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Magnetic and thermoelectric properties of AOs_4Sb_{12} (A = Sr, Ba)

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Recently, alkaline-earth filled skutterudites AFe_4Sb_{12} (A = Ca, Sr, Ba) have been found to be nearly ferromagnetic metals [1]. These compounds show a maximum in both magnetic susceptibility and thermopower at 50 K. A large electronic specific heat coefficient γ of 100 mJ/mol K^2 implies that the Fe $3d$ derived flat bands are located near the Fermi level. Furthermore, a remanent moment of the order of 10^{-3} μ_B/Fe was observed below 54, 48, and 40 K for A = Ca, Sr, and Ba, respectively. On the other hand, ARu_4Sb_{12} (A = Sr and Ba) are diamagnetic metals with a smaller γ of 10 mJ/mol K^2 [2]. This fact indicates that the Ru $4d$ bands are deeper and broadened than the Fe $3d$ bands.

We report here the magnetic and transport properties of the $5d$ counterparts AOs_4Sb_{12} (A = Sr and Ba). These compounds were synthesized by the reaction of alkaline-earth antimonide and osmium antimonide. The powder samples were densified as high as 90% of the ideal density by the spark-plasma sintering (SPS) method for the transport measurements.

Fig. 1 shows the temperature dependence of magnetic susceptibility $\chi = M/B$ of powder samples. Positive $\chi(T)$ means that the Os $5d$ electrons contribute to the paramagnetism. However, the value is one order of magnitude smaller than that of $BaFe_4Sb_{12}$, and temperature dependence does not obey the Curie-Weiss law. Thus, unlike Fe $3d$ electrons in AFe_4Sb_{12}, the Os $5d$ electrons have no magnetic moment. Since the γ values are modestly large, 48 mJ/mol K^2, the Os $5d$ electrons should contribute to the density of state near the Fermi level. No magnetic transition was observed in both powder and SPS samples down to 2 K.

Fig. 2(a) shows the temperature dependence of electrical resistivity $\rho(T)$ that is normalized by the value at 292 K. A shoulder appears around 100 K in $\rho(T)$ for both A = Sr and Ba. In the case of $BaFe_4Sb_{12}$, more pronounced shoulder around 70 K was explained by the scattering of the conduction electrons by the spin fluctuations of Fe $3d$ electrons [1]. Because such scattering is unlike to be dominant in the Os compounds, the shoulder can be attributed to strong electron scattering by rattling motion of Sr and Ba ions in the oversized cage [3].

The thermopower $S(T)$ at 500 K of A = Sr and Ba are 35 and 46 μV/K, respectively, which are intermediate between that of $BaFe_4Sb_{12}$ and $BaRu_4Sb_{12}$ as shown in Fig. 2(b). This result is consistent with the fact that the γ values of A = Sr and Ba are also intermediate between Fe compounds (100 mJ/mol K^2) and Ru compounds (10 mJ/mol K^2).

[1] E. Matsuoka et al., cond-mat/0412094.
[2] K. Hayashi et al., The second workshop on skutterudite (July, 2004), PS14.
[3] H. Kusunose and K. Miyake, J. Phys. Soc, Jpn. **65** (1996) 3032.

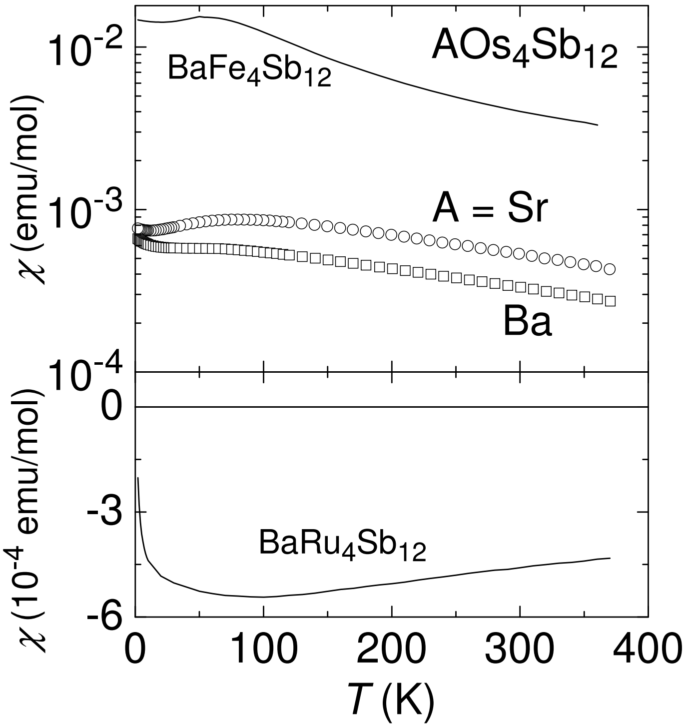

Figure 1: Temperature dependence of the magnetic susceptibility of AOs_4Sb_{12}, $BaFe_4Sb_{12}$, and $BaRu_4Sb_{12}$.

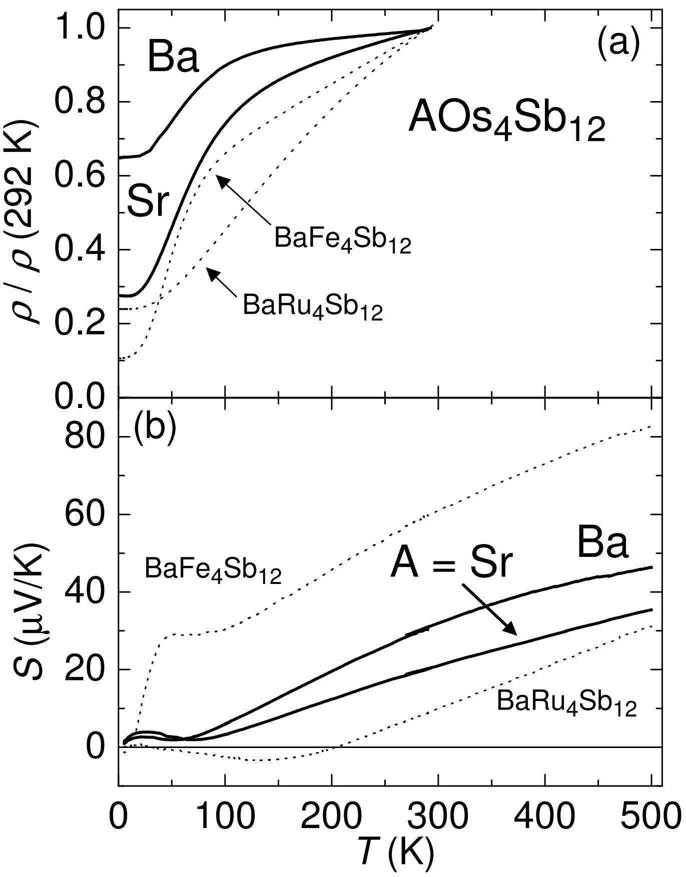

Figure 2: Temperature dependence of (a) electrical resistivity and (b) thermopower of AOs_4Sb_{12}, $BaFe_4Sb_{12}$, and $BaRu_4Sb_{12}$.