Strong Orbital Fluctuations in Multipolar Ordered States of $\text{PrV}_2\text{Al}_{20}$

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Abstract

$\text{PrT}_2\text{Al}_{20}$ ($T = \text{Ti, V}$) are ideal systems to study the quadrupole Kondo effect and quantum criticality arising from orbital degrees of freedom. The both systems have the nonmagnetic cubic Γ_3 crystal electric field ground doublet with the well separated excited state. In particular, $\text{PrV}_2\text{Al}_{20}$ exhibits anomalous metallic behavior above and below the multipolar ordering temperatures, reflecting the even stronger hybridization between f and conduction electrons possibly due to a proximity to an orbital quantum critical point. Here we report the anomalous metallic behaviors found in a pure single crystal of $\text{PrV}_2\text{Al}_{20}$. Our detailed analyses revealed that the resistivity indicates power law temperature dependence proportional to T^3 . Furthermore, we pointed out that the $4f$ electron contribution to the specific heat also exhibits power law behavior proportional to T^4 . Both observations are in a sharp contrast to the gapped behavior found in $\text{PrTi}_2\text{Al}_{20}$ and indicate the strong c - f hybridization and strong orbital fluctuations in $\text{PrV}_2\text{Al}_{20}$. In addition, the $4f$ electron contribution to the entropy in $\text{PrV}_2\text{Al}_{20}$ reaches only 50% of $R\ln 2$ at an orbital ordering at $T = 0.75$ K, suggesting another 50% of $R\ln 2$ expected for Γ_3 doublet is already released at higher temperature possibly due to quadrupole Kondo effect.

Keywords: orbital fluctuations, multipole ordering, quadrupole Kondo effect, unconventional superconductivity

1. Introduction

Novel quantum phases may arise from f electrons' orbital degrees of freedom through hybridization with conduction electrons. For instance, quadrupole Kondo effect, originally suggested as a single ion effect in the nonmagnetic cubic Γ_3 crystal electric field (CEF) doublet[1], has attracted a lot of attention because of its non-Fermi liquid ground state. It is an interesting open question what happens in the lattice systems. On the other hand, it is a further non-trivial open issue to search for a novel quantum phase arises in the vicinity of a quantum critical point (QCP) of an orbital ordering. So far, a lot of interesting quantum states, such as exotic superconductivity (SC) with highly enhanced effective mass and non-Fermi liquid, have been reported in the vicinity of magnetic QCPs[2, 3, 4]. Similarly exotic states may also be found at the border of orbital QCPs.

In order to study this, it is important to choose a material which has purely orbital degrees of freedom in the

ground state. Such examples can be found in cubic Γ_3 doublet systems of a f^2 configuration. Intensive studies have revealed various interesting phenomena in the cubic Γ_3 doublet systems, in particular, in Pr-based intermetallic compounds[5, 6, 7, 8, 9]. However, Pr-based cubic Γ_3 doublet systems usually have well-localized f moments, and thus there has been no prototypical system where we can study the orbital instability by tuning the hybridization between f and conduction electrons.

On the other hand, recent studies have revealed that $\text{PrT}_2\text{Al}_{20}$ ($T = \text{Ti, V}$) are ideal systems to study the quadrupole Kondo effect and quantum criticality arising from orbital degrees of freedom, where we can tune the c - f hybridization by chemical substitution and by pressure[10, 11]. The both systems have the nonmagnetic cubic Γ_3 CEF doublet with the well separated excited state at $\Delta_{\text{CEF}} \sim 60$ K (Ti) and 40 K (V)[10, 12, 13]. In addition, strong hybridization is expected in these compounds, from a caged structure with 16 Al ions surrounding T ions. Indeed, the hybridization is strong as is evident in many physical properties[10, 14, 15, 16]. In particular, $\text{PrV}_2\text{Al}_{20}$ exhibits anomalous metallic behavior above and below the multipolar ordering temperatures, reflecting the even stronger hybridization due to

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a smaller size of the cage.

$\text{PrV}_2\text{Al}_{20}$ exhibits double multipolar orderings at temperatures $T = 0.65$ and 0.75 K, which has been revealed by our recent specific heat measurements using a pure single crystal[17, 16]. The double transition becomes more prominent in the samples with better quality, which indicates that the double transition is the intrinsic property of $\text{PrV}_2\text{Al}_{20}$. However, the origin of the double transition is not clear so far. On the other hand, a ferro-quadrupole (FQ) ordering (at $T = 2.0$ K) has been suggested for $\text{PrTi}_2\text{Al}_{20}$ [10, 12, 18]. Interestingly, both of these compounds exhibit heavy fermion SC inside the multipole ordering phases[19, 11, 16]. In $\text{PrTi}_2\text{Al}_{20}$, the SC is highly enhanced under pressure of 8 GPa, where the ferro-quadrupole ordering starts to be suppressed. The transition temperature (T_c) at ambient pressure (~ 0.2 K) increases up to ~ 1 K and the effective mass reaches $m^*/m_0 \sim 110$ [11]. This strongly suggests a novel heavy fermion SC emerges in the vicinity of the orbital QCP. Moreover, in the case of $\text{PrV}_2\text{Al}_{20}$, the SC appears at $T_c = 0.05$ K with the highly enhanced effective mass ($m^*/m_0 \sim 140$) even at ambient pressure[16]. This observation indicates the first realization of the novel SC arising from the orbital fluctuation of the f electrons at ambient pressure.

Here, we discuss the anomalous metallic behaviors found below and above the multipole ordering temperatures in a pure single crystal of $\text{PrV}_2\text{Al}_{20}$ [16, 20], focusing on how they are different from those found in $\text{PrTi}_2\text{Al}_{20}$. We revealed that the resistivity of $\text{PrV}_2\text{Al}_{20}$ indicates power law temperature dependence proportional to T^3 , which is in contrast to the gapped behavior found in $\text{PrTi}_2\text{Al}_{20}$. Likewise, the $4f$ electron contribution to the specific heat of $\text{PrV}_2\text{Al}_{20}$ exhibits power law behavior proportional to T^4 , which is again in contrast to the gapped behavior in $\text{PrTi}_2\text{Al}_{20}$. In addition, the $4f$ electron contribution to the entropy in $\text{PrV}_2\text{Al}_{20}$ reaches only 50% of $R\ln 2$ at the orbital ordering at $T = 0.75$ K, suggesting another 50% of $R\ln 2$ expected for Γ_3 doublet is already released at higher temperature possibly due to quadrupole Kondo effect. All these observations indicate the strong c - f hybridization and strong orbital fluctuations which may arise from the proximity to a orbital QCP in $\text{PrV}_2\text{Al}_{20}$. Note that a part of the results presented in this paper has been already discussed in the previous works[10, 16, 20].

2. Experimental

Single crystals of $RT_2\text{Al}_{20}$ ($R = \text{Pr, La}$, $T = \text{Ti, V}$) were grown by the Al self-flux method[10]. We have succeeded in growing high quality single crystals,

whose residual resistivity ratio (RRR) reaches ~ 20 , by tuning the starting ratio. Single and powder X-ray diffraction and SEM-EDX measurements indicate high quality and single phase of obtained crystals. The electrical resistivity $\rho(T)$ was measured by a low-frequency ac four-terminal method below ~ 1 K using ^3He - ^4He dilution refrigerator. A dc four-terminal method was used above $T = 0.3$ K in conjunction with a commercial system (PPMS, Quantum Design). In this paper, RRR is roughly defined as the ratio between the values at 300 K and lowest temperature above T_c of the SC transition. The specific heat was measured using a relaxation method for the pure single crystal with RRR ~ 20 (0.111 mg), which is the same sample as used for the ac- and dc-susceptibility measurements in the previous studies[16]. For the measurements above $T = 0.9$ K, a commercial system (PPMS, Quantum Design) was used. Measurements below $T \sim 1$ K were made by using a specific heat cell installed in a ^3He - ^4He dilution refrigerator. The details were already discussed in the previous work[16].

3. Results and Discussions

First we present the low temperature resistivity ρ for various single crystals of $\text{PrV}_2\text{Al}_{20}$ with different sample qualities in Figure 1 (a). Here, the data for the single crystal of $\text{PrTi}_2\text{Al}_{20}$ (RRR ~ 150) are also shown for comparison. As clearly seen in the figure, $\text{PrV}_2\text{Al}_{20}$ with RRR ~ 19 exhibits a resistivity drop due to the SC transition at $T_c = 0.05$ K, as indicated by an arrow. The SC transition was also found in another sample (RRR ~ 18) at a slightly lower $T_c = 0.039$ K. However, we did not find any sign of SC at least down to 38 and 43 mK for the sample with RRR ~ 4 and 7, respectively. The results suggest that T_c strongly depends on RRR, which may indicate an unconventional character of the SC. Apparently, further measurements are required to confirm the RRR dependence.

On the other hand, there is another relatively steep drop of the resistivity at $T_O \sim 0.7$ K, which is associated with multipolar orderings of $\text{PrV}_2\text{Al}_{20}$. Here, we defined T_O as a peak temperature of $d\rho/dT$, which is dependent on the RRR of the sample[16]. While clear double transitions were found in the specific heat at $T_Q = 0.75$ K and $T^* = 0.65$ K, $d\rho/dT$ indicates only one peak even for the sample with RRR ~ 19 at least in zero-magnetic field. T_O increases from 0.5 to 0.67 K if the RRR is increased from ~ 4 to ~ 19 . In the case of $\text{PrTi}_2\text{Al}_{20}$, T_O is found at ~ 2.0 K, which is associated with the FQ ordering.

Figure 1: (a) Temperature dependence of the resistivity ρ for various single crystals of $\text{PrV}_2\text{Al}_{20}$ with different sample qualities (filled circles). The data for the single crystal of $\text{PrTi}_2\text{Al}_{20}$ (RRR ~ 150) are also shown for comparison (open circles). Note that the data for $\text{PrTi}_2\text{Al}_{20}$ are multiplied by a factor of two for clarity. The dashed line represents a fit by a $\ln T$ curve at $T > T_0$ to the data for RRR ~ 19 . (b) Full logarithmic plot of $\Delta\rho$ against normalized temperature T/T_0 for $\text{PrV}_2\text{Al}_{20}$ and $\text{PrTi}_2\text{Al}_{20}$. Here, $\Delta\rho$ is defined as $\Delta\rho \equiv \rho - \rho_0$, where ρ_0 is the residual resistivity estimated above T_c . The broken line indicates a power law temperature dependence of $\Delta\rho \propto T^3$. Note that the same symbols as (a) are used here. For details, see text.

In order to discuss the temperature dependence of the resistivity below T_0 more in detail, we present a full logarithmic plot of $\Delta\rho$ against normalized temperature T/T_0 for $\text{PrV}_2\text{Al}_{20}$ and $\text{PrTi}_2\text{Al}_{20}$ in Figure 1 (b). Here, $\Delta\rho$ is defined as $\Delta\rho \equiv \rho - \rho_0$, where ρ_0 is the residual resistivity estimated above T_c . Remarkably, all the data collapse on top for $\text{PrV}_2\text{Al}_{20}$ with RRR $\sim 7, 18$

Figure 2: Temperature dependence of C/T (closed circles). Here, the lattice contribution is subtracted by using C/T for $\text{LaV}_2\text{Al}_{20}$. C_{4f}/T obtained after subtracting the nuclear contribution C_n/T is also shown (open circles). The broken line at $T > T_Q$ indicates the anomalous temperature dependence with $C/T \propto T^{-3/2}$. Inset shows $\Delta C/T$ obtained after subtracting γ from C_{4f}/T at the lowest temperatures around T_c . In addition to the zero-field data, those in the weak magnetic field along the $[110]$ direction are also shown. See text for details.

and 19. Furthermore, these exhibit a power law temperature dependence of $\Delta\rho \propto T^{3.0 \pm 0.1}$ at low temperatures below $T/T_0 \sim 0.5$, which is indicated by the broken line in the figure. On the other hand, $\text{PrTi}_2\text{Al}_{20}$ exhibit a convex curve in the full logarithmic plot, reflecting the gapped temperature dependence arising from the FQ ordering as discussed in the previous work[19]. Even though the temperature range for the power law behavior is somewhat narrow, the contrasting behavior between $\text{PrV}_2\text{Al}_{20}$ and $\text{PrTi}_2\text{Al}_{20}$ is evident. The power law behavior in $\text{PrV}_2\text{Al}_{20}$ suggests strong orbital fluctuation even in the multipolar ordered state. Interestingly, $\text{PrTi}_2\text{Al}_{20}$ under high pressure of 8.7 GPa also indicates the same T^3 power law behavior[11]. Note that, for the sample with RRR 18 and 19, $\Delta\rho$ deviates upward from the power law behavior just below T_0 , while it is not observed in the sample RRR with 5. This is supposed to reflect the fact that the multipolar transitions become sharper as RRR increases.

Next, we present the temperature dependence of the specific heat (C) divided by T in Figure. 2. Here, the lattice contribution is already subtracted by using C/T for $\text{LaV}_2\text{Al}_{20}$. At the high temperatures, as we mentioned above, there are two peaks at $T_Q = 0.75$ K and $T^* = 0.65$ K corresponding to the multipole orderings. Note that these temperatures are designated following the previous work[17] and we do not intend to refer to

any specific order parameter in this paper. In the low temperatures, the data exhibit a slight increase on cooling below 0.12 K and finally indicate an anomaly due to SC transition at $T = 0.046$ K. The low T upturn in C/T becomes evident in the normal state stabilized under the magnetic field of 20 mT and is found to follow $C/T \sim T^2$ down to the lowest T of 30 mK. As already discussed in the previous work[16], this contribution is most likely arising from a hyperfine enhanced nuclear magnetism. The $4f$ -electron contribution of the specific heat divided by T , C_{4f}/T is obtained after subtracting $C_n/T \sim T^{-2}$ coming from the nuclear magnetism. The zero-field SC anomaly is clearly seen in C_{4f}/T , which is shown in the figure by the open circles. This confirms the bulk character of the SC. The nearly constant C_{4f}/T in the normal state provides an estimate of the large electronic coefficient $\gamma \sim 0.9$ J/molK², consistent with the effective mass $m^*/m_0 \sim 140$ estimated from the B_{c2} curve[16].

The SC jump in C/T is evaluated using the temperature dependence of $\Delta C/T \equiv C_{4f}/T - \gamma$, as is shown in the inset of Figure 2. The jump reaches 0.3 J/molK², which provides direct evidence of the heavy-fermion SC. The ratio obtained from the jump, $\Delta C/\gamma T_c \sim 0.3$ is much smaller than the BCS value of 1.43. As already discussed in the previous work[16], the above estimated jump can be further increased if the sample quality is improved, which is the one of the important future issues. In the inset of Figure 2, $\Delta C/T$ in weak magnetic fields are also shown. The SC anomaly is slightly suppressed at $B = 0.5$ mT and is not observed at $B = 20$ mT. T_c at $B = 0.5$ mT is roughly estimated to be 0.042 K, which is consistent with the previously reported B_{c2} curve[16].

Corresponding to the contrasting behavior in the resistivity below T_0 , the specific heat data also exhibit contrasting behavior in $\text{PrTi}_2\text{Al}_{20}$ and $\text{PrV}_2\text{Al}_{20}$ below the multipolar ordering temperatures. In order to demonstrate this, we present the full logarithmic plot of C_{4f} versus temperature for both $\text{PrV}_2\text{Al}_{20}$ and $\text{PrTi}_2\text{Al}_{20}$ in Figure 3(a). As clearly seen in the figure, $\text{PrTi}_2\text{Al}_{20}$ shows the exponential T dependence with the convex curve below $T_0 \sim 2.0$ K, which is arising from an excitation gap expected for FQ ordering. On the other hand, $\text{PrV}_2\text{Al}_{20}$ exhibits a clear power law behavior in $\Delta C \equiv C_{4f} - \gamma T$ with $\Delta C \propto T^4$ below T^* [16, 20] as shown by open circles. This power law behavior indicates the existence of a gapless mode corresponding to the so-called “orbital” proposed for various transition metal based systems[21]. The gapless mode in the multipolar ordered state in $\text{PrV}_2\text{Al}_{20}$ would be the signature of strong orbital fluctuations arising from the proximity

Figure 3: (a) Temperature dependence of C_{4f} for $\text{PrV}_2\text{Al}_{20}$ (closed circles) and $\text{PrTi}_2\text{Al}_{20}$ (closed squares) in full logarithmic scale. $\Delta C \equiv C_{4f} - \gamma T$ for $\text{PrV}_2\text{Al}_{20}$ is also shown by open circles. The dashed line below T^* represents the T^4 behavior in ΔC . The broken line above T_Q represents the $T^{-1/2}$ behavior in C_{4f} . (b) Temperature dependence of the $4f$ electron contribution to the entropy S_{4f} for $\text{PrV}_2\text{Al}_{20}$ (solid line) and $\text{PrTi}_2\text{Al}_{20}$ (dashed line). For details, see text.

to an orbital QCP. This gapless character should be the results of the strong c - f hybridization effect, which is also seen in the temperature dependence of $4f$ electron contribution to the entropy S_{4f} obtained after the integration of C_{4f}/T vs T . As shown in Figure 3(b), while S_{4f} for $\text{PrTi}_2\text{Al}_{20}$ reaches 90% of $R \ln 2$ at $T_0 \sim 2.0$ K, S_{4f} for $\text{PrV}_2\text{Al}_{20}$ reaches only 50% of $R \ln 2$ at $T_Q \sim 0.75$ K. Another 50% of $R \ln 2$ expected for Γ_3 doublet is already released at higher temperature range from T_Q to $T \sim 4$ K. Interestingly, in the same temperature range, C_{4f} in $\text{PrV}_2\text{Al}_{20}$ exhibits the anomalous power law behavior of $C_{4f} \propto T^{-1/2}$ (Fig. 3(a)), as already discussed in the previous work[16, 20]. In addition, in this temperature range, we have seen various anomalous metallic properties far different from the Fermi liquid behavior, such as the magnetic susceptibility proportional to $-T^{1/2}$ [10] and the resistivity proportional to $\ln T$ as indicated by

the dashed line in Figure 1 (a). One possible scenario for the origin of the anomalous metal is the quadrupolar Kondo effect. Note that a residual entropy of $R/2\ln 2$ is expected for the quadrupolar Kondo effect, which is consistent with our observation of S_{4f} reaches only 50% of $R\ln 2$ in $\text{PrV}_2\text{Al}_{20}$ at T_Q .

4. Conclusion

Here, we discussed the anomalous metallic behaviors found below and above the multipole ordering temperatures in a pure single crystal of $\text{PrV}_2\text{Al}_{20}$. We revealed that the resistivity of $\text{PrV}_2\text{Al}_{20}$ indicates power law temperature dependence proportional to T^3 , which is in contrast to the gapped behavior found in $\text{PrTi}_2\text{Al}_{20}$. Likewise, the $4f$ electron contribution to the specific heat of $\text{PrV}_2\text{Al}_{20}$ exhibits power law behavior proportional to T^4 , which is again in contrast to the gapped behavior in $\text{PrTi}_2\text{Al}_{20}$. This suggests the existence of a gapless mode corresponding to “ orbiton ” in $\text{PrV}_2\text{Al}_{20}$. In addition, the $4f$ electron contribution to the entropy in $\text{PrV}_2\text{Al}_{20}$ reaches only 50% of $R\ln 2$ at the orbital ordering at $T = 0.75$ K, suggesting another 50% of $R\ln 2$ expected for Γ_3 doublet is already released at higher temperature possibly due to quadrupole Kondo effect. All these observations indicate the strong c - f hybridization and strong orbital fluctuations which may arise from the proximity to a orbital QCP in $\text{PrV}_2\text{Al}_{20}$. In order to clarify these possibilities, a lot of studies are required for both experiments and theories. In particular, the origin of the double transition found at $T_Q = 0.75$ and $T^* = 0.65$ K should be clarified through detailed studies of thermodynamics, transport and microscopic measurements. Further improvement of the sample quality is also the important future issue.

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