

## The ferromagnetic Kondo-lattice compound $\text{SmFe}_4\text{P}_{12}$

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2003 J. Phys.: Condens. Matter 15 L229

(<http://iopscience.iop.org/0953-8984/15/14/101>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.119

The article was downloaded on 19/05/2010 at 08:37

Please note that [terms and conditions apply](#).

## LETTER TO THE EDITOR

**The ferromagnetic Kondo-lattice compound  $\text{SmFe}_4\text{P}_{12}$** **Naoya Takeda and Masayasu Ishikawa**Institute for Solid State Physics, University of Tokyo, Kashiwanoha, Kashiwa,  
Chiba 277-8581, Japan

E-mail: ntakeda@issp.u-tokyo.ac.jp

Received 10 February 2003

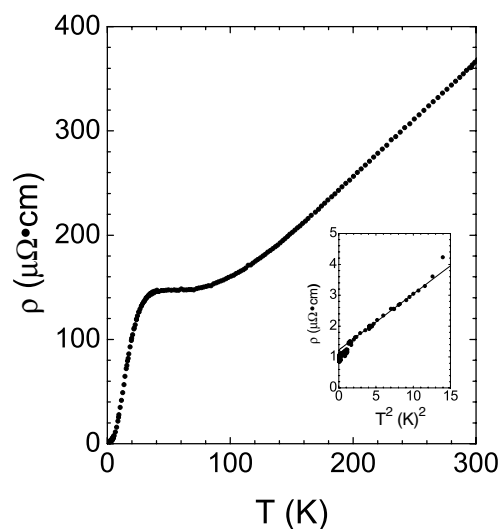
Published 31 March 2003

Online at [stacks.iop.org/JPhysCM/15/L229](http://stacks.iop.org/JPhysCM/15/L229)**Abstract**

We report on the magnetic properties of a filled skutterudite compound,  $\text{SmFe}_4\text{P}_{12}$ . Magnetic susceptibility and specific heat measurements revealed a ferromagnetic transition at 1.6 K. The temperature dependence of the electrical resistivity exhibits a Kondo-lattice behaviour and the electronic specific heat coefficient attains values as large as  $370 \text{ mJ mol}^{-1} \text{ K}^{-2}$ . This compound is thereby the first Sm-based heavy-fermion system found with a ferromagnetic ground state. The Kondo temperature is estimated to be about 30 K.

Filled skutterudite compounds with a general formula  $\text{RT}_4\text{X}_{12}$  (R = alkaline earth, rare earth, Th and U; T = Fe, Ru and Os; X = P, As and Sb) crystallize in a unique body-centred cubic structure of space group  $Im\bar{3}$  (No 204) [1]. It is now known that these compounds exhibit various interesting physical properties at low temperatures, such as non-Fermi-liquid behaviour, metal–insulator transitions, heavy fermions and the recently found heavy-fermion superconductivity [2–6]. One of the most striking features of this series of compounds is the occurrence of heavy-fermion behaviour in Pr compounds. The magnetic properties of ordinary Pr-based compounds have been recognized as a combination of the effect of well localized 4f electrons and a crystalline electric field (CEF) effect. It was recently reported that  $\text{PrFe}_4\text{P}_{12}$  and  $\text{PrOs}_4\text{Sb}_{12}$  showed heavy-fermion behaviours and, in fact, an extremely large cyclotron mass reaching as much as  $81 m_0$  was observed in  $\text{PrFe}_4\text{P}_{12}$  by means of the de Haas–van Alphen (dHvA) effect [5, 6]. It is pointed out that the interactions of quadrupole moments of Pr ions and conduction electrons may play an important role, in contrast with the case for Ce- or Yb-based heavy-fermion compounds, where the magnetic Kondo effect is important. Sm compounds, such as SmS, exhibit a mixed-valence state, unlike Pr compounds, which has been attracting a lot of attention for a long time, but only a few compounds, such as  $\text{SmSn}_3$ , are known as heavy-fermion systems [7, 8].

The magnetic susceptibility and electrical resistivity of  $\text{SmFe}_4\text{P}_{12}$  were reported by Jeitschko *et al* [9]. They concluded that  $\text{SmFe}_4\text{P}_{12}$  is a Van Vleck paramagnet. This implies that the Sm ions are divalent and hence that the Hund's-rule ground state is a non-magnetic



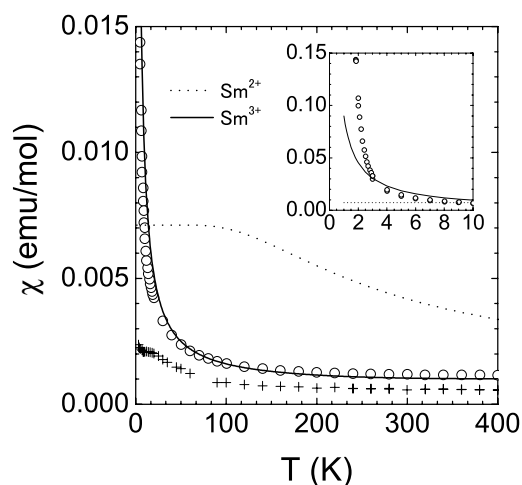
**Figure 1.** The temperature dependence of the electrical resistivity. The inset displays a  $\rho(T)-T^2$  plot showing a  $T^2$ -law below 3.4 K. The slight drop at low temperatures is due to the ferromagnetic transition. The solid line is the least-squares fit.

singlet. In fact, the lattice parameter is slightly larger than the value expected for trivalent  $\text{RFe}_4\text{P}_{12}$  ( $\text{R} = \text{La}, \text{Pr}$  and  $\text{Nd}$ ). On the other hand, the electrical resistivity indicates that there is Kondo-lattice behaviour rather than a simple paramagnetic metallic behaviour. If this is the case, Sm ions must be trivalent or have mixed valence and hence carry a local moment. In order to clarify this inconsistency, we prepared high-quality samples and explored the ground state down to very low temperatures by specific heat, electrical resistivity and magnetization methods.

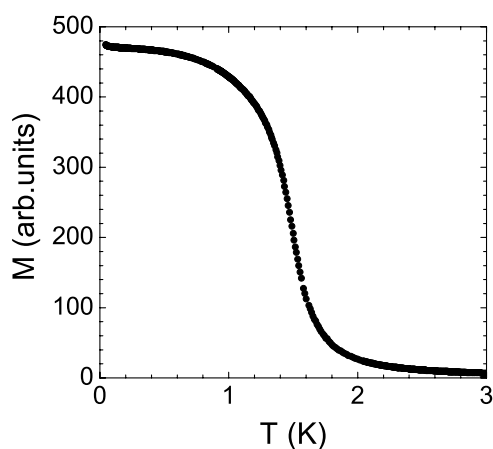
The samples were prepared by a molten-metal-flux growth method with a Sn flux [10]. Powder x-ray diffraction examination does not reveal any impurity profiles. The lattice parameter determined using  $\text{Cu K}\alpha_1$  radiation is  $7.7990(1) \text{ \AA}$  which is very close to the reported value [9]. The magnetic susceptibility between 1.8 and 400 K was measured using a Quantum Design MPMS. The magnetization at the magnetic field of 210 Oe was measured by a SQUID magnetometer between 45 mK and 3 K. The specific heat was measured by a semi-adiabatic heat-pulse method in a dilution refrigerator. The electrical resistivity was measured by a four-probe dc method in a  $^3\text{He}$  cryostat and a dilution refrigerator.

Figure 1 shows the temperature dependence of the electrical resistivity,  $\rho(T)$ , which agrees qualitatively with the previous report [9].  $\rho(T)$  decreases linearly with  $T$  and deviates below about 130 K; this is followed by a sharp decrease. The characteristic feature of Kondo lattices can be seen in the shoulder below 100 K. A distinct minimum, usually observed for Kondo compounds, cannot be seen in  $\rho(T)$ , but if the phonon part was subtracted from the total resistivity, the magnetic part would have a negative  $d\rho/dT$  above 50 K. Therefore this shoulder cannot be ascribed to a simple spin-disorder scattering caused by a crystalline field splitting; a Kondo-type scattering has to be involved.  $\rho(T)$  follows a  $T^2$ -law below 3.4 K and shows a small drop below 1.3 K, as shown in the inset of figure 1.

The temperature dependence of the magnetic susceptibility between 1.8 and 400 K is shown in figure 2 and the low-temperature part below 10 K is replotted in the inset. The theoretical curves for free  $\text{Sm}^{3+}$  and  $\text{Sm}^{2+}$  are also included. The experimental data increase monotonically with decreasing temperature and follow the curve for  $\text{Sm}^{3+}$  fairly well down



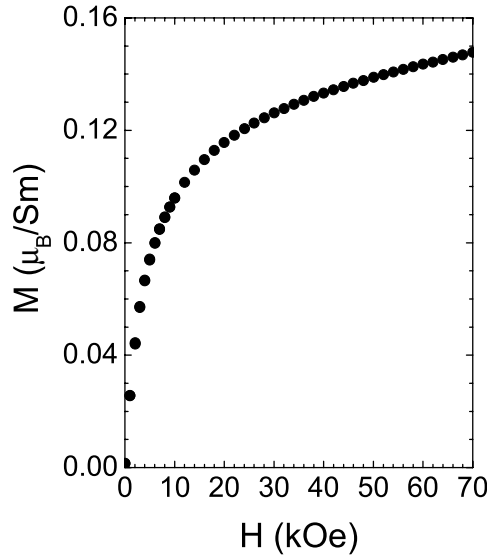
**Figure 2.** The temperature dependence of the magnetic susceptibility for  $\text{SmFe}_4\text{P}_{12}$  (O) and  $\text{La}_{0.4}\text{Sm}_{0.6}\text{Fe}_4\text{P}_{12}$  (+). The solid and dotted curves are the theoretical curves for free  $\text{Sm}^{3+}$  and  $\text{Sm}^{2+}$ . The inset emphasizes the low-temperature steep increase for  $\text{SmFe}_4\text{P}_{12}$ .



**Figure 3.** The thermal variation of the magnetization at  $H = 210$  Oe, clearly indicating a ferromagnetic transition.

to 3 K; there is no indication of a Van Vleck magnetism. Although the slightly large lattice parameter implies admixture of divalent states, it is conjectured from the present results that the majority of Sm ions are trivalent. The temperature dependence of the magnetization at  $H = 210$  Oe depicted in figure 3 shows a steep increase below about 1.6 K and is nearly constant at low temperatures, which clearly indicates a ferromagnetic transition.  $T_c$  is determined as 1.6 K from the linear extrapolation of the  $1/\chi-T$  plot between 1.8 and 4.0 K.

The Kondo compounds usually have a negative Weiss temperature at high temperatures, but a  $1/\chi-T$  plot is not available for Sm systems at high temperatures due to the large contribution of the low-lying  $J$ -multiplet. Evidence for a Kondo effect in  $\chi$ , such as a negative Weiss temperature and  $T$ -independent Pauli paramagnetic susceptibility at low temperatures, is not apparently seen for this compound. We therefore measured the magnetic susceptibility of  $\text{La}_{0.4}\text{Sm}_{0.6}\text{Fe}_4\text{P}_{12}$ , where the ferromagnetic transition is suppressed; the result is shown in



**Figure 4.** The magnetization at  $T = 1.8$  K. The magnetization at  $H = 70$  kOe is much smaller than the value  $0.71 \mu_B$  for free  $\text{Sm}^{3+}$ .

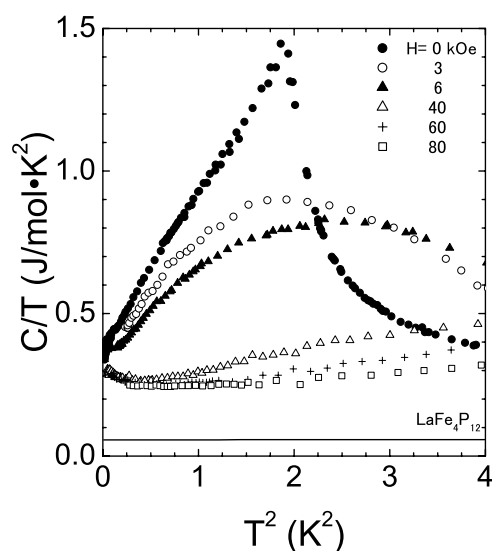
figure 2. It can be seen clearly that  $\chi(T)$  tends to saturate below about 30 K. The Kondo temperature is therefore estimated to be about 30 K.

We show the magnetic field dependence of the magnetization at  $T = 1.8$  K in figure 4. The magnetization shows a steep increase in low fields and a slow increase above about 20 kOe, but does not saturate even at 70 kOe. The magnetization of  $0.15 \mu_B$  at 70 kOe is much smaller than the value  $0.71 \mu_B$  for the free  $\text{Sm}^{3+}$  ion, implying a magnetic moment screened by the Kondo effect, as often observed for Ce-based Kondo systems.

The ferromagnetic transition is also evidenced by the  $T$ -dependence of the specific heat. We show the  $C/T - T^2$  plot in figure 5. The peak due to the phase transition shows a pronounced broadening and shifts to higher temperatures on applying a magnetic field. The value of  $C/T$  extrapolated to  $T = 0$  K is  $370 \text{ mJ mol}^{-1} \text{ K}^{-2}$  at zero field. This value is 6.5 times that for  $\text{LaFe}_4\text{P}_{12}$ , for which a large cyclotron mass is observed by means of the dHvA effect [5, 11]. The magnetic entropy evaluated by subtracting the specific heat of  $\text{LaFe}_4\text{P}_{12}$  reaches about  $1 \text{ J mol}^{-1} \text{ K}^{-1}$  at 1.6 K, which is only about 17% of  $R \ln 2$ . It is worthy of note that the magnetic entropy of the isostructural compound  $\text{SmOs}_4\text{P}_{12}$  reaches  $R \ln 4$  at  $T_N = 4.6$  K, so the CEF ground state is a quartet [12]. We cannot conclude that there is a doublet ground state for  $\text{SmFe}_4\text{P}_{12}$  from just the present data, but, if this is indeed the case, some mechanism such as the anisotropic  $p$ - $f$  mixing in Ce monopnictides, where the  $\Gamma_7$  doublet ground state is realized in CeP and CeAs and the  $\Gamma_8$  quartet ground state in CeSb and CeBi, has to be involved [13].

It should be noted here that the specific heat in zero field below 0.8 K is well described by a  $T^3$ -law, which is different from the theoretical prediction from ordinary ferromagnetic spin wave theory: a  $T^{3/2}$ -law. Such a  $T$ -dependence is also reported for ferromagnetic  $\text{NdFe}_4\text{P}_{12}$  with the same crystal structure [14].

In summary, we have described the magnetic and transport properties of the filled skutterudite  $\text{SmFe}_4\text{P}_{12}$ . The magnetic measurements clearly show that the compound is not a Van Vleck paramagnet but a ferromagnet below 1.6 K. The electrical resistivity, small saturation moment, reduced magnetic entropy and large  $\gamma$ -value strongly suggest that  $\text{SmFe}_4\text{P}_{12}$  is a



**Figure 5.** The thermal variation of  $C/T$  under magnetic fields. The peak shifts to higher temperatures with a pronounced broadening on applying a small magnetic field, as expected in a ferromagnetic transition. A nearly linear behaviour at high fields suggests a heavy Fermi liquid.

Kondo system. The Kondo temperature is estimated to be about 30 K from the magnetic susceptibility of  $\text{La}_{0.4}\text{Sm}_{0.6}\text{Fe}_4\text{P}_{12}$ .

This work was supported by a Grant-in-Aid for Scientific Research(C) from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

## References

- [1] Braun D J and Jeitschko W 1977 *Acta Crystallogr.* **33** 3401
- [2] Takeda N and Ishikawa M 1999 *Physica B* **259–291** 92
- [3] Takeda N and Ishikawa M 2001 *J. Phys.: Condens. Matter* **13** 5971
- [4] Sekine C, Uchiumi T and Shirohani I 1997 *Phys. Rev. Lett.* **79** 3218
- [5] Sugawara H, Matsuda T D, Abe K, Aoki Y, Sato H, Nojiri S, Inada Y, Settai R and Ōnuki Y 2002 *Phys. Rev. B* **66** 134411
- [6] Bauer E D, Frederick N A, Ho P-C, Zapf V S and Maple M B 2002 *Phys. Rev. B* **65** 100506(R)
- [7] Jayaraman A 1979 *Handbook on the Physics and Chemistry of the Rare Earths* vol 2, ed K A Gschneidner Jr and L R Eyring (Amsterdam: North-Holland) p 575
- [8] Kasaya M, Liu B, Sera M, Kasuya T, Endoh D, Goto T and Fujimura T 1985 *J. Magn. Magn. Mater.* **52** 289
- [9] Jeitschko W, Foecker A J, Paschke D, Dewalsky M V, Evers Ch B H, Künnen B, Lang A, Kotzyba G, Rodewald U Ch and Möller M H 2000 *Z. Anorg. Allg. Chem.* **626** 1112
- [10] Takeda N and Ishikawa M 2003 *Proc. LT23 Int. Conf.* (Amsterdam: Elsevier) at press
- [11] Meisner G P, Stewart G R, Torikachvili M S and Maple M B 1984 *Proc. LT17 Int. Conf.* (Amsterdam: Elsevier) p 171
- [12] Giri R, Shimaya Y, Sekine C, Shirohani I, Matsuhira K, Doi Y and Hinatsu Y 2003 *Proc. LT23 Int. Conf.* (Amsterdam: Elsevier) at press
- [13] Takahashi H and Kasuya T 1985 *J. Phys. C: Solid State Phys.* **18** 2697
- [14] Torikachvili M S, Chen J W, Dalichaouch Y, Guertin R P, McElfresh M W, Rossel C, Maple M B and Meisner G P 1987 *Phys. Rev. B* **36** 8660