Thermopower measurement under high pressure using “seesaw heating method”

M Hedo¹, D Nakamura¹, Y Takaesu¹,², T Fujiwara², K Uchima³, K Yagasaki¹ and T Nakama¹
¹Faculty of Science, University of the Ryukyus, Nishihara, Okinawa, 903-0213, Japan
²Faculty of Science, Yamaguchi University, Yamaguchi, Yamaguchi, 753-8512, Japan
³Okinawa Christian Junior College, Nishihara, Okinawa, 903-0207, Japan
E-mail: hedo@phys.u-ryukyu.ac.jp

Abstract. We have developed a set-up with modified “seesaw heating method” for the thermopower measurement under pressures \( P \) up to 3 GPa at the temperature range between 2 K and 300 K. By using this set-up, the thermopower and electrical resistivity of the single crystalline YbMn\(_2\)Ge\(_2\) under high pressure were measured with enough accuracy. \( S(T) \) curve shows the characteristic feature at the magnetic transition in all pressure range, while no evidence of the magnetic phase transition is observed in \( \rho(T) \) at \( P > 1.25 \) GPa. The measurement results indicate that the simultaneous measurement of the thermopower and electrical resistivity is a useful tool to study the pressure-induced phase transitions.

1. Introduction
In the heavy-fermion system, the temperature coefficient of thermopower \( S/T \) at low temperatures has a universal correlation with the coefficient of the electronic specific heat \( \gamma \), \( S/T \propto \gamma \) [1], which is similar to the Kadowaki-Woods relation of \( A \propto \gamma^2 \), here \( A \) is the temperature coefficient of resistivity \( (\rho = \rho_0 + AT^2) \) [2]. The values of \( S/T \), \( \gamma \) and \( A \) are enhanced due to a large electronic density of states at the Fermi level. Thermopower \( S \) is an important transport coefficient to investigate the properties of the conductive materials because of its high sensitivity against the electronic structure and the scattering mechanism, which means that \( S \) is a good probe to observe susceptible changes of the electronic structure in the vicinity of the Fermi level under several physical conditions. Nowadays the measurements of \( S \) have been carried out extensively to study the ground state of the strongly correlated electrons systems [3, 4]. Few measurements of \( S \) under high pressure, however, have been performed because of the problems of the measurement stability and the accuracy.

We reported the system for thermopower measurement with a unique DC heating procedure called “seesaw heating method” in previous paper [5]. The driving force (temperature gradient, in this case) reversal to reduce spurious voltages effectively and to obtain accurate experimental data is conventional technique in measurements of transport properties, such as electrical resistivity and Hall effect. This method can be adopted easily to the measurement under multiple environments of high magnetic field and high pressure, due to the flexibility of the accommodation time and the heating power according with heat capacity of sample and of environment, which depend strongly on temperature.
In this paper, we demonstrate our developing set-up for the simultaneous measurements for $S$ and $\rho$ under high pressure at the temperature range from 2 K and 300 K. And we present the results on $S$ and $\rho$ of YbMn$_2$Ge$_2$ under pressures up to 2 GPa, as a test material.

2. Set-up with modified “Seesaw heating method” under high pressure

The thermopower measurement technique called “seesaw heating method” using two gradient heaters on both sides of the sample has been developed for the high sensitive and stable measurement under a strong magnetic field [5]. Due to the limitation of the sample space, we employed the modified “seesaw heating method” with one gradient heater, installed on a glass epoxy sample holder plate. Figure 1 shows the schematic diagram of the sample holder plate. The gradient heater (1) and the sample (2) are glued on the thin Cu layer (3) on a glass epoxy plate (4), and the other side of plate is fixed on the arch-like Cu wire (5), connected to the plug (6) in order to establish steady state of the heat flow.

Chromel-constantan thermocouples were used as the probes for thermopower measurement because of the small temperature dependence of the pressure effect [6]. The junctions of the thermocouples were made by spot welding, and glued directly to the sample by using Ag paste. The cold ends of the thermocouples are wrapped around the arch-like Cu wire. Thus the temperature differences between the junctions and the fluctuation of the sample temperature are negligible during a thermopower measurement due to a large heat capacity of the pressure cell.

Figure 2 shows the time dependences of thermal voltages $V_{\text{chx}}$ and $V_{\text{cox}}$ in the thermocouple - sample circuit at room temperature. The modulation time is 100 sec for example in this figure.
Figure 3. Temperature dependences of $S$ and $\rho$ of YbMn$_2$Ge$_2$ under several pressures. The arrows indicate the magnetic transition temperature $T_{N2}$ at ambient pressure.

Figure 4. Pressure dependences of the magnetic transition temperatures of $T_{N1}$ and $T_{N2}$. The symbols of $\times$ and $\triangle$ are obtained from $S(T)$ and $\rho(T)$, respectively. The values of $T_{N1}(\square)$ and $T_{N2}(\bigcirc)$ are taken from the literature [9].

thermoresistor put on outside of high pressure cell, and the sample temperature $T_x$ was obtained as $T_x = T_0 + \Delta T/2$.

The accuracy of our high pressure set-up was checked by Ni and TbCo$_2$ as the reference materials. The temperature dependences of thermopower of Ni and TbCo$_2$ show almost the same behavior. The differences between our data and literatures [7, 8] are within 2 $\mu$V/K, which may be attributed to the difference of the sample size (in other words, the difference of the distance between the measuring probes), which is about 10 times smaller (shorter) than that in usual measurements. Therefore we concluded that the thermopower under high pressure can be obtained with enough precision by means of this method. In addition, for the resistivity measurement, a pair of Cu wires as current lead was attached to the both edges of the sample, and a thermocouple wire was utilized for the voltage measurement of the resistivity. Then the electrical resistivity and thermopower can be measured simultaneously in the temperature range between 1.5 K and 300 K under high pressure.

3. Measurement results

As an example, we have measured the electrical resistivity $\rho$ and thermopower $S$ of YbMn$_2$Ge$_2$ under pressures up to 2 GPa. YbMn$_2$Ge$_2$ is an antiferromagnet (AFM-I phase) with the Neel temperature of $T_{N1} \approx$400 K, and indicates an additional magnetic transition at $T_{N2} \approx$165 K of AFM-II phase. It was reported that Yb ion in this compound is in a mixed valence state between divalent and trivalent at ambient pressure, and shows the pressure-induced valence transition at the critical pressure $P_c=1.25$ GPa [9, 10]. A single crystalline sample of YbMn$_2$Ge$_2$ was prepared by In flux method [9]. We used a hybrid piston cylinder type high pressure cell up to 3 GPa [11], with Daphne 7373 oil as a pressure transmitting medium [12], for the measurements of $S$ and $\rho$. The directions of the current and the temperature gradient were made perpendicular
to the c-axis.

Figure 3 shows the temperature dependences of $S$ and $\rho$ of YbMn$_2$Ge$_2$ at the temperature range between 1.5 K and 300 K under pressures up to 2 GPa. At ambient pressure, $S$ and $\rho$ show a kink at $T \approx 165$ K, indicated by arrows in Fig. 3, which is corresponding to the lower magnetic phase transition temperature of $T_{N2}$. The temperature dependences of $S$ and $\rho$ change their feature at the critical pressure $P_c$. Although $S(T)$ curve indicates the characteristic behavior at the magnetic transition temperature in the whole pressure range, no clear evidence of the magnetic transition between AFM-III and AFM-IV phases is observed in $\rho(T)$ at $P > P_c$. The magnetic transition temperatures of $T_{S2}^N$ and $T_{R2}^N$ are determined as the temperatures where the temperature derivatives of $S$ and $\rho$ take maxima, respectively. As shown in Fig. 4, the obtained $T_{S2}^N$ and $T_{R2}^N$ are in good agreement with the results in ref. [9, 13].

In summary, we have developed the set-up for the simultaneous measurements of thermopower and electrical resistivity under pressures up to 3 GPa in the temperature range between 1.5 K and 300 K. We have measured, as an example, the electrical resistivity and thermopower of YbMn$_2$Ge$_2$. The magnetic transition temperature of YbMn$_2$Ge$_2$ can be obtained from $S(T)$ curve in the whole pressure range, while no evidence of the magnetic transition was detected in $\rho(T)$ at $P > P_c$. These results indicate that the simultaneous measurement of thermopower and electrical resistivity is a useful tools to understand the change of the electronic structure near the Fermi level.

**Acknowledgements**

Part of this work was supported by a Grant-in-Aid for Young Scientists (B) (19740214) from Japan Society for the Promotion of Science.

**References**

[1] Behnia K, Jaccard D and Flouquet J 2004 *J. Phys.:Condens. Matter* 16 5187
[2] Kadowaki K and Woods S B 1986 *Solid State Commun.* 58 507
[3] Sales B C, Mandrus D and Williams R K 1996 *Science* 272 1325
[4] Terasaki I, Sasago Y and Uchinokura K 1997 *Phys. Rev. B* 56 R12685
[5] Resel R, Gratz E, Burkov A T, Nakama T, Higa M and Yagasaki K 1996 *Rev. Sci. Instrum.* 67 1970
[6] Choi E S, Kang H, Jo Y J and Kang W 2002 *Rev. Sci. Instrum.* 73 2999
[7] Balet F J, Flood D J, Rowe V, Schroeder P A and Cox J E 1967 *Phys. Rev. Lett.* 18 395
[8] Gratz E and Zuckermann M J 1982 *J. Magn. Magn. Mater.* 29 181
[9] Fujiwara T, Fujii H, Uwatoko Y, Royama K, Motokawa M and Shigeoka T 2003 *Acta. Phys. Pol. B* 34 1541
[10] Nakama T, Hedo M, Nakamura D, Takaesu Y, Yagasaki K, Fujiwara T and Uwatoko Y 2009 *J. Phys.: Conf. Series* 150 042135
[11] Uwatoko Y, Todo S, Ueda K, Uchida A, Kosaka M, Mori N and Matsumoto T 2002 *J. Phys.: Condens. Matter* 14 11291–11296
[12] Murata K, Yoshino H, Yadav H O, Honda Y and Shirakawa N 1997 *Rev. Sci. Instrum.* 69 2490
[13] Fujiwara T, Uwatoko Y, Fujii H, Koyama K, Motokawa M and Shigeoka T 2004 *J. Magn. Magn. Mater.* 310 1877