

Metamagnetism of DyPd₂Si₂ Single Crystal

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Magnetic characteristics of a DyPd₂Si₂ single crystal with the ThCr₂Si₂-type structure have been studied from measurements of magnetic susceptibility $\chi(T)$ and magnetization $M(B)$. It was found that DyPd₂Si₂ orders antiferromagnetically below $T_N = 9.2$ K with the basal-plane [100] axis being the easy magnetization direction at high magnetic fields. Upon increasing magnetic field, the [100] magnetization curve exhibits at least six metamagnetic transitions at 0.2, 1.8, 3.0, 3.7, 4.2 and 6.1 T before reaching the saturation magnetization $M_s = 10 \mu_B$. Five metamagnetic transitions were detected on decreasing fields. For the [110] magnetization process, three metamagnetic transitions were evidenced. In contrast, the $M(B)$ curve along the [001] axis, which is the hard magnetization direction, displays no metamagnetic transition; it shows a monotonous increase with a concave curvature up to 18 T. Finally, we constructed a comprehensive B_{100} - T magnetic phase diagram consisting of six magnetic ordered phases.

KEYWORDS: DyPd₂Si₂, Antiferromagnetism, Multi-step metamagnetic transition, Magnetic phase diagram.

1. Introduction

The ternary compounds RM₂X₂ (R=rare earth, M=transition metal, X=Si, Ge and so on) with the tetragonal ThCr₂Si₂-type structure have been extensively studied over the last three decades because of their large variety of physical properties [1]. Most of these compounds show antiferromagnetic ordering, and many of them exhibit Ising-like behaviors with the [001] direction (*c*-axis) being the easy magnetization axis. Much less study has been made on the RM₂X₂ compounds with the easy magnetization direction in the basal plane. Such a study is interesting since unique and new behaviors in distinct from the *c*-axis easy-direction compounds are expected. It has been reported that on the RPd₂Si₂ (R = rare earth) compounds have an anisotropy with the easy direction in the basal plane [1, 2]. Recently, we performed detailed studies on several RPd₂Si₂ compounds, including GdPd₂Si₂ without crystalline electric field (CEF) effects, a heavy-rare-earth compound TbPd₂Si₂ and a light-rare-earth compound PrPd₂Si₂ with CEF effects have been already investigated, and observed interesting magnetic properties, such as novel magnetic behaviors and peculiar *B*-*T* magnetic phase diagrams [3, 4], multi-step metamagnetic behaviors in the basal plane magnetization processes, as well as a strong magnetic anisotropy within the basal plane [5, 6]. The main features of the anisotropic magnetic behavior can be well explained by considering the CEF effects

[5, 6]. In this work, a DyPd₂Si₂ single crystal having a Kramers ion, is studied by magnetic measurements. It is expected that the Dy-compound shows different behavior from those of non-Kramers Tb- and Pr-compounds.

2. Experimental

The DyPd₂Si₂ single-crystal was grown by using the tri-arc Czochralski method. The tetragonal ThCr₂Si₂-type structure and the single-phase nature were confirmed by using X-ray powder diffraction. The quality of the single crystal was checked and the crystallographic orientations were determined by back Laue method. Low-field magnetizations below 7 T were measured using a Physical Property Measurement System (PPMS, Quantum Design). High-field magnetization measurements up to 18 T were performed using a superconducting magnet by a sample extracting magnetometer at the High Field Laboratory for Superconducting Materials, the Institute for Material Research, Tohoku University.

3. Results and Discussion

The temperature dependences of magnetic susceptibility $\chi(T)$ along the main symmetry axes of the tetragonal cell are shown in Fig.1. A cusp which indicates an antiferromagnetic transition is seen at 9.2 K in each $\chi(T)$ curve. The magnetic susceptibilities follow the Curie-Weiss law for high temperatures; the inverse susceptibility $\chi^{-1}(T)$ curves are linear and almost parallel to each other above 100 K (see in the inset of Fig. 1). The effective moment μ_{eff} estimated from the gradient of the line is 10.8 μ_B , which agrees with the expected value of 10.6 μ_B for the Dy³⁺ free-ion, indicating localized *f*-electrons in this compound.

Magnetization curves along the main symmetry axes of the tetragonal cell at 1.6 K up to 18 T are shown in Fig. 2. It is clear that the easy magnetization axis is the basal-plane [100] direction. The [100] magnetization is almost saturated above 15 T and reaches $M_s = 10.0 \mu_B$, which is in good agreement with the Dy³⁺ free ion moment ($gJ=10.0 \mu_B$). It is noticeable that multi-step metamagnetic behaviors appear. To examine this behavior in more detail, the differential magnetization curve, dM/dB vs. B on the ascending process, is shown together with the magnetization curve $M(B)$ in Fig. 3 as an example. Hereafter, a metamagnetic transition field is defined by a peak field in the dM/dB curve.

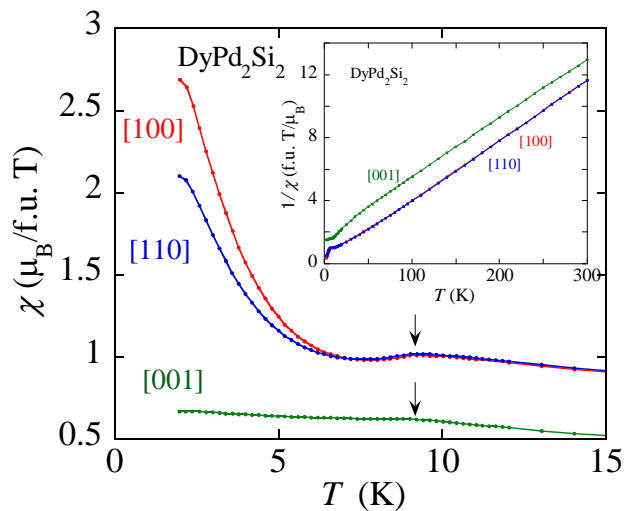


Fig. 1. Temperature dependences of magnetic susceptibilities along the main symmetry axes of a tetragonal cell on a DyPd₂Si₂ single crystal. The inset shows the inverse susceptibilities.

Six peaks at $B_{a1} = 0.2$, $B_{a2} = 1.8$, $B_{a3} = 3.0$, $B_{a4} = 3.7$, $B_{a5} = 4.2$, and $B_{a5} = 6.1$ T are evidenced, which shows one-to-one correspondence to the metamagnetic transitions in the $M(B)$ curve. A six-step metamagnetic process appears on the ascending process. On the descending process, five metamagnetic transitions appear; that is, dM/dB shows five peaks at $B_{d5} = 6.1$, $B_{d4} = 4.5$, $B_{d3} = 2.6$, $B_{d2} = 1.0$, and $B_{d1} = 0.2$ T although the $dM/dB(B)$ curve is not shown here. Two hysteresis loops are seen between B_{a4} and B_{d3} , and between B_{a2} and B_{d2} .

The [110] magnetization process is a three-step metamagnetic one; three transitions appear at $B_{b1} = 0.2$, $B_{b2} = 2.8$, and $B_{b3} = 5.3$ T. The $M_{[110]}$ curve is almost parallel to the $M_{[100]}$ curve at high fields and $M_{[110]}$ cannot reach saturation up to 18 T, indicating that the magnetic anisotropy is very strong even within the basal plane.

In contrast, along the [001] direction, i.e., the c -axis, which is the hard magnetization direction, the magnetization increases monotonously with a concave curvature. A strong magnetic anisotropy between the c -axis and the basal plane is also evidenced. Although all metamagnetic transitions disappear in the paramagnetic region, the strong magnetic anisotropy still persists. To understand such anisotropic behavior, we carried out an analysis of CEF effect by using the same procedure as described in the previous works [5, 6]. Unfortunately, satisfactory result cannot be obtained, indicating that additional effects such as quadrupolar effect may play an important role.

Finally, the B_{100} - T magnetic phase diagram was constructed from the temperature dependences of magnetization at various magnetic fields and the magnetization processes at various temperatures. As shown in Fig. 4, six magnetically ordered phases referred to as phase I, II, III, IV, V, and VI in the order of increasing magnetic field were obtained. Above 6 K, some phase boundaries cannot be determined unambiguously because transitions are smeared out by the thermal effects. This complex magnetic diagram may come from competing magnetic interactions. To achieve a better

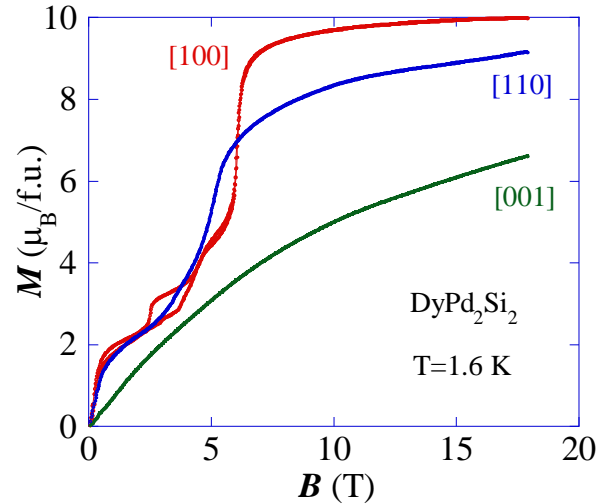


Fig. 2. Magnetization curves along the main symmetry axes at 1.6 K on the DyPd_2Si_2 single crystal.

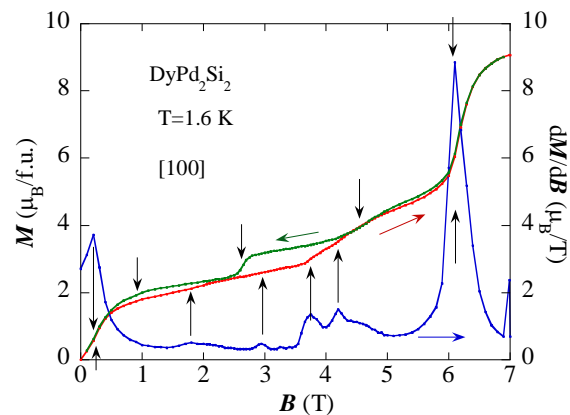


Fig. 3. Magnetization curve at 1.6 K, and differential magnetization dM/dB curve on the ascending process. Up- and down-ward arrows show transition positions on the ascending and descending process, respectively.

understanding on these magnetic behaviors, it is required to determine the magnetic structures under magnetic fields.

3. Summary

Magnetic characteristics of a DyPd_2Si_2 single crystal were studied. This compound orders antiferromagnetically below $T_N = 9.2$ K. The [100] magnetization is saturated and reaches the expected value of $10.0 \mu_B$ for the Dy^{3+} free ion moment, indicating well localized $4f$ electrons, which indicated that the easy magnetization axis is the [100] direction. A six-step metamagnetic behavior appears in the [100] process. The [110] magnetization process is a three-step one. Strong magnetic anisotropy is observed between the [100] and the [110] directions within the basal plane for high magnetic fields, and between the [001] and directions in the basal plane. The $B_{[100]}-T$ magnetic phase diagram was constructed, in which six magnetic ordered phases exist. Information on magnetic structures under magnetic fields is required in order to understand this complex behavior. Further studies, such as neutron diffraction, are planned.

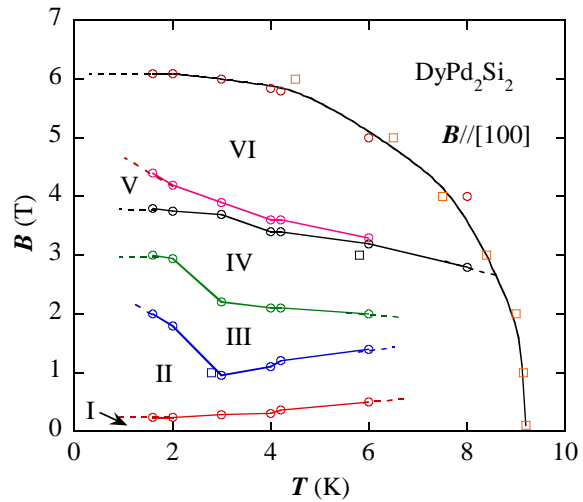


Fig. 4. $B_{[100]}-T$ magnetic phase diagram. Circles and squares are transition points determined from M vs. B and M vs. T curves, respectively.

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