Non-Fermi Liquid Properties in a Cubic Pr-Based Compound $PrPb_3$ under Magnetic Fields

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Abstract. We have studied the electrical resistivity ρ of the Pr-based cubic compound $PrPb_3$ with the Γ_3 doublet ground state. The temperature dependence of the resistivity is found to exhibit a non-Fermi liquid behavior with convex curve at high temperatures, as suggestive of the putative realization of the quadrupole Kondo effect. At low temperatures under magnetic fields, we observe anomalies associated with antiferroquadrupole (AFQ) ordering, incommensurate/commensurate transition inside the AFQ phase, and field-induced phase transition. The constructed phase diagram based on these observations well reproduces the one determined by the specific heat and magnetization measurements. Deep inside each phase, the resistivity turns to show the Fermi liquid behavior with the quadratic temperature dependence. The estimated slope of the T^2 term is almost independent of field in contrast to our previous work of $PrIr_2Zn_{20}$ and $PrRh_2Zn_{20}$ in which the slope is largely enhanced at the edge of the AFQ phase.

1. Introduction

Investigations of non-Kramers systems with an even number of f-electrons such as Pr^{3+} are of great significance in the field of condensed matter physics. Especially in the case of Γ_3 doublet lying on the crystal electric field (CEF) ground state, novel electronic states irrelevant to the time-reversal symmetry are expected to emerge at low temperatures in contrast to Kramers systems. In such Γ_3 systems, the local quadrupole moment of f-electron can be scattered by conduction electrons via two equivalent scattering channels, and the channel frustration leads to the imperfect screening of the quadrupole moments, preventing the quasi-particle formation. Therefore, the breakdown of the Landau's Fermi liquid (FL) theory allows us to expect the realization of non-trivial physics. Indeed, the two-channel picture suggested the emergence of a quadrupole Kondo effect with non-Fermi liquid (NFL) properties of $\rho \propto T^{0.5}$ and $C/T \propto -\log T$ [1, 2]. In addition, the imperfect screening results in partial releasing of the entropy by only a half of $R \log 2$ even at 0 K, so that the quadrupole Kondo state cannot be the ground state, and instead another state must be realized. Several possibilities are proposed as a way of releasing the residual entropy, such as the ordering of localized Majorana particles by spontaneous channel symmetry breaking [3] and the NFL-FL crossover arising from the channel anisotropy due to external and/or internal perturbations [4].

However, the experimental confirmations of these proposals have not been done due to lack of suitable materials. One of a few candidates is the Heusler type compounds $PrMg_3$ and $PrAg_2In$. Although these compounds were reported to show $-\log T$ dependence of specific heat and $T^{0.5}$ like dependence of resistivity [5, 6] as expected from the quadrupole Kondo effect, the effect of lattice distortions cannot be neglected because the Heusler structure has a possibility for the random site exchange leading to the local splitting of Γ_3 doublet and partial quenching of the quadrupole degrees of freedom. Thus the intrinsic features arising from the Γ_3 doublet may be masked. Recently the cage compound $PrIr_2Zn_{20}$ with the Γ_3 doublet ground state was discovered as a new candidate and was reported to show the $T^{0.5}$ like dependence of the resistivity [7]. However, the convex temperature dependence of the resistivity can arise from the anharmonic phonon characteristic of the cage compounds as discussed in the β-pyrochlore oxide [8, 9]. Thus, whether such phenomenon is originated from the quadrupole Kondo effect remains elusive. Another candidate is $La_xPr_{1-x}Pb_3$ with the Γ_3 doublet ground state in which the specific heat was reported to exhibit $-\log T$ dependence, and is expected to be a first example of the realization of quadrupole Kondo effect [10, 11]. However, the arguments are based on the experiments using the diluted sample by La on the Pr sites, so that the local breaking of the cubic CEF symmetry around Pr site similar to the Heusler type compounds cannot be excluded. In contrast, the physical properties of the non-doped $PrPb_3$ have barely been explored except for the quadrupole ordered phase, even though the system is suitable for studying quadrupole Kondo physics arising from Γ_3 doublet without the randomness and the cage structure.

In this paper, we report the electrical resistivity of $PrPb_3$, which has seldom been reported ever, as a part of systematic understanding of Γ_3 systems.

2. $PrPb_3$

$PrPb_3$ is the $AuCu_3$ type cubic compound (space group: $Pm\bar{3}m$) with the Γ_3 doublet ground state. The first excited Γ_4 triplet is separated by about 15 K [12] and thus the Γ_3 doublet is isolated at low temperatures. The ground state carries two types of quadrupole moment $O_2^0 \equiv (3J_z^2 - \boldsymbol{J}^2)/2$ and $O_2^2 \equiv \sqrt{3}(J_x^2 - J_y^2)/2$. The O_2^0 type antiferroquadrupole (AFQ) ordering was observed at 0.4 K by the specific heat and elastic modulus measurements [13, 14, 15]. The constructed phase diagram from these experiments was well reproduced by the simplified two-sublattice mean field calculation [12]. In addition, it was also reported that this AFQ phase has two modulated structures: commensurate (C) and incommensurate (IC) phase [16]. Although nature of the ordered state has been extensively studied, the effect of coupling between the quadrupole moments and conduction electrons has not been fully examined.

The electrical resistivity is one of promising probes to unveil such effect, but there has been no satisfactory report devoted to the resistivity of $PrPb_3$. This is mainly because Pb thin layer tends to form on the surface of the host crystal by exposing the air and/or applying stresses [17, 18]. In addition, since Pb undergoes a superconducting transition at about 7 K, the Pb layer gives a fatal error in the precise estimation of the intrinsic transport properties.

3. Experiments

The electrical resistivity ρ was measured by a standard DC four-probe method under magnetic fields by using a single crystal of $PrPb_3$. The current and field were applied in parallel with the [100] direction. The Au wires with a diameter of $\phi = 25\mu$m were attached to the sample by a spot-welding technique for the electrical contacts. The measurements were performed in the temperature range of $0.04 < T < 10$ K and the field range of $0 < B < 9$ T. The sample geometry is $2.25 \times 1.15 \times 1.25$ mm, where the largest length is parallel to the [100] direction. The residual resistivity ratio is estimated to be ~ 300, indicating a good sample quality.

In order to avoid the Pb layer formation on the sample surfaces, the sample was shaped by cleavage. We also attempted to shorten the exposing time to the air within about one hour during the sample setting.

4. Results and discussion

First, let us discuss the resistivity obtained at relatively high temperatures. From Figure 1(a), one immediately notice that there is a clear discrepancy below T_C of Pb pointed by the black arrow between our data and the previous data taken from Ref. [18] in which the Pb formation on the surface was recognized. In addition, since the field dependence of the resistivity at 4 K shown in Fig. 1(b) displays no anomaly corresponding to the superconducting transition, the Pb contribution is likely to be negligibly small in our sample. Therefore, we can succeed to prepare the setting to uncover the intrinsic electrical resistivity of $PrPb_3$ with negligible contributions of the Pb layer for the first time.

Now, we shall move on to the discussion of the low temperature resistivity. Fig. 2(a) shows the temperature dependence of the resistivity below 2 K under several fields. Interestingly, the resistivity exhibits an unusual behavior with a convex curve in the temperature range above around 0.5 K. This is obviously different from the expectation of the FL theory with $\rho \propto T^2$. As shown in Fig. 2(b), the exponent of the temperature dependence in this region is close to 0.5. A power of the temperature dependence of the resistivity less than unity indicates that the system does not reach the coherent state, implying that the equivalence of two scattering channels for the quadrupole moments of Pr^{3+} strongly enhances the fluctuations and prevents the formation of the quasi-particles. In this sense, ρ is supposed to follow $T^{0.5}$ dependence for the single-impurity quadrupole Kondo model as mentioned Section 1. It is interesting to note that the NFL behavior was also observed in the specific heat measurements as a logarithmic temperature dependence, which is also consistent with the single-impurity quadrupole Kondo effect [10]. From these experimental facts, one may expect that the quadrupole Kondo effect

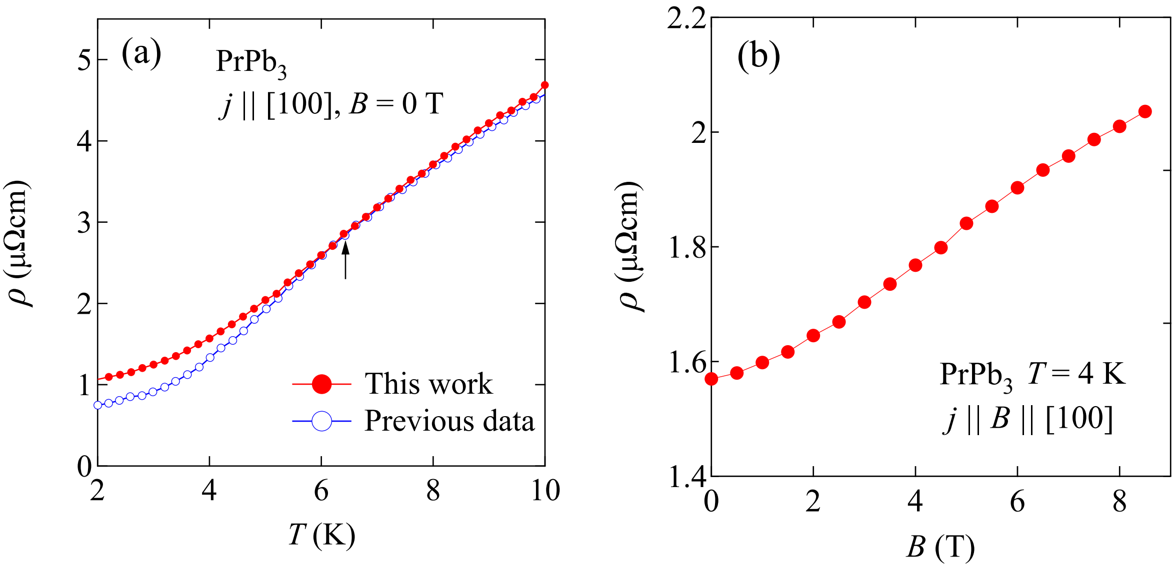

Figure 1. (a) The temperature dependence of the resistivity at 0 T above 2 K together with the previous data, in which Pb layer formation was recognized [18]. The black arrow indicates the superconducting transition temperature of Pb. There is a discrepancy between the two data below T_C of Pb because the resistivity is affected by the superconductivity of Pb in the previous report. (b) The field dependence of the resistivity at 4 K. There is no anomaly accompanied by the superconductivity of Pb.

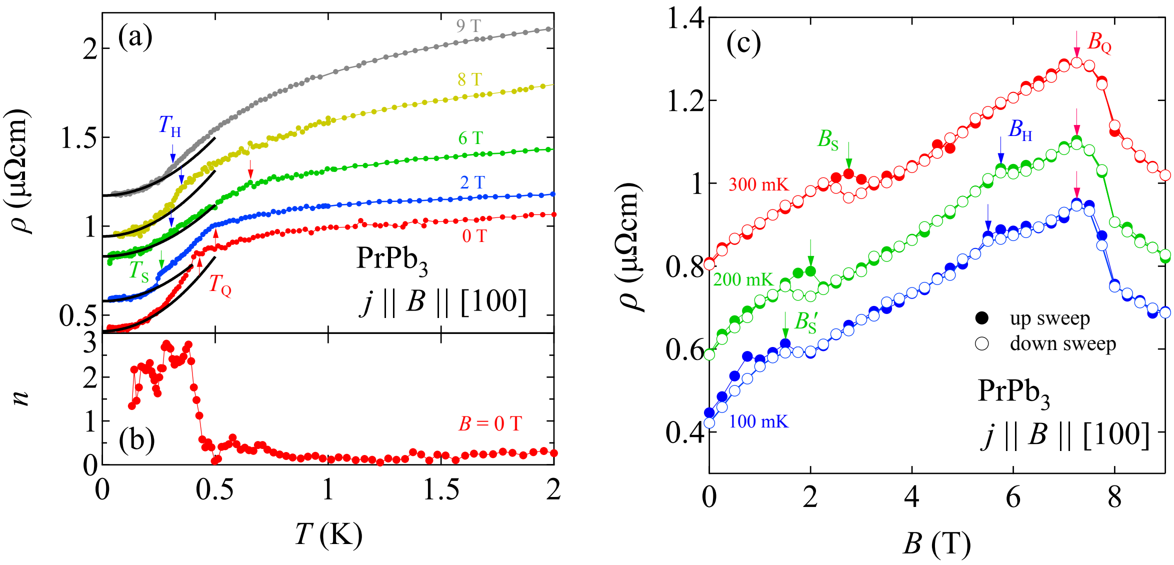

Figure 2. (a) The temperature dependence of the resistivity under several fields. The red, blue and green arrows indicate the transition temperature of the AFQ phase T_Q, high-field phase T_H and incommensurate/commensurate T_S, respectively. The solid lines represent the results of the fitting by a form of $\rho = \rho_0 + AT^2$. (b) The temperature dependence of the exponent n of the resistivity at $B = 0$ T which is estimated from the equation $\rho = \rho_0 + AT^n$. (c) The field dependence of the resistivity below 0.3 K. The closed and open circles represent the up and down sweep measurement, respectively. The anomalies observed at B_Q, B_H and B_S correspond to those at T_Q, T_H and T_S in the temperature dependence (a), respectively. B_S' represents the shoulder like anomaly.

is the case in $PrPb_3$. However such expectation is not conclusive because the single-impurity model is obviously not appropriate to the present material. Therefore, further discussions are required in order to specify the origin of the NFL behavior in $PrPb_3$.

On further cooling, the resistivity shows kinks or anomalies under each field as indicated by arrows in Fig. 2(a). The red, green and blue arrows correspond to the AFQ ordering T_Q, incommensurate/commensurate (IC-C) transition T_S, and transition to the high-field (HF) phase T_H. The corresponding anomalies are also found at B_Q, B_S and B_H in the field dependence of the resistivity as shown in Fig. 2(c) and these characteristic temperatures and fields are summarized in Fig. 3(a). We note that the obtained phase diagram is consistent with the one determined from the specific heat and magnetization measurements [13]. Our resistivity measurement first detects the anomalies associated with HF phase transition. However, the nature of this phase including the order parameter is unclear at this stage. The clear hysteresis accompanied by the IC-C transition is observed in the field dependence of the resistivity (Fig. 2(c)), as already reported by the magnetization measurements and the neutron diffraction experiments [19, 16]. The hysteresis becomes ambiguous below 0.1 K and changes to a shoulder like anomaly at B_S' because the phase boundary reaches the critical end point around 0.1 K. Curiously enough, the shoulder like anomalies corresponding to the crossover temperatures seem to intersect with $T = 0$ line at finite field, although the entropy of the IC phase is larger than the C phase. The IC phase is forbidden to exist at $T = 0$ and must change to another state in order to release the residual entropy. This unknown state is suggested to exist below 0.2 K by the specific heat and magnetization measurements [13, 20], but we cannot detect any anomalies corresponding to this state from the resistivity measurements within our experimental resolution.

Well below the anomalies, the resistivity exhibits the quadratic temperature dependence

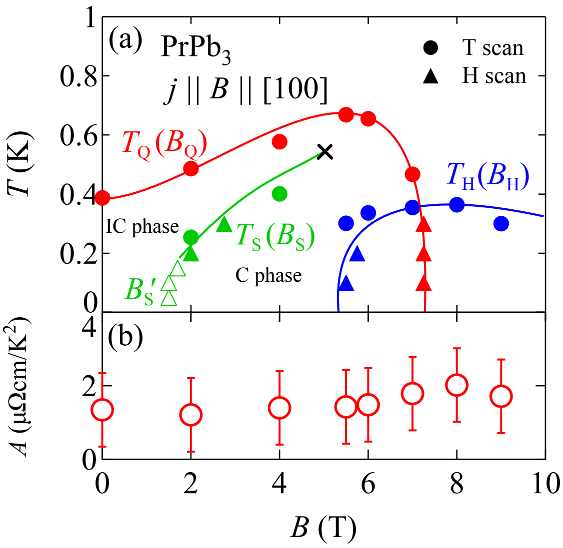

Figure 3. (a) The phase diagram of $PrPb_3$ obtained from the resistivity measurements. The red, green and blue marks represent the antiferroquadrupole, incommensurate/commensurate and high-field phase transitions, respectively, The open green triangles indicate the shoulder like anomaly B'_S without accompanying the hysteresis around T_S (B_S). The black cross mark indicates the termination of the phase boundary referred from Ref. [13]. The solid lines are guide to the eyes. (b) The field dependence of the A coefficient determined by fitting the resistivity data to the form of $\rho = \rho_0 + AT^2$.

$\rho = \rho_0 + AT^2$ as indicated by the solid lines in Fig. 2(a). This is because the FL behavior is attained due to freezing the quadrupole degree of freedom by ordering. Fig. 2(b) clearly shows the attainment of the FL behavior with the power $n \sim 2$ below T_Q. The estimated A coefficient under each field is insensitive to the applied magnetic field within the experimental error as shown in Fig. 3(b). With knowledge of the large specific heat $C/T \sim 6$ J/K^{2}mol at 0 T reported in Ref. [10], the Kadowaki-Woods ratio A/γ^2 is estimated to be $\sim 4 \times 10^{-3} a_0$, which is much less than the universal value a_0. This means that the large C/T is not due to the mass enhancement of itinerant quasi-particles, consistent with the small cyclotron mass $\sim 2m_0$ estimated by the dHvA experiments [21].

We have previously studied the electrical transport coefficients of $PrIr_2Zn_{20}$ and $PrRh_2Zn_{20}$ which have a Γ_3 doublet at the CEF ground state as well as $PrPb_3$ [22, 23, 24]. These compounds have a common feature with $PrPb_3$ that the resistivity exhibits the $T^{0.5}$ like NFL behavior above the AFQ ordering and the T^2 like FL behavior below the AFQ ordering. Taking into account this commonality, we can claim that the NFL behavior with convex temperature dependence of the resistivity is widely realized in non-Kramers systems with Γ_3 doublet ground state, likely due to the quadrupole Kondo effect regardless of the crystal structure as long as the cubic symmetry is guaranteed. On the other hand, these compounds have the different feature that the A coefficient is greatly enhanced when the AFQ ordering is collapsed by the magnetic field, while it is nearly field insensitive in $PrPb_3$ irrespective of the ground state. Given these differences, the HF phase in $PrPb_3$ emerged near the AFQ phase boundary would not be linked to the non-trivial heavy Fermion state in $PrIr_2Zn_{20}$ and $PrRh_2Zn_{20}$.

5. Summary

We have succeeded to measure the intrinsic electrical resistivity of $PrPb_3$ with the Γ_3 doublet ground state by avoiding the formation of Pb thin layer on the host crystal. We found the

NFL behavior of the resistivity with a convex curve in the wide temperature and field range, as suggestive of the realization of the quadrupole Kondo effect. Well below the ordering temperatures of the AFQ phase or the field-induced HF phase, the FL behavior with the T^2 dependence is found to be realized. From the field dependence of the resistivity, the transition to the field-induced HF phase is clearly detected for the first time in the resistivity measurements. The constructed phase diagram based on the resistivity measurements is consistent with the previous one determined by the specific heat and magnetization measurements. The A coefficient of the T^2 dependence of the resistivity is almost independent of field.

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References

[1] Nozières P and Blandin A 1980 *J. Phys. (Paris)* **41** 193
[2] Cox D L and Zawadowski A 1998 *Adv. Phys.* **47** 599
[3] Hoshino S, Otsuki J and Kuramoto Y 2013 *J. Phys. Soc. Jpn.* **82** 044707
[4] Yotsuhashi S and Maebashi H 2002 *J. Phys. Soc. Jpn.* **71** 1705
[5] Tanida H, Suzuki H, Takagi S, Onodera H and Tanigaki K 2006 *J. Phys. Soc. Jpn.* **75** 073705
[6] Yatskar A, Beyermann W P, Movshovich R and Canfield P C 1996 *Phys. Rev. Lett.* **77** 3637
[7] Onimaru T, Matsumoto K T, Inoue Y F, Umeo K, Sakakibara T, Karaki Y, Kubota M and Takabatake T 2011 *Phys. Rev. Lett.* **106** 177001
[8] Dahm T and Ueda K 2007 *Phys. Rev. Lett.* **99** 187003
[9] Hiroi Z, Yonezawa S, Nagao Y and Yamaura J 2007 *Phys. Rev. B* **76** 014523
[10] Kawae T, Shimogai M, Mito M, Takeda K, Ishii H and Kitai T 2001 *Phys. Rev. B* **65** 012409
[11] Kawae T, Yamamoto T, Yurue K, Tateiwa N, Takeda K and Kitai T 2003 *J. Phys. Soc. Jpn.* **72** 2141
[12] Tayama T, Sakakibara T, Kitami K, Yokoyama M, Tenya K, Amitsuka H, Aoki D, Onuki Y and Kletowski Z 2001 *J. Phys. Soc. Jpn.* **70** 284
[13] Sato Y, Morodomi H, Ienaga K, Inagaki Y, Kawae T, Suzuki H S and Onimaru T 2010 *J. Phys. Soc. Jpn.* **79** 093708
[14] Niksch M, Assmus W, Lüthi B, Ott H R and Kjems J K 1982 *Helv. Phys. Acta.* **55** 688
[15] Onimaru T, Sakakibara T, Harita A, Tayama T, Aoki D and Onuki Y 2004 *J. Phys. Soc. Jpn.* **73** 2377
[16] Onimaru T, Sakakibara T, Aso N, Yoshizawa H, Suzuki H S and Takeuchi T 2005 *Phys. Rev. Lett.* **94** 197201
[17] Kletowski Z, Sławiński P and Cichorek T 1996 *J. Magn. Magn. Mater.* **162** 277–279
[18] Morie T 2007 *Ph.D. Thesis* The University of Tokyo
[19] Sakakibara T, Tayama T, Onimaru T, Aoki D, Onuki Y, Sugawara H, Aoki Y and Sato H 2003 *J. Phys.: Condens. Matter* **15** S2055
[20] Sato Y, Makiyama S, Kawae T, Onimaru T, Suzuki H S, Jackson M, Paulsen C, Amara M and Galéra R 2014 *JPS Conf. Proc.* **3** 011052
[21] Aoki D, Katayama Y, Settai R, YInada, Onuki Y, Harima H and Kletowski Z 1997 *J. Phys. Soc. Jpn.* **66** 3988
[22] Ikeura T, Matsubara T, Machida Y, Izawa K, Nagasawa N, Matsumoto K T, Onimaru T and Takabatake T 2014 *JPS Conf. Proc.* **3** 011091
[23] Machida Y, Yoshida T, Ikeura T, Izawa K, Nakama A, Higashinaka R, Aoki Y, Sato H, Sakai A, Nakatsuji S, Nagasawa N, Matsumoto K, Onimaru T and Takabatake T 2015 *J. Phys: Conf. Ser.* **592** 012025
[24] Yoshida T, Machida Y, Izawa K, Shimada Y, Nagasawa N, Onimaru T and Takabatake T 2015 *Physics Proceedia* accepted