Very Low Temperature Magnetoresistance in the Quadrupole Ordered System PrV$_2$Al$_{20}$

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Abstract. We measured magnetoresistance of the quadrupole ordered system PrV$_2$Al$_{20}$ with a $\Gamma_3$ doublet ground state under high-DC magnetic field up to 30 T. The field dependence of the magnetoresistance is strongly different between $H \parallel [111]$ and $H \parallel [110]$ in spite of the cubic crystal structure. For $H \parallel [110]$, we observed a jump with distinct hysteresis between 13 and 16 T and a shoulder structure in the field of 8 T at 23 mK.

1. Introduction

Cubic Pr-based compounds with $\Gamma_3$ non-magnetic doublet ground state recently attracts much interest. The $\Gamma_3$ doublet does not carry magnetic moments but only multipole moments, two electric quadrupole moments $O_{2}^{2}$, $O_{2}^{0}$ and magnetic octapole moments $T_{xyz}$. Therefore, we can study pure multipole contribution at low temperature if the $\Gamma_3$ doublet ground state is sufficiently separated from the crystal-electric-field excited state. In usual case, the degeneracy of the $\Gamma_3$ doublet is lifted by the quadrupole ordering at low temperature. For example, PrPb$_3$, PrIr$_2$Zn$_{20}$ and PrRh$_2$Zn$_{20}$ are found to exhibit antiferro-quadrupolar ordering at 0.4 K, 0.11 K, and 0.06 K, respectively [1, 2, 3]. These magnetic phase diagrams are anisotropic among [100], [110] and [111] in spite of the cubic symmetry. The anisotropy of the magnetic phase diagram gives us important clue to determine the quadrupolar order parameter.

PrV$_2$Al$_{20}$ with the $\Gamma_3$ doublet ground state also exhibits an antiferro-quadrupole ordering at 0.65 - 0.75 K [4, 5]. In the quadrupole ordered state, superconducting transition with heavy quasi-particle mass was recently found at 0.05 K [5]. The phase diagram of the quadrupole ordered state is little anisotropic for three principal field directions [100], [110] and [111] below 9 T [4]. However, above 9 T, the magnetic phase diagram is strongly anisotropic. For $H \parallel [111]$, the quadrupole phase diagram is closed at the critical field of 11 T [6]. Near the critical field, the field dependence of magnetoresistance $\rho(H)$ exhibits a peak, suggesting the enhancement of the residual resistivity. In addition, the field dependence of $A$ coefficient in $\rho(T) = \rho_0 + AT^2$, where $\rho_0$ is residual resistivity, divergently increases by approaching the critical field from 30 T. These results suggest the emergence of the field-induced quantum critical point at the critical field of 11 T. By contrast, for $H \parallel [100]$, another high-field ordered phase was found above 11 T [7]. This high-field phase transition probably is due to the switching of the quadrupolar ordered
Figure 1. The field dependence of magnetoresistance $\rho(H)$ of Pr$V_2$Al$_{20}$ (sample 1), measured at the various temperature points in the field aligned for $I \parallel H \parallel [111]$.

parameter between $O^2_2$ and $O^0_2$. Thus, the phase diagram is strongly anisotropic under high magnetic field. The results about magnetic phase diagram above 9 T for $H \parallel [110]$ has not been reported in magnetoresistance as well as we know.

2. Experimental

Single crystal samples of Pr$V_2$Al$_{20}$ were grown by Al-self-flux method [4, 5, 6]. In this paper, we show results of the magnetoresistance for two samples. For one sample (sample 1), residual resistive ratio (RRR) and the current direction are RRR = 10 and $I \parallel [111]$, respectively. Those for the other sample are RRR = 12 and $I \parallel [110]$, respectively. For both samples, emergence of the multipole ordered transition at $\sim 0.6$ K was confirmed.

3. Results and Discussion

Figure 1 shows the field dependence of magnetoresistance $\rho(H)$ in Pr$V_2$Al$_{20}$ (sample 1) at various temperatures between 0.41 K and 0.92 K for the magnetic field and current parallel to the [111] direction. Below $\sim 10$ T, the value of the resistivity dramatically increases from 0.46 K to 0.71 K. This change is due to the quadrupole ordered transition observed at $\sim 0.6$ K. In the field sweep, two characteristic anomalies were observed. One is a small peak in the field of $\sim 1$ T, and the other is the peak observed at $\sim 11$ T. While the former peak is observed in the temperature only below $T_Q = 0.6$ K, the latter peak remains even above the order temperature. The emergence of the peak even above $T_Q$ is evidence of the development of the critical quantum fluctuation at $\sim 11$ T. Above 11 T, no-anomalies due to the transition were detected in the high-magnetic field up to 30 T. This result suggests that para-quadrupole state remains above 11 T. The detailed temperature dependence above 11 T and the analysis have been already reported [6].
Figure 2. $\rho(H)$ in PrV$_2$Al$_{20}$ (sample 2) for the $I || [110] \perp H || [111]$ (triangle) and $I || H || [110]$ (circle) at 23 mK. Open (close) symbols indicate the field-increasing (decreasing) process. Two open arrows show a small anomaly at $\sim 1$ T and a shoulder structure at $\sim 8$ T for $I || H || [110]$.

Figure 2 exhibits $\rho(H)$ in PrV$_2$Al$_{20}$ (sample 2) for the current parallel to [110] at the very low temperature of 23 mK, deep in the quadrupolar ordered phase. We measured $\rho(H)$ for two field directions of $H || [111]$ (triangle) and $H || [110]$ (circle). For $H || [111]$, two peaks at $\sim 1$ T and $\sim 11$ T were also observed, consistent with the results as shown in Fig. 1. For these peaks, distinct hysteresis was not detected. Under the high magnetic field for $H || [111]$, the magnetoresistance in Fig. 2 is enhanced, compared with that in Fig. 1. Difference of these two data is mainly arising from the relation between current direction and magnetic field direction ($H$ parallel or perpendicular to the current direction) For $H || [110]$, $\rho(H)$ is dramatically different from that for $H || [111]$. $\rho(H)$ for $H || [110]$ shows a jump accompanied with distinct hysteresis in the field between 13 - 16 T, in addition to the small anomaly at $\sim 1$ T and a shoulder structure at $\sim 8$ T. These two anomalies at $\sim 1$ T and $\sim 8$ T remain only inside the quadrupole ordered state (not shown). The field where jump is observed for $H || [110]$ is higher than the critical field of 11 T for $H || [111]$. This result suggests that the critical field of the ordered phase for $H || [110]$ is higher than that for $H || [111]$. The anisotropy of the magnetization indicates that $\Gamma_3$ magnetic triplet is the first-excited state lying above the $\Gamma_3$ doublet ground state [8]. In that case, the gap in the $\Gamma_3$ doublet for $H || [111]$ is larger than that for $H || [110]$ under the magnetic field. The difference of the gap size under magnetic field is probably associated with the anisotropic critical field. The detailed field/temperature dependence and the phase diagram for $H || [110]$ will be published elsewhere.

Anisotropic phase diagram has been studied in detail in the cubic PrPb$_3$ with a $\Gamma_3$ doublet ground state, indicating an antiferro-quadrupole ordering due to the $O_2^3$ quadrupole moments at 0.4 K [1, 9]. For $H || [110]$, above 7 T, another high-field phase was recently found by specific heat measurements [10]. This ordered phase remains, at least, up to 13 T [11]. By contrast, for $H || [111]$, the quadrupole ordered phase closes at 6 T, and the high-field ordered phase as
observed for $H \parallel [110]$ was not observed [12].

In the case of PrV$_2$Al$_20$, as shown in Fig. 2, a shoulder structure at 8 T probably due to the transition was observed for $H \parallel [110]$. This result suggests emergence of the high-field ordered state in the field between 8 T and 15 T. Such an anomaly was not found for $H \parallel [111]$. Noted that both of PrV$_2$Al$_20$ and PrPb$_3$ exhibit high-field ordered phases for $H \parallel [100]$ above ~ 11 T and ~ 6 T, respectively [13, 7]. Thus, for both systems, the high-field ordered state is observed only for $H \parallel [110]$ and $H \parallel [110]$. In PrPb$_3$, for $H \parallel [110]$, neutron scattering measurements revealed that the order parameter of the high-field phase above 7 T is $O^2_2$ quadrupole moments, which is different from that of $O^2_2$ moments at zero field [9, 14]. From the analogy to the phase diagram in PrPb$_3$, a shoulder structure at 8 T in PrV$_2$Al$_20$ for $H \parallel [110]$ may be due to switching of the ordered parameter between $\Gamma_3$-type quadrupole moments $O^0_2$ and $O^2_2$.

4. Conclusion

We reported the magnetoresistance in the quadrupole ordered system PrV$_2$Al$_20$ with $\Gamma_3$ doublet ground state at very low temperature of 23 mK under DC-high magnetic field. The field dependence between $H \parallel [111]$ and $H \parallel [110]$ is strongly anisotropic. The critical field of ordered phase for $H \parallel [110]$ is higher than that for $H \parallel [111]$.

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