Low temperature properties of Sm-based filled skutterudite phosphides

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Abstract

Filled skutterudite phosphides $SmFe_4P_{12}$ and $SmOs_4P_{12}$ have been prepared at high temperature and high pressure. Electrical resistivity, magnetic susceptibility and specific heat measurements indicate that $SmOs_4P_{12}$ is an anti-ferromagnet with a T_N of 4.6K, whereas $SmFe_4P_{12}$ is paramagnetic down to 2K.

Key words:
Filled skutterudite; specific heat; magnetic ordering

1. Introduction

Ternary compounds with the general formula MT_4P_{12} (M=alkaline earth, rare earth and actinide; T=Fe, Ru and Os; X=P, As and Sb) crystallize in the filled skutterudite structure. [1]. This MT_4X_{12} skutterudite compounds are attracting a great deal of attention, because they show various physical properties like superconductivity [2], metal-insulator transition [3], magnetic ordering [4], a heavy fermion metal [5].

$PrRu_4P_{12}$ shows an anomalous metal-insilator (MI) transition at 60K [3]. The MI transition is caused by a structural phase transition that doubles the unit cell and the nesting of the Fermi surface. $SmRu_4P_{12}$ also shows MI transition at 16K and behaves metallic above this temperature [6]. In fact, the MI transition occurs in two successive steps and could be due to an orbital ordering and magnetic ordering. The interesting property found in $SmRu_4P_{12}$ motivated us to further study in Sm-based skutterudite phospides by substituting transition metals. In this paper we report the low temperature properties of $SmFe_4P_{12}$ and $SmOs_4P_{12}$.

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2. Experiment

Single phase polycrystalline $SmFe_4P_{12}$ and $SmOs_4P_{12}$ have been synthesized by using high pressure and high temperature method [7]. The samples were characterized by powder X-ray diffraction using $CuK\alpha_1$ radiation and silicon as a standard. Resistivity was measured by using standard four-prove direct current method over a temperature range from 2 to 300K. The DC magnetic susceptibility was measured in the range from 2 to 300K with a Quantum Design MPMS SQUID magnetometer. The specific heat was measured by thermal relaxation method (PPMS, Quantum Design Inc.).

3. Experimental results and discussion

The cubic lattice parameters of $SmFe_4P_{12}$ and $SmFe_4P_{12}$ are determined from x-ray powder diffractometry to be a=7.7986$\dot{A}$, 8.0752$\dot{A}$, respectively.

The electrical resistivity versus temperature for $SmFe_4P_{12}$ is plotted in Fig. 1. The resistivity decreases linearly down to 100K and the decreasing rate

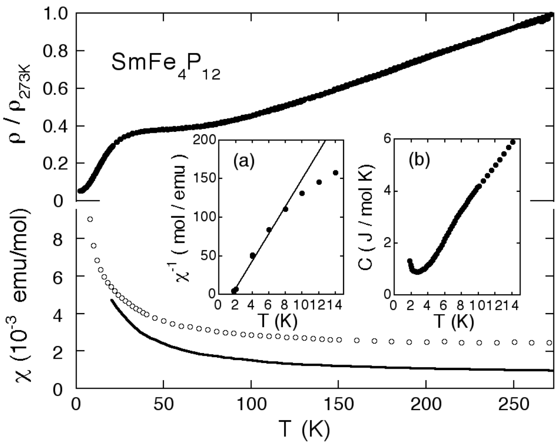

Fig. 1. Normalized electrical resistivity ρ/ρ_{273K} and magnetic susceptibility χ of $SmFe_4P_{12}$ as a function of temperature. Low temperature inverse magnetic susceptibility χ^{-1} and spesific heat coefficient are shown in inset (a) and (b), respectively.

of resistivity becomes very small down to 30K followed by a steep decrease. This behavior resembles that of Kondo compounds. The resistivity of $SmOs_4P_{12}$ exhibits a weak T-dependence above 40K and a broad minimum near 40K, followed by a sharp drop at about 5K, which is associated with the onset of magnetic order as shown in Fig. 2. The magnetic susceptibility χ for $SmFe_4P_{12}$ and $SmOs_4P_{12}$ are plotted versus temperature in Fig. 1 and Fig.2, respectively. In the figures, the solid curves show the theoretical magnetic susceptibility of Van Vleck-Frank Sm^{3+} [8]. The observed susceptibility of $SmFe_4P_{12}$ does not follow the theoretical model.This fact suggests that the Sm ion is in the valence fluctuation state in $SmFe_4P_{12}$. On the other hand,the observed susceptibility of $SmOs_4P_{12}$ is similar to the theoretical model. This fact suggests that Sm ion is nearly trivalent state. An distinct cusp anomaly found in the susceptibility at 4.5K, which indicates antiferromagnetic ordering. The specific heat coefficient of $SmFe_4P_{12}$ shows an upturn, which is still increasing at 2K and the intersection of χ^{-1} is positive at around 2K. These facts indicate a possibility of ferromagnetic ordering at lower temperature (Inset (a) and (b) of Fig. 1). The specific heat coefficient of $SmOs_4P_{12}$ shows a pronounced peak at $T_N = 4.6K$, that correlates well in temperature with the cusp of χ and sharp drop in ρ (Inset (a) and (b) of Fig. 2). The magnetic entropy estimated at zero field reches $Rln4$ at T_N. This suggests that the crystalline electric field (CEF) ground state in cubic symmetry is a quartet. As magnetic field increases, the anomaly shifts to lower temperature. This also indicates the magnetic phase transition is antiferromagnetic one.

In Summary, we have investigated the low temperature properties of Sm-based filled skutterudite

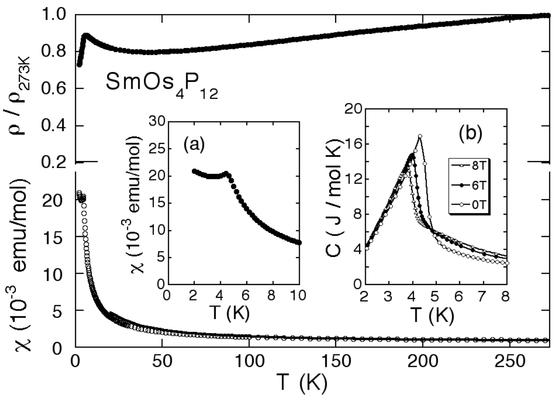

Fig. 2. Normalized electrical resistivity ρ/ρ_{273K} and magnetic susceptibility χ versus temperature for $SmOs_4P_{12}$. Low temperature magnetic susceptibility χ and specific heat coefficient are shown in inset (a) and (b), respectively.

SmT_4P_{12}(T=Fe,Os) compounds. We have found that, $SmOs_4P_{12}$ is an antiferromagnet with transition temperature $T_N = 4.6K$, while, no phase transition was found in $SmFe_4P_{12}$.

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