Specific Heat and Electrical Resistivity Measurements in $Pr_{0.03}La_{0.97}Pb_3$

Tatsuya Kawae [a,1], Tatsuharu Yamamoto [a], Kenji Yurue [a], Masaki Mito [a], Kazuyoshi Takeda [a], Tetsuo Kitai [b]

[a] *Department of Applied Quantum Physics, Kyushu University, Hakozaki, Fukuoka 812-8581, Japan*

[b] *Faculty of Engineering, Kyushu Institute of Technology, Kitakyushu 804-8585, Japan*

Abstract

We have measured specific heat C and electrical resistivity ρ for $Pr_{0.03}La_{0.97}Pb_3$ with the Γ_3 doublet in the crystalline-electric-field ground state to examine the Kondo effect arising from the correlation between the quadrupolar moments and the conduction electrons. It is found that C/T increases monotonically with lowering temperature below 1.5 K, which is clearly different from that in the Pr concentrated region. Moreover, ρ shows a clear drop below 3 K.

Key words: quadrupolar ordering; La impurity effect; specific heat

1. Introduction

Recently, several Pr-based compounds have been reported to show the heavy Fermion behavior with a large T-linear coefficient in the specific heat [1] or the heavy electron mass in the de Haas-van Alphen effect [2]. One of the possible candidate to explain the behavior is the quadrupolar Kondo effect, which originates from the interaction between the quadrupolar fluctuations of a nonmagnetic Γ_3 doublet and the charge of the conduction electrons, as introduced by Cox to explain the unusual magnetic properties in UBe_{13} [3]. Because the crystal-electric-field (CEF) level scheme of Pr^{3+} is the same as that of U^{4+} as long as they are in the same crystal field symmetry. The quadrupolar Kondo theory predicts that a non-Fermi liquid behavior such as $C/T \propto -ln(T/T_0)$, $\chi \propto T^{-\beta}$ and $\rho \propto -T$. However, a non-Fermi liquid behavior has not been observed in Pr-based compounds. Therefore the origin is not clarified.

We have studied the low temperature properties of $Pr_{1-x}La_xPb_3$ to clarify the heavy Fermion behavior in Pr-based compounds [4]. The magnetic properties of $PrPb_3$ has been studied extensively so far. The CEF ground state is Γ_3 doublet and the first excited state is Γ_4 with an energy difference of 19 K. The fluctuation of the Γ_3 moment is entirely depressed below T=0.4 K due to a quadrupolar ordering (QPO). When La is substituted for Pr ions, it is clarified that the low temperature behavior of the Γ_3 moment is largely modified. QPO is expected to disappear only for the La concentration of x=0.02. For a wide range of La concentration for 0.1≤ x≤0.8 where the ordering is absent, the specific heat shows a T-linear variation, which is in an excellent agreement with the result obtained by the model for amorphous materials with a random configuration of two level system of the two Γ_3 moments. This indicates that the CEF ground state with the Γ_3 doublet does not change for the Pr concentration. The Kondo effect is explained as single-site properties of the Γ_3 doublet. Therefore we measured the specific heat and electrical resistivity in a very diluted region of the Γ_3 moments with x= 0.03.

[1] Corresponding author. Present address:Department of Applied Quantum Physics, Kyushu University, Hakozaki, Fukuoka 812-8581, Japan E-mail: kawaetap@mbox.nc.kyushu-u.ac.jp

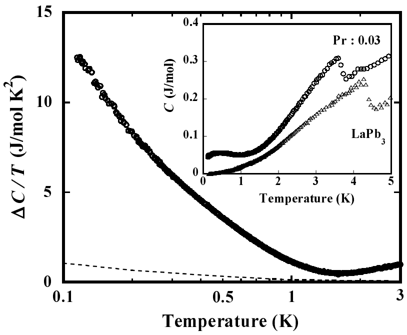

Fig. 1. $\Delta C/T$ plotted in logarithmic temperature scale for $x=0.03$, where the back ground contribution estimated from the specific heat of $LaPb_3$ below 4 K is subtracted and the specific heat is normalized by the Pr concentrations. The dashed line is the theoretical calculation for T_K=3 K [5]. Inset: Temperature dependence of the specific heat C for $Pr_{0.03}La_{0.97}Pb_3$ and $LaPb_3$.

2. Experimental

The polycrystalline sample is prepared by the Bridgeman method. The specific heat is measured by a semiadiabatic method using a dilution refrigerator. A small amount of Apiezon grease is mixed into some pieces of the sample crystal to keep good thermal contact. The resistivity is measured by a four probe method using ac resistance bridge.

3. Results

We plot the temperature dependence of the specific heat for x=0.03 and $LaPb_3$ in the inset of Fig. 1. A distinct bump is clearly seen below 1.5 K. $LaPb_3$ shows a superconducting transition around T=4 K. In Fig. 1, a jump of the specific heat is observed at $T_C=4.4$K, and decreases down to at $T_C=3.5$ K for $x=Pr_{0.03}La_{0.97}Pb_3$. This indicates that the bump of the specific heat below 1.5 K comes from Pr moments, not due to the superconductivity.

In Fig. 1, we show $\Delta C/T$ plots in the logarithmic scale of temperature, where the back ground contribution is estimated from the specific heat of $LaPb_3$. $\Delta C/T$ shows a non-Fermi liquid behavior with a monotonical enhancement below 1 K. This is entirely different from that for the case of Pr concentration larger than x=0.8. When we remember that the CEF ground state in $Pr_{1-x}La_xPb_3$ is the Γ_3, this NFL behavior may arise from the quadrupolar Kondo effect. We compare the present results to the theoretical calculations based on the $S=1/2$ two-channel Kondo system [5]. When the Kondo temperature T_K is taken to be 3 K at which the resistivity starts to drop as shown in Fig. 2, the estimation is about eight times smaller than the observed results at the lowest temperature. The entropy change for x=0.03 from 0.1 K to 2 K is about $0.8R\ln2$ which is much larger than that of $1/2R\ln2$, which is of the theoretical prediction. The observed results can not be considered as the pure two-channel Kondo system. The distortion of the CEF is observed in the higher concentration of Pr ions, which may play an important for the difference from the theoretical calculation.

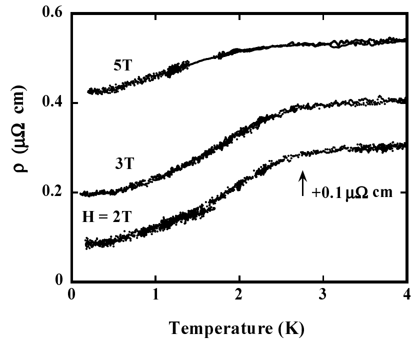

Fig. 2. Temperature dependence of ρ below 4 K under the magnetic fields of 2 T, 3 T and 5 T. The data at 3 T and 5 T are shifted by 0.1 ($\mu\Omega$cm).

In Fig. 2, we show the temperature dependence of the electrical resistivity ρ in the magnetic field of 2T, 3T and 5T which is applied to break the superconductivity of $LaPb_3$ and the Pb phase formed on the surface by oxidization. A remarkeble drop of ρ is seen below $T\sim3$ K. The origin of the drop, however, is not clear. Further experimental studies in the other concentrations are needed.

4. Conclusion

We observed that C/T in $Pr_{0.03}La_{0.97}Pb_3$ increases monotonically with lowering temperature below 1.5 K, which may arise from the quadrupolar Kondo effect. A drop of ρ is seen below 3 K. The origin, however, is not clear.

References

[1] A. Yatskar et al., Phys. Rev. Lett. **77** (1996) 3637.

[2] H. Sugawara et al., J. Magn. Magn. Mat, 226-230 (2001) 48.

[3] D. L. Cox, Phys. Rev. Lett. **59** (1987) 1240.

[4] T. Kawae et al., Phys. Rev. B**65** (2002) 012409.

[5] P.D. Sacramento et al., Phys. Rev. B**43** (1991) 13294.