Thermoelectric Power of PrMg\textsubscript{3}

Yosikazu Isikawa, Kazuya Somiya, Huruto Koyanagi, Toshio Mizushima, Tomohiko Kuwai, Takashi Tayama
Department of Physics, University of Toyama, Toyama 930-8555, Japan
E-mail: isikawa@sci.u-toyama.ac.jp

Abstract. PrMg\textsubscript{3} is supposed to be one of the strongly correlated electron systems originated from the hybridization between the Pr 4f and conduction electrons, because the gigantic electronic specific heat coefficient $C/T$ was observed at low temperatures. However, a typical behaviour of $-\ln T$ dependence was not observed in the temperature dependence of the electrical resistivity. The thermoelectric power $S$ is a powerful tool to investigate the density of states at the Fermi energy. We measured carefully the thermoelectric power of PrMg\textsubscript{3} in the temperature range between 2 and 300 K. $S$ is extremely small, ranged within $\pm 1\ \mu V/K$ over the whole temperature. The value of $S/T$ at low temperature limit was also significantly smaller than expected from the specific heat results. We therefore conclude that the density of state at the Fermi level is not enhanced in PrMg\textsubscript{3}.

1. Introduction
PrMg\textsubscript{3} is supposed to be one of the strongly correlated electron systems originated from the hybridization between the Pr 4f and the conduction electrons [1]. Typical experimental results obtained so far are as followed. The gigantic electronic specific heat coefficient $C/T$ was observed at low temperatures, and the value of $C/T$ grows up to 2.8 J/mol K\textsuperscript{2} at 0.54 K. No trace of cooperative phase transitions was observed both in the temperature dependence of $C/T$ and in the magnetic susceptibility $\chi(T)$ [1]. At low temperatures, $\chi(T)$ shows mostly the Van Vleck-type $T$ independent behaviour accompanied with a small contribution of $-\log T$. The energy scheme due to the crystalline electric field (CEF) in the cubic crystal structure of PrMg\textsubscript{3} has been determined by the inelastic neutron scattering experiment [2, 3]; the ground state is nonmagnetic doublet $\Gamma_3$ and the excited states are triplet $\Gamma_4$, singlet $\Gamma_1$ and triplet $\Gamma_5$ located at 56 K, 135 K and 183 K above $\Gamma_3$, respectively. The doublet ground state is consistent with the result inferred from the entropy below 5 K. Recently, the magnetization has been measured down to 60 mK by Morie \textit{et al.} [4]. According to them, the $\chi(T)$ continuously increases as $-\log T$ or $-\sqrt{T}$ on cooling below 3 K, and they suggested this unusual increase of $\chi(T)$ is originated from the presence of the hybridization effect between the Pr 4f and conduction electrons. More recently, Yoshida \textit{et al.} [5] observed a small peak around 0.1 K in the $\chi(T)$ curve measured down to 0.3 mK. They proposed that the peak following the $-\log T$ dependence in $\chi(T)$ is due to the quenching of the multipole degree of freedom in $\Gamma_3$.

An important issue to be solved in PrMg\textsubscript{3} is how the ground-state doublet, the degree of freedom, has disappeared at the absolute zero temperature. The entropy should be zero at 0 K, which is enforced by the third law of the thermodynamics. Magnetic order is excluded as a mechanism because the ground state is nonmagnetic. Quadrupole order is one possibility, but
no strict anomaly due to the quadrupole order has been observed so far except a small peak around 0.1 K appeared in $\chi(T)$. Another possibility to quench the degree of freedom is the hybridization between the Pr 4f and conduction electrons as suggested by Tanida et al. [1] and Morie et al. [4].

The thermoelectric power $S$ is a powerful tool to investigate the density of state of conduction electrons at the Fermi energy. Thermoelectric power coefficient $S/T$ has been shown to be proportional to the $C/T$ in the typical heavy-Fermion compounds at low temperature limit [6, 7, 8]. Thus, we measured $S$ of PrMg$_3$ because $S/T$ is expected to be significantly enhanced at low temperature limit if the extremely enhanced $C/T$ of PrMg$_3$ is originated from the hybridization between the Pr 4f and conduction electrons.

2. Experiment procedure

Polycrystalline sample of PrMg$_3$ was prepared by arc melting in an argon gas atmosphere. Crystal structure of the cubic Fe$_3$Al type was confirmed by the X-ray powder diffraction pattern. As shown in Fig. 1, there is no trace of impurity phases. The thermoelectric power was carefully measured from 2 to 300 K by detecting small electric potential $\Delta V$ induced by small temperature gradient $\Delta T$ across a sample rod. The magnetization $M$, the magnetic susceptibility $\chi$, the electrical resistivity $\rho$ and the specific heat $C$ were also measured to compare to those data previously reported. $M$ and $\chi$ were measured by using a SQUID magnetometer, $\rho$ was measured by a conventional four terminal DC method, and $C$ was measured by using the Physical Properties Measurement System (PPMS).

![Figure 1](image1.png)  \hspace{1cm}  \textbf{Figure 1.} X-ray powder diffraction pattern of PrMg$_3$. (a) Experimental result and (b) simulation.

![Figure 2](image2.png)  \hspace{1cm}  \textbf{Figure 2.} Magnetization curve of PrMg$_3$ at 2 K.

3. Experimental results and discussion

Figure 2 shows the magnetic field dependence of $M(H)$ at 2 K. As shown in the figure, $M$ is paramagnetic and passes through the origin, indicating that there is no magnetic impurity such as PrMg$_2$. PrMg$_2$ is ferromagnetic with $T_c$ of 9.6 K [9]. Figure 3 shows $\chi(T)$ under $H$ of 1 T. Below 10 K, $\chi(T)$ is nearly independent on temperature, which is consistent with that the ground state of PrMg$_3$ is a non-magnetic $\Gamma_3$, although a small contribution of $-\log T$ dependence is superposed in $\chi(T)$. Both $M(H)$ and $\chi(T)$ coincide quantitatively with those reported so far [1, 4]. $\rho(T)$ is shown in Fig. 4. The temperature dependence of $\rho(T)$ is similar to those of
normal metals without Kondo effects such as $-\log T$ dependence. The residual resistivity is 17 $\mu\Omega\text{cm}$, which is somewhat larger than the value reported [10].

The temperature dependence of $C(T)/T$ and $C(T)$ are shown in Fig. 5. $C(T)$ has a broad peak with a value of 1.8 J/mol K at 1.0 K, and $C(T)/T$ increases with decreasing temperature and goes up to 2.7 J/mol K$^2$ at 0.52 K. Both $C/T$ and $C$ are quantitatively agreement with those reported previously [1].

As mentioned before, the thermoelectric power $S$ is powerful tool to know whether the conduction electrons at the Fermi energy hybridize with the 4f electrons, in consequence, whether the mass of the conduction electrons increases. The temperature dependence of $S$ is shown in Fig. 5. In the whole temperatures, $S$ is extremely small, and similar to the result reported by Sakurai et al. [7]. The characteristic behaviours of $S$ are summarized as follows. (i) $S(T)$ is approximately linear in temperature through the origin above 120 K, and the slope $S/T$ is
\(-2.7 \times 10^{-3} \ \mu V/K^2\). (ii) A negative peak appears around 50 K, which is presumably due to the phonon-drag effect. (iii) Below 20 K, the sign of \(S\) changes from negative to positive, and the value of \(S\) is \(+0.1 \ \mu V/K\) with the error of \(\pm 0.1 \ \mu V/K\). No enhancement associated with, so-called, the dilute-Kondo effect, was detected at low temperatures, which is in contrast to \(S\) of the noble metals with magnetic impurities, for example, Cu and Au [11], and also in contrast to \(S\) of the concentrated Kondo compound, i.e., Ce-based and Yb-based compounds.

The thermoelectric power is given by the Mott’s equation, 

\[
S = -\frac{\pi^2 k_B T}{3e} \left( \frac{\partial \ln \sigma(\epsilon)}{\partial \epsilon} \right)_{E_F},
\]

where \(\sigma(\epsilon) = \tau(\epsilon)D(\epsilon)\), and \(D(\epsilon)\) is the density of state of conduction electrons, and the other parameters are conventional. The heavy-fermion compounds have, in general, large values of both \(C/T\) and \(S/T\), because the 4f electrons hybridize with the conduction electrons, thus, the density of state at \(E_F\) is enhanced by this hybridization. Behnia, Jaccard and Flouquet (BJF) derived the relationship between \(S/T\) and \(C/T\) at low temperature limit as

\[
S/T (\mu V/K^2) \approx 10 C/T (J/mol K^2)
\]

based on a simple model, and they succeeded to summarize many data of heavy-fermion compounds using this relationship [6]. According to this relationship, \(S/T\) of \(\text{PrMg}_3\) is predicted to be about 28 \(\mu V/K^2\). However, \(S/T\) at low temperatures is significantly smaller than predicted from the \(C/T\) of \(\text{PrMg}_3\). We, therefore, conclude that the Pr 4f electrons in \(\text{PrMg}_3\) do not hybridize with the conduction electrons at \(E_F\).

We can find similar examples in other Pr-based compounds, for example, \(\text{PrInCu}_2\), \(\text{PrInAg}_2\), and \(\text{PrPd}_3\) [12, 13, 14]. Former two compounds, Heusler compounds, have a \(\Gamma_3\) ground state of non-magnetic doublet. These compounds show a huge \(C/T\) at low temperatures without any phase transition. In contrast, \(S/T\) are not enhanced at low temperatures. Moreover, the Kondo-like behaviour of \(-\log T\) dependence was not observed in \(\rho(T)\). We rather suggest, instead of the hybridization or heavy fermion as an origin of the huge \(C/T\), a possibility of the quadrupole fluctuations of 4f electrons discussed in ref. [13] or the lattice instability originated from the peculiarity of the cubic crystal structure. Local lattice distortions or randomness of the atom positions are presumably important to understand the huge \(C/T\) of \(\text{PrMg}_3\).

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