Physics of Strongly Correlated Electron Systems JJAP Series 11, pp. 24-26

# Metamagnetic Transition of RSb<sub>2</sub> at High Pressure

Tomoko KAGAYAMA, Gendo OOMI, Yasuhiro IYE<sup>1</sup>, S.L. BUD'KO<sup>2</sup>, and P.C. CANFIELD<sup>2</sup>

Department of Mechanical Engineering and Materials Science, Kumamoto University, Kumamoto 860–8555 <sup>1</sup> The Institute for Solid State Physics, The University of Tokyo, Minato-ku, Tokyo 106–8666 <sup>2</sup> Ames Laboratory, Iowa State University, Iowa, NM 50011, USA

(Received Nobember 9, 1998)

The field dependences of the electrical resistance are reported for  $RSb_2$  (R=La, Ce, Pr, Nd and Sm) single crystals. Three of them (R=Ce, Pr and Nd) shows some metamagnetic transitions, which are briefly compared with the magnetization reported previously. Pressure effects up to 2 GPa are also shown for R=La, Ce, Pr.

KEYWORDS: metamagnetism, electrical resistance, high pressure

## §1. Introduction

The light rare-earth diantimonides  $RSb_2$  (R = La–Nd, Sm) crystallize in the orthorhombic  $SmSb_2$  structure.<sup>1, 2)</sup> This structure is highly anisotropic, having two unique Sb sites and one unique R site. The magnetic and transport properties of these compounds have not been well known for several decades. Only a little was known as follows: LaSb<sub>2</sub> is a superconductor of  $T_c \sim 0.4 \text{ K}$ ,<sup>3)</sup> CeSb<sub>2</sub> is a ferromagnet having  $T_C=15 \text{ K}^4$  and PrSb<sub>2</sub> is a antiferromagnet of  $T_N=5.1 \text{ K}$ . Successful growth of high quality single crystals of these compounds allows us to proceed in detailed investigations and make clear that these have a lot of interesting electronic and magnetic properties such as magnetic order or metamagnetism at low temperature.

These properties are highly anisotropic reflecting the two dimensional orthorhombic crystal structure. The magnetism in RSb<sub>2</sub> compounds depends strongly on the R-R distance. These facts imply that the electronic structure of RSb<sub>2</sub> is affected largely by an application of pressure. Recently we have reported the results of the electrical resistance measurement at various pressures and magnetic fields for PrSb<sub>2</sub>.<sup>6,7</sup> Main results are that (1) some metamagnetic transitions are found and tend to be smeared out at high pressure above 2 GPa like an antiferromagnetic transition, (2) hump-like anomaly in R(T) curve at  $T^* \sim 100$  K, possibly originating from the charge-density wave, and its large pressure dependence are observed.

Here we report the recent results of zero-pressure magnetoresistance up to 15 T and its pressure effect up to 5 T for some RSb<sub>2</sub> compounds.

### §2. Experimental Method

Single crystals of RSb<sub>2</sub> were grown out of antimony flux.<sup>4,5)</sup> Typical sample size for measurement was a rectangle of  $2 \times 1 \times 0.3 \text{ mm}^3$ , which was cut from the plate which grows perpendicular to the *c*-axis. The long side of the rectangle was one of *a*- and *b*-axis, though it was impossible to detect exact crystal axis because of difficulty of Laue diffraction. The electrical resistance was measured by a standard dc four probe method down to 0.5 K at ambient pressure and down to 2 K at pressures. The magnetic field was applied in perpendicular to both of plane-axis (*c*-axis) and current-axis (*a*- or *b*-axis) up to 15 T at ambient pressure and to 5 T at high pressures. Hydrostatic pressure up to 21 kbar was generated by means of Cu-Be cylinder and WC piston. The detail of the high pressure apparatus was described previously.<sup>8)</sup>

## §3. Results and Discussion

Figure 1 shows  $\rho(H)$  curves at various temperatures at ambient pressure for RSb<sub>2</sub> (R=Ce, Pr, Nd, Sm). It is clear seen that sharp changes in resistance occur at certain magnetic fields for R=Ce, Pr, Nd. These seem to almost correspond to the step like changes in magnetization reported by Bud'ko et al. for one direction of the magnetic field in the *ab*-plane except for the value of magnetic field at which metamagnetic transition occurs.<sup>9</sup> The difference is considered to be due to the difference of the direction of magnetic field in *ab*-plane.

CeSb<sub>2</sub> is not a simple ferromagnet but is one having complex spin structures clearly observed as anomalies in the temperature derivative of the resistivity  $d\rho/dT$ and the magnetization d(MT)/HdT at 15.5, 11.7 and 9.3 K.<sup>9</sup> It shows some metamagnetic transitions reflecting such complexity in magnetic ordering. The field dependence of the resistance for CeSb<sub>2</sub> is interpreted as follows; the sharp increase at lowest field and the drop or the kink at about 1 T are possibly associated with some kind of spin-flip ordering processes to 0.2 and 0.4  $\mu_{\rm B}/{\rm Ce}$ , respectively. The next change until *R* drops at about 3 T may be due to the spin rotation where magnetization gradually increases. The ordered state above 3 T is expected to have some fun-type structure having the net magnetization of 1.1  $\mu_{\rm B}/{\rm Ce}$ .

 $PrSb_2$  orders antiferromagnetically at 5.1 K showing a resistance drop accompanied by a slight upturn which may due to a gap formation at the Fermi level. The R(H) curve of  $PrSb_2$  at 1.5 K shows two clear discontinuous changes at about 0.5 T and 1.5 T. These metam-



Fig. 1. Magnetoresistance at ambient pressure for (a) CeSb<sub>2</sub>, (b) PrSb<sub>2</sub>, (c) NdSb<sub>2</sub> and (d) SmSb<sub>2</sub>.

agnetic transitions becomes less clear at higher temperature 4.2 K but the R(H) curve above 5 T is almost the same for different temperatures. Other interesting feature occurs at 0.5 K (not shown here): the R(H) curve shows hysteresis at a range below the magnetic field of large drop at 1.5 T.

Most exotic variation of the magnetoresistance is seen for  $NdSb_2$ , however, it is difficult to make clear the correspondence to the metamagnetic transition observed in the magnetization.

There is no metamagnetic transition in the field range of  $H \leq 15$  T for SmSb<sub>2</sub>. The magnetoresistance of SmSb<sub>2</sub> becomes as large as over 15 times that at zero field, reflecting the high purity of the sample. In addition to this, the Shubnikov-de Haas oscillation is observed above 10 T.

Figure 2 shows the electrical resistance of LaSb<sub>2</sub> at 4.2 K as a function of the magnetic field at various pressures. At ambient pressure (P=0 kbar), R increases almost linearly with the field up to 5 T becoming larger than double that at zero-field. With increasing pressure R decreases and the change in R by magnetic field  $\Delta R$  also becomes small.

Effect of pressure on the field dependences of the electrical resistance of CeSb<sub>2</sub> at 4.2 K is illustrated in Fig. 3. R shows a sharp increase below 0.5 T and drops at 1 and 3 T corresponding to the metamagnetic transitions as mentioned above. The drops in R(H) curve are sifted to higher field and disappeared above 12 kbar (the data at pressures below 12 kbar are not shown here). The first metamagnetic transition also shifts to high H of



Fig. 2. Field dependent electrical resistance R(H) at 4.2 K for LaSb<sub>2</sub> at various pressure. The data above 13 kbar (not shown here) are little different from that at 9 kbar.

1 T and is smeared at 2.1 kbar. These are considered to have close relation with the pressure dependence of the temperatures at which the R(T) curve has anomalies. The anomaly at 15.5 K shifts to higher temperature and that at 17.7 K opposingly goes down with increasing pressure. Both of these anomalies in R(T) become less clear at 21 kbar. The whole dependence against the magnetic field tends to be strong at high pressures: Rat 2.1 kbar decreases initially showing quadratic behavior and becomes almost a half of that at zero-field. The striking enhancement of the value of R at low tempera-

#### 26 JJAP Series 11



Fig. 3. The electrical resistance R of CeSb<sub>2</sub> as a function of magnetic field H at high pressure.

ture by pressure is unique properties in RSb<sub>2</sub> (R=Ce), which may be associated with the Kondo effect. Similar change of R(T) at low temperature induced by pressure is reported in monoantimonide CeSb.

Figure 4 shows the R(H) of PrSb<sub>2</sub> at 4.2 K for various



Fig. 4. The magnetoresistance of PrSb<sub>2</sub> at 4.2 K for various pressures.

pressures. The discontinuous changes observed at 2 K are less clear at 4.2 K but these are seen as a small hump and a sharp peak, respectively. The higher transition field seems to increase with pressure up to 10 kbar and to decrease with further pressurising. The peak becomes smeared at high pressures.

## §4. Conclusion

We report in this paper the data of magnetoresistance of high quality single crystals of  $RSb_2$  (R=La, Ce, Pr,

#### T. KAGAYAMA et al.

Nd and Sm) and their pressure dependences. It is obvious that the magnetic structures of these compounds are very complex. The study of the magnetization at high pressure is highly desired to make the anomalous magnetoresistance clear.

On the other hand, it becomes obvious from recent experiments that there are pressure-induced large hysteresis behaviors in the R(T) curved for R=La, Ce and Pr. The high pressure X-ray diffraction study is now on progress to clarify the origin of such anomalies.

- 1) R. Wang and H. Steinfink: Inorg. Chem, 6 (1967) 1685.
- F. Hullinger: Handbook of Physics and Chemistry of Rare Earths, eds. K.A. Gschneidner and L. Eyring, (North-Holland, Amsterdam, 1979), Vol. 4, p. 153.
- F. Hullinger and H.R. Ott: J. Less-Common Met. 55 (1977) 103.
- P.C. Canfield, J.D. Thompson and Z. Fisk: J. Appl. Phys. 70 (1991) 5992.
- 5) P.C. Canfiled and Z. Fisk: Philos. Mag. B 65 (1992) 1117.
- G. Oomi, T. Kagayama, K. Kawaguchi, P.C. Canfield and S.L. Bud'ko: Physica B 230-232 (1997) 776.
- K. Kawaguchi, T. Kagayama, G. Oomi, P.C. Canfield and S.L. Bud'ko: Physica B 237-238 (1997) 587.
- 8) G. Oomi and T. Kagayama: Physica B 239 (1997) 191.
- S.L. Bud'ko, P.C. Canfield, C.H. Mielke and A.H. Lacerda: Phys. Rev. B 57 (1998) 13624.