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Magnetic and transport properties of alkaline-earth filled skutterudites AFe_4Sb_{12} (A = Ca, Sr, Ba); nearly ferromagnetic metals

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The discovery of itinerant ferromagnetism in alkali-metal filled skutterudites $NaFe_4Sb_{12}$ and KFe_4Sb_{12} [1] has shed light on the role of $3d$ electrons in the physical properties of skutterudites with nonmagnetic fillers. We report here the magnetic and transport properties of alkaline-earth filled skutterudites AFe_4Sb_{12} (A = Ca, Sr, Ba). The compounds were synthesized by the reaction of alkaline-earth antimonide and iron 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. It is found that X-ray powder diffraction lines of these sintered samples are broadened compared with those of the untreated powder samples. This broadening may originate from the deformation of the lattice induced by SPS.

The magnetic susceptibility χ of the powder samples measured in a field of 5 T follows the Curie-Weiss law above 200 K with the effective magnetic moments and Weiss temperatures θ of 1.52, 1.47, and 1.50 μ_B/Fe and 54, 53, and 31 K for A = Ca, Sr, and Ba, respectively. The largely positive value of θ indicates the ferromagnetic interaction among Fe $3d$ electrons. The appearance of the broad maximum in $\chi(T)$ around 60 K implies that AFe_4Sb_{12} compounds are nearly ferromagnetic. This T-dependence of $\chi(T)$ is consistent with the ^{123}Sb-NQR experiment for $SrFe_4Sb_{12}$. In fact, the nuclear spin-lattice relaxation time T_1 follows the relation $(T_1T)^{-1} \propto \chi(T)$ [2]. In a weak field of 0.01 T, however, the data of M/B of powder samples for A = Ca and Sr abruptly increase on cooling below 20 and 35 K, respectively (see Fig. 1). At 5 K, the magnetization curves show a clear hysteresis loop with the remanent moment about 1.0×10^{-3} μ_B/Fe, as shown in the inset. It is noteworthy that no spontaneous moment appears for the powder sample of $BaFe_4Sb_{12}$, but the sintered sample shows the ferromagnetic behavior below 40 K. Furthermore, sintered samples of $CaFe_4Sb_{12}$ and $SrFe_4Sb_{12}$ have higher T_C and larger remanent moments compared with the powder samples as shown in the Fig. 1. The systematic increase of T_C from 40 K for A = Ba, 48 K for A = Sr to 54 K for A = Ca is the order of the decreasing in the size of the A ion. These facts suggest that the lattice defect and/or deformation caused by SPS may induce the ferromagnetism in $BaFe_4Sb_{12}$ and raise both the T_C and remanent moment in $CaFe_4Sb_{12}$ and $SrFe_4Sb_{12}$. Above 0.1 T, the magnetization increases linearly with increasing field up to 5 T. These behaviors can be understood if a part of the sample orders ferromagnetically below T_C and the rest remains paramagnetic. This conjecture has been corroborated by the μSR experiment of a sintered sample of $BaFe_4Sb_{12}$ [3]. The inhomogeneous nature of the ferromagnetism is also consistent with the fact that no sign of phase transition at T_C was observed in the specific heat measurement. The electronic specific heat coefficients are estimated to be approximately 100 mJ/mol K^2, which indicates the enhanced electronic density of states derived from the Fe atoms. Fig. 2 shows the temperature dependence of the electrical resistivity and thermopower. The abrupt decreases in both quantities below 50 K coincide with the ferromagnetic transition. Above 100 K, the thermopower increases linearly with increasing temperature, and reachs 100 μV/K at 500 K.

[1] A. Leithe-Jasper et al., Phys. Rev. Lett. **91** (2003) 037208.
[2] M. Matsumura, PS13 of this meeting.
[3] W. Higemoto, 30b2 of this meeting.

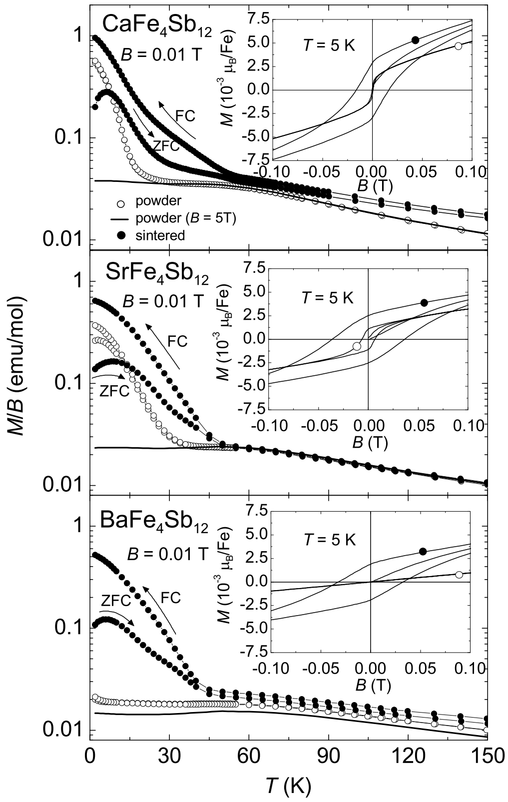

Figure 1: Temperature dependence of the magnetic susceptibility of AFe_4Sb_{12}. Solid and open circles represent the data for the sintered sample by SPS method and powder sample, respectively. The insets show the magnetization curves.

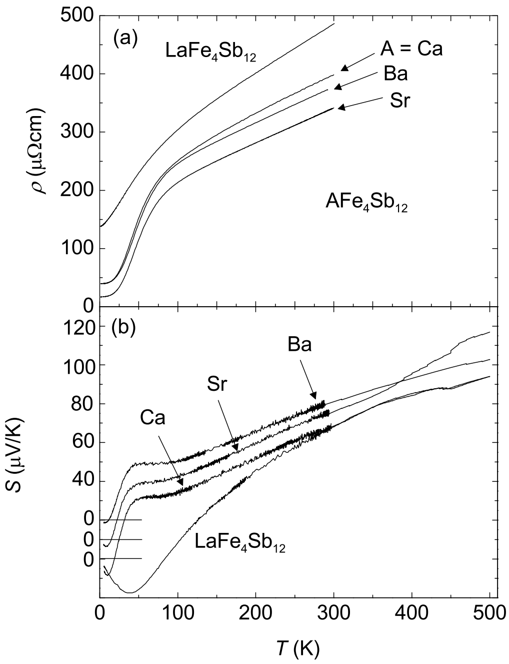

Figure 2: Temperature dependence of (a) electrical resistivity and (b) thermopower for AFe_4Sb_{12} (A = Ca, Sr, Ba) and $LaFe_4Sb_{12}$.