**Susceptibility measurements in Pr$_x$La$_{1-x}$InAg$_2$ with $\Gamma_3$ doublet ground state**

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**Abstract.** We have measured the susceptibility and magnetization of Pr$_x$La$_{1-x}$InAg$_2$ at Pr concentrations $x=1$ and 0.1 down to $T \sim 0.5$ K by SQUID magnetometer with a home-made $^3$He insert. The susceptibility above $T = 15$ K is well reproduced by the crystal-electric-field level scheme with a non-Kramers $\Gamma_3$ doublet in the ground state for each concentration, while that below $T = 15$ K shows a non-Fermi-liquid (NFL) behavior with $-\ln T$-dependence at low magnetic field. With increasing magnetic field, $-\ln T$-dependence is suppressed and $-T^{1/2}$-dependence appears at $H = 7$ T. The magnetization at $T = 0.5$ K increases with increasing magnetic fields up to $H = 7$ T, indicating that the increase of the susceptibility does not come from impurity ions. These results suggest that the quadrupolar Kondo effect is responsible for NFL behavior of the susceptibility.

1. **Introduction**

Pr-based compounds with the cubic symmetry have attracted much attention because of the discovery of their fascinating features, e.g., a coexistence of quadrupolar ordering and superconductivity [1], and a quadrupolar ordering triggered by octupole moments induced by magnetic fields [2]. A Pr$^{3+}$ ion with the 4$f^2$ configuration at a site of cubic symmetry can have a non-Kramers $\Gamma_3$ doublet with the quadrupolar moment in the crystalline-electric-field (CEF) ground state. Hence, it is believed that these features are related to the quadrupolar Kondo effect, which is proposed by Cox to explain anomalous heavy Fermion behavior in UBe$_{13}$ under magnetic fields [3]. According to Cox, the quadrupolar Kondo effect is expected to be realized in a U$^{4+}$ ion with the 5$f^2$ configuration under the cubic symmetry with the CEF ground state of a $\Gamma_3$ doublet, which is the same situation as the Pr-based compounds. In order to provide the experimental evidence of the quadrupolar Kondo effect, we have investigated the low-temperature properties of Pr$_x$La$_{1-x}$Pb$_3$ with the $\Gamma_3$ doublet in the CEF ground state with Pr concentration changing between $0 \leq x \leq 1$ [4], and showed that NFL behavior for $x \leq 0.05$ can be understood by the quadrupolar Kondo effect [5]. In the course of the study, we observed that the susceptibility increases below 5 K deviating from the CEF model with the $\Gamma_3$ doublet [6].

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We have studied the low-temperature properties of Pr$_x$La$_{1-x}$InAg$_2$ for a wide concentration range of Pr ions. We reported that the susceptibility above $T = 15$ K is well reproduced by the CEF level scheme with a non-Kramers $\Gamma_3$ doublet in the ground state for each concentration, while that $2$ K $\leq T \leq 15$ K shows NFL behavior with $-\ln T$-dependence [7]. This would be explained by the theoretical prediction based on the two-channel Kondo model given by Kusunose et al. [8]. Recently, the temperature dependence of the magnetic susceptibility is reported in other $\Gamma_3$ doublet compounds. In PrV$_2$Al$_2$, the susceptibility shows $-T^{1/2}$ dependence for $2$ K $\leq T \leq 30$ K [9], which is consistent with the temperature dependence predicted from the ”3-4-7-8” model where the $\Gamma_3$ ground state and the $\Gamma_4$ first excited state of $f^2$, the $\Gamma_7$ state of an excited $f^1$ configuration and the $\Gamma_8$ quartet of conduction electrons are taken into account [10]. In PrMg$_3$, it is interesting that the susceptibility exhibits $-\ln T$-dependence for $1$ K $\leq T \leq 15$ K while the temperature dependence seems to approach $-T^{1/2}$ at lower temperatures [11]. These studies indicate that the susceptibility in Pr-compounds with the $\Gamma_3$ doublet exhibit NFL behavior at low temperatures, which is likely due to the quadrupolar Kondo effect.

In the present study, we measure the dc magnetic susceptibility and magnetization down to $T \sim 0.5$ K by SQUID magnetometer with a home-made $^3$He insert in order to investigate the temperature dependence of the susceptibility in Pr$_x$La$_{1-x}$InAg$_2$ below $T = 2$ K. It is well known that PrInAg$_2$ has the Heusler structure, where a random site exchange is possible. This may affect the low-temperature susceptibility of PrInAg$_2$. Thus, we study the very dilute region of Pr with $x=0.1$, where the single ion effect is observed in the specific heat measurements, together with a pure PrInAg$_2$ [7].

2. Experimental

A polycrystal of PrInAg$_2$ and a single crystal of Pr$_{0.1}$La$_{0.9}$InAg$_2$ were prepared by the Bridgman method. In PrInAg$_2$, the stoichiometric amounts of Pr, In and Ag of the required quantities in a Ta crucible were heated in a closed quartz tube under Ar atomosphere. A single crystal of Pr$_{0.1}$La$_{0.9}$InAg$_2$ were heated in a molybdenum crucible sealed by electric beam welding in a vacuum. Both PrInAg$_2$ and LaInAg$_2$ have the same Heusler structure with the lattice parameters $a = 7.075$ Å and 7.156 Å, respectively [10], which means that Pr ions are substituted for La ions without a change of the crystal symmetry.

Magnetic susceptibility is measured down to $T = 0.5$ K by a Quantum Design SQUID magnetometer with a home-made $^3$He insert. The sample and $^3$He gas are inserted through a stainless steel pipe with the outer diameter of 6.4 mm. The liquid $^3$He is condensed in a copper container with the inner diameter of 6 mm and length of 150 mm, which is connected with the stainless pipe. The vacuum jacket made of a copper pipe with the outer diameter of 8.5 mm is soldered to the stainless pipe, which is also used for the heat exchange between $^3$He gas and $^4$He bath at $T \sim 1.7$ K. The temperature was measured by RuO$_2$ thermometer mounted on the $^3$He container.

3. Results and Discussion

We show the temperature dependence of the magnetic susceptibility and magnetization for PrInAg$_2$ in Figs. 1(a) and 1(b), respectively. The dc susceptibility is measured in three fields, $H = 0.25$ T, 3 T and 7 T. The susceptibility above 15 K is reproduced by the CEF level scheme with a $\Gamma_3$ doublet in the ground state as plotted in the inset of Fig. 1(a), where the first excited state is a $\Gamma_4$ triplet with the energy gap of 73 K. In contrast, the susceptibility represents a $-\ln T$-dependence for 2 K $\leq T \leq 15$ K. As the magnetic field is increased, the sharp increase of the susceptibility below 20 K is suppressed. At $H = 7$ T, it approaches a $-T^{1/2}$ dependence. From Fig. 1(b), it is seen that the magnetization increases gradually with increasing magnetic field up to $H = 7$ T, reflecting the non-magnetic ground state.
We have measured the susceptibility and magnetization of PrInAg₄. Conclusion
The present results indicate that the NFL behavior below 1 K at T=0.5 K and at T=1.7 K, the susceptibility at lower fields increases with decreasing the temperature from T=1.7 K to T=0.5 K as shown in Figs. 1(a) and 2(a).

From these facts, we can conclude that −lnT-dependence of the susceptibility is an intrinsic feature of PrₓLa₁₋ₓInAg₂ with the Γ₃ doublet in the CEF ground state. As the magnetic field is increased, the enhancement of susceptibility is suppressed. Finally, the susceptibility shows a T¹/²-dependence as in the case of the pure system. We observed the suppression of NFL behavior below 1 K at H = 5 T in the specific heat measurements. Although the relation between the present results and the theoretical models is not obvious, it is supposed that −lnT-dependence at low fields and T¹/²-dependence at high fields in PrₓLa₁₋ₓInAg₂ are connected with the quadrupolar Kondo effect.

4. Conclusion
We have measured the susceptibility and magnetization of PrₓLa₁₋ₓInAg₂ at Pr concentrations x=1 and 0.1 down to T ~ 0.5 K by SQUID magnetometer with a home-made ³He insert.

Figure 1. (a) The temperature dependence of the magnetic susceptibility for PrInAg₂. The inset is a fitting by the CEF model with x = −0.070, W=−1.29 K. (b) The field dependence of the magnetization at T = 0.5 K and 1.7 K.
Figure 2. (a) The temperature dependence of the magnetic susceptibility for $x=0.1$. The inset is a fitting by the CEF model with $x = -0.065$, $W = -1.10 [K]$. (b) The field dependence of the magnetization for $T = 0.5 \text{K}$ and $1.7 \text{K}$.

susceptibility above $T = 15 \text{K}$ is well reproduced by the CEF model with a non-Kramers $\Gamma_3$ doublet in the ground state. On the other hand, the dc susceptibility measured in $H = 0.25 \text{T}$ shows NFL behavior with $-\ln T$-dependence for both concentrations below $T = 15 \text{K}$ at low field. With increasing magnetic field, $-\ln T$-dependence of the susceptibility is suppressed and $-T^{1/2}$-dependence appears. The magnetization at $T = 0.5 \text{K}$ increases with increasing magnetic fields up to $H = 7 \text{T}$, indicating that the increase of the susceptibility does not come from impurity ions. From these facts, we can conclude that NFL behavior of the susceptibility is an intrinsic feature of $\text{Pr}_x\text{La}_{1-x}\text{InAg}_2$ with the $\Gamma_3$ doublet in the CEF ground state. These results suggest that the quadrupolar Kondo effect is responsible for NFL behavior of the susceptibility.

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