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Magnetic and Electrical Properties of Pr_7Ni_3 Single Crystal

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Abstract. Magnetic and electrical properties of Pr_7Ni_3 have been studied using single crystals. Pr_7Ni_3 possesses an ferrimagnetic or canted state below $T_N = 4.2$ K and another phase transition has been found at $T_t = 2.2$ K. A large magnetic anisotropy was observed between *c*-axis and *c*-plane below T_N. At 1.9 K, the magnetization curve in the low magnetic field shows a ferromagnetic like behavior along the *c*-axis and antiferromagnetic one in the *c*-plane. However, magnetization at 70 kOe in the *c*-plane (1.27 μ_B/Pr) is larger than that along the *c*-axis (0.75 μ_B/Pr). That is, Pr_7Ni_3 has a small ferromagnetic component in the direction of *c*-axis, and magnetic moments tend to orient in the *c*-plane. Electrical resistivity shows metallic conduction and a small anomaly at T_t.

1. Introduction

The intermetallic compounds R_7Ni_3 (R = La, Ce, Pr, Nd) crystallize in the Th_7Fe_3-type hexagonal structure with the space group $P6_3mc$ in which the R atom occupies three non-equivalent sites [1, 2]. Several investigations have been carried out on these compounds. La_7Ni_3 behaves as a Pauli paramagnet and therefore Ni does not carry the magnetic moment [1]. Thus it is accepted that the *sd* shell of Ni is filled and magnetic properties originate from rare earth ions in the R_7Ni_3 compounds. The Ce_7Ni_3 is a heavy-fermion antiferromagnet with a geometrically frustrated structure under hydrostatic pressures and magnetic fields, and it undergoes two magnetic transitions at $T_{N1} = 1.9$ K and $T_{N2} = 0.7$ K [3]. Below T_{N1}, Ce_7Ni_3 has an incommensurate magnetic structure and transfers to another antiferromagnetic structure at T_{N2} [3-6]. The Nd_7Ni_3 orders magnetically at $T_R = 7.8$ and $T_C = 11.5$ and $T_N = 25$ K. Below T_R, a commensurate conical magnetic structure has been concluded with the propagation vector $\boldsymbol{Q} = (0\ 0\ 1/3)$. From T_R to T_C, Nd_7Ni_3 has an incommensurate helical structure, and above T_C, the antiferromagnetic state [7-9]. However, there seems to be no data for Pr_7Ni_3 except those for the paramagnetic properties of polycrystalline specimens above 77 K [1]. In this work, we have succeeded to grow Pr_7Ni_3 single crystals and performed magnetic and electrical measurements. In this report, we will discuss the magnetic and electrical properties of single crystalline Pr_7Ni_3.

2. Experimental

Polycrystalline ingots of Pr_7Ni_3 were prepared by arc-melting the constituent elements of 99.9% Pr and 99.993% Ni under a high-purity argon atmosphere. In this process, the ingots were turned over and re-melted several times to ensure homogeneity. Single crystals were obtained by remelting the ingots at 800 °C in an electric furnace and cooling slowly to room temperature. The powder X-ray diffraction analysis indicated that the samples were in single phase with Th_7Fe_3 type hexagonal structure and the lattice parameters a and c were in good agreement with those determined by Olcese[1]. The single crystals were formed into spheres and rectangular shape and annealed at 300 °C for 24 h in an evacuated quartz tube. The crystal orientation was determined by the back reflection Laue method. Measurements of magnetization M and magnetic susceptibility χ were carried out using a PPMS (Quantum Design) in the magnetic fields up to 70 kOe in the temperature range 1.9 - 300 K and a vibrating sample magnetometer in magnetic fields up to 17 kOe. The electrical resistivity ρ was measured by a conventional DC four-terminal method in the temperature range 1.8 - 300 K. Measurements of the specific heat C were carried out by a relaxation method using the PPMS in the temperature range 1.9 - 90K.

3. Results and Discussion

Figure 1 shows the magnetization curves for Pr_7Ni_3 at several temperatures in the c-plane and along the c-axis. At 1.9 K, the magnetization curves show a ferromagnetic like behavior along the c-axis indicating that Pr_7Ni_3 has a ferromagnetic component. No remanent magnetization was observed after removing the external magnetic field. However, although the magnetization value along the c-axis is larger than that in the c-plane in the low-field region, the magnetization

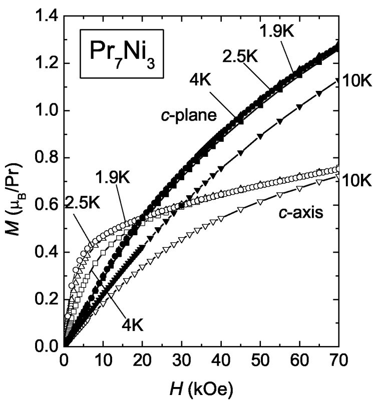

Figure 1. Magnetization curves for Pr_7Ni_3 at several temperatures in the c-plane (solid symbols) and along the c-axis (open symbols).

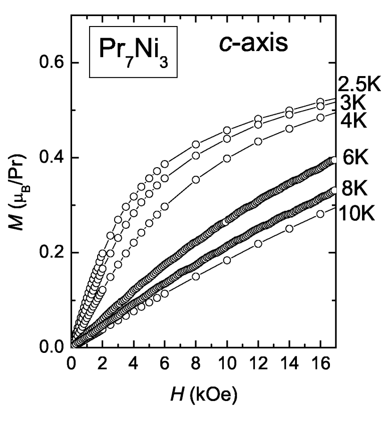

Figure 2. The low field magnetization curves for Pr_7Ni_3 at several temperatures along the c-axis.

value in the c-plane becomes larger than that along the c-axis above 20 kOe. This suggests that magnetic moment is easy to rotate in the c-plane. Magnetization value at 70 kOe is 0.75 μ_B/Pr and 1.27 μ_B/Pr along the c-axis and in the c-plane, respectively. These values are small compared to the Pr^{3+} free ion gJ value of 3.20 μ_B. Since the magnetization values at 70 kOe is smaller than the gJ value, some metamagnetic transitions may occur in the higher magnetic field. It should be noted that the ferromagnetic compound Pr_7Rh_3 which has the same crystal structure as Pr_7Ni_3 shows a low saturation magnetization value of 1.38 μ_B/Pr which is reduced by the crystal electric field (CEF) effect[10]. Therefore, the magnetization measurements in higher field is required in order to reveal the influence of the CEF effect and the metamagnetic transitions. The magnetization curves above 30 kOe at 2.5 and 4 K are similar to those at 1.9 K. However, at 10 K, the magnetization value along the c-axis is smaller than that in the c-plane in the all observation magnetic field range. Figure 2 shows the low field magnetization curves along the c-axis. The M-H curves up to 4 K indicate a large initial increase of magnetization but the curves at 6 K shows a paramagnetic like behavior. Thus, it is considered that a ferrimagnetic or antiferromagnetic state takes place between 4 K and 6 K. Figure 3 shows the magnetic

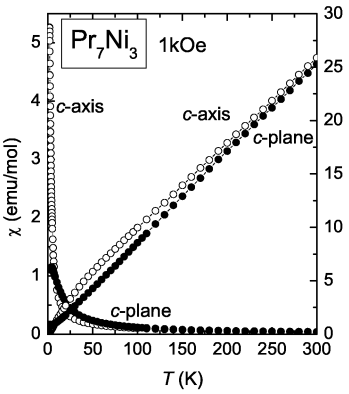

Figure 3. Magnetic susceptibility χ and reciprocal susceptibility χ^{-1} for Pr_7Ni_3 as a function of temperature.

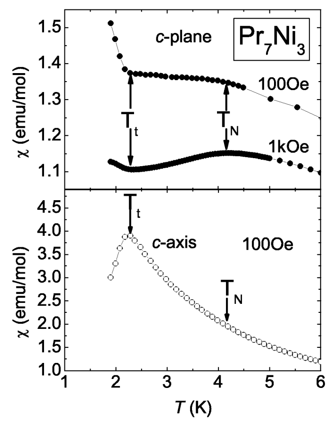

Figure 4. Magnetic susceptibility χ in low temperature at 100 Oe and 1 kOe.

susceptibility χ and the reciprocal susceptibility χ^{-1} as a function of temperature T. In both axes, paramagnetic susceptibility obeys Curie-Weiss law. Obtained effective magnetic moments $\mu_{\rm eff}$ are 3.67 and 3.78 $\mu_{\rm B}$/Pr in the c-plane and along the c-axis, respectively. The $\mu_{\rm eff}$ are in reasonable agreement with the theoretical value of 3.58 $\mu_{\rm B}$ for Pr^{3+}. This result advocates that the Ni ion is non-magnetic in Pr_7Ni_3 compound. The asymptotic Curie temperatures $\theta_{\rm P}$ are -1.35 and -24.3 K in the c-plane and along the c-axis, respectively. The χ in the c- plane is a little larger than that along the c-axis in high temperature region. However, a crossing of the χ - T curves is observed around 5 K; a large magnetic anisotropy is observed between c-axis and c-plane below this temperature. This can be originated by the easy axis change from the c-plane to the c-axis. Figure 4 shows the low-temperature χ-T curves measured at 100 Oe and 1 kOe in the c-plane and along the c-axis. In the c-plane, the χ-T curves show a shoulder (100 Oe) and a peak (1 kOe) at 4.2 K and anomalies at 2.2 K. On the other hand, χ shows a peak at 2.2 K along the c-axis. Thus, it is considered that Pr_7Ni_3 shows ferrimagnetic or canted phase below $T_{\rm N}$ = 4.2 K and the magnetic transition to another ferrimagnetic or canted state at $T_{\rm t}$ = 2.2 K. Figure 5 shows the electrical resistivity ρ for Pr_7Ni_3 as a function of temperature T. The inset shows the low-temperature part. The ρ-T curves show a metallic conduction below room temperature in the c-plane and along the c-axis, and a small anomaly at $T_{\rm t}$ in the c-plane; no anomaly is observed at $T_{\rm N}$ in both axes. The ρ in the c-plane is larger than that along the c-axis. Figure 6 shows the specific heat C of Pr_7Ni_3 as a function of temperature T. The inset shows the low-temperature part. Although a distinct peak is shown at $T_{\rm t}$, no anomaly is observed at $T_{\rm N}$. A small anomaly at 3.5 K can be seen, but the origin of this anomaly is not clear. As shown above, all magnetic, electrical and thermal data for Pr_7Ni_3 indicate that

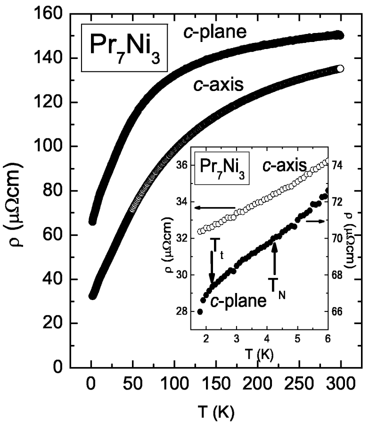

Figure 5. Electrical resistivity ρ for Pr_7Ni_3 as a function of temperature. The inset shows ρ-T curve in the low temperature.

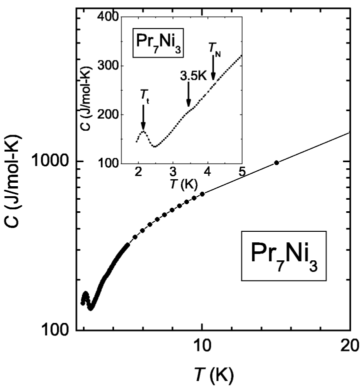

Figure 6. Specific heat C of Pr_7Ni_3 as a function of temperature. The inset shows the low temperature part.

the transition at T_N is obscure. It is unusual that the distinct anomaly is not observed at the transition temperature from paramagnetic to magnetically ordered state in the C - T, χ - T and ρ - T curves. In our preliminary results of the neutron diffraction study using single crystal, magnetic scattering peaks having the propagation vector $\boldsymbol{Q} = (0.5\ 0\ 0)$ were observed below T_N. Moreover, the magnetic scattering was observed at the nuclear position at (100) below T_t. Thus, a long range antiferromagnetic structure exists below T_N; at least low temperature phase has a ferromagnetic component along the c-axis below T_t. Since the Pr ion occupies three non-equivalent crystallographic sites in Pr_7Ni_3, the some of the Pr ions may not order at T_N but at T_t.

In conclusion, we have investigated the magnetic and electric properties of Pr_7Ni_3 using single crystals. Two ferrimagnetic or canted states were found below $T_t = 2.2$ K and $T_N = 4.2$ K. A small ferromagnetic component can exist along the c-axis in these two ordered phase. However, the magnetic phase transition at T_N is not clear. For further investigations, the higher-field magnetization measurements and the more detailed neutron diffraction studies using a single crystal are now in progress.

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