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Non-Fermi liquid behavior in Pr-based dilute quadrupolar system

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Abstract

We have studied the low temperature properties of $PrPb_3$ and $PrInAg_2$ with the crystal-electric-field ground state of a non-Kramers Γ_3 doublet diluted by non-magnetic La ions to examine the correlation between the Γ_3 quadrupolar moments and the conduction electrons. In $Pr_xLa_{1-x}Pb_3$, a non-Fermi liquid (NFL) behavior is observed at Pr concentrations of $x \leq 0.05$. The specific heat C/T increases monotonically below $T = 1.5$ K, which can be scaled with a characteristic temperature T^* defined at each concentration x. Application of magnetic field raises C/T rapidly, which demonstrates the presence of the residual entropy at lower temperatures. In $Pr_xLa_{1-x}InAg_2$, NFL behavior is confirmed for $x = 0.05$ and 0.1 in the susceptibility below 15 K and in the specific heat below 7 K. It is supposed that the Kondo effect arising from the correlation between the dilute Γ_3 moments and the conduction electrons gives rise to NFL behavior in both systems.
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1. Introduction

A non-Fermi-liquid (NFL) behavior observed in U- and Ce-based compounds has attracted much interest during the past decade. The γ-coefficient of the specific heat and the magnetic susceptibility show the unusual temperature dependence such as $C/T \propto -ln(T/T_0)$, which are clearly inconsistent with conventional Fermi-liquid behavior. A theoretical model leading to NFL state is the single-ion multichannel Kondo effect arising from the overscreening of the spin S of the 4*f* or 5*f* atom by the conduction electrons N, with $N > 2S$. Cox showed that the two-channel Kondo effect can be realized in $U^{4+}(5f^2)$ ions at a site of cubic symmetry with the crystal-electric-field (CEF) ground state of non-Kramers Γ_3 doublet, which is referred to as the quadrupolar Kondo effect [1]. Γ_3 doublet has no magnetic moments, but it has electric quadrupolar moments which can be described by pseudospin moments with $S = 1/2$. Thus, the interaction between Γ_3 doublet and the charge of the conduction electrons involves two electron channels, giving an overcompensation of the localized quadrupolar moment by conduction electrons below a characteristic temperature T_K in the quadrupolar Kondo effect. This leads to NFL behavior such as logarithmic temperature dependence in the electronic specific heat and the quadrupolar susceptibility. Moreover, the residual entropy in the ground state is $1/2R\ln 2$.

A lot of experimental studies have been done to confirm the quadrupolar Kondo effect [2,3]. However, there remain several questions to understand NFL behavior observed in U-compounds within the framework of the quadrupolar Kondo effect [4,5].

Pr-based compounds are good candidates to study the quadrupolar Kondo effect, because the CEF level scheme of Pr^{3+} is the same as that of U^{4+} as long as it is in the same crystal field symmetry. Thus, we have studied the low temperature properties of Pr-based compounds with the CEF ground state of a non-Kramers Γ_3 doublet diluted by nonmagnetic La ions to examine the quadrupolar Kondo effect [6–8].

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2. Experimental

The samples of $Pr_xLa_{1-x}Pb_3$ and $Pr_xLa_{1-x}InAg_2$ were prepared by the Bridgeman method. The susceptibility was measured down to 2 K by using a Quantum Design SQUID magnetometer. The specific heat measurements were carried out by a adiabatic method using a dilution refrigerator below 4 K and a ^{3}He-refrigerator above 1 K.

3. Results

3.1. $Pr_xLa_{1-x}Pb_3$

The temperature dependence of the susceptibility for $Pr_xLa_{1-x}Pb_3$ for $x \geq 0.01$ is qualitatively the same as that for $PrPb_3$. It can be fitted by the theoretical estimation with the Γ_3 doublet in the CEF ground state.

$PrPb_3$ exhibits a quadrupolar ordering at $T = 0.4$ K. By addition of La impurity, the ordering disappears only for $x \sim 0.98$. In the wide concentration for $0.1 \leq x \leq 0.95$, $\Delta C/T$ is almost constant below $T = 0.5$ K with a large T-linear coefficient [6]. Here $\Delta C = (C - C_l)/(1 - x)$, where C is the specific heat of $Pr_{1-x}La_xPb_3$, and C_l is the back-ground contribution of $LaPb_3$. This feature is well reproduced by the model of the specific heat for amorphous materials with a random configuration of two-level system. By taking account of the entropy, it is found that the random two-levels come from the Γ_3 moments, which splits due to the distortion of CEF by the difference of the ion radius between La and Pr [6].

In Fig. 1, we show the temperature dependence of $\Delta C/T$ in the Pr concentration of $x \leq 0.1$. For $x = 0.05$, 0.03 and 0.01, $\Delta C/T$ increases monotonically below 1 K with decreasing temperature, which is entirely different from that for $x \geq 0.1$ [7,8]. It is apparent that anomalous $\Delta C/T$ is not caused by impurity ions or magnetic ordering of Pr ions, because the temperature dependence of the anomaly has not been observed by changing the concentration. A Schottky specific heat by the splitting of Pr nuclear spin state due to strong hyperfine magnetic field is also ruled out, since the temperature dependence of the anomaly is different from the expected Schottky one.

It is noteworthy that the data is scaled with a characteristic temperature T^* as shown in the inset of Fig. 1, where T^* is taken to be 1 K for $x = 0.05$, 0.65 K for $x = 0.03$ and 0.40 K for $x = 0.01$ [8]. This indicates that the NFL behavior with the enhancement in $\Delta C/T$ for $x \leq 0.05$ is understood as a single-ion effect of the Γ_3 moment and does not come from the superposition of the Schottky specific heat due to the splitting of the Γ_3 moments. Moreover, the increase in T^* with the concentration x is understood as the increase in hybridization width due to the compression of the lattice by increasing the concentration x from $-\ln T$-dependence of the volume compression [8].

When we compare the experimental results to the numerical calculations based on the $S = 1/2$ two-channel Kondo

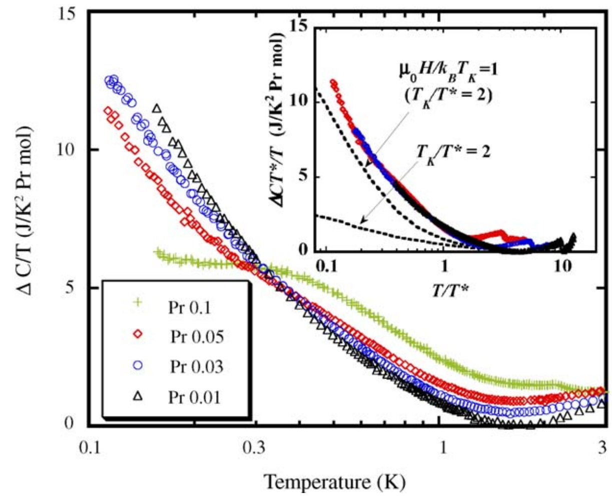

Fig. 1. The temperature dependence of $\Delta C/T$ plotted on a logarithmic temperature scale for $x = 0.1$, 0.05, 0.03 and 0.01. Inset: $\Delta CT^*/T$ vs. $\log(T/T^*)$. T^* is taken to be 1 K for $x = 0.05$, 0.65 K for $x = 0.03$ and 0.40 K for $x = 0.01$. The lower dashed line is the theoretical calculation for T_K/T^*=2 in the two-channel Kondo theory and the upper dashed line that for $\mu_0 H/k_B T_K = 1$ with $T_K/T^* = 2$ obtained from ref. [9].

system [9], $\Delta C/T$ is much larger than the theoretical calculation. This is due to the distortion of the CEF by La impurity because of the difference of the ion radius between Pr^{3+} and La^{3+}. Thus the present results should be compared with the two-channel Kondo theory under strong magnetic fields. When we assume the magnitude of the CEF distortion is comparable to the Kondo temperature, the data are in reasonable agreement with the theoretical calculation [8].

We show the magnetic field dependence of the specific heat for the polycrystal in the inset of Fig. 2. As the magnetic field is increased, the peak due to the superconducting transition of $LaPb_3$ at ∼3 K disappears. Moreover, at lower temperatures, an upturn with approximately $1/T^2$-dependence appears and grows. This upturn is ascribed to a Schottky specific heat due to Pr nuclei. By subtracting the back ground contribution due to the nuclei and the phonon, ΔC originating from the fluctuating quadrupolar moments is obtained as shown in Fig. 2. As the magnetic field is increased, the slope of $\Delta C/T$ increases rapidly. This indicates the presence of the residual entropy in the lower temperature region, which is one of the most important features in the quadrupolar Kondo effect. At $H = 4$ T, a clear suppression is seen around 0.7 K and at $H = 6$ T, $\Delta C/T$ is almost a constant. It seems that application of magnetic fields collapses the NFL state, which is consistent with the magnetic field dependence expected by the theory [9,10].

We found that the electrical resistivity $\rho(T)$ in the corresponding temperature region shows a marked decrease deviating from a Fermi-liquid behavior $\rho(T) \propto T^2$ below 5 T [7,8]. At 6 T, approximately T^2-dependence of $\rho(T)$ is obtained in the whole temperature region. The above results indicate that the Fermi-liquid ground state appears above 6 T.

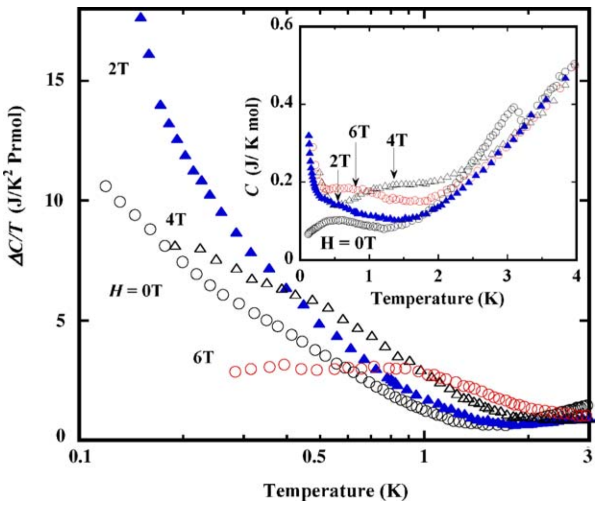

Fig. 2. The magnetic field dependence of $\Delta C/T$ for $x = 0.05$. Inset: the magnetic field dependence of C.

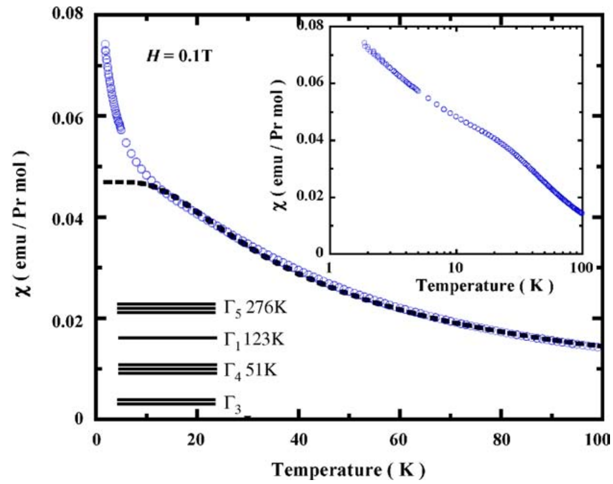

Fig. 3. The temperature dependence of the magnetic susceptibility. The dashed line represents the susceptibility calculated by the CEF level scheme given in the figure. Inset: the susceptibility plotted on a logarithmic temperature scale.

The observed results strongly suggest that the quadrupolar Kondo effect arising from the correlation between the Γ_3 moments and conduction electrons gives rise to NFL behavior in $Pr_xLa_{1-x}Pb_3$ for $x \le 0.05$.

3.2. $Pr_xLa_{1-x}InAg_2$

Next we show the experimental results in $Pr_xLa_{1-x}InAg_2$ for $x = 0.1$ and 0.05. $PrInAg_2$ has a non-Kramers Γ_3 doublet in the CEF ground state. Contrary to $PrPb_3$, it shows no quadrupolar ordering down to 50 mK, but has the Fermi-liquid like ground state with a very large value of $\gamma \sim 6.5\,J/mol\,K^2$ [11]. Furthermore, the field dependence of the specific heat is very weak compared to the conventional heavy Fermion materials arising from the Kondo effect. The interaction between the quadrupolar fluctuations of the Pr ions and the conduction electrons is discussed as the origin of a heavy fermion ground state. In order to clarify the problem, we have studied the low-temperature properties of $Pr_xLa_{1-x}InAg_2$ for $x = 0.1$ and 0.05.

In Fig. 3, we show the magnetic susceptibility of $Pr_{0.05}La_{0.95}InAg_2$. The susceptibility above 15 K is well reproduced by the theoretical estimation with the Γ_3 ground state in the CEF level scheme given in the figure, which is almost the same as that of $PrInAg_2$. This indicates that the CEF level does not change by La substitution. In fact, the susceptibility for $x = 0.1$ is in good agreement with that for $x = 0.05$. In the temperature region below 15 K, the susceptibility increases exponentially, which is probably related with the fluctuation of the quadrupolar moments.

We show the temperature dependence of $\Delta C/T$ in $Pr_xLa_{1-x}InAg_2$ for $x = 0.1$ and 0.05 in Fig. 4, where the phonon contribution estimated from $LaInAg_2$ is subtracted. NFL behavior with the monotonic enhancement of $\Delta C/T$ is revealed below 7 K and $\Delta C/T$ is almost the same for each concentration, indicating that the NFL behavior is explained by the single-ion physics.

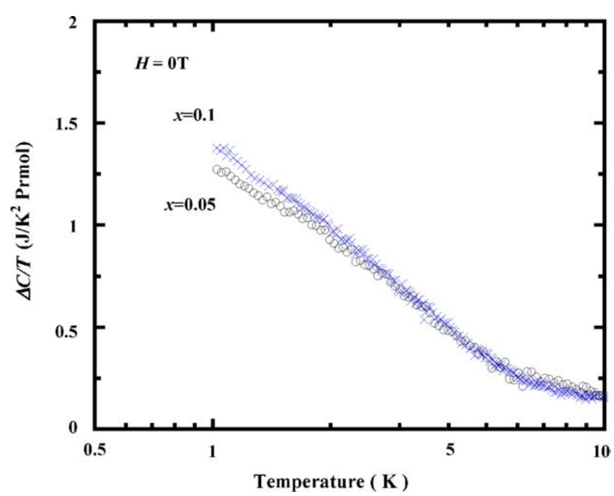

Fig. 4. The temperature dependence of $\Delta C/T$ plotted on a logarithmic temperature scale in $Pr_xLa_{1-x}InAg_2$ for $x = 0.1$ and 0.05.

From the susceptibility and specific heat measurements, it is supposed that NFL behavior in $Pr_xLa_{1-x}InAg_2$ is caused by the correlation between the Γ_3 moments and the conduction electrons. In order to confirm this prediction, we are preparing the resistivity measurements by using high quality samples with low residual resistivity at low temperatures.

4. Conclusion

We have studied the low temperature properties of $Pr_xLa_{1-x}Pb_3$ and $Pr_xLa_{1-x}InAg_2$ with the CEF ground state of a non-Kramers Γ_3 doublet. In $Pr_xLa_{1-x}Pb_3$, NFL behavior is seen at Pr concentrations of $x \leq 0.05$. $\Delta C/T$ increases monotonically below $T = 1.5$ K, which can be scaled with a characteristic temperature T^* defined at each concentration x. Application of magnetic field raises $\Delta C/T$ rapidly, which demonstrates the presence of the residual entropy at lower temperatures. In $Pr_xLa_{1-x}InAg_2$, NFL behavior is confirmed for $x = 0.05$ and 0.1 in the susceptibility below 15 K and in the specific heat below 7 K. We suppose that the Kondo effect arising from the correlation between the dilute Γ_3 moments and the conduction electrons gives rise to NFL behavior in both systems.

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