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Unconventional superconductivity of $NpPd_5Al_2$

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Abstract

The high quality single crystals of $NpPd_5Al_2$ with the body-centered tetragonal structure were grown by the Pb flux method. $NpPd_5Al_2$ was found to be the first Np-based heavy fermion superconductor with the relatively high critical temperature $T_{sc} = 4.9$ K. The upper critical field H_{c2} is large and highly anisotropic. Corresponding to the heavy electronic state, the initial slope of H_{c2} is large, but H_{c2} at low temperatures is suppressed by the magnetic field, indicating a strong Pauli paramagnetic effect and the first-order transition at H_{c2}. These results imply that $NpPd_5Al_2$ is located at the proximity of the antiferromagnetic ordering, which might be hidden by the superconductivity. The d-wave superconductivity with a spin singlet state is most likely realized in $NpPd_5Al_2$.

(Some figures in this article are in colour only in the electronic version)

Unconventional superconductivity is one of the most interesting topics among the strongly correlated electron systems. After the discovery of superconductivity in $CeCu_2Si_2$ [1], many unconventional superconductors have been reported, and their novel physical phenomena, for example, superconductivity without inversion symmetry or the coexistence of ferromagnetism and superconductivity, have opened the frontiers of heavy fermion physics. The discovery of superconductivity in $PuCoGa_5$ and $PuRhGa_5$ has also provided a new perspective on the physics of transuranium compounds [2, 3]. It is important to clarify experimentally the electronic state of transuranium compounds including Np and Pu.

In general it is believed that the 5f electrons of neptunium compounds have an itinerant nature. Recently we grew many single crystals of neptunium compounds and clarified the Fermi surface properties by the de Haas–van Alphen (dHvA) effect. For example, the dHvA oscillations were observed in the antiferromagnetic state of $NpRhGa_5$ with tetragonal structure [4]. The four kinds of cylindrical Fermi surfaces detected were well explained by the spin- and orbital-polarized band calculation based on the 5f-itinerant model. The cyclotron masses are extensively enhanced, indicating that the itinerant 5f electrons contribute to the conduction band and the heavy electronic state is realized.

We continued investigating neptunium compounds and obtained a new ternary compound $NpPd_5Al_2$. Surprisingly, $NpPd_5Al_2$ was found to be the first Np-based heavy fermion superconductor [5]. Novel electronic and superconducting properties were observed in this compound.

The Np metal as a starting material was prepared by electrolysis [6]. The single crystals of $NpPd_5Al_2$ was grown by the Pb flux method. The starting materials were put into an alumina crucible, which was sealed in a quartz ampule with Ar atmosphere gas. The quartz ampule was heated up to the maximum temperature of 1050 °C and was maintained for 2 days by the electric furnace. Then the temperature was decreased at a slow cooling rate of 0.5–1.5 °C h^{-1}. At 600 °C, the furnace was switched off. The Pb flux was removed by spinning off the ampule in the centrifuge. Many small single crystals were obtained. The typical size is $1 \times 1 \times 0.5$ mm^3, as shown in figure 1. The crystals were analyzed by x-ray diffraction and EPMA measurements. As shown in figure 2, $NpPd_5Al_2$ crystallizes in the body-centered tetragonal structure with $ZrNi_2Al_5$-type (space group: $I4/mmm$) [7]. It is

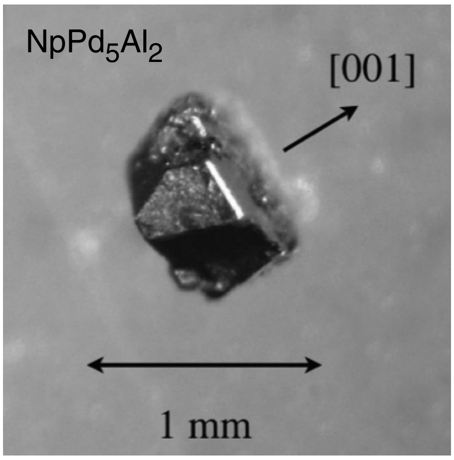

Figure 1. Photograph of $NpPd_5Al_2$.

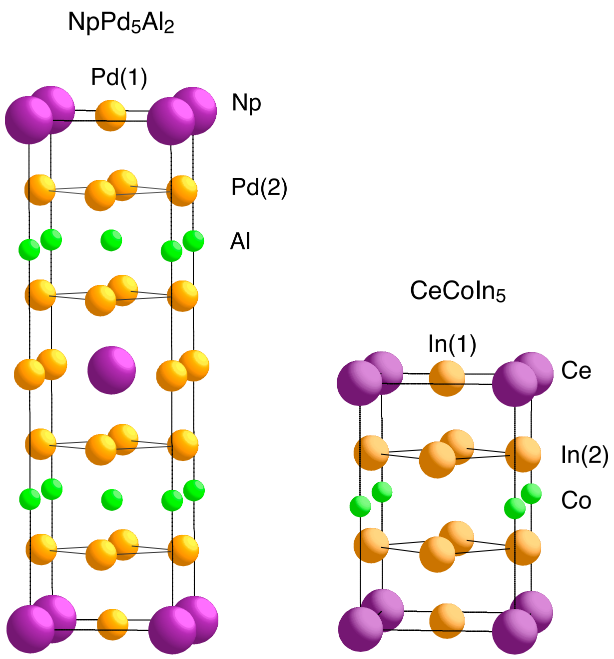

Figure 2. Crystal structures of $NpPd_5Al_2$ and $CeCoIn_5$.

characteristic that the lattice parameter c (=14.716 Å) is much larger than the value of a (=4.148 Å). The crystal structure of $NpPd_5Al_2$ can be compared to that of the well-known heavy fermion superconductor $CeCoIn_5$ [8]. At first glance, there is a similarity between $NpPd_5Al_2$ and $CeCoIn_5$. In $CeCoIn_5$, the uniaxially distorted $AuCu_3$-type layers of $CeIn_3$ and $CoIn_2$ layers are stacked sequentially along the c direction. On the other hand, the $NpPd_3$ layers and the Pd_2Al_2 layers are stacked sequentially along the c direction in $NpPd_5Al_2$. The lattice parameter c value of $NpPd_5Al_2$ is almost twice that of $CeCoIn_5$. It is noted that there were no reports of rare earth and actinide compounds with the same structure before $NpPd_5Al_2$. Recently other compounds, namely UPd_5Al_2, $ThPd_5Al_2$, $PuPd_5Al_2$ and $CePd_5Al_2$, were reported [7, 9, 10]. Interestingly, the antiferromagnet $CePd_5Al_2$ also becomes superconductive at around 10 GPa with the paramagnetic effect of the upper critical field [11].

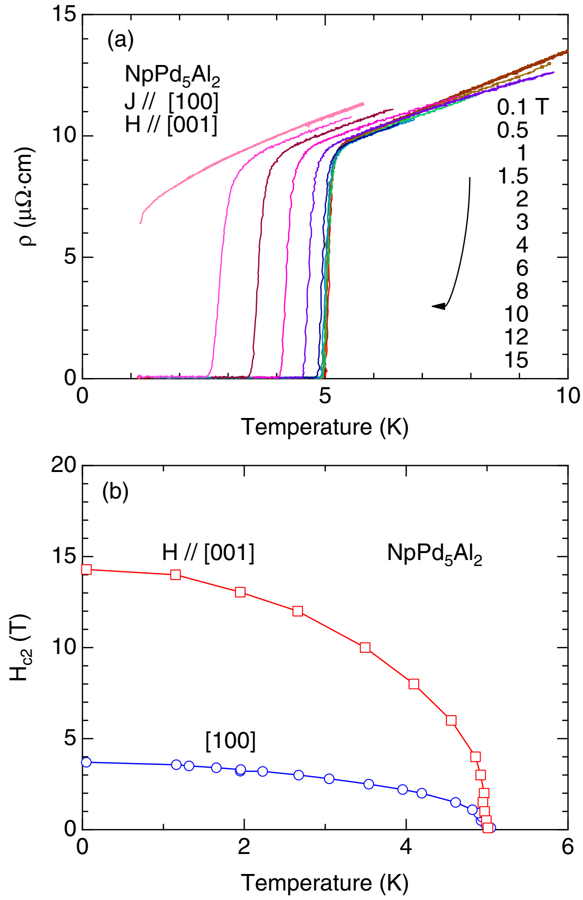

Figure 3. (a) Temperature dependence of the resistivity under magnetic fields for $H \parallel [001]$ and (b) the temperature dependence of the upper critical field H_{c2} for $H \parallel [001]$ and [100] in $NpPd_5Al_2$.

Figure 3(a) shows the temperature dependence of the electrical resistivity under magnetic fields for $H \parallel [001]$. The resistivity at 1 kOe follows the T linear dependence at high temperatures, indicating the non-Fermi liquid behavior. The superconductivity is realized below $T_{sc} = 4.9$ K with a sharp drop of resistivity. With increasing fields, T_{sc} shifts to lower temperatures. We show in figure 3(b) the temperature dependence of the upper critical field H_{c2} for $H \parallel [001]$ and [100]. Here the critical temperature was defined as a midpoint of the resistivity drop. The phase diagram demonstrates that the H_{c2} is strongly suppressed by the magnetic field for both field directions, indicating the strong Pauli paramagnetic effect with the spin singlet state of Cooper pairing. The values of H_{c2} at 0 K are 14.3 and 3.7 T for $H \parallel [001]$ and [100], respectively. The initial slopes of H_{c2} at T_{sc}, namely $-dH_{c2}/dT_{sc}$ (= H_{c2}'), are 31 and 6.4 T K^{-1} for $H \parallel [001]$ and [100], respectively, which are compared to the values of the typical heavy fermion superconductor $CeCoIn_5$ ($H_{c2}' = 24$ and 11 T K^{-1} for $H \parallel$ [100] and [001], respectively)

The Pauli paramagnetic limiting field is defined as $H_P = \frac{\sqrt{2}\Delta}{g\mu_B}$. On the basis of the weak-coupling BCS model, H_P is written as $H_P = 1.86T_{sc}$, assuming that the gyromagnetic ratio g is equal to 2. In $NpPd_5Al_2$, one can obtain the value of H_P as 9.1 T, which cannot explain the larger experimental value of 14.3 T for $H \parallel [001]$. This implies the large superconducting gap Δ and/or small g value. In fact, the electronic specific heat shows the large jump with the value of $\Delta C/\gamma T_{sc} = 2.33$ at T_{sc}, which is larger than the weak-coupling BCS value of 1.43, indicating the strong-coupling superconductivity in $NpPd_5A_2$ [5], which is also confirmed by the NMR measurement [12].

Here we can simply estimate the so-called Maki parameter $\alpha = \sqrt{2}H_{orb}/H_P$. The orbital limiting field H_{orb} can be written as $H_{orb} = 0.73H_{c2}'T_{sc}$. Corresponding to the heavy electronic state of $NpPd_5Al_2$, one can obtain the large orbital limiting fields, $H_{orb} = 111$ and 23 T for $H \parallel [001]$ and [100], respectively. If we assume that H_{c2} is determined by the Pauli limiting field H_P, the Maki parameters are deduced as $\alpha = 31$ and 8.8 for $H \parallel [001]$ and [100], respectively. Theoretically, the first-order transition at H_{c2} is expected when α is larger than 1.8 [13, 14]. The extremely large Maki parameters in $NpPd_5Al_2$ suggest the existence of the first-order transition at H_{c2} for both field directions, as observed in $CeCoIn_5$. In fact, the magnetization curves at low temperatures for $H \parallel [100]$ in $NpPd_5Al_2$ shows the step-like behaviors at H_{c2}, indicating the first-order transitions [5].

In summary, we grew high quality single crystals of a new compound $NpPd_5Al_2$ with a body-centered tetragonal structure. $NpPd_5Al_2$ is the first Np-based heavy fermion superconductor with the relatively high critical temperature, $T_{sc} = 4.9$ K. The upper critical field H_{c2} is large and highly anisotropic. The strong Pauli paramagnetic effect was detected, together with the first-order transition at H_{c2}.

Acknowledgments

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