

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

Y Shimura<sup>1</sup>, M Tsujimoto<sup>1</sup>, B Zeng<sup>2</sup>, Q Zhang<sup>2</sup>, L Balicas<sup>2</sup>,  
A Sakai<sup>1,3</sup>, and S Nakatsuji<sup>1</sup>

<sup>1</sup>Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan

<sup>2</sup>National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida  
32310, USA

<sup>3</sup>I. Physikalisches Institut, Georg-August-Universität Göttingen, 37077 Göttingen, Germany

E-mail: simu@issp.u-tokyo.ac.jp

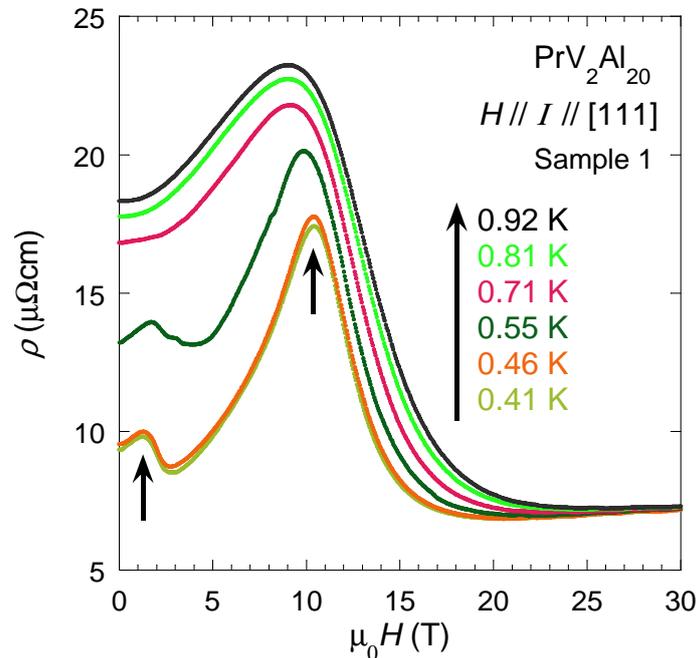
**Abstract.** We measured magnetoresistance of the quadrupole ordered system  $\text{PrV}_2\text{Al}_{20}$  with a  $\Gamma_3$  doublet ground state under high-DC magnetic field up to 30 T. The field dependence of the magnetoresistance is strongly different between  $H \parallel [111]$  and  $H \parallel [110]$  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,  $\text{PrPb}_3$ ,  $\text{PrIr}_2\text{Zn}_{20}$  and  $\text{PrRh}_2\text{Zn}_{20}$  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.

$\text{PrV}_2\text{Al}_{20}$  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  $\text{PrV}_2\text{Al}_{20}$  (sample 1), measured at the various temperature points in the field aligned for  $I \parallel H \parallel [111]$ .

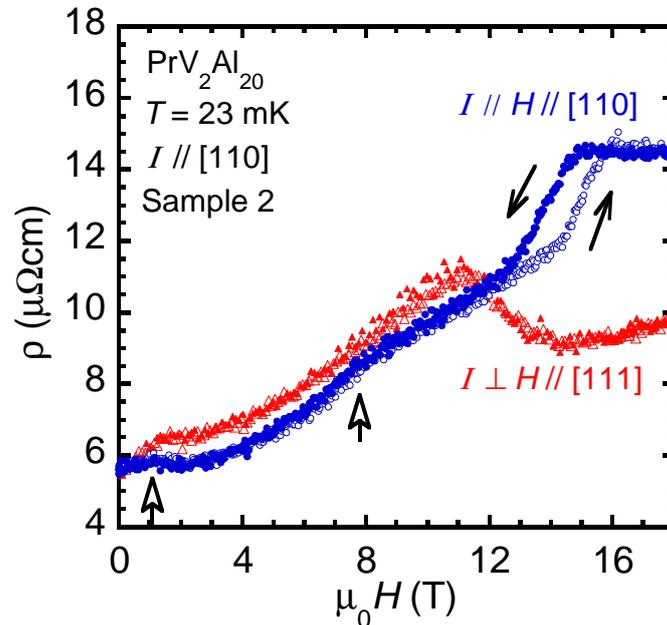
parameter between  $O_2^2$  and  $O_2^0$ . Thus, the phase diagram is strongly anisotropic under high magnetic field. The results about magnetic phase diagram above 9 T for  $H \parallel [110]$  has not been reported in magnetoresistance as well as we know.

## 2. Experimental

Single crystal samples of  $\text{PrV}_2\text{Al}_{20}$  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  $\text{RRR} \sim 10$  and  $I \parallel [111]$ , respectively. Those for the other sample are  $\text{RRR} \sim 12$  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  $\text{PrV}_2\text{Al}_{20}$  (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  $\text{PrV}_2\text{Al}_{20}$  (sample 2) for the  $I \parallel [110] \perp H \parallel [111]$  (triangle) and  $I \parallel H \parallel [110]$  (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  $I \parallel H \parallel [110]$ .

Figure 2 exhibits  $\rho(H)$  in  $\text{PrV}_2\text{Al}_{20}$  (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  $H \parallel [111]$  (triangle) and  $H \parallel [110]$  (circle). For  $H \parallel [111]$ , 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  $H \parallel [111]$ , 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  $H \parallel [110]$ ,  $\rho(H)$  is dramatically different from that for  $H \parallel [111]$ .  $\rho(H)$  for  $H \parallel [110]$  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  $H \parallel [110]$  is higher than the critical field of 11 T for  $H \parallel [111]$ . This result suggests that the critical field of the ordered phase for  $H \parallel [110]$  is higher than that for  $H \parallel [111]$ . The anisotropy of the magnetization indicates that  $\Gamma_5$  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  $H \parallel [111]$  is larger than that for  $H \parallel [110]$  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  $H \parallel [110]$  will be published elsewhere.

Anisotropic phase diagram has been studied in detail in the cubic  $\text{PrPb}_3$  with a  $\Gamma_3$  doublet ground state, indicating an antiferro-quadrupole ordering due to the  $O_2^0$  quadrupole moments at 0.4 K [1, 9]. For  $H \parallel [110]$ , 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  $H \parallel [111]$ , 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  $\text{PrV}_2\text{Al}_{20}$ , 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  $\text{PrV}_2\text{Al}_{20}$  and  $\text{PrPb}_3$  exhibit high-field ordered phases for  $H \parallel [100]$  above  $\sim 11$  T and  $\sim 6$  T, respectively [13, 7]. Thus, for both systems, the high-field ordered state is observed only for  $H \parallel [110]$  and  $H \parallel [100]$ . In  $\text{PrPb}_3$ , 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  $O_2^0$  moments at zero field [9, 14]. From the analogy to the phase diagram in  $\text{PrPb}_3$ , a shoulder structure at 8 T in  $\text{PrV}_2\text{Al}_{20}$  for  $H \parallel [110]$  may be due to switching of the ordered parameter between  $\Gamma_3$ -type quadrupole moments  $O_2^0$  and  $O_2^2$ .

#### 4. Conclusion

We reported the magnetoresistance in the quadrupole ordered system  $\text{PrV}_2\text{Al}_{20}$  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  $H \parallel [111]$ .

#### Acknowledgments

This work is partially supported by PRESTO, Japan Science and Technology Agency, Grants-in-Aid for Scientific Research (No. 25707030, 15J08663 and 25887015), by Grants-in-Aids for Scientific Research on Innovative Areas (15H05882, 15H05883) and Program for Advancing Strategic International Networks to Accelerate the Circulation of Talented Researchers (No. R2604) from the Japanese Society for the Promotion of Science. Y.S. is partially supported by the Institute of Complex Adaptive Matter (ICAM). The NHMFL is supported by NSF through NSF-DMR-0084173 and the State of Florida. L.B. is supported by DOE-BES through award DE-SC0002613. This work was supported in part by NSF Grant No. PHYS-1066293 and the hospitality of the Aspen Center for Physics.

#### References

- [1] Bucher E, Andres K, Gossard A and Maita J 1974 *J. Low Temp. Phys.* **2** 322
- [2] Onimaru T, Matsumoto K T, Inoue Y F, Umeo K, Sakakibara T, Karaki Y, Kubota M and Takabatake T 2011 *Phys. Rev. Lett.* **106**
- [3] Onimaru T, Nagasawa N, Matsumoto K T, Wakiya K, Umeo K, Kittaka S, Sakakibara T, Matsushita Y and Takabatake T 2012 *Phys. Rev. B* **86** 184426
- [4] Sakai A and Nakatsuji S 2011 *J. Phys. Soc. Jpn.* **80** 063701
- [5] Tsujimoto M, Matsumoto Y, Tomita T, Sakai A and Nakatsuji S 2014 *Phys. Rev. Lett.* **113** 267001
- [6] Shimura Y, Tsujimoto M, Zeng B, Balicas L, Sakai A and Nakatsuji S 2015 *Phys. Rev. B* **91** 241102
- [7] Shimura Y, Ohta Y, Sakakibara T, Sakai A and Nakatsuji S 2013 *J. Phys. Soc. Jpn.* **82** 043705
- [8] Araki K, Shimura Y, Kase N, Sakakibara T, Sakai A and Nakatsuji S 2014 *JPS Conf. Proc.* **3** 011093
- [9] Onimaru T, Sakakibara T, Aso N, Yoshizawa H, Suzuki H S and Takeuchi T 2005 *Phys. Rev. Lett.* **94** 197201
- [10] Sato Y, Makiyama S, Morodomi H, Inagaki Y, Kawae T, Suzuki H S and Onimaru T 2012 *J. Phys.: Conf. Ser.* **391** 012060
- [11] Onimaru T 2005 *Ph.D. Thesis, The University of Tokyo*
- [12] Tayama T, Sakakibara T, Kitami K, Yokoyama M, Tenya K, Amitsuka H, Aoki D, Ōnuki Y and Kletowski Z 2001 *J. Phys. Soc. Jpn.* **70** 248
- [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] Onimaru T, Aso N, Prokes K, Suzuki H, Sato T and Sakakibara T 2007 *J. Phys. Chem. Soli.* **68** 2091