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## LETTER TO THE EDITOR

## Thermoelectric power of the superconducting pyrochlore $\text{Cd}_2\text{Re}_2\text{O}_7$

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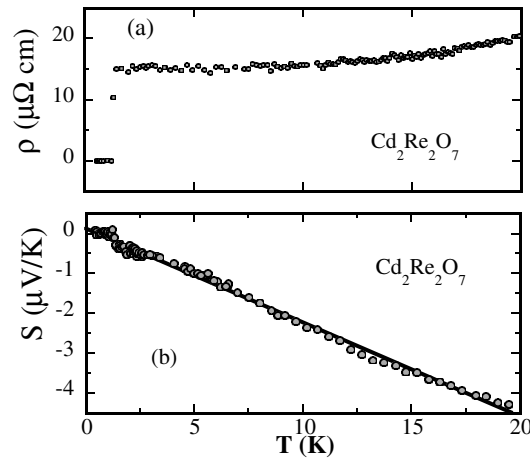
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### Abstract

We report the thermoelectric power ( $S$ ) of the recently discovered superconducting pyrochlore  $\text{Cd}_2\text{Re}_2\text{O}_7$  with  $T_c = 1.4$  K. In the normal state, this compound shows a continuous structural phase transition at  $T^* = 200$  K.  $S$  is negative from room temperature down to 1.4 K. Above  $T^*$ ,  $S$  shows small absolute values and approximately linear dependence on temperature.  $S(295 \text{ K}) = -2.2 \mu\text{V K}^{-1}$ . The absolute value of  $S$  increases rapidly below  $T^*$ . A kink and a minimum were observed round 125 and 60 K, respectively.  $S(60 \text{ K}) = -8.2 \mu\text{V K}^{-1}$ . In the low-temperature range  $1.4 \text{ K} < T < 20 \text{ K}$ , another linear portion was observed. As regards the  $T^2$ -dependent electrical resistivity in this temperature range, a Fermi liquid ground state is suggested for this compound. No change of  $S$  in a magnetic field up to 2 T was observed at 77 and 295 K.

The recent discovery of superconductivity in  $\text{Cd}_2\text{Re}_2\text{O}_7$  [1, 2] has stimulated much interest in transition-metal (TM) oxides in the vicinity of a metal–insulator transition (MIT). Although its transition temperature  $T_c$  is low (1.4 K), it is of interest because it is the first pyrochlore superconductor.  $\text{Cd}_2\text{Re}_2\text{O}_7$  belongs to the large family of pyrochlore compounds with formula  $\text{A}_2\text{B}_2\text{O}_7$ , where A represents a rare earth or late TM and B represents a TM. The measurements of the electrical resistivity  $\rho$ , specific heat  $C$ , and Hall coefficient  $R_H$  for  $\text{Cd}_2\text{Re}_2\text{O}_7$  in the normal state suggested that there is a structural phase transition around  $T^* = 200$  K [1–4]. The high-temperature phase crystallizes in an ideal cubic pyrochlore structure (space group  $Fd\bar{3}m$ ). Below  $T^*$ , it changes to another cubic structure (space group  $F\bar{4}3m$ ) [3]. There must be a drastic change of the electronic state from the high-temperature phase to the low-temperature phase. Furthermore, all of the reported measurements of electrical resistivity showed an anomaly around 100 K, but its origin has not been understood.



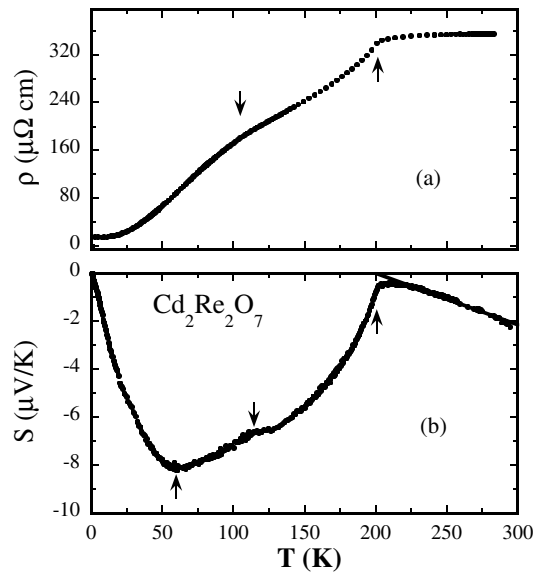
**Figure 1.** Electrical resistivity  $\rho$  (a) and thermoelectric power  $S$  (b) of  $\text{Cd}_2\text{Re}_2\text{O}_7$  plotted as a function of  $T$  at low temperatures ( $T < 20$  K).

In attempts to understand the mechanism of the superconductivity in this material, theoretical and experimental studies have been carried out extensively. Among the transport properties, the thermoelectric power ( $S$ ) is known to be very sensitive to the band structure near the Fermi level and the scattering mechanism. In this letter, we present experimental studies of  $S$  and  $\rho$  for a single crystal,  $\text{Cd}_2\text{Re}_2\text{O}_7$ , over a wide temperature range, carried out in order to get some insight into the electronic structure and scattering process in the normal state. We find that  $S$  shows very different behaviours above and below  $T^*$ .

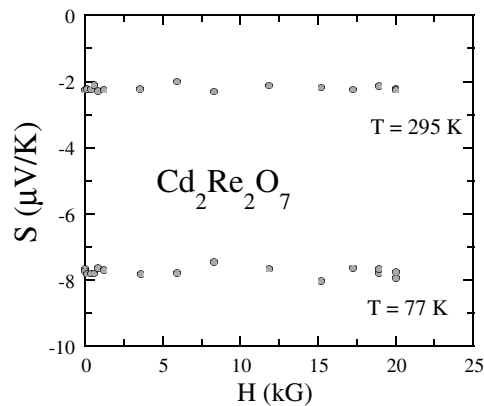
The single-crystal sample of  $\text{Cd}_2\text{Re}_2\text{O}_7$  was synthesized by solid-state reaction [5]. It is confirmed by powder XRD at room temperature (RT) that the sample crystallizes in the cubic pyrochlore structure with a lattice constant of 10.223 Å. Electrical resistivity was measured by using a standard four-probe technique from 0.3 K to RT. Thermoelectric power was measured from 2 K to RT with a slowly varying gradient technique by using chromel/Au + 0.07 at.% Fe thermocouples. The measurement of  $S$  from 3 to 0.3 K was carried out with a steady-state heat-flow technique employing a  $^3\text{He}$ - $^4\text{He}$  dilute refrigerator [6]. A superconducting wire of Nb was used for measuring the thermoelectric voltage.

The temperature dependences of  $S$  and  $\rho$  for  $\text{Cd}_2\text{Re}_2\text{O}_7$  are shown in figures 1 and 2. No hysteresis was observed in our cooling and heating runs. It is interesting that  $S$  is negative above its superconducting transition temperature, although the Hall coefficient changes sign at 125 K [7]. The difference in sign between  $S$  and the Hall coefficient was also observed for the related compound  $\text{Cd}_2\text{Os}_2\text{O}_7$  [8] above its MIT temperature  $T_{\text{MIT}}$ . Both electrons and holes participate in the electrical transport in these compounds. There is a competition between the positive contribution from hole-type carriers and the negative contribution from electron-type carriers. The electron-type carriers probably dominate throughout our measurement temperature range. Recently, a band calculation suggested multiband character near the Fermi surface for  $\text{Cd}_2\text{Re}_2\text{O}_7$  [9, 10].

In figure 1,  $S(T)$  and  $\rho(T)$  for  $\text{Cd}_2\text{Re}_2\text{O}_7$  at low temperatures are shown. Both of them drop to zero due to the superconducting transition at  $T_c = 1.4$  K, which is higher than the value for a polycrystalline sample [2], but close to the value for a single crystal reported by Jin *et al* [4]. Recently, Hiroi and Hanawa [11] pointed out that there is a sample dependence of  $T_c$ , as deduced from resistivity measurements for this material. The transition temperature



**Figure 2.** Thermoelectric power  $S$  (a) and electrical resistivity  $\rho$  (b) of  $\text{Cd}_2\text{Re}_2\text{O}_7$  plotted as a function of temperature.



**Figure 3.** The magnetic field dependence of the thermoelectric power of  $\text{Cd}_2\text{Re}_2\text{O}_7$  measured at 77 and 295 K.

seems to be sensitive to the surface treatment. Above  $T_c$ ,  $|S|$  (the absolute value of  $S$ ) increases linearly with temperature up to 20 K. The solid line shows a fit to the straight line passing the origin,  $S = aT$  with  $a = -0.234 \mu\text{V K}^{-2}$ .

$S(T)$  and  $\rho(T)$  of  $\text{Cd}_2\text{Re}_2\text{O}_7$  for overall temperature range are shown in figure 2.  $\rho$  at RT and just above  $T_c$  takes the values 355 and 15  $\mu\Omega$  cm, respectively, giving a residual resistivity ratio  $\text{RRR} = 24$ . Below 20 K,  $\rho$  shows a  $T^2$ -dependence,  $\rho = \rho_0 + AT^2$  with  $\rho_0 = 14.6 \mu\Omega$  cm and  $A = 0.012 \mu\Omega$  cm  $\text{K}^{-2}$ . The  $T^2$ -dependent  $\rho$  below 60 K was also reported by Jin *et al* [4], although the coefficient  $A$  is only half of the value reported by them. As regards the linear  $T$ -dependent  $S$  in the same temperature range, a correlated Fermi liquid state for  $\text{Cd}_2\text{Re}_2\text{O}_7$  is suggested.  $S$  deviates from the linear dependence above 20 K, gradually. It shows a minimum around 60 K and a kink at 125 K. Above 200 K, where a continuous structural

phase transition occurs,  $|S|$  is small and shows linear temperature dependence again. The two linearly  $T$  dependent portions can be explained as arising from the diffusion contribution to the thermoelectric power for normal metals. However, the linear portion above  $T^*$  gives an intercept of  $+4.6 \mu\text{V K}^{-1}$ . Interestingly,  $S$  for the related compound  $\text{Cd}_2\text{Os}_2\text{O}_7$  also shows small values and nearly linear temperature dependence above its MIT temperature 226 K [8]. Correspondingly, the electrical resistivity for these two materials shows very weak temperature dependence in the temperature range from  $T_{\text{MIT}}(T^*)$  to RT, which suggests saturation of the mean free path of carriers. The absolute values of  $S$  at RT are almost the same for  $\text{Cd}_2\text{Os}_2\text{O}_7$  ( $S(295 \text{ K}) = +2 \mu\text{V K}^{-1}$ ) [8] and  $\text{Cd}_2\text{Re}_2\text{O}_7$  ( $S(295 \text{ K}) = -2.2 \mu\text{V K}^{-1}$ ).

Since  $\text{Cd}^{2+}$  and  $\text{Re}^{5+}$  have  $4d^{10}$  and  $5d^2$  outer electron configurations, respectively,  $\text{Re}^{5+}$  alone is expected to be responsible for the magnetic properties of  $\text{Cd}_2\text{Re}_2\text{O}_7$ . However, it was reported that the system undergoes a continuous structural phase transition at  $T^*$  without magnetic order [1–4]. In order to see the magnetic field effect on  $S$ , we measured  $S$  in magnetic fields up to 2 T at 77 K and RT (295 K). As can be seen in figure 3, there is no change due to the fields in our measurements for the high-temperature phase or low-temperature phase. It is suggested that spin-dependent scattering could be unimportant in  $\text{Cd}_2\text{Re}_2\text{O}_7$ .

In summary, the thermoelectric power of  $\text{Cd}_2\text{Re}_2\text{O}_7$  was reported for the first time.  $S$  is negative in the temperature range from 1.4 K to RT. The different behaviours of  $S$  below and above the structural transition temperature  $T^*$  reflect the change in electronic structure for the two phases. We have not observed any magnetic field effect on  $S$  at 77 and 295 K in magnetic fields up to 2 T.

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