IRREVERSIBLE MAGNETIZATION PROCESS OF A $PrCu_2Ge_2$ SINGLE CRYSTAL*

T. Shigeoka, Y. Taneda

Faculty of Science, Yamaguchi University, Yoshida, Yamaguchi 753-8512, Japan

M. Hedo, and Y. Uwatoko

Institute for Solid State Physics, University of Tokyo, Chiba 277-8581, Japan

(Received July 10, 2002)

Measurements of magnetization and magnetic susceptibility have been carried out on a tetragonal ternary $PrCu_2Ge_2$ single crystal compound. It orders antiferromagnetically below T_N=14.4 K which is an anomalously high transition temperature. The magnetic behavior in the virgin state is different from one in the state after magnetization saturation process along the c-axis in low temperatures. The susceptibility of the virgin state is enhanced below 3.5 K while one after saturation becomes very small. An irreversible magnetization process appears in the virgin state while it becomes reversible after saturation, which has been never seen yet.

PACS numbers: 75.30.–m, 75.30.Cr, 75.20.Hr

1. Introduction

The ternary compound $PrCu_2Ge_2$, crystallizing in the tetragonal $ThCr_2Si_2$-type structure, shows interesting magnetic behaviors [1]. It orders antiferromagnetically below T_N=16 K which is an anomalously high transition temperature. The Néel temperature of the corresponding Gd compound is 12 K [2]. The antiferromagnetic structure reported is the AF-type I; a simple collinear structure with the wave vector (0,0,1), consisting of ferromagnetic c-planes coupled antiferromagnetically with the $+ - + -$ sequence [3, 4]. The existence of another magnetic transition at 4.2 K was suggested by behavior of heat-capacity, magnetic susceptibility and electrical resistivity. The details are, however, unknown yet. In order to elucidate this

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

magnetic behavior, measurements of magnetic susceptibility and magnetization have been carried out on a $PrCu_2Ge_2$ single crystal compound grown by the tri-arc Czochralski method. The magnetic measurements have been carried out for 1.8–300 K and 0–7 T by a sample extracting magnetometer at the Institute of Solid State Physics, University of Tokyo.

2. Result and discussion

The temperature dependence of magnetic susceptibility along the three symmetry directions of tetragonal cell is shown in Fig. 1.

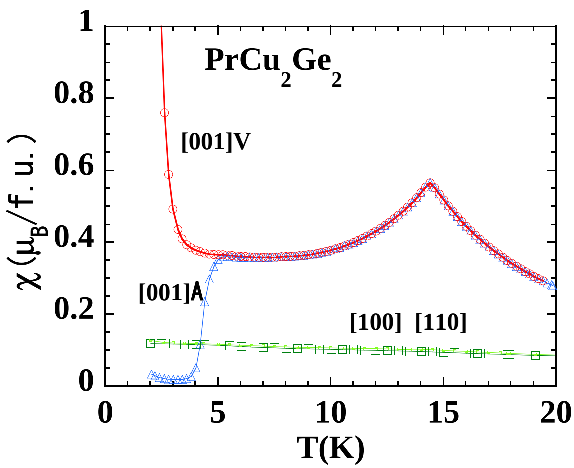

Fig. 1. The temperature dependence of magnetic susceptibility along the main symmetry directions on a $PrCu_2Ge_2$ single crystal compound. Here, $[001]_V$ and $[001]_A$ means a virgin state and one after saturation, respectively.

The susceptibility is anisotropic; the *c*-axis susceptibility is larger than those in the basal plane, indicating the easy magnetization axis is the *c*-axis. The susceptibility within the basal plane is almost isotropic. Along the *c*-axis, a cusp is observed at 14.4 K, attributed to the ordering temperature, which is slightly lower than the previous report, 16 K [1]. In low temperatures, the susceptibility shows a very peculiar behavior; the susceptibility of a virgin state is different from one after a saturation magnetization process. Below 3.5 K, a strong increase in virgin susceptibility can be observed with decreasing temperature. After the magnetization is saturated by an applied field above 1.5 T at low temperature of 2 K, the susceptibility becomes very small (the detail will be mentioned below). Then with increasing temperature, it persists the small value below 3.5 K, increases rapidly around 4 K, reaches the value of the virgin susceptibility at 5 K and at last above 5 K, behaves similarly to one of the virgin state. This behavior suggests a magnetic transition around 4 K. On the other hand, there is no detectable anomaly at the both temperatures along the [100] and [110] directions in the basal plane. The high temperature paramagnetic susceptibility follows the Curie–Weiss law along and perpendicular to the c-axis. The effective magnetic moment estimated is 3.5 μ_B which is nearly equal to the theoretical value for a Pr^{3+} free ion. The paramagnetic Curie temperature is 9 K and $--7$ K along and perpendicular to the c-axis, respectively. The difference of 16 K is not so large comparing to those of family compounds PrM_2X_2 (M=Ru, Co, X=Si, Ge) which have a huge magnetic anisotropy [5, 6].

Magnetization as a function of applied magnetic field along the main symmetry axes at 2 K is shown in Fig. 2. Along the [100] and [110] directions in the basal plane, magnetization is linear and small, showing that these axes are the hard magnetization directions. Within the basal plane, magnetization is almost isotropic in a low field region.

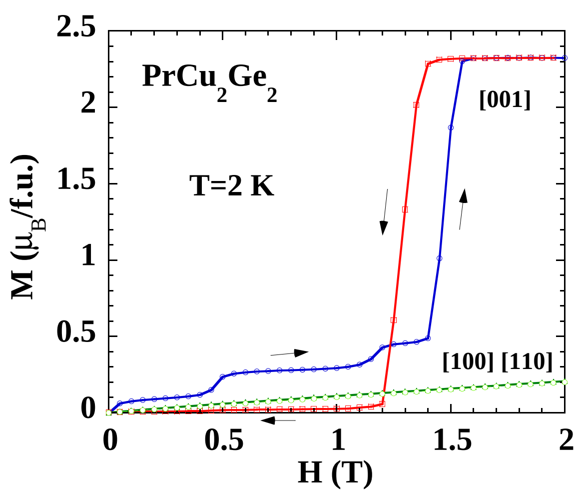

Fig. 2. Magnetization curves along the main symmetry axes at 2 K on the $PrCu_2Ge_2$ single crystal compound.

On the other hand, magnetization along the easy c-axis shows a very peculiar behavior: *an irreversible magnetization process*; in the ascending process, a four-step metamagnetic process appears. Magnetization increases rapidly around 0.2 T, 0.45 T, 1.15 T and 1.45 T, and reaches the saturation value of 2.34 μ_B with increasing field. Then in the descending process after saturation, it decreases rapidly around 1.3 T, crosses over one of the ascending process and reaches a very small value below 1.2 T. This peculiar irreversible process appears only in the virgin state (which means the first magnetization measurement reached the saturation after cooling down.) and has been never seen yet. The process in the second run becomes a one-step metamagnetic one similar to one of the descending process in the virgin state with small hysteresis; it becomes a reversible process. The irreversible process can be observed up to 3.5 K. Above 4 K, the process becomes reversible; an ascending process becomes a one-step one with a larger initial sloop than one of the hard magnetization curve. The descending process is similar to one of the ascending process. Here, a small hysteresis can be seen. The one-step process persists up to T_{N}. The temperature dependence of magnetic susceptibility changes corresponding to change of magnetization process. After a saturation process at a low temperature, the initial susceptibility becomes very small for low temperatures and recovers above about 4 K with increasing temperature. This dependence is in contrast to one of the virgin state (see Fig.1). This behavior indicates the existence of a magnetic phase transition around 4 K. In the four-step metamagnetic process, the magnetization of each plateau is 0.09 μ_{B}, 0.27 μ_{B}, 0.45 μ_{B} and 2.34 μ_{B}, corresponding to $1/26\mathrm{M_s}$, $3/26\mathrm{M_s}$, $5/26\mathrm{M_s}$ and $\mathrm{M_s}$(= the saturation moment), respectively. It is worth noting that the ratios have a common denominator (= 26), suggesting this metamagnetic process occurs by spin-flip transitions. The saturation moment is in good agreement with one of the previous neutron diffraction report [3,4]. It is, however, difficult to understand this process on the basis of a simple antiferromagnetic structure reported.

Although the origin of the irreversible process is unknown yet, orbital effects, quadrupolar effects may be responsible. Further study is now in progress.

REFERENCES

[1] E.V. Sampathakumaran, I. Das, R. Vijayaraghavan, *Solid State Commun.* **83**, 609 (1992).

[2] A. Szytula, J. Leciejewicz, *Handbook on the Physics and Chemistry of Rare Earths*, **12**, Elsevier, Amsterdam 1989, p. 133.

[3] A. Szytula, V. Bazela, J. Leciejewicz, *Solid State Commun.* **48**, 1053 (1983).

[4] V. Ivanov, M. Kolenda, J. Leciejewicz, N. Stusser, A. Szytula, *J. Alloy. Compd.* **234**, L4 (1996).

[5] T. Shigeoka, N. Iwata, H. Fujii, *J. Magn. Magn. Mater.* **104-107**, 1229 (1992).

[6] T. Shigeoka, H. Fujii, K. Yonenobu, K. Sugiyama, M. Dates, *J. Phys. Soc. Jpn.* **58**, 394 (1989).