Specific heat and anisotropy of the nonconventional superconductors $PuCoGa_5$ and $PuRhGa_5$

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We report on specific-heat measurements under a magnetic field on oriented single crystals of the nonconventional superconductors $PuCoGa_5$ and $PuRhGa_5$ with superconducting transition temperature $T_c \sim 18$ and 9 K, respectively. In the superconducting state, the electronic specific heat follows a quadratic temperature dependence, which indicates an axial state with line nodes in the superconducting gap structure. The specific heat measured under a magnetic field applied along different crystallographic directions reveals anisotropic behavior. For both compounds, T_c decreases faster for fields applied along the c axis than for fields perpendicular to it. The value of the slope of the critical field at T_c is higher in the case of $PuCoGa_5$, while the anisotropy effect is stronger in $PuRhGa_5$. Further results are discussed in the context of the isostructural Ce- and Np-based materials. The possibility of the appearance of a Fulde-Ferrell-Larkin-Ovchinnikov state and/or strong spin-orbit coupling, related to the observed low-temperature linear shape of the critical field deviating from the common d-wave superconductor behavior, is considered, especially in the $PuRhGa_5$ case.

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I. INTRODUCTION

The actinide-based $ATGa_5$ (A=actinide, T=transition metal) intermetallic compounds have attracted much interest since the discovery of superconductivity in $PuCoGa_5$ (Ref. 1) and $PuRhGa_5$ (Ref. 2) with critical temperatures one order of magnitude higher than the maximum seen in U- and Ce-based heavy-fermion systems up to now. These $ATGa_5$ materials crystallize in the tetragonal $HoCoGa_5$-type structure with the space group $P4/mmm$. $PuCoGa_5$ is a superconductor displaying a remarkably high critical temperature $T_c = 18.5$ K and upper critical field $H_{c2}(0)$ estimated at ~ 74 T.[1] $PuRhGa_5$ has lower critical parameters, $T_c = 8.7$ K and $H_{c2}(0) \sim 21$ T.[2] Both materials appear to be close to a magnetic phase instability[3,4] and to display unconventional superconductivity as recently suggested by NMR studies.[5,6] The magnetic susceptibility of both compounds in the normal state indicates local-moment (Curie-Weiss) behavior close to that expected for Pu^{3+}.

The uranium isostructural materials $UTGa_5$ are nonsuperconducting paramagnets for T from the iron and cobalt column, but order antiferromagnetically for T from the nickel column.[7–9] Among the Np-based compounds, $NpCoGa_5$, $NpRhGa_5$, $NpFeGa_5$, and $NpNiGa_5$ have been investigated so far and they all exhibit antiferromagnetic order.[10–12] Recent studies on the $Pu(Co,Rh)Ga_5$ and other substituted $ATGa_5$ compounds[13,14] revealed a correlation between the c/a ratio and T_c in analogy to the $CeMIn_5$ (M=Co,Rh,Ir) heavy-fermion d-wave superconductors,[15] pointing to a similarity of the superconducting mechanism in both Pu- and Ce-based 115 materials. Unconventional superconductivity is corroborated by the absence of superconductivity in isostructural compounds with U, Np, or a rare earth other than Ce. Moreover, Pu substitution by nonmagnetic U or magnetic Np leads to comparably strong suppression of superconductivity.[14]

An important contribution to the understanding of the electronic properties of these materials is obtained from the specific-heat data. Results can be compared with parameters obtained by other techniques, such as magnetization and resistivity, especially in the case of substitution of the transition metals (Co–Rh). Moreover, this technique allows one to access important information, such as an estimation of the density of states at the Fermi level, and to categorize the materials in the superconductor family. It is a bulk measurement indicating the quality of the superconductivity.

In this paper, we present results for the specific heat of $PuCoGa_5$ and $PuRhGa_5$ single crystals measured in magnetic fields up to 9 T. For clarity and coherency of the data presented, we include also some previously reported experimental data that were repeated on new samples. However, the present work focuses on two major additional results. First, the low-temperature specific heat in the superconducting state of $PuRhGa_5$ down to 3 K is presented (previous measurements were limited to 7 K, i.e., to only a short range below T_c). The extended range of measurements allows evidencing the T^2 power law of the electronic specific heat in the superconducting state of $PuRhGa_5$, like that observed for $PuCoGa_5$.[13] Additionally, comparison of the specific heats of $PuCoGa_5$ and $PuRhGa_5$ with those of their uranium analogs in a wide temperature range up to 100 K allows us to estimate the γ value of the electronic specific heat. The second major result is the observation of the anisotropic response of both $PuCoGa_5$ and $PuRhGa_5$ versus an applied magnetic field. Contrary to previously reported specific-heat data (for $PuCoGa_5$ only),[16] our present results provide clear evidence of the anisotropy in $PuCoGa_5$. The anisotropy in $PuRhGa_5$, so far observed only by magnetization measurements,[17] is now confirmed by another technique and on another sample. Finally, the temperature dependence of the critical fields and the Maki parameter were determined, and a possible Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state in these compounds is discussed.

II. EXPERIMENT

Polycrystals and large single crystals of $PuRhGa_5$ and $PuCoGa_5$ were synthesized. The single crystals were grown by a Ga flux method. Polycrystalline ingots were prepared by arc-melting stoichiometric amounts of the constituent elements under an atmosphere of high-purity argon on a water-cooled copper hearth, using a Zr getter. To ensure homogeneity, the arc-melted buttons were turned over and remelted three times. Weight losses were checked during the synthesis process and found to be negligibly small. The pellets were annealed at 750 °C for one week under high vacuum. The crystal structure and the purity of the polycrystalline samples obtained were then checked from x-ray powder diffraction data (Cu $K\alpha$ radiation) collected on a Bragg-Brentano Siemens D500 diffractometer. Single-crystal x-ray diffraction data were collected on an Enraf-Nonius CAD-4 four-circle diffractometer employing monochromated Mo $K\alpha$ radiation. The data processing was carried out using the MOLEN package.[18]

The specific heat was measured by the relaxation method on a PPMS-9 system (Quantum Design). For $PuCoGa_5$, we measured a 30 mg polycrystalline sample and a 1.4 mg single crystal. For $PuRhGa_5$, we examined a 20 mg polycrystalline sample above 6 K (the lowest temperature was limited by self-heating effects) and two small single crystals (1.2 and 0.8 mg) measured down to 3.3 K. The large polycrystalline samples were measured to achieve satisfactory precision at higher temperatures. All the samples were measured in magnetic fields up to 9 T with different orientations with respect to the crystallographic axes. The results on both $PuRhGa_5$ single crystals were identical within the experimental errors: we report data for the larger one.

The self-heating, mainly due to the ^{239}Pu, (i) limits the lowest achievable temperatures and (ii) causes the different parts of the measured system (sample, platform with thermometer, wires) to be at different temperatures. These differences increase with decreasing thermal conductivity of the wires at low temperatures. The standard data acquisition and analysis software does not take this effect into account, but measured data can be corrected for these errors.[19] In our case, the samples measured at low temperatures were relatively small and well thermally coupled to the platform, reducing the corrections to less than 0.5% of the total C_p value. The data presented include these corrections although the corrections do not have any significant influence.

III. RESULTS

The specific heat of $PuCoGa_5$ measured in zero field shows a clear anomaly at $T_c = 17.8$ K (Fig. 1). The T_c value is slightly lower than the originally reported value of $T_c = 18.5$ K,[1] presumably due to aging effects. The $C_p(T)$ data measured on both the single-crystal and polycrystalline samples are almost identical and are in good agreement with results published previously.[1,13] The specific heat of $PuRhGa_5$ and $URhGa_5$ is shown in Fig. 1 up to 100 K and in Fig. 2 with focus on the low-temperature part. The transition to the superconducting state is accompanied by a well-pronounced anomaly at $T_c \approx 9$ K.

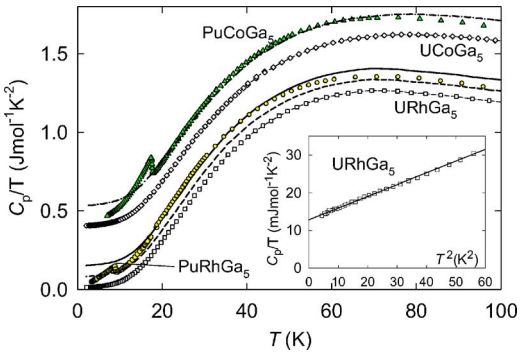

FIG. 1. (Color online) Specific heat of $PuCoGa_5$ (△), $UCoGa_5$ (◇, taken from Ref. 10), $PuRhGa_5$ (○), and $URhGa_5$ (□). Data for $ACoGa_5$(A=U,Pu) are shifted upward by 0.4 mJ mol^{-1} K^{-2} to avoid overlap with $ARhGa_5$ data. The lines correspond to a shift of C_p/T of the uranium analog to get the γ value of 130 mJ mol^{-1} K^{-2} in $PuCoGa_5$ (dash-dotted line), 150 mJ mol^{-1} K^{-2} in $PuRhGa_5$ (full line), and 80 mJ mol^{-1} K^{-2} in $PuRhGa_5$ (dashed line), respectively. Inset shows the low-temperature C_p/T vs T^2 plot for $URhGa_5$.

As $URhGa_5$ does not order magnetically, its specific heat consists of the lattice and electronic contributions only. At low temperatures (below 8 K), the lattice part follows a T^3 power law and the specific heat can be described by the simple formula

$$C_p = C_{latt} + C_{el} = \beta T^3 + \gamma T. \tag{1}$$

The fit to experimental data below 8 K allows us to extract the value of the Sommerfeld coefficient $\gamma = 13 \pm 1$ mJ mol^{-1} K^{-2} (somewhat higher than the 7 mJ mol^{-1} K^{-2} reported for a $URhGa_5$ single crystal[20]) and the Debye temperature $\Theta_D = 184$ K, assuming that it describes three acoustic phonon branches.

In a first approximation, we take the lattice specific heat of $URhGa_5$ as an estimation of this contribution also in $PuRhGa_5$. The electronic contribution in $PuRhGa_5$ is then calculated as

$$C_{el} = C_{\mathrm{Pu}} - C_{latt} = C_{\mathrm{Pu}} - C_{\mathrm{U}} + \gamma_{\mathrm{U}} T \tag{2}$$

and its low-temperature part is shown in the inset of Fig. 2. In the superconducting phase (i.e., below T_c), it can be well described by the power-law formula

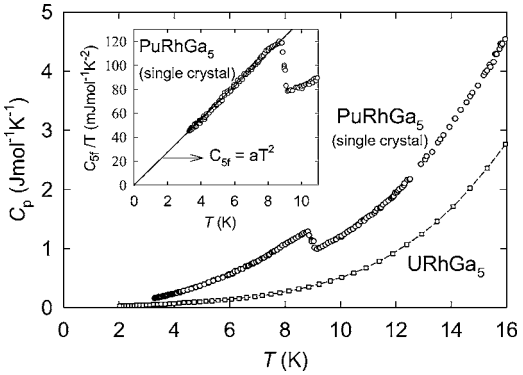

FIG. 2. Low-temperature specific heat of $PuRhGa_5$ single crystal and $URhGa_5$. Inset shows C_{el}/T of $PuRhGa_5$; the line is a fit to Eq. (3).

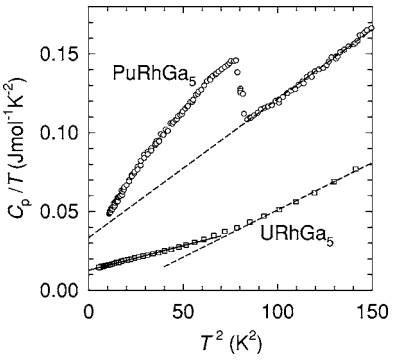

FIG. 3. C_p/T vs T^2 plot for $PuRhGa_5$ and $URhGa_5$ at low temperatures; the solid line is a linear fit to $URhGa_5$ data below 8 K (as in Fig. 1) where the simplification of the lattice specific heat to the T^3 power law (1) is correct, whereas the dashed lines representing linear extrapolation from the T^2 region between 80 and 150 K^2 (i.e., above T_c of $PuRhGa_5$) give clearly different slopes and would lead to wrong Debye temperatures and γ values.

$$C_{el} = aT^2 + \gamma_0 T \tag{3}$$

with $a = 13.8$ mJ mol^{-1} K^{-3} and γ_0 close to zero (0 ± 1 mJ mol^{-1} K^{-2}) as seen from Fig. 2.

The normalized specific heat jump at T_c, $\Delta C_{el}/T = 45 \pm 5$ mJ mol^{-1} K^{-2}, obtained for the three studied samples, is about three times smaller than that in $PuCoGa_5$.[1,13] Using the BCS relation $\Delta C_{el}/\gamma T_c = 1.43$, it yields a Sommerfeld γ coefficient ≈ 30 mJ mol^{-1} K^{-2}. Roughly the same γ value is extracted when applying Eq. (1) to the temperature region 9–12 K (i.e., just above T_c). However, this application is not valid in this case, as demonstrated in Fig. 3 for $URhGa_5$ data. The full line here corresponds to the correct application of (1) at $T \ll \Theta_D$ (see above), whereas the extrapolation from higher temperatures above 9 K clearly follows a different slope and leads to wrong values of γ and Θ_D. The phonon spectrum is, in this particular case, too complex to describe its specific heat by the simple T^3 law above ~ 8 K.

Considerably higher γ values are derived when comparing $URhGa_5$ and $PuRhGa_5$ data above the critical temperature: the value of γ is estimated between ≈ 80 and 150 mJ mol^{-1} K^{-2} (dashed and full line, respectively, in Fig. 1). The assumption of the same lattice contribution in Pu and U compounds is only a first approximation, but possible differences in the phonon spectra should not play a significant role when comparing $URhGa_5$ and $PuRhGa_5$ at low temperatures around T_c, where the lattice part is only a minor contribution in $PuRhGa_5$ (see Fig. 2). The value of $\gamma \approx 80$ mJ mol^{-1} K^{-2} inferred from our data just above T_c can thus be taken as a lower limit for γ estimation in the nonsuperconducting phase, more than twice higher than the value derived from experimental $\Delta C_{el}/T_c$ and the BCS relation.

Analogous analysis for $PuCoGa_5$ and $UCoGa_5$ (see Fig. 1) shows that $UCoGa_5$ gives a rather good estimation of the lattice specific heat in $PuCoGa_5$ (as well as in $NpCoGa_5$),[11] and we infer a value of $\gamma \approx 130$ mJ mol^{-1} K^{-2} in $PuCoGa_5$ (dash-dotted line in Fig. 1), which is finally surprisingly close to that (≈ 95 mJ mol^{-1} K^{-2}) derived from the BCS relation.[1,13]

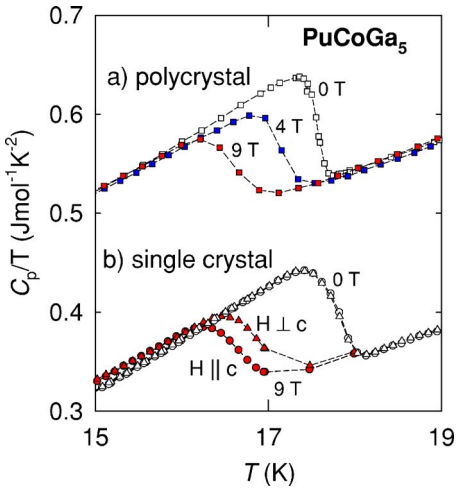

FIG. 4. (Color online) Specific heat of $PuCoGa_5$. (b) Single crystal with $H \| c$ (circles) and $H \perp c$ (triangles), and (a) polycrystal, shifted upward by 0.2 J mol^{-1} K^{-2} to avoid overlapping.

The effect of an applied field on the specific heats of $PuCoGa_5$ and $PuRhGa_5$ is demonstrated in Figs. 4 and 5, respectively. In both compounds, the results obtained for H along the a and b axes are almost identical, as expected. We show only one set of curves and denote $H \perp c$. The very high slope at T_c is determined in $PuCoGa_5$: $\mu_0 dH_{c2}/dT|_{Tc} \cong -8$ and -10 T/K for H parallel to c and perpendicular to it, respectively (see Fig. 6). In the case of $PuRhGa_5$, the critical temperature decreases with increasing field much faster. The inferred temperature dependencies of the critical field are represented in Fig. 6. We plot here also data derived from magnetization measurements by Haga *et al.*[17] In

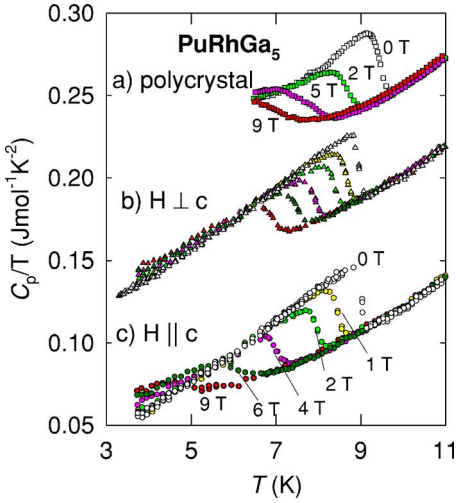

FIG. 5. (Color online) Specific heat of $PuRhGa_5$. (a) Polycrystal; and the single crystal with (b) $H \perp c$ and (c) $H \| c$. The individual sets of curves are shifted upward by 0.08 J mol^{-1} K^{-2} to avoid overlapping. The applied fields in (b) and (c) are identical. The critical temperature is slightly higher for the polycrystalline sample than for the single crystal, essentially due to self-irradiation damage (Ref. 21), as the single crystal was measured with one month aging, whereas the polycrystalline sample was measured with only three days aging.

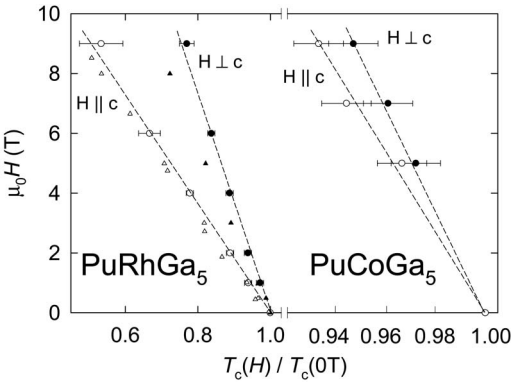

FIG. 6. Critical field $\mu_0 H_{c2}(T)$ determined by specific heat in $PuCoGa_5$ and $PuRhGa_5$ for field applied along the c axis (open circles) and the a axis (filled circles); the dashed lines are to guide the eye; the triangles represent data derived from magnetization measurements (Ref. 17). Due to the very high slope at T_c in these materials, very high fields are required to restore the normal state ($\mu_0 H > 10$ T). In the case of $PuRhGa_5$, we observe a linear variation of the upper critical field down to $T/T_c \sim 0.5$ for $H \| c$. Magnetization measurements (Ref. 17) indicate that this regime is present also for $H \| a$ in the same temperature range, and is even maintained down to $T/T_c \sim 0.1$ for $H \| c$ ($\mu_0 H = 14$ T).

$PuRhGa_5$, we find approximately an initial slope of $\mu_0 dH_{c2}/dT_{|Tc} \cong -2.0$ and -4.0 T/K for the field applied along c and perpendicular to it, respectively.

IV. DISCUSSION

The Pu-based 115 compounds are naturally compared with the CeTIn$_5$ superconductors. Several analogies, including the correlation of T_c with c/a, point to similar underlying physics in both groups of materials.[13] We find now another similarity when inspecting the electronic specific heat below T_c. The T^2 power law in the specific heat below T_c seems to be a common behavior in all these compounds—it is also found at low temperatures in $PuCoGa_5$,[1,13] and in isostructural Ce superconductors $CeRhIn_5$ under pressure,[22] $CeCoIn_5$, and $CeIrIn_5$.[23] Such a T^2 dependence indicates an axial state with line nodes in the superconducting gap structure, as in many unconventional d-wave superconductors, including $YBa_2Cu_3O_{6.95}$,[24] La_2CuO_4,[25] or UPd_2Al_3.[26] Our present $PuRhGa_5$ study reveals the T^2 dependence in a broad temperature region between the low-temperature limit of 3.3 K and up to the critical temperature. The fact that $C_p/T \sim T$ for $0.4 < T/T_c < 1$ is rather unusual. More often the power law behavior is seen below $\sim 0.5 T_c$ only. The former C_p data on $PuCoGa_5$ showed the T^2 dependence of C_{el} in the limited temperature range from 5.2 up to 7.2 K only[13] (the lower limit due to self-heating). In $CeCoIn_5$ also the power law occurs at lower T/T_c.[13] We have no clear explanation at this moment as to why $PuRhGa_5$ is an exception, showing the $C_p/T \sim T$ behavior up to T_c. The conclusion about the line nodes in the superconducting gap in $PuRhGa_5$ is furthermore corroborated by a recent nuclear quadrupole resonance study.[6]

The value of the coefficient $a \simeq 14$ mJ mol^{-1} K^{-3} in formula (3) is higher than the value of 5 mJ mol^{-1} K^{-3} found in $PuCoGa_5$, but still one order of magnitude lower than values observed in CeTIn$_5$ heavy-fermion superconductors ($a = 0.25$ and 3.6 J mol^{-1} K^{-3} in $CeCoIn_5$ and $CeIrIn_5$, respectively).[22,23] The $\gamma_0 \approx 0$ below T_c obtained in our case reveals that the Fermi surface is fully gapped (except for the line nodes) and the superconductivity is bulk and complete.

The gradual enhancement of C_p/T seen in $PuRhGa_5$ at the lowest measured temperatures when increasing the magnetic field (Fig. 5) might be due to an increase of the linear term in (3) by a certain value $\gamma(H)$, indicating a field dependence of the density of states at the Fermi level. The lower temperature limit of our measurement does not allow any further quantitative analysis that could be compared with theoretical predictions as discussed, e.g., for $YBa_2Cu_3O_{6.95}$.[24]

The γ value in the normal state of $PuRhGa_5$, although estimated with higher uncertainty, is comparable to that of $PuCoGa_5$. The twice lower critical temperature in $PuRhGa_5$ could be due to a weaker pairing potential, which might depend on the c/a ratio.[4] The strong sensitivity of T_c to the c/a ratio with almost unchanged γ coefficient when substituting Co or Pu corroborates this assumption.[14]

The decrease of T_c when applying a magnetic field is stronger for $PuRhGa_5$ than for $PuCoGa_5$. It corresponds well to the different upper critical field H_{c2} in both compounds as inferred from the resistivity data.[1,2] The extracted value of $\mu_0 dH_{c2}/dT_{|Tc}$ in $PuCoGa_5$ is practically the same as the one reported in previous work.[16] More interesting is the anisotropy of the field influence on T_c. It is clearly observed in $PuRhGa_5$ that T_c is suppressed approximately twice faster if the magnetic field is applied along the tetragonal c axis rather than perpendicular to it. Our observation is corroborated also by recent magnetization measurements,[17] which reveal the slope of $-dH_{c2}/dT = 2.0$ and 3.5 T/K for $H \| c$ and $H \| a$, respectively, well comparable with our values. A similar size of the T_c suppression for $H \| c$ is inferred also from NMR measurements:[6] T_c decreases to ~ 4 K in 11 T. The same tendency for anisotropy is found in $PuCoGa_5$, although the anisotropy here is lower and less evident, as the maximum available field extends our $PuCoGa_5$ data to $T_c(H)/T_c(0) \sim 0.93$ only. The observed anisotropy is another strong analogy to the Ce-based isostructural superconductors $CeCoIn_5$ and $CeIrIn_5$.[27] The anisotropy of H_{c2} is here well explained by the anisotropic effective mass, reflecting a quasi-two-dimensional ellipsoidal Fermi surface. Our measurements, in agreement with magnetization data,[17] would imply an ellipsoidal Fermi surface with the main axis ratio $k_c/k_a \simeq 2.0$ in $PuRhGa_5$, slightly lower than in $CeTIn_5$ compounds. The anisotropy of dH_{c2}/dT as inferred from our data is considerably lower in $PuCoGa_5$, implying roughly $k_c/k_a \simeq 1.3$. This value should be taken with care as the data are limited to $T_c(H)/T_c(0) \sim 0.93$, and may differ in higher fields.

It is worth stressing that two different techniques (specific heat and magnetization[17]) applied on different samples both show exactly the same unusual behavior of the $PuRhGa_5$ upper critical field $H_{c2}(T)$, confirming this intrinsic behavior of the material. This deviation of $H_{c2}(T)$ from saturation when approaching 0 K is commonly observed in d-wave superconductors[28,29] such as the related compound

TABLE I. Critical temperature T_c, initial slope $\mu_0 dH_{c2}/dT$ obtained for $PuCoGa_5$ and $PuRhGa_5$ in both $H\|c$ and $H\perp c$ directions, Maki parameter α, and inferred critical fields: Pauli limited $\mu_0 H_p(0)$, orbital limited $\mu_0 H^*_{c2}(0)$, and combined (Pauli and orbital limited, using Maki formula) $\mu_0 H^{**}_{c2}(0)$.

	$PuCoGa_5$		$PuRhGa_5$	
	$H\|c$	$H\perp c$	$H\|c$	$H\perp c$
T_c (K)	17.8	17.8	9.0	9.0
$-\mu_0 dH_{c2}/dT$ (T/K)	8	10	2	4
$\mu_0 H_p(0)$ (T)	33	33	17	17
$\mu_0 H^*_{c2}(0)$ (T)	98.7	123.4	12.5	25.0
α	4.22	5.28	1.06	2.11
$\mu_0 H^{**}_{c2}(0)$ (T)	22.8	23.0	8.6	10.7

$CeCoIn_5$,[30] which develops a FFLO (Refs. 31 and 32) state.[33,34] This state can in principle occur only in *d*-wave superconductors in the clean limit for strongly Pauli-limited systems with a high Maki parameter and presenting low or reduced dimensionality (one or two dimensions). The possibility of such behavior in $PuTGa_5$ (T=Co,Rh) systems is discussed below.

The Pauli-limited critical field[35] can be inferred from

$$\mu_0 H_p(0) = 1.84 T_c, \tag{4}$$

whereas the orbital-limited critical field,[36] the Maki parameter,[37] and the combined orbital and paramagnetic limited critical field[38] can be inferred from the initial dH_{c2}/dT slope by

$$\mu_0 H^*_{c2}(0) = -0.693 T_c (\mu_0 dH_{c2}/dT)_{T=T_c}, \tag{5}$$

$$\alpha = -0.528 (\mu_0 dH_{c2}/dT)_{T=T_c}, \tag{6}$$

and

$$H^{**}_{c2}(0) = H^*_{c2}(0)(1+\alpha^2)^{-1/2}, \tag{7}$$

respectively. The values obtained for $PuCoGa_5$ and $PuRhGa_5$ in different directions are listed in Table I. For both compounds, the anisotropy of the $(\mu_0 dH_{c2}/dT)_{T=T_c}$ slope is reflected in the H^*_{c2} field. The values obtained for the Maki parameter and the *d*-wave character previously reported[5,6] would qualify both compounds for a possible FFLO phase. However, several considerations have to be taken into account. First, the true value of the upper critical field at 0 K of $PuCoGa_5$, in both the Pauli and orbital limits (33 T and, depending on the orientation, 98/123 T, respectively) are experimentally inaccessible. Nonetheless, Table I shows that $PuCoGa_5$ is probably controlled by the orbital limit, which should prevent the occurrence of a FFLO state in this compound. Only future measurements in very high fields ($\mu_0 H > 30$ T) could confirm or refute this assumption. Second, the FFLO state is in principle very sensitive to (suppressed by) impurities, which would make it difficult to observe in a plutonium-based system where the self-decay of ^{239}Pu permanently creates defects. This phenomenon is more pronounced in $PuRhGa_5$, which rapidly becomes a dirty superconductor.[21] Nevertheless, the Rh compound would present a more favorable configuration for the magnetic field applied along the *c* axis, where the paramagnetic limit plays a role, and the weak negative curvature of the critical field for H perpendicular to the *c* axis could be ascribed to the beginning of a FFLO effect (which would then occur around $0.55T_c$, as theoretically predicted). However, such an assumption remains very speculative for the reasons discussed above, and also because we do not see any evolution from second- to first-order character of the superconducting transition in the specific heat, which should occur in the vicinity of the FFLO state.[39]

Another reason for the linear regime at low temperature of $H_{c2}(T)$ could be the possibility of a strong electron-phonon coupling effect, implying phonon modes at low energies. This phenomenon, commonly observed for BCS-like superconductors, has also been revealed for some unconventional superconductors.[40,41] The main consequence of this strong coupling is an enhanced critical field at low temperatures, which is more or less what we observe, especially in $PuRhGa_5$. Recent predictions[42] and measurements[43] achieved on $PuCoGa_5$ by synchrotron radiation would point to this possibility. Nevertheless, none of these has been performed yet on $PuRhGa_5$, and we can only assume a similar behavior between these two systems.

Finally, the revealed anisotropy of the superconductivity suppression in $PuTGa_5$ is very comparable to the magnetic anisotropy observed in the isostructural Np-based compounds. The antiferromagnetic order in both $NpCoGa_5$ (Ref. 44) and $NpRhGa_5$ (Ref. 45) is suppressed more rapidly for H parallel to *c* than perpendicular to it. It is tempting to conclude that there is a correlation between the magnetic and superconducting properties of both the $PuTGa_5$ and $NpTGa_5$ series. Indeed, band-structure calculations predict that antiferromagnetic interactions can be expected to be present in $PuTGa_5$ and that antiferromagnetic (AF) spin fluctuations play a role in Cooper pair formation.[4] The suppression of superconductivity could then be related to the suppression of these AF fluctuations.

V. CONCLUSIONS

$PuCoGa_5$ and $PuRhGa_5$ undergo a transition to the superconducting state at $T_c \sim 18$ and 9 K, respectively, the critical temperature depending also on sample aging. The electronic specific heat below T_c follows a quadratic temperature dependence that indicates an axial state with line nodes in the superconducting gap structure. The Sommerfeld coefficient γ is estimated in $PuRhGa_5$ between 80 and 150 mJ mol^{-1} K^{-2} from $C_p(T)$ data above T_c. The reduction of T_c under applied external magnetic field is faster in $PuRhGa_5$ than in $PuCoGa_5$ and anisotropic in both compounds. The anisotropy is larger for $PuRhGa_5$, where the suppression of T_c is twice stronger for the field applied along the *c* axis than perpendicular to it. In $PuCoGa_5$, we notice that for both directions the orbital-limited critical field H^*_{c2} is much higher than the Pauli-

limited one. The difference between H_{c2}^{*} and other critical fields is less obvious in $PuRhGa_5$ and, for $H\|c$, H_{c2}^{*} is lower than H_p. We can conclude that the paramagnetic (spin) limitation does not affect $PuCoGa_5$ in which the $5f$ Cooper pair breaking is driven by the orbital limitation. On the contrary, it plays a role in $PuRhGa_5$, where the values of the Maki parameter, anisotropy, and Pauli limitation for $H\|c$ could suggest the occurrence of a FFLO state. Deviation from the saturation of H_{c2} observed in this direction for different samples and by different techniques would lead us to consider this phenomenon as intrinsic, and related effectively to the possible development of a FFLO state or strong electron-phonon coupling.

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