# Discovery of new heavy fermion superconductor and related compounds: $RPd_2Al_3$ (R = U, Ce)

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A new family $RPd_2Al_3$ (R: rare earth and actinide elements) crystallized with the hexagonal $PrNi_2Al_3$ type structure were synthesized. Among them, $UPd_2Al_3$ is a new heavy fermion superconductor with a record superconducting transition temperature $T_c$ of 2 K. In addition, it shows a transition to long-range antiferromagnetic order at Néel temperature $T_N = 14$ K and heavy fermion superconductivity coexists with antiferromagnetism below $T_c$. On the other hand, $CePd_2Al_3$ is a new heavy fermion antiferromagnet. The sample dependence of magnetic properties observed at low temperatures suggests that the Kondo interaction competes well with the RKKY interaction in $CePd_2Al_3$.

## Introduction

In recent years, a lot of intermetallic cerium and uranium compounds exhibiting unusual electronic and magnetic properties at low temperatures have attracted much attention in the solid state physics. At low temperatures, the hybridization between the f-electrons and the conduction electrons leads to the screening of the local f magnetic moment, to the formation of the Kondo lattice and to the formation of quasi particles with effective masses from 100 to 1000 times the mass of the free electron, the "heavy fermions" (HF). Especially, the quasi particles play important roles of superconductivity and magnetism in HF-superconductors. The origin of HF-superconductivity, however, is still completely uncertain.

Therefore the discovery of a new HF-superconductor, $UPd_2Al_3$ ($T_c = 2$ K and $T_N = 14$ K),[1)] by the collaboration of the author and Prof. Steglich's group at the beginning of 1991 brought new insight into these phenomena. During the search for a new HF-superconductor, we found a new HF antiferromagnet $CePd_2Al_3$ in the same crystal structure.[2)] It is important to compare $UPd_2Al_3$ with nonsuperconductng $CePd_2Al_3$.

This article reviews our recent work on the superconducting and magnetic properties of $UPd_2Al_3$ and $CePd_2Al_3$ in collaboration with Prof. Stegich's group.

Polycrystalline samples were synthesized by melting the pure elements in an arc furnace. As-cast samples were annealed at 900 °C for 120 h at a pressure of about $1 \times 10^{-5}$ Torr. Single crystalline samples of $CePd_2Al_3$ were grown by the Czochralski method using a triarc furnace. The crystal structure of $RPd_2Al_3$ was confirmed as the hexagonal $PrNi_2Al_3$ type structure (P6/mmm) by the X-ray diffractometry.

## Coexistence of superconductivity and antiferromagnetism in $UPd_2Al_3$

The bulk superconductivity of $UPd_2Al_3$ was confirmed by the observation of sharp drops at $T_c = 2$ K to an unresolvably small value in the temperature dependence of the electrical resistivity $\rho(T)$ and to a diamagnetic value in the temperature dependence of the magnetic susceptibility $\chi(T)$. A Meissner volume fraction was determined to be 67% by the $\chi(T)$ measurement in the field-cooling process under $B = 0.05$ mT. The specific heat $C(T)$ under zero field shows a huge and sharp jump $\Delta C$ at $T_c$ (see Fig. 1a). In the normal state well below $T_N$, the specific heat can be approximated by a straight line when plotted as $C/T$ vs $T^2$. The electronic specific heat coefficient $\gamma$ in the normal state is estimated to be 150 mJ/mol $K^2$ by this plot. Both large $\Delta C$ and $\gamma$ imply that heavy electrons are ascribed to the formation of Cooper pairs because $\gamma$ is approximately proportional to the effective mass of conduction electrons. $T_c = 2$ K is the highest value which has ever been reported.

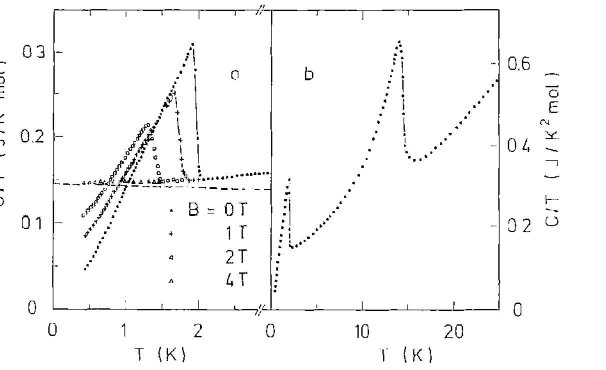


Fig. 1. Specific heat of $UPd_2Al_3$ as $C/T$ vs $T^2$, (a) in the temperature range between 0.4 K and 3 K under several magnetic fields and (b) in the temperature up to 25 K under zero field (after Ref. 1). Dashed curve in (a) represents electronic contribution, $C_{el}(T)/T$. Thin solid lines are guide to the eyes.

On the other hand, the anomaly of $C(T)$ at 14 K as shown in Fig. 1b indicates that $UPd_2Al_3$ undergoes the antiferromagnetic ordering state below $T_N = 14$ K. In fact, the recent experimental result of neutron diffraction[3)] revealed the magnetic structure as follows: (1) the ordered magnetic moment is 0.85 $\mu_B$, which is the largest value observed for any HF superconductor, (2) the magnetic moment is ferromagnetically aligned in the basal plane and each plane is antiferromagnetically coupled.

Thus $UPd_2Al_3$ is the first compound in which HF superconductivity coexists with antiferromagnetically ordering between local U-derived moments of ordinary size.

## Large sample dependence in heavy fermion $CePd_2Al_3$

The temperature dependence of the reciprocal magnetic susceptibility $1/\chi(T)$ for single-crystalline samples reveals that the magnetic easy axis lies in the basal plane as in the case of $UPd_2Al_3$[4)] as shown in Fig. 2. Each effective magnetic morment $\mu_{eff}$ obtained from a Curie - Weiss (C-W) fitting in the temperature range 100 K-300 K is close to the value of the free $Ce^{3+}$-ion, 2.54 $\mu_B$. Each curve deviates from the C-W

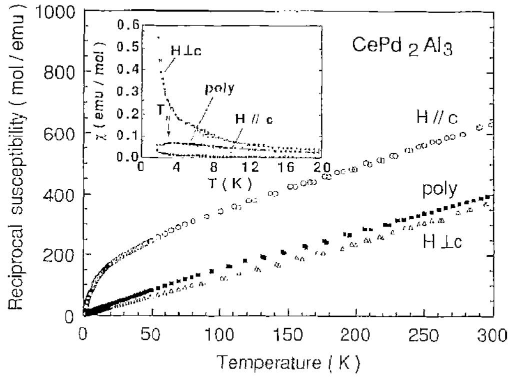


Fig. 2. Temperature dependence of the reciprocal magnetic susceptibility of the polycrystalline and the single-crystalline $CePd_2Al_3$ for $B = 0.1$ T. The inset shows the temperature dependence of the magnetic susceptibility at low temperatures.

law at lower temperatures due to the crystalline electrical field effect. However, a large difference of magnetic properties at low temperatures between polycrystalline and single-crystalline samples was observed as shown in the inset. The $\chi_{poly}(T)$ for the polycrystalline sample has a kink at $T_N = 2.8$ K due to the antiferromagnetic ordering. The neutron scattering measurement[5)] confirmed the antiferromagnetic structure below $T_N$ which is similar to that of $UPd_2Al_3$. On the other hand, the $\chi_{//c}(T)$ and $\chi_{\perp c}(T)$ for the single-crystalline sample have no anomaly and increase monotonously down to 1.7 K. In fact, recent experimental results of electrical resistivity and specific heat exhibit prominently different magnetic ground states at low temperatures. The $\gamma$ value in the polycrystalline sample is 380 mJ/mol $K^2$, which is 42 times larger than 8.9 mJ/mol $K^2$ of nonmagnetic $LaPd_2Al_3$. The single crystalline sample does not undergo any magnetic phase ordering down to 1.5 K. Especially the $\gamma$ value in the single-crystalline sample increases to 1.0 J/mol $K^2$ at 1.5 K with decreasing temperature.

Thus, these results indicate a strong competition between the Kondo and RKKY interactions ($T_K \approx T_{RKKY}$) and that this system is close to the transition from a magnetic Kondo lattice to a nonmagnetic heavy fermion compound. The origin of the sample dependence at low temperatures is uncertain. Further measurements to clarify the real ground state of $CePd_2Al_3$ are in progress.

## References


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