Journal of Physics: Conference Series

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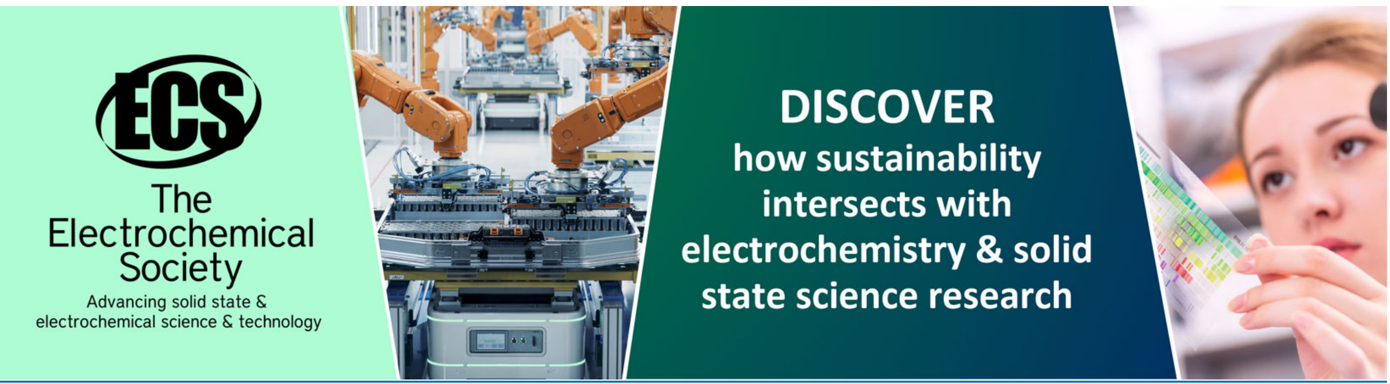
To cite this article: Y Ikeda *et al* 2015 *J. Phys.: Conf. Ser.* **592** 012013



Journal of Physics: Conference Series **592** (2015) 012013 doi:10.1088/1742-6596/592/1/012013

Characterization of Ferromagnetic Order in $CePd_2P_2$

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Abstract. We examine magnetic ordering in $CePd_2P_2$ by neutron-scattering experiment and confirmed a ferromagnetic order at low temperatures. From the neutron-scattering data, the ordered moment is evaluated to be $\sim 1.3(3)$ μ_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 $CePd_2P_2$ was observed in both ferromagnetic and paramagnetic phases. To obtain further insight to the large Curie temperature T_C of $CePd_2P_2$, we have examined the magnetic properties of $GdPd_2P_2$ 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 $GdPd_2P_2$. By comparing the transition temperatures of the Ce and Gd compounds, we argue the origin of the large Curie temperature of $CePd_2P_2$.

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 $CePd_2P_2$ [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 $CePd_2P_2$, 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 $CePd_2P_2$ [5].

We note that $CePd_2P_2$ 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 $CePd_2P_2$ by neutron-scattering experiments, and examine the FM transition through the DC magnetization measurements in further detail. As a reference material, $GdPd_2P_2$ has been also investigated by measuring DC magnetization to verify the de Gennes scaling on $REPd_2P_2$ (RE: rare earth) system.

2. Experimental

Polycrystalline samples of both $CePd_2P_2$ and $GdPd_2P_2$ 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 $CePd_2P_2$ and $GdPd_2P_2$ was confirmed to be the $ThCr_2Si_2$-type structure with the powder x-ray diffraction measurement (Rigaku; MiniFlex). The lattice constants of $CePd_2P_2$ and $GdPd_2P_2$ 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 $CePd_2P_2$, 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 $CePd_2P_2$ 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 $CePd_2P_2$ 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 μ_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 $CePd_2P_2$ are described elsewhere [7].

In Fig. 2(a), we show isothermal magnetization curves of $CePd_2P_2$ 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 $CePd_2P_2$ at low temperatures. The saturation magnetization is evaluated as about 1 μ_B and 0.8 μ_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

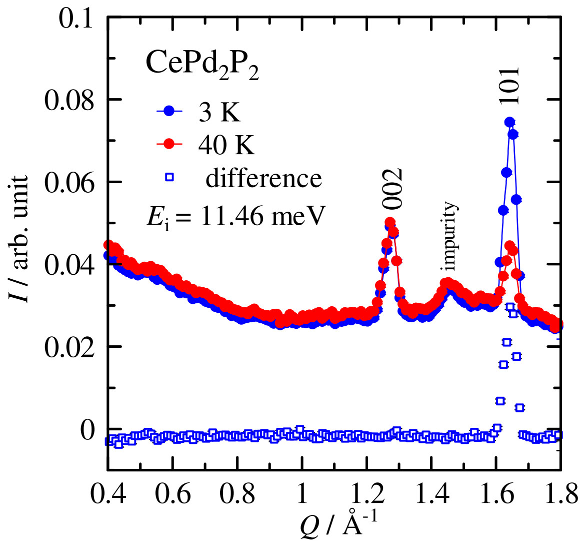

Figure 1. Neutron diffraction pattern at 3 and 40 K. The difference of the neutron-scattering intensity between 3 and 40 K is depicted with the open squares. The numbers on the peaks are the Miller indices.

experiment ($\sim 1.3\ \mu_B$). The slight discrepancy may be due to imperfection of field orientation for the powder sample. In any case, the difference between the oriented and no-oriented powder sample is indicative of the magnetic anisotropy in $CePd_2P_2$ in the FM ordering state.

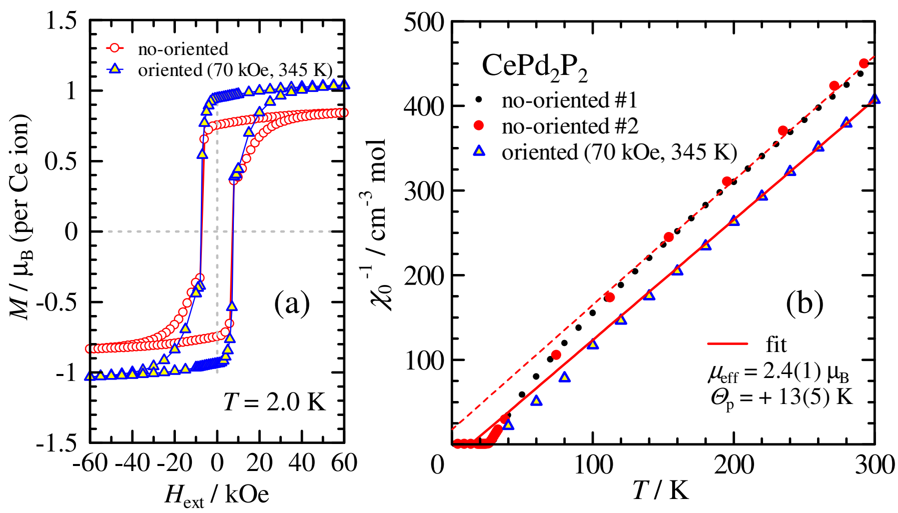

Figure 2. (a) Magnetic field dependence of the magnetization for $CePd_2P_2$ measured at 2 K. (b) Reciprocal molar magnetic susceptibility of $CePd_2P_2$ as a function of the temperature. Here χ_0^{-1} indicated by "oriented" (triangles) were measured with the powder sample, which was aligned in the 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 χ_0^{-1} of $CePd_2P_2$ for both "no-oriented" and "oriented" samples. As seen in Fig. 2(b), χ_0^{-1} of the oriented sample is smaller than that of the no-oriented sample. This difference in χ_0^{-1} in a paramagnetic state is also indicative of the magnetic anisotropy in $CePd_2P_2$ owing to the CEF effect.

At high temperatures, χ_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 $CePd_2P_2$ are evaluated as $\mu_{\rm eff} = 2.4(1)\mu_{\rm B}$/Ce and $\Theta_{\rm p} = +13(5)$ K, respectively. The evaluated $\mu_{\rm eff}$ is close to the free ion moment (2.54 $\mu_{\rm B}$) and consistent with the reported values [4, 5] within the experimental accuracy. On the other hand, $\Theta_{\rm p}$ differs from their evaluated value ($\Theta_{\rm p}^{\rm Tran} = -2$ K [5]). This discrepancy in $\Theta_{\rm p}$ can be attributed to a magnetic anisotropy owing to the CEF effect. In fact, a similar negative value of $\Theta_{\rm p}$ was also evaluated for χ_0^{-1} of our no-oriented sample. In contrast to their interpretation in Ref.[5], the positive $\Theta_{\rm p}$ of our oriented sample is in reasonable agreement with the ferromagnetic transition temperature of $T_{\rm C} \sim 28$ 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 $CePd_2P_2$ 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 $CePd_2P_2$.

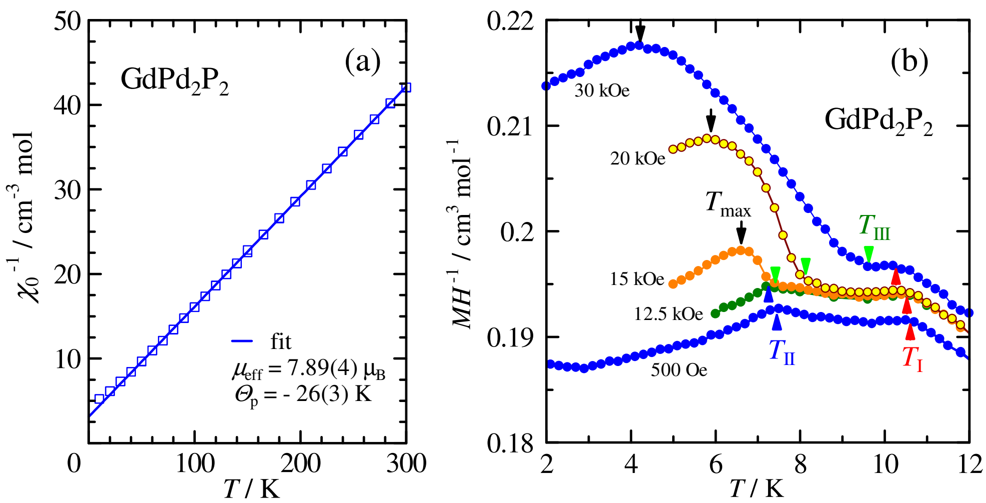

Figure 3. (a) Reciprocal molar magnetic susceptibility of $GdPd_2P_2$ (open-rectangles) as a function of the temperature. The solid lines are results of a fit. (b) Temperature dependence of M/H of $GdPd_2P_2$ at several fields.

Finally, we compare the magnetic properties between Ce and Gd compounds. For $GdPd_2P_2$, the reciprocal susceptibility shows the Curie-Weiss behavior in a wide temperature range. By fitting high-temperature data, $\mu_{\rm eff}$ and $\Theta_{\rm p}$ are evaluated as $\mu_{\rm eff} = 7.89(4)\mu_{\rm B}$/Gd and $\Theta_{\rm p} = -26(3)$ K, respectively. The evaluated $\mu_{\rm eff}$ is consistent with the theoretical value of Gd^{3+}-ion (7.94 $\mu_{\rm B}$ for $J = 7/2$) within the experimental accuracy. To examine the magnetic transition temperature of $GdPd_2P_2$, we depict the temperature dependence of M/H at lower temperatures in Fig. 3(b). As one can expect from the negative $\Theta_{\rm p}$, M/H at 0.5 kOe exhibits a distinct cusp anomaly at $T_{\rm I} = 10.6$ K, indicating an antiferromagnetic ordering. Furthermore, one can see a successive magnetic transition at $T_{\rm II} = 7.6$ K, and a complex temperature-field dependence of M/H below T_{I}. These results indicate the existence of complex magnetic interactions in $GdPd_2P_2$.

Although such complex magnetic ordering on $GdPd_2P_2$ 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 $T_{\mathrm{N}} \propto (g_J - 1)^2 J(J+1) \cdot J_{\mathrm{RKKY}}(Q)$, where g_J is the Lande factor, J is the total angular momentum, and J_{RKKY} is the RKKY exchange interaction [8]. The first term $(g_J - 1)^2 J(J+1)$, which is called as de Gennes factor $G_{\mathrm{RE}}(J)$, only depends on J, while J_{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 $G_{\mathrm{RE}}(J)$, where $G_{\mathrm{Ce}}(J = 5/2) = 0.18$ for Ce^{3+}, and $G_{\mathrm{Gd}}(J = 7/2) = 15.75$ for Gd^{3+} [9]. Thus, the transition temperature of $CePd_2P_2$ is expected to be $\sim 1/100$ smaller than that of $GdPd_2P_2$. Contrary to this expectation, T_{C} of $CePd_2P_2$ is about three times larger than T_{N} of $GdPd_2P_2$. This result indicates that the Curie temperature of $CePd_2P_2$ is substantially enhanced. The enhancement of T_{C} on $CePd_2P_2$ may be attributed to the enhanced FM interaction through $J_{\mathrm{RKKY}}(Q = 0)$. For further understanding of the enhancement of T_{C}, it is important to investigate the band structure of $CePd_2P_2$. 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 $CePd_2P_2$ and confirmed that a FM order is realized below T_{C} in $CePd_2P_2$. The ordered moment was evaluated as $\sim 1.3(3)$ μ_{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 $CePd_2P_2$ 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 $CePd_2P_2$. For the oriented sample, the saturation moment at 2 K of $CePd_2P_2$ is about 1 μ_{B} per Ce ion, and it is in reasonable agreement with the neutron-scattering data. From χ_0^{-1} of the oriented sample, the effective magnetic moment and the paramagnetic Curie-Weiss temperature of $CePd_2P_2$ are evaluated as $\mu_{\mathrm{eff}} = 2.4(1)\mu_{\mathrm{B}}$ per Ce ion and $\Theta_{\mathrm{p}} = +13(5)$ K, respectively. The positive value of Θ_{p} is in reasonable agreement with T_{C} of $CePd_2P_2$ and clearly indicates strong FM correlations between the magnetic moment of Ce ions.

To obtain further insight of the large T_{C} of $CePd_2P_2$, we also examined the magnetic properties of $GdPd_2P_2$ as a reference material. From the results of the DC magnetization measurement, we showed that $GdPd_2P_2$ exhibits the successive antiferromagnetic transitions at $T_{\mathrm{I}} = 10.6$ K and $T_{\mathrm{II}} = 7.4$ K. By comparing the magnitude of the transition temperature between Ce and Gd compounds, we conclude that T_{C} of $CePd_2P_2$ is substantially enhanced.

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