of parameters. The GL functional of the coupled heavy (1) and light (2) bands are written as $F = F_1 + F_2 + F_{12}$, with $F_i = a_i(T)|\psi_i|^2 + (1/2)b_i|\psi_i|^4 + \kappa_i|(\nabla - i(e/\hbar c)\mathbf{A})\psi_i|^2$, and coupling term $F_{12} = \eta(\psi_1\psi_2^* + \psi_1^*\psi_2 - |\psi_1|^2 - |\psi_2|^2)$. The GL parameters are $a_i(T) = \alpha_i[T - T_i(1 - \nu_i H^2)]$ and $b_i = \alpha_i\beta$. Here T_i is the critical transition temperature in the absence of coupling, $\nu = 0$, while $\mathbf{H} = \nabla \times \mathbf{A}$ is the magnetic field inside the superconductor. The factor $(1 - \nu_i H^2)$ accounts for the Zeeman effect and the spin-split conduction bands with $\nu_i = \frac{7\zeta(3)}{4\pi^2}((g/2)\mu_B/T_i)^2$.

Minimization of the free energy results in the coupled GL equations. The transition temperature is obtained by solving the linearized GL equations at $\mathbf{H}=0$ for a uniform superconductor, which gives $T_c = [T_1 + T_2 + \gamma_1 + \gamma_2 \pm T_0]/2$, with $\gamma_i = \eta/\alpha_i$ and $T_0 = \sqrt{(T_1 - T_2)^2 + 4\gamma_1\gamma_2}$.

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

$$H_{c2}(T) = \frac{h_1 h_2}{2}\left[t_1 + t_2 \pm \sqrt{(t_2 - t_1)^2 + 4\gamma_1\gamma_2/(h_1 h_2)}\right] \quad (1)$$

where $t_1 = (T_1 + \gamma_1 - T)/h_2$ and $t_2 = (T_2 + \gamma_2 - T)/h_1$, 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

$$T_c(H) = \frac{1}{2}\left[T_0(H) \pm \sqrt{T_0(H)^2 + 4D(H)}\right] \quad (2)$$

where $D(H) = \gamma_1\gamma_2 + \bar{T}_1\bar{T}_2 + H((\bar{T}_1/h_2) + (\bar{T}_2/h_1)) - (H^2/h_1 h_2)$, $T_0(H) = \bar{T}_1 + \bar{T}_2 + (H/h_1) + (H/h_2)$, and $\bar{T}_i = T_i(1 - \nu_i H^2) + \gamma_i$. Fig. 3 shows the upper critical fields for a specific choice of GL parameters with a small coupling term $\eta = 0.06$ K and $\alpha_1 = 10\alpha_2 = 10$ K (density of states $N_f^i \propto \alpha_i$). 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 β_i and κ_i 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 $H/H_{c2}(0)$ vs. T/T_{c0}. 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

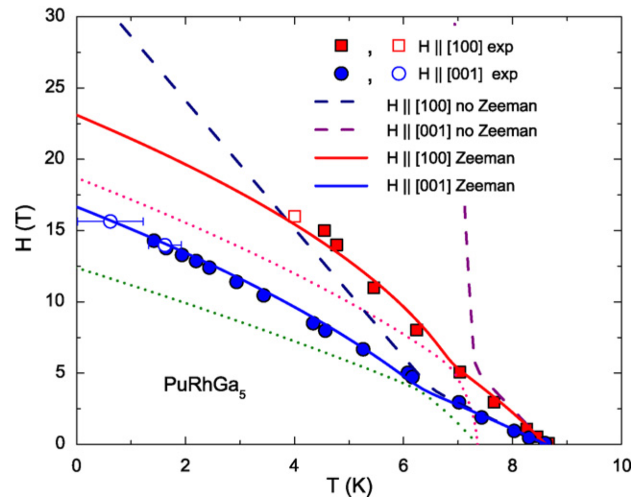

Fig. 3. Upper critical field within two-band Ginzburg-Landau model with bare transition temperatures $T_1 = 8.55$ K, $T_2 = 7.3$ K and slopes $h_1 = -1.8$ T/K, $h_2 = -4.5$ T/K for $H||c$ and $h_1 = -4.0$ T/K, $h_2 = -70$ T/K for $H||ab$. The solid (dashed) lines are with (without) the Zeeman effect. 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.).

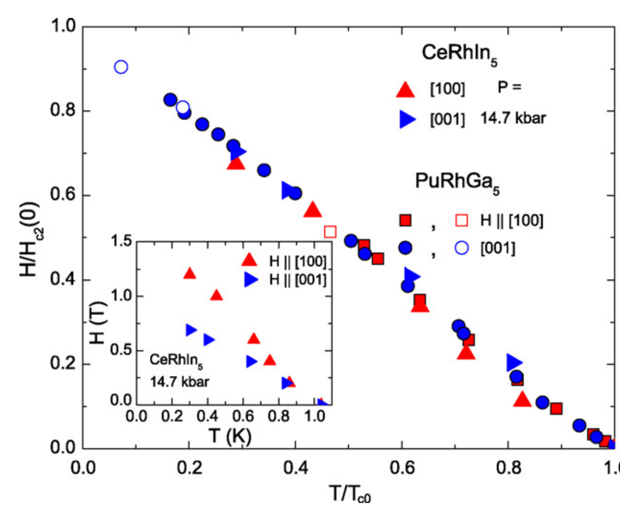

Fig. 4. Upper critical field, scaled by the zero-temperature value $H/H_{c2}(0)$vs. T/T_{c0} of $PuRhGa_5$ for $H||c$ (blue circles) and $H||ab$ (red squares) [data (closed symbols) from Ref. [14]] and $CeRhIn_5$ at $P = 14.7$ kbar for $H||c$ (blue triangles) and $H||ab$ (red triangles). Inset: Anisotropic 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 $PuRhGa_5$ 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 $PuMGa_5$ superconductors and to fully characterize the nature of their 5f-electrons.

5. Conclusions

The upper critical field H_{c2} of $PuRhGa_5$ 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 $PuRhGa_5$ and the pressure-induced superconductor $CeRhIn_5$ suggests that $PuRhGa_5$ may be more localized than its isostructural counterpart $PuCoGa_5$.

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