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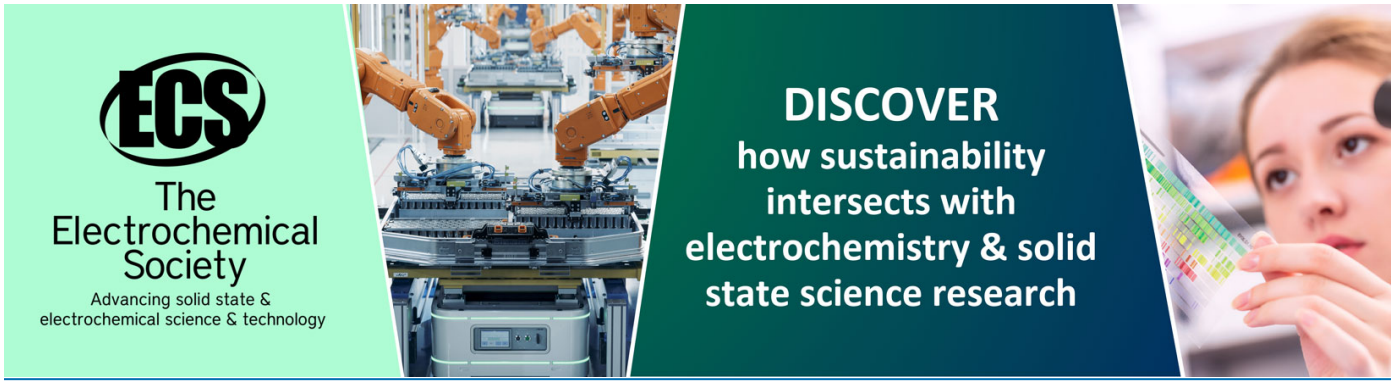

Journal of Physics: Conference Series **592** (2015) 012044 doi:10.1088/1742-6596/592/1/012044

Electrical resistivity and thermopower of $Y_{1-x}Pr_xCo_2$ Compounds

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Abstract. Electrical resistivity ρ and thermopower S of the pseudo-binary compounds of $Y_{1-x}Pr_xCo_2$ have been measured in the temperature range between 2 and 300 K under magnetic fields up to 10 T, together with the pressure measurements of ρ and S in $Y_{0.4}Pr_{0.6}Co_2$. The Curie temperature decreases with decreasing x, and vanishes at the critical composition $x_c \approx 0.4$, where the residual resistivity attains a maximum value. The Curie temperature and the residual resistivity of $Y_{0.4}Pr_{0.6}Co_2$ show the same pressure dependence as those of the heavy-rare-earth based compounds. These behaviors of ρ and S indicate the inhomogeneous distribution of the Co 3d magnetization. The magnetoresistance of the light-rare earth $Y_{1-x}Pr_xCo_2$ system is negative in the whole range of x, except for $x = 0$ and 1, which is a characteristic behavior related with magnetic state and magnitude of the effective field acting on the Co 3d subsystem.

1. Introduction

The pseudo-binary $Y_{1-x}R_xCo_2$ (R stays for rare-earth element) compounds belong to the RCo_2 family with a cubic Laves phase $MgCu_2$-type structure [1]. The characteristic feature of their electronic structure is responsible for their magnetic instability and unusual transport properties. When a magnetic R is substituted by non-magnetic Y, the Curie temperature T_C of the $Y_{1-x}R_xCo_2$ system decreases with decreasing x and vanishes at a critical composition x_c. It is reported that numerous observations of anomalous magnetic and transport properties near the phase boundary x_c between the paramagnetic and magnetically ordered states in $Y_{1-x}R^HCo_2$ compounds with heavy-rare-earths R^H [2, 3, 4, 5, 6]. It is found that the anomalous behaviors of low-temperature conduction with the composition x, magnetic field B, and pressure P are strongly connected with a non-uniform magnetization of the Co subsystem, which is induced by a spatial fluctuating exchange field owing to the structural disorder of the R^H subsystem in the $Y_{1-x}R^H_xCo_2$ system [5]. Recently, the similar behaviors of the magnetic and transport properties have been observed in the light-rare-earth $Y_{1-x}Nd_xCo_2$ system [7].

In order to clarify the magnetic and transport properties in the light-rare-earth system of $Y_{1-x}Pr_xCo_2$, the measurements of the electrical resistivity ρ and thermopower S have been

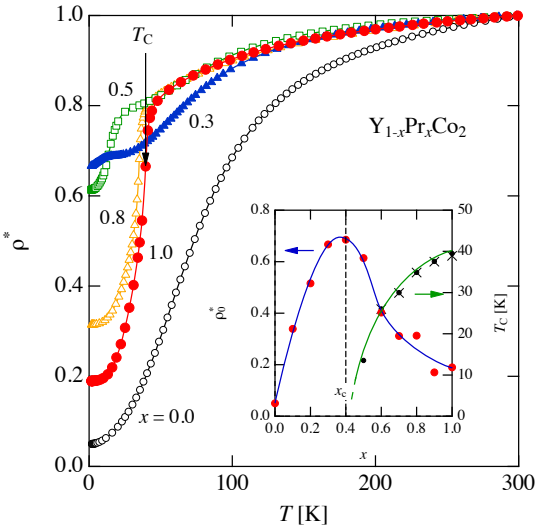

Figure 1. Temperature dependence of the electrical resistivity $\rho^\star$ of $Y_{1-x}Pr_xCo_2$. The arrow indicates the Curie temperature T_C of $PrCo_2$. The inset shows the composition x dependence of the Curie temperature T_C and the residual resistivity $\rho_0^\star$ of $Y_{1-x}Pr_xCo_2$. The symbols of • and × are the data obtained from $\rho(T)$ and $S(T)$ curves, respectively.

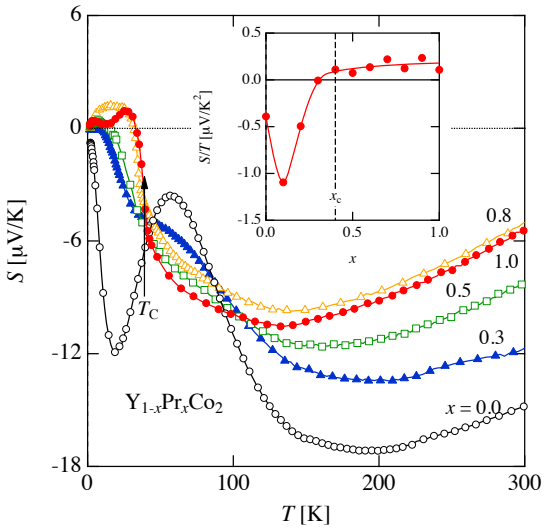

Figure 2. Temperature dependence of thermopower S of $Y_{1-x}Pr_xCo_2$. The arrow indicates the Curie temperature T_C of $PrCo_2$. The inset shows the composition x dependence of the low-temperature thermopower gradient S/T of $Y_{1-x}Pr_xCo_2$. The vertical dashed line indicates the phase boundary between the paramagnetic and the magnetically ordered states.

performed at temperatures from 2 to 300 K under pressures up to 3 GPa.

2. Experimental procedures

Polycrystalline samples of $Y_{1-x}Pr_xCo_2$ were prepared by arc-melting their constituents with an ratio R:Co of 1:1.93, which was chosen to avoid the formation of the magnetic impurity RCo_3 phase. The cubic Laves phase structure was confirmed by the powder X-ray diffraction measurement. The electrical resistivity ρ and thermopower S were measured by using a standard four-probe dc method and a differential method with the seesaw heating procedure [8], respectively. ρ and S were measured simultaneously at temperatures from 2 to 300 K in magnetic fields up to 10 T. A piston-cylinder-type pressure-cell with Daphne 7373 oil as the pressure transmitting medium was utilized for the measurements of ρ and S under pressures up to 3 GPa. The direction of the external magnetic field was parallel to both the current and the temperature gradient in order to reduce the influences of additional galvanomagnetic and thermomagnetic effects.

3. Results and Discussion

RCo_2 compounds are well known for their poor mechanical properties due to the presence of pores and micro-cracks. Throughout this paper, we use the normalized resistivity, $\rho^\star = \rho(T)/\rho(300)$, to eliminate the geometrical uncertainties for the electrical resistivity of the RCo_2 system. Accordingly, the residual resistivity $\rho_0^\star$ is determined as $\rho_0^\star = \rho(2)/\rho(300)$. Figure 1 shows the normalized electrical resistivity $\rho^\star$ of the selected $Y_{1-x}Pr_xCo_2$ compounds in the temperature range from 2 to 300 K. $\rho^\star$ decreases with decreasing temperature at higher temperature region,

and shows a sharp drop at the Curie temperature $T_{\rm C}$ in the magnetic compounds. We determined $T_{\rm C}$ as a temperature where $\mathrm{d}\rho/\mathrm{d}T$ takes maximum value. As seen in Fig. 1, $T_{\rm C}$ and $\rho^{\star}(T)$ at low temperatures strongly depend on the Pr composition x. The composition x dependences of the Curie temperature $T_{\rm C}$, inferred from $\rho(T)$ curve, and the residual resistivity $\rho_0^{\star}$ are shown in the inset of Fig. 1. $T_{\rm C}$ decreases with decreasing x and vanishes at a critical composition $x_{\rm c} \approx 0.4$, which is the phase boundary between the paramagnetic phase and the magnetically ordered state. $\rho_0^{\star}$ increases with increasing x, attains a maximum at $x \approx x_{\rm c}$, and decreases with further increase of x. These behaviors of $T_{\rm C}$ and $\rho_0^{\star}$ in the $Y_{1-x}Pr_xCo_2$ system are qualitatively similar to those found in the heavy-rare-earth based system [5, 9] and a light-rare-earth based $Y_{1-x}Nd_xCo_2$ system [7]. As reported previously, these anomalous composition x dependences of $T_{\rm C}$ and $\rho_0^{\star}$ in the vicinity of $x_{\rm c}$ are attributed to the distribution of the inhomogeneous 3d magnetization in the Co subsystem [5, 10].

Figure 2 depicts the temperature dependence of the thermopower S of the selected $Y_{1-x}Pr_xCo_2$ compounds at temperatures from 2 to 300 K. The feature of S shows large variations with the composition x at low temperatures. In the magnetic compounds with $x > 0.4$, $S(T)$ shows a sudden increase at $T_{\rm C}$ with decreasing temperature. The obtained $T_{\rm C}$ is coincident with the data inferred from $\rho(T)$, as shown in the inset of Fig. 1. At low temperatures, S for metallic conductors is expressed by Mott's expression:

$$S = \frac{\pi^2 k_{\rm B}^2}{3e} T \left[\frac{1}{\sigma} \frac{\partial \sigma}{\partial \varepsilon} \right]_{\varepsilon=\varepsilon_{\rm F}},$$

where e, $k_{\rm B}$ and σ are the electronic charge, the Boltzmann constant and the spectral electronic conductivity, respectively. This formula predicts a linear temperature dependence for S in low temperature limit. The inset of Fig. 2 indicates the composition x dependence of the low-temperature thermopower gradient S/T. The vertical dashed line indicates the critical composition $x_{\rm c}$. S/T for the compounds of $x > x_{\rm c}$ shows the almost x independent behavior. In the range of $x < x_{\rm c}$, S/T shows a large composition variation, and changes its sign from positive to negative at a composition just below $x_{\rm c}$. It has been shown that due to dominating s-d scattering in RCo_2 series, $\sigma \propto 1/\ N_{\rm d}(\varepsilon)$ [11]. where $N_{\rm d}(\varepsilon)$ is Co 3d electronic state density (DOS). Thus, S/T is expected to relate with $N_{\rm d}(\varepsilon)$ as follows:

$$S/T \propto \left[\frac{1}{N_{\rm d}(\varepsilon)} \frac{\partial N_{\rm d}(\varepsilon)}{\partial \varepsilon} \right]_{\varepsilon=\varepsilon_{\rm F}}.$$

Then, the low-temperature S/T in the ferromagnetic state indicates that the electronic state of the Co 3d electron subsystem near the Fermi level is almost constant in the composition range of $x > x_{\rm c}$.

The high pressure measurements of ρ and S for the ferromagnetic compound $Y_{0.4}Pr_{0.6}Co_2$ are carried out under pressures up to 3 GPa. Figure 3 shows the temperature dependence of ρ of $Y_{0.4}Pr_{0.6}Co_2$ at temperatures from 2 to 300 K. ρ increases in magnitude with increasing pressure in the whole temperature range. It seems that $\rho(T)$ shows the almost the same temperature dependence under all applied pressures. The residual resistivity, however, increases with increasing pressure. As shown in Fig. 4. $S(T)$ shift toward lower ones with increasing pressure, indicating the similar temperature behavior, except for the low temperature region below about 10 K. A broad minimum in $S(T)$ curve is observed at $T_{\rm min} \approx 150$ K. $T_{\rm min}$ increases with increasing pressure, which means a broadening of the Co-3d DOS [11, 12]. The pressure dependence of the low-temperature gradient of thermopower S/T is shown in the inset of Fig. 4. S/T decreases with increasing pressure, and changes its sign from positive to negative around $P = 2$ GPa. From the same analogy as the x dependence of S/T shown in the inset of Fig. 2,

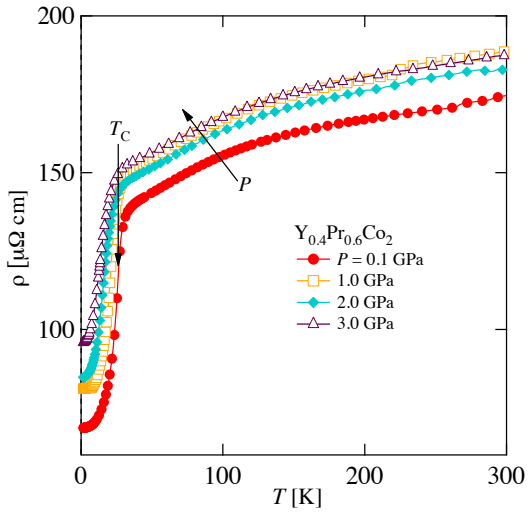

Figure 3. Temperature dependence of ρ_0 of $Y_{0.4}Pr_{0.6}Co_2$ under pressures up to 3 GPa.

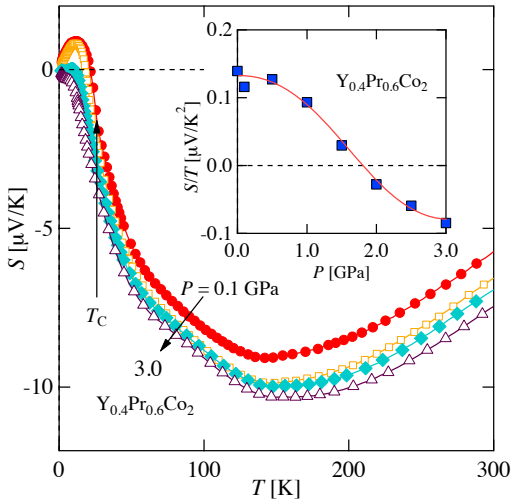

Figure 4. Temperature dependence of thermopower S of $Y_{0.4}Pr_{0.6}Co_2$ under pressures up to 3 GPa. The inset shows the pressure dependence of S/T of $Y_{0.4}Pr_{0.6}Co_2$.

it is considered that magnetic ground state changes from ferromagnetic to paramagnetic at $P \approx 2$ GPa.

In our theoretical model, the low-temperature transport properties of the $Y_{1-x}R_xCo_2$ system are related to the conduction electron scattering due to static magnetic disorder in the itinerant Co 3d electron subsystem [5, 10]. At low temperatures, the total resistivity ρ of the $Y_{1-x}Pr_xCo_2$ system can be expressed as

$$\rho = \rho_0 + \rho_{4f} + \rho_{3d} + \rho_m.$$

ρ_0 is the conventional residual resistivity. ρ_{4f} and ρ_{3d} are the resistivity due to fluctuations of 4f and 3d magnetic moments, respectively. The last term means a conduction electron scattering due to the non-uniform magnetization of the Co 3d subsystem, depending on the volume fraction y of the high-magnetization in the Co-subsystem as $\rho_m \propto y(1-y)$. The application of pressure, in a first approximation, has the same effect on the low-temperature conductions as the substitution Y for Pr, i.e., decrease of Pr composition x, accordingly decrease of y. Figure 5 shows the pressure dependences of the residual resistivity ρ_0 and the Curie temperature T_C of $Y_{0.4}Pr_{0.6}Co_2$, which is the compound with $x > x_c$. T_C decreases, and ρ_0 increases linearly with increasing pressure. These pressure variations of T_C and ρ_0 are in agreement with the theoretical prediction for the compounds of $x > x_c$ [5, 7].

On the other hand, the effect of magnetic field on the low-temperature conductions of the $Y_{1-x}Pr_xCo_2$ system should be the same as that of an increase of x (y). Then, a positive magnetoresistance (MR) for the compounds of $x < x_c$ and negative for $x > x_c$ can be expected, if the effect of magnetic field on ρ_{4f} and ρ_{3d} can be ignored. Figure 6 shows the x dependence of MR at $T = 2$ K, where MR$= \Delta\rho/\rho(0)$ ($\Delta\rho = \rho(10) - \rho(0)$), of the $Y_{1-x}Pr_xCo_2$ system. As seen in Fig. 6, MR is negative in the whole range of x, except the pure compounds of YCo_2 and $PrCo_2$. The similar behavior of MR was observed in that of a light-rare-earth $Y_{1-x}Nd_xCo_2$ system [13]. The different behavior of MR between the heavy- and light-rare-earths systems might be related with the differences of magnetic structure and of magnitude of the effective field acting on the Co 3d electrons.

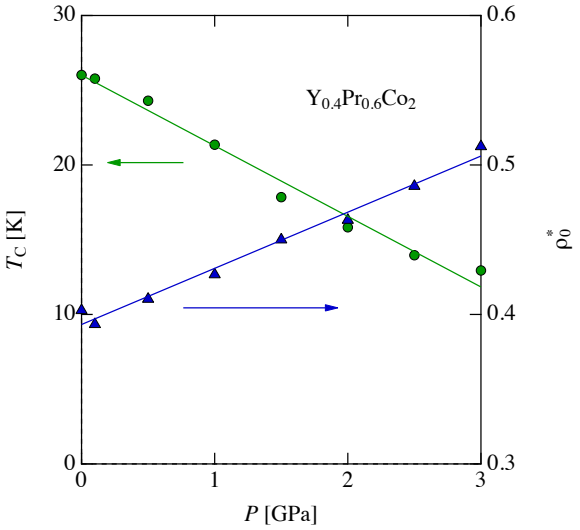

Figure 5. Pressure dependence of T_C and ρ_0 of $Y_{0.4}Pr_{0.6}Co_2$.

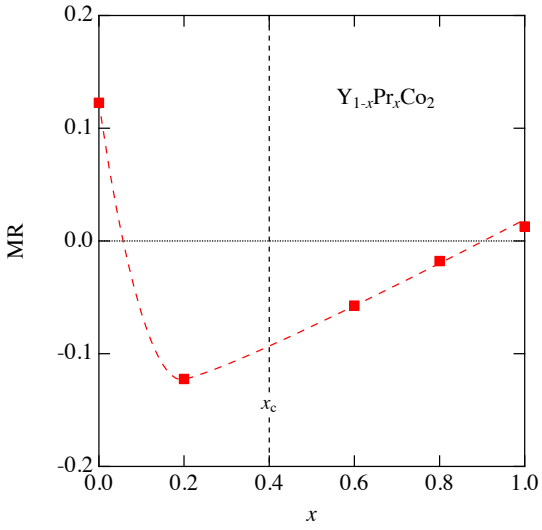

Figure 6. Composition x dependence of the magnetoresistance of $Y_{1-x}Pr_xCo_2$. The vertical dashed line indicates the critical composition x_c.

4. Summary

We have measured the electrical resistivity and thermopower of the light-rare-earth based $Y_{1-x}Pr_xCo_2$ system at temperatures from 2 to 300 K under magnetic fields up to 10 T, together with the pressure measurements of the electrical resistivity and thermopower in $Y_{0.4}Pr_{0.6}Co_2$. The Curie temperature and the residual resistivity show the same behaviors against the substitution Y for Pr and the application of pressure for the magnetically ordered compound as those of the heavy-rare-earth based compounds, revealing the inhomogeneous distribution of the Co 3d magnetization. This inhomogeneous distribution of the static Co-3d magnetization plays a dominant role for the low-temperature electron conduction. The magnetoresistance shows the negative value in the whole range of x, except for $x = 0$ and 1. It is plausible that a characteristic behavior of magnetoresistance of the $Y_{1-x}Pr_xCo_2$ system is related to magnetic state and magnitude of the effective field acting on the Co 3d subsystem.

Acknowledgment

This work was supported by Grants-in-Aid for the Promotion of Science from the Uruma Fund.

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