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Anomalous field dependent heat capacity in UPt_3 below 1 K

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Abstract

Specific heat measurements of polycrystalline UPt_3 have been carried out in temperatures between 60 mK and 4.2 K and in magnetic fields up to 2.5 T in order to reveal the nature of the anomalous peak in the specific heat found at 18 mK [1]. In zero magnetic field, γ, the linear term of the specific heat in temperature is nearly constant, being 420 mJ mol^{-1} K^{-2} below 1 K. In magnetic fields, however, it is observed that the specific heat tends to increase sharply as temperature is lowered and amounts to a value even larger than 10 J mol^{-1} K^{-2} at 75 mK in 2.5 T. This observation is reminiscent of the zero field anomaly found by Schuberth *et al.* [1] and is attributed to the electronic cooperative phase transition in the heavy quasi-particles in UPt_3.

1. Introduction

The ground state of the heavy fermions has been known as either a normal metal down to low temperatures with heavy quasi-particles in a Fermi liquid, magnetically ordered state with partially reduced Fermi surfaces due to the localization effect of itinerant electrons or as a superconducting state formed in heavy quasi-particles. In some cases, it has been claimed that a magnetic order with small moments co-exists with normal metallic behavior with heavy quasi-particles, where the system may eventually undergo superconducting state. In $CeAl_3$, $CeCu_6$ and $CeRu_2Si_2$ for example, weak magnetic order with ordered moment of 0.01-0.03 μ_B has been observed by muon spin rotation experiments. In UPt_3, co-existence of the weak magnetic order at 5 K [2,3] with the low temperature superconducting state, which exhibits a clear double transition at 0.54 K and 0.488 K in the specific heat, has been observed [4,5]. The multiple superconducting phases as well as antiferromagnetic spin correlations persisting in the superconducting state in UPt_3 have been thought to be evidence of d-wave superconductivity as a ground state in this system.

In 1991, Schuberth *et al.* [1] have reported for the first time the specific heat measurements of UPt_3 down to 6.5 mK using the nuclear demagnetization technique. The most striking feature observed in their measurements was the huge peak in the specific heat at 18 mK. They argued that the most probable origin of the anomaly is electronic, and they excluded the possibility of either the nuclear Schottky contribution of Pt or the nuclear quadrupole effect of ^{235}U.

2. Experimental details

The specific heat was measured by a semi-adiabatic pulse heating method in a dilution refrigerator below 4.2 K. The lowest temperature reached is about 25 mK but actual reliable measurements were pursued only above about 50 mK. The calibrated carbon glass thermometer was used in zero and in magnetic fields.

The sample was prepared in a polycrystalline form by arc-melting of highly depleted ^{235}U metal and high purity platinum metal (99.995%). The depletion level of ^{235}U is as high as 100 ppm, as determined by neutron absorption measurements [6]. The sample of UPt_3 with a mass of 47.3 mg was cut from a large ingot into a thin plate in order to reduce the Kapitza heat resistance between sample and sample holder as much as possible and was annealed at 900 °C for 1 week before the experiments. The magnetic field up to 2.5 T was generated by a small superconducting solenoid.

3. Results and discussion

The specific heat in zero field of UPt_3 above the superconducting transition temperature $T_c \approx 0.53$ K is well established with a value of $\gamma \approx 420$ mJ mol^{-1} K^{-2}.

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Below T_c, it is observed that there exists a clear double superconducting transition in the specific heat in certain samples [4,5]. In addition, in approximate T^2-dependence of the specific heat in the superconducting state below T_c down to 0.1 K has repeatedly been claimed as evidence of anisotropic unconventional superconductivity.

Here, our results of the specific heat measurements are presented in Fig. 1. The specific heat in zero field agrees fairly well with the previous reports, except the superconducting transition at $T_c \approx 0.46$ K is less clear, being considerably lower and broader than those with the highest T_c values with a clear double transition. From previous studies, the reason is known to be due mostly to the impure uranium in our case [6,7], although the elemental analysis of uranium has not been carried out. Nevertheless, our results below T_c down to 50 mK reproduce very well the power law $T^{1.2}$, which is a somewhat lower power than that reported previously, and agrees well with part of the results of Schuberth *et al.* [1].

In magnetic fields, however, the specific heat begins to show a remarkable upturn below ~200 mK for 1 T and 400 mK for 2.5 T. As shown in Fig. 1, the specific heat at 63 mK in 2.5 T amounts to a value even higher than 0.7 J mol^{-1} K^{-1}, which corresponds to $\gamma \geqslant 10$ J mol^{-1} K^{-2}. It is rather surprising that it seems to increase even further at lower temperatures. This astonishing feature is more clearly seen in Fig. 2, where C/T ($=\gamma$) is plotted as a function of temperature. It is noted that this peak height is again comparable to the result of Schuberth *et al.* [1].

Considering the properties of uranium and platinum nuclei, firstly it is clear that ^{238}U in the present sample with more than 99.99% abundance cannot possibly contribute such a huge anomaly of the specific heat, because of possession of neither spins nor quadrupolar moments. It is highly doubtful that only the 100 ppm level (or less) of ^{235}U, which carries a nuclear magnetic moment of -0.35 μ_N and nuclear quadrupolar moment of 4.1 barn, may cause such an anomaly.

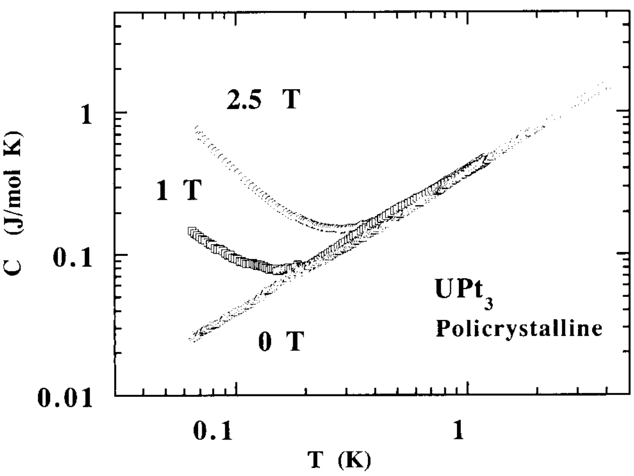

Fig. 1. The specific heat of polycrystalline UPt_3 down to 65 mK in zero field (○), 1 T (□) and 2.5 T (△).

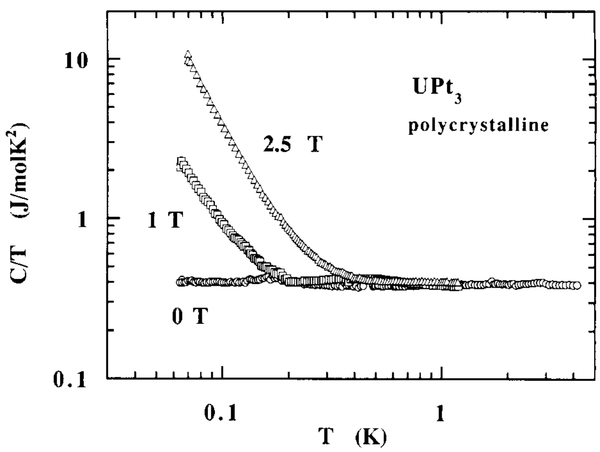

Fig. 2. The electronic term of the specific heat, γ ($=C/T$), of a polycrystalline sample of UPt_3 in zero magnetic field (△), 1 T (□) and 2.5 T (○) shown in Fig. 1.

On the other hand, there exist four stable isotopes of platinum: ^{194}Pt, ^{195}Pt, ^{196}Pt and ^{198}Pt. Among them, only ^{195}Pt possesses a nuclear magnetic moment(of magnitude 0.6060 μ_N without quadrupolar moment). Taking into account the hyperfine constant of the Pt nucleus, an extremely large internal field of $\sim 7.5 \times 10^4$ T would be required to induce this anomalous transition. Therefore, it is natural to exclude such a possibility. As expected from Figs. 1 and 2, the anomaly at 18 mK in zero field does not move much to higher fields, since the maximum is not reached even at 65 mK in 2.5 T. Although the entire curve of the anomaly is not revealed in the present experiment, it is unlikely that it is due to the field dependent behavior of a Schottky anomaly of the nuclear spins. The possibility of some type of additional phase transition in the superconducting state can also be excluded, because the anomaly still exists above $H_{c2}(0) \approx 1.9$ T [7]. Therefore, it is concluded that the anomalous increase in the specific heat observed here is naturally attributed to an instability of the electronic state in the heavy quasi-particles.

In summary, the specific heat of UPt_3 has been measured down to 50 mK in magnetic fields up to 2.5 T. The growing magnitude of the low temperature specific heat anomaly in magnetic fields cannot be attributed to the spurious nuclear origin of the elemental constituents of uranium and platinum, and requires a new explanation, which strongly suggests instability of the electronic state of the heavy fermion ground state in UPt_3.

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