Characterization of Ferromagnetic Order in CePd$_2$P$_2$

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Abstract. We examine magnetic ordering in CePd$_2$P$_2$ by neutron-scattering experiment and confirmed a ferromagnetic order at low temperatures. From the neutron-scattering data, the ordered moment is evaluated to be $1.3(3)$ $\mu_B$ per Ce ion at 3 K and is considered to be parallel to the $c$-axis. In addition, no evidence of the change in the magnetic structure was observed below $T_C$ within the experimental accuracy. In DC magnetization measurement with an as-grown powder sample and a magnetically aligned sample, a clear magnetic anisotropy in CePd$_2$P$_2$ was observed in both ferromagnetic and paramagnetic phases. To obtain further insight to the large Curie temperature $T_C$ of CePd$_2$P$_2$, we have examined the magnetic properties of GdPd$_2$P$_2$ as a reference material. In DC magnetization measurement, successive magnetic transitions at $T_{I} = 10.6$ K and $T_{II} = 7.4$ K were observed in GdPd$_2$P$_2$. By comparing the transition temperatures of the Ce and Gd compounds, we argue the origin of the large Curie temperature of CePd$_2$P$_2$.

1. Introduction

A competition between the Kondo effect and magnetic ordering is a subject of great interest in $f$-electron systems, and frequently result in the occurrence of exotic quantum critical phenomena. In this context, a number of Ce compounds were investigated to clarify how the antiferromagnetic ordering competes with the Kondo effect, but a clear example of the ferromagnetic (FM) ordering in a Ce compound is hardly known. This is a striking contrast to the case of the uranium-based compounds such as UGe$_2$, URhGe and UCoGe [1]. In order to explore the essential features of the FM quantum critical behavior and related phenomena, it would be intriguing to search a new FM compound.

Recently, we found a new candidate of a Ce-based FM compound CePd$_2$P$_2$ [2]. Although this material was prepared by Jeitschko et al. in 1983, detailed physical properties were not reported except for the crystal structure [3] at that time. Very recently, two groups independently examined detailed magnetic properties of CePd$_2$P$_2$, and established the FM ordering with macroscopic measurements [4, 5]. Especially, Tran et al. reported an anomalous critical behavior and discussed a possible competition between the ferromagnetism and the Kondo effect in CePd$_2$P$_2$ [5].

We note that CePd$_2$P$_2$ possesses several interesting features: e.g. (i) a large Curie
temperature in spite of a large Ce-Ce distance, (ii) anomalous critical behavior as reported in ref. [5], and more interestingly (iii) a possible candidate to examine FM quantum critical phenomena in the Ce-based compound. In the present study, we investigate the magnetic structure of CePd$_2$P$_2$ by neutron-scattering experiments, and examine the FM transition through the DC magnetization measurements in further detail. As a reference material, GdPd$_2$P$_2$ has been also investigated by measuring DC magnetization to verify the de Gennes scaling on REPd$_2$P$_2$ (RE: rare earth) system.

2. Experimental

Polycrystalline samples of both CePd$_2$P$_2$ and GdPd$_2$P$_2$ were prepared through the solid state reaction in an evacuated silica tube as described in Ref. [3]. To improve the homogeneity of the sample, the sintered products were ground and reheated several times. The crystal structure of both CePd$_2$P$_2$ and GdPd$_2$P$_2$ was confirmed to be the ThCr$_2$Si$_2$-type structure with the powder x-ray diffraction measurement (Rigaku; MiniFlex). The lattice constants of CePd$_2$P$_2$ and GdPd$_2$P$_2$ were evaluated as $a = 4.159$ Å, $c = 9.897$ Å, and $a = 4.083$ Å, $c = 9.861$ Å, respectively, and they are consistent with the reported values [3].

Neutron scattering experiment was performed using the High Resolution Chopper spectrometer (HRC) [6] installed at BL12 of the Material and Life Science Experimental Facility (MLF) in Japan Proton Accelerator Research Complex (J-PARC), Tokai, Japan. Experimental details of the neutron-scattering study is described elsewhere [7].

DC Magnetization was measured in a temperature range of 2-300 K and a field range of ±70 kOe with an AC/DC magnetometry system (ACMS) installed on a physical property measurement system (Quantum Design). To examine the magnetic anisotropy of CePd$_2$P$_2$, magnetic susceptibility was measured with two samples; one is an as-grown powder sample, the other one is a magnetically aligned powder sample, which was mixed with paraffin and aligned in a magnetic field of $H = 70$ kOe at 345 K. In this study, we do not make a correction for the demagnetization field.

3. Results and Discussion

To begin with, we confirm the ferromagnetic ordering in CePd$_2$P$_2$ by the neutron-scattering experiment. Figure 1 exhibits neutron diffraction patterns measured at 3 K in the magnetically ordered phase (filled-blue circles) and at 40 K in the paramagnetic phase (filled-red circles). In order to extract magnetic signals, the intensity measured at 40 K was subtracted from that of 3 K, and the residual intensity is depicted in Fig. 1 with open-blue rectangles. In a low-$Q$ region, no Bragg peak was observed except for that at fundamental Bragg positions. From this difference pattern, it is obvious that the magnetically ordered phase of CePd$_2$P$_2$ is not a complex antiferromagnetic order but a simple ferromagnetic order. Furthermore, no significant change of the intensity at the 002 position clearly indicates that an ordered moment is parallel to the c-axis. By analyzing the neutron scattering intensity at the 101 position, a magnitude of the ordered moment has been evaluated as $\sim 1.3(3)\mu_B$ per Ce ion at 3 K. A reduction in the ordered moment from the full moment (2.14 $\mu_B$ for $J = 5/2$) can be attributed to a crystalline electric field (CEF) effect and/or a Kondo effect, as discussed in ref. [5]. Details of the CEF effect on CePd$_2$P$_2$ are described elsewhere [7].

In Fig. 2(a), we show isothermal magnetization curves of CePd$_2$P$_2$ at 2 K. Here magnetization data labeled as “no-oriented” (circles) and “oriented” (triangles) in Fig. 2(a) were measured with an as-grown powder sample and a magnetically aligned powder sample, respectively. A clear hysteresis in the magnetization curves is also indicative of a FM ordering in CePd$_2$P$_2$ at low temperatures. The saturation magnetization is evaluated as about 1 $\mu_B$ and 0.8 $\mu_B$ per Ce ion for oriented and no-oriented powder sample, respectively. The value of the oriented sample is in reasonable agreement with the evaluated ordered moment from the neutron-scattering
Figure 1. Neutron diffraction pattern at 3 and 40 K. The difference of the neutron-scattering intensity between 3 and 40 K is depicted with the open squares. The numbers on the peaks are the Miller indices.

Figure 2. (a) Magnetic field dependence of the magnetization for CePd$_2$P$_2$ measured at 2 K. (b) Reciprocal molar magnetic susceptibility of CePd$_2$P$_2$ as a function of the temperature. Here $\chi^{-1}$ indicated by “oriented” (triangles) were measured with the powder sample, which was aligned in the magnetic field of 70 kOe at 345 K. The solid lines are results of a fit. The dash line is the guide to the eye.

Next, we examine magnetic properties in a paramagnetic region. Figure 2(b) shows the temperature dependence of the reciprocal magnetic susceptibility $\chi^{-1}$ of CePd$_2$P$_2$ for both “no-
oriented” and “oriented” samples. As seen in Fig. 2(b), $\chi_0^{-1}$ of the oriented sample is smaller than that of the no-oriented sample. This difference in $\chi_0^{-1}$ in a paramagnetic state is also indicative of the magnetic anisotropy in CePd$_2$P$_2$ owing to the CEF effect.

At high temperatures, $\chi_0$ of the oriented sample shows Curie-Weiss behavior. From the result of a fit (red solid line), the effective magnetic moment and the paramagnetic Curie-Weiss temperature of CePd$_2$P$_2$ are evaluated as $\mu_{\text{eff}} = 2.4(1) \mu_B$/Ce and $\Theta_p = +13(5)$ K, respectively. The evaluated $\mu_{\text{eff}}$ is close to the free ion moment (2.54 $\mu_B$) and consistent with the reported values [4, 5] within the experimental accuracy. On the other hand, $\Theta_p$ differs from their evaluated value ($\Theta_p^{\text{Tran}} = -2$ K [5]). This discrepancy in $\Theta_p$ can be attributed to a magnetic anisotropy owing to the CEF effect. In fact, a similar negative value of $\Theta_p$ was also evaluated for $\chi_0^{-1}$ of our no-oriented sample. In contrast to their interpretation in Ref.[5], the positive $\Theta_p$ of our oriented sample is in reasonable agreement with the ferromagnetic transition temperature of $T_C \approx 28$ K, and indicates strong FM correlations between magnetic moments of Ce ions. Additionally, a deviation from the Curie-Weiss law at low temperatures could be attributed to a CEF effect and/or a development of ferromagnetic correlations.

Our neutron-scattering data indicate that the magnetic moments are parallel to the $c$ axis, so that the magnetic easy axis of CePd$_2$P$_2$ can be considered to be the $c$ axis. Considering the magnetic anisotropy observed in the magnetization measurement, a uniaxial (Ising like) FM ordering is likely to be realized in CePd$_2$P$_2$.

![Figure 3](image-url) (a) Reciprocal molar magnetic susceptibility of GdPd$_2$P$_2$ (open-rectangles) as a function of the temperature. The solid lines are results of a fit. (b) Temperature dependence of $M/H$ of GdPd$_2$P$_2$ at several fields.

Finally, we compare the magnetic properties between Ce and Gd compounds. For GdPd$_2$P$_2$, the reciprocal susceptibility shows the Curie-Weiss behavior in a wide temperature range. By fitting high-temperature data, $\mu_{\text{eff}}$ and $\Theta_p$ are evaluated as $\mu_{\text{eff}} = 7.89(4) \mu_B$/Gd and $\Theta_p = -26(3)$ K, respectively. The evaluated $\mu_{\text{eff}}$ is consistent with the theoretical value of Gd$^{3+}$ ion (7.94 $\mu_B$ for $J = 7/2$) within the experimental accuracy. To examine the magnetic transition temperature of GdPd$_2$P$_2$, we depict the temperature dependence of $M/H$ at lower temperatures in Fig. 3(b). As one can expect from the negative $\Theta_p$, $M/H$ at 0.5 kOe exhibits a distinct cusp anomaly at $T_I = 10.6$ K, indicating an antiferromagnetic ordering. Furthermore, one can see a successive magnetic transition at $T_{II} = 7.6$ K, and a complex temperature-field dependence.
of $M/H$ below $T_1$. These results indicate the existence of complex magnetic interactions in GdPd$_2$P$_2$.

Although such complex magnetic ordering on GdPd$_2$P$_2$ itself is also interesting, we would like to focus on the de Gennes scaling behavior. Usually, a magnetic ordering of rare-earth compounds originates from the RKKY interaction. Accordingly, the magnetic transition temperature $T_N$ can be described as $T_N \propto (g_J - 1)^2 J(J + 1) \cdot J_{\text{RKKY}}(Q)$, where $g_J$ is the Lande factor, $J$ is the total angular momentum, and $J_{\text{RKKY}}$ is the RKKY exchange interaction [8]. The first term $(g_J - 1)^2 J(J + 1)$, which is called as de Gennes factor $G_{\text{RE}}(J)$, only depends on $J$, while $J_{\text{RKKY}}$ is closely correlated with the conduction-band properties through the $cf$ exchange interaction. When we assume the same density of state at Fermi energy between Ce and Gd compounds, $T_N$ can be scaled with $G_{\text{RE}}(J)$, where $G_{\text{Ce}}(J = 5/2) = 0.18$ for Ce$^{3+}$, and $G_{\text{Gd}}(J = 7/2) = 15.75$ for Gd$^{3+}$ [9]. Thus, the transition temperature of CePd$_2$P$_2$ is expected to be $\sim 1/100$ smaller than that of GdPd$_2$P$_2$. Contrary to this expectation, $T_C$ of CePd$_2$P$_2$ is about three times larger than $T_N$ of GdPd$_2$P$_2$. This result indicates that the Curie temperature of CePd$_2$P$_2$ is substantially enhanced. The enhancement of $T_C$ on CePd$_2$P$_2$ may be attributed to the enhanced FM interaction through $J_{\text{RKKY}}(Q = 0)$. For further understanding of the enhancement of $T_C$, it is important to investigate the band structure of CePd$_2$P$_2$. At the same time, further experimental efforts on the growth of a single crystal are also useful.

4. Conclusion

We have performed neutron scattering experiment to investigate magnetic ordering in CePd$_2$P$_2$ and confirmed that a FM order is realized below $T_C$ in CePd$_2$P$_2$. The ordered moment was evaluated as $\sim 1.3(3) \mu_B$ per Ce ion at 3 K. No significant change in the neutron-scattering intensity at the 002 position indicates that the ordered moment is parallel to the $c$-axis. In particular, no evidence of the change in the magnetic structure was observed below $T_C$ within the experimental accuracy.

In DC magnetization measurement with an as-grown powder sample and a magnetically aligned powder sample, a clear magnetic anisotropy was observed in CePd$_2$P$_2$ in both FM and paramagnetic phases. Considering the result of the neutron-scattering experiment, the magnetic easy axis can be considered to be the $c$ axis for CePd$_2$P$_2$. For the oriented sample, the saturation moment at 2 K of CePd$_2$P$_2$ is about 1 $\mu_B$ per Ce ion, and it is in reasonable agreement with the neutron-scattering data. From $\chi_0^{-1}$ of the oriented sample, the effective magnetic moment and the paramagnetic Curie-Weiss temperature of CePd$_2$P$_2$ are evaluated as $\mu_{\text{eff}} = 2.4(1) \mu_B$ per Ce ion and $\Theta_p = +13(5) \text{K}$, respectively. The positive value of $\Theta_p$ is in reasonable agreement with $T_C$ of CePd$_2$P$_2$ and clearly indicates strong FM correlations between the magnetic moment of Ce ions.

To obtain further insight of the large $T_C$ of CePd$_2$P$_2$, we also examined the magnetic properties of GdPd$_2$P$_2$ as a reference material. From the results of the DC magnetization measurement, we showed that GdPd$_2$P$_2$ exhibits the successive antiferromagnetic transitions at $T_1 = 10.6 \text{K}$ and $T_{1f} = 7.4 \text{K}$. By comparing the magnitude of the transition temperature between Ce and Gd compounds, we conclude that $T_C$ of CePd$_2$P$_2$ is substantially enhanced.

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