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Magnetic and Thermal Properties of Heavy Fermion Superconductor UPd₂Al₃

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Abstract

1. Introduction

The magnetism and the superconductivity of the heavy-fermion (HF) superconductors (UBe₁₃, UPt₃, URu₂Si₂, etc.) is one of the most interesting problems of condensed matter physics. But the origin of these phenomena is still unsolved. New uranium compound UPd₂Al₃ (hexagonal) was added to the HF superconductors ($T_C \sim 2\text{K}$, $T_N \sim 14\text{K}$). In the superconducting state, it shows large magnetic moments ($0.85\mu_B/\text{U}$) in comparison with UPt₃ ($0.03\mu_B/\text{U}$) or URu₂Si₂ ($0.02\mu_B/\text{U}$). Therefore UPd₂Al₃ is a proper compound to study the magnetism and superconductivity of HF systems. In this paper, I study the properties of 5f electrons of the superconducting and normal states of UPd₂Al₃ by the specific heat, the electrical resistivity, and the magnetic susceptibility. I also investigate the dilution effect of U atom by La atom (U_xLa_{1-x}Pd₂Al₃) on these magnetic properties.

2. Experimental

Single crystals of nominal composition of UPd_{2+x}Al_{3+y} and U_xLa_{1-x}Pd₂Al₃ were made by the Czochralski pulling method using a triarc furnace. I made the specific heat measurement system by an adiabatic heat pulse method using a ³He-⁴He dilution refrigerator. The specific heat measurement was made in a ³He-⁴He dilution refrigerator ($0.2\text{K} \leq T \leq 1\text{K}$) and ³He cryostat ($0.8\text{K} \leq T \leq 20\text{K}$). The magnetic susceptibility was measured using a SQUID magnetometer (Quantum Design Co. LTD.) in a ⁴He cryostat and the induction method in a ³He cryostat. The electrical resistivity was measured by the DC-4 probe method.

3. Results and discussion

In order to obtain the pure single crystal, I made 4 samples; UPd₂Al₃(#0), UPd₂Al_{3.03}(#1), UPd₂Al_{3.06}(#6) and UPd_{2.02}Al_{3+y}(#H2). #1 and #H2 were annealed for 5h at 900°C and 800°C, respectively. In the electrical resistivity measurement, #H2 shows highest T_C and RRR. Therefore I can decide #H2 is the best sample quality of all.

The superconducting upper critical field $H_{C2}(T)$ of #0 was measured by the electrical resistivity in the magnetic field applied parallel to the current. The $H_{C2}(T)$ shows little anisotropy for $I//c$ and $I \perp c$. And the $H_{C2}(T)$ curves show remarkable flattening, in contrast to those of other Uranium superconductors. This feature suggests that UPd_2Al_3 reveals strong Pauli limiting effect. The experimental result of $H_{C2}(T)$ of UPd_2Al_3 is explained by the WHH theory which takes the spin paramagnetic effect into consideration. This result suggests that UPd_2Al_3 is a singlet pairing state.

NMR (Kitaoka) and μ^+SR (Amato) studies suggest that the spin susceptibility decreases below T_C , therefore the electrons of the superconducting state form singlet pairs. These results coincide with that of the $H_{C2}(T)$.

The specific heat of $UPd_{2+x}Al_{3+y}$ in the superconducting state has been measured and it reveals T^n dependence. For $0.2K \leq T \leq 1K$ the specific heat is roughly proportional to $T^{2.5}$. However, for $0.2K \leq T \leq 0.5K$, it shows T^3 dependence clearly. In order to investigate the gap structure, we must study the temperature dependence of specific heat much lower than T_C . And the coefficient of cubic term was about 100 times larger in the superconducting state than that in the normal state, therefore I consider the ground state of the specific heat is proportional to T^3 and it reveals points node on the Fermi surface.

Kitaoka et.al. measured the $^{27}(1/T_1)$ of poly crystal UPd_2Al_3 by ^{27}Al -NMR. The result below T_C shows T^3 dependence and they suggest that the energy gap vanishes along lines on the Fermi surface, which is different with my specific heat measurement. Some heavy fermion superconductors reveals difference between the result of specific heat and NMR measurement, and now, this problem is open question.

In order to make further investigation of the property of 5f electrons, I made single crystal $U_xLa_{1-x}Pd_2Al_3$ and studied the magnetic susceptibility and the electrical resistivity.

The electrical resistivity of $U_xLa_{1-x}Pd_2Al_3$ increases with decreasing temperature. It looks like the Kondo effect. As the U atom is replaced by the La atom, the Neel temperature T_N is lowered (14.5K for $X=1$, 9K for $X=0.9$, 3K for $X=0.75$). For $X=0.6$ and $X=0.5$, the inverse magnetic susceptibility $1/\chi$ shows zero around 5K and the magnetization shows hysteresis below 4.2K, which reveals the ferromagnetic state. As the U atom is diluted with the La atom further, the magnetic susceptibility shows anisotropy similar to UPd_2Al_3 . But the gradient of $1/\chi$ becomes smaller with decreasing the U concentration, which differs from $Ce_xLa_{1-x}B_6$. The origin of this matter is, I think, the polarization of the spins of the electrons by the magnetic moments of U atom, or the existence of the constant term χ_0 independent with the temperature and U concentration. In order to solve this problem, we must make more investigation of the magnetic property of $U_xLa_{1-x}Pd_2Al_3$.