de Haas-van Alphen effect in SmOs$_4$P$_{12}$

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Abstract. We have succeeded in growing single crystals of SmOs$_4$P$_{12}$ and measured the de Haas-van Alphen effect. An almost spherical Fermi surface, whose cyclotron effective masses are ranging 2.0-2.3 $m_0$, has been observed.

1. Introduction
The filled skutterudite compounds RT$_4$X$_{12}$ (R= rare earth, T= Fe, Ru, Os, and X= pnictogen) have attracted much attention because of the wide variety of transport and magnetic properties, such as unconventional superconductivity, Kondo semiconducting behavior, metal-insulator (M-I) transition, multipole ordering, and heavy fermion (HF) behavior [1]. Among the filled skutterudites, Sm-based compounds have been most intensively investigated next to the Pr-based system, because some of them exhibit unprecedented features, such as the first Sm-based HF ferromagnetism in SmFe$_4$P$_{12}$ [2], the new type of multipolar ordered state in SmRu$_4$P$_{12}$ [3, 4], and the HF behavior robust against magnetic fields in SmOs$_4$Sb$_{12}$ with a ferromagnetic ground state [5]. In order to understand the mechanism of unusual properties, the information of physical properties in related compounds may be helpful.

SmOs$_4$P$_{12}$ was reported to be an antiferromagnetic (AFM) metal with a Néel temperature $T_N = 4.6$ K [6]. The temperature dependence of magnetic susceptibility $\chi(T)$ shows a distinct cusp at $T_N$, which is a typical feature of AFM transition, although the $\chi(T)$ shows upturn below $\sim 3.5$ K. The specific heat shows a pronounced $\lambda$-type peak at $T_N$ that agrees with temperatures of the cusp in $\chi(T)$. The magnetic entropy estimated in zero field reaches $R \ln 4$ at $T_N$, which suggests that the crystalline electric field (CEF) ground state is a quartet $\Gamma_{67}$ [6, 7]. Meanwhile the temperature dependence of electrical resistivity $\rho(T)$ shows Kondo like anomaly; the resistivity increases logarithmically with decreasing temperature from far above $T_N$, which is common feature of most Sm-based skutterudites [8].

In the previous works on SmOs$_4$P$_{12}$, the physical properties were investigated on the polycrystalline samples synthesized by the high-pressure technique. Recently, we have succeeded in growing single crystals of SmOs$_4$P$_{12}$ for the first time. In this work, we report on the measurements of the de Haas-van Alphen (dHvA) effect to deepen the understanding of electronic state.
2. Experiment

Single crystals of SmOs$_4$P$_{12}$ were grown by a tin-flux method which is basically the same as described in elsewhere. The raw materials were 3N (99.9% pure)-Sm, 4N-Os, 6N-P and 5N-Sn. The size of single crystals (∼0.2 mm in length) are rather smaller than those of other ROs$_4$P$_{12}$. Crystal structure of the single crystal was confirmed by a back Laue X-ray diffraction method. The dHvA experiment was performed by the torque method using the micro-cantilever with the modulation field of $h \sim 100$ Oe (a modulation frequency of $\omega/2\pi = 11$ Hz) in a top loading dilution refrigerator with a 17 T superconducting magnet [9, 10].

3. Results and discussion

Figure 1(a) shows a typical recorder trace of the dHvA oscillations and Fig. 1(b) shows its fast Fourier transformation (FFT) spectrum at 59 mK for $H \parallel [001]$ in SmOs$_4$P$_{12}$.

One dHvA branch named as $c$ with the dHvA frequency $F \sim 1.1 \times 10^3$ T was observed over the whole field angles with a slight angular dependence, indicating almost spherical Fermi surface (FS). The angular dependence is almost the same as that of $\gamma$-branch in LaOs$_4$P$_{12}$ except the $\sim 17\%$ smaller magnitude [11]. The $\gamma$-branch in LaOs$_4$P$_{12}$ is well identified as the 47th-band FS by the band structure calculation [12]. The closeness of the FS to that in LaOs$_4$P$_{12}$ suggests the localized nature of 4$f$-electrons in SmOs$_4$P$_{12}$, that is consistent with the trivalent state of Sm ions suggested by the result of magnetic susceptibility [6]. However, any other branches found in LaOs$_4$P$_{12}$ were not observed. Such a result is the same as that in SmFe$_3$P$_{12}$ [13], in which...
the multiply-connected 48th-band FS with heavy mass of \( \sim 59m_0 \) was expected. Differently from \( \text{SmFe}_4\text{P}_{12} \), \( \text{SmOs}_4\text{P}_{12} \) orders antiferromagnetically below 4.6 K. The \( H - T \) phase diagram was determined by the specific heat measurements under the magnetic fields \([6, 3]\), in which the AFM ordered state survives up to 8 T at 4 K. Therefore, it is inferred that the topology of FS in paramagnetic state changes in the AFM state due to the magnetic Brillouin zone. In the present dHvA experiments, we observed an evident metamagnetic anomaly at the limited filed angles (52° \( \sim \) 61°) around \( H \parallel [111] \) (not shown); e.g. the metamagnetic anomaly was observed at \( H_m = 13.2 \) T for field along the \( H \parallel [111] \) at 97 mK. However, the dHvA spectrum shows no essential change below and above \( H_m \) except the enhancement of amplitude above \( H_m \), suggesting that the reconstruction of magnetic Brillouin zone is not the essential reason for undetecting 48th-band FS in \( \text{SmOs}_4\text{P}_{12} \).

From the temperature dependence of the dHvA amplitude, we have determined the cyclotron effective mass \( m_c^* \) for the three principal directions as shown in Fig. 3. The \( m_c^* \) of \( \text{SmOs}_4\text{P}_{12} \) is roughly twice as large as that of \( \text{LaOs}_4\text{P}_{12} \) as shown in Table 1. Such a enhancement of \( m_c^* \) is frequently observed in the magnetic materials and ascribable to the electron-magnon interaction \([14]\). In \( \text{LaOs}_4\text{P}_{12} \) the larger FS (\( \alpha \)-branch) with \( m_c^* = 4.7m_0 \), which corresponds to the open orbit of 48th-band FS, was observed around \( H \parallel [001] \). If we simply estimate the effective mass of 48th-band FS for \( H \parallel [001] \) in \( \text{SmOs}_4\text{P}_{12} \) assuming the same mass enhancement; i.e., \( (m_c^*)_{\text{SmOs}_4\text{P}_{12}}/(m_c^*)_{\text{LaOs}_4\text{P}_{12}} = (m_c^*)_{\text{LaOs}_4\text{P}_{12}}/(m_c^*)_{\text{LaOs}_4\text{P}_{12}} \), the larger mass of \( m_c^* \sim 9.4m_0 \) is expected in \( \text{SmOs}_4\text{P}_{12} \), which may marginal value for detecting the dHvA signal in the present experimental condition.

### Table 1. Comparison of the dHvA frequency \( F \) and the cyclotron effective mass \( m_c^* \) between \( \text{LaOs}_4\text{P}_{12} \) (\( \gamma \)-branch) and \( \text{SmOs}_4\text{P}_{12} \) (\( c \)-branch). Data of \( \text{LaOs}_4\text{P}_{12} \) are sited from Ref. [11].

| \( H \parallel [101] \) | \( H \parallel [001] \) | \( H \parallel [111] \) |
|---|---|---|
| \( F \times 10^3 \) T | \( m_c^*(m_0) \) | \( F \times 10^3 \) T | \( m_c^*(m_0) \) | \( F \times 10^3 \) T | \( m_c^*(m_0) \) |
| \( \text{LaOs}_4\text{P}_{12} \) | 1.37 | 1.3 | 1.29 | 1.1 | 1.39 | 1.2 |
| \( \text{SmOs}_4\text{P}_{12} \) | 1.14 | 2.3 | 1.10 | 2.2 | 1.17 | 2.0 |

### Figure 3. The semi-logarithmic plot of the dHvA amplitude \( A \) vs. temperature for \( c \)-branch in \( \text{SmOs}_4\text{P}_{12} \). \( \lambda \) in the vertical-axis label is a constant \( \lambda = 2\pi^2ck_B/eh \).

### Figure 4. Dingle plot for \( c \)-branch in \( \text{SmOs}_4\text{P}_{12} \). \( J_1(x) \) in the vertical-axis label is the Bessel function with \( x = 2\pi Fh/H^2 \) due to the modulation method.
We have also determined the Dingle temperature $T_D(=\hbar/2\pi k_B\tau)$, which is inversely proportional to the scattering lifetime $\tau$, from the field dependence of dHvA amplitude as shown in Fig. 4. The Dingle temperature is $T_D=2.0$ K for $c$-branch from which we can estimate the mean free path $\ell=580\text{Å}$ using the relations: $S_F=\pi k_F^2$, $\hbar k_F=m^*_c v_F$, and $\ell=v_F\tau$, where $k_F$ is half of the caliper dimension of a circular $S_F$ and $v_F$ is the Fermi velocity. For the comparison, we have also estimated the Dingle temperature and mean free path in LaOs$_4$P$_{12}$ for the similar condition at 100 mK. The estimated values are $T_D=0.093$ K and $\ell=27000\text{Å}$, respectively. The mean free path in SmOs$_4$P$_{12}$ is roughly 1/50 of that in LaOs$_4$P$_{12}$, that is also the reason why the 48th-band FS could not be detected in SmOs$_4$P$_{12}$. Taking into account the same manner of sample preparation, such a large difference of mean free path between these compounds is not understandable only from the difference of sample quality due to the impurity and/or defect in lattice. The magnetic scattering associated with Kondo like behavior in $\rho(T)$ may be a possible origin for the smaller mean free path in SmOs$_4$P$_{12}$.

In summary, we have succeeded in observing the dHvA effect in SmOs$_4$P$_{12}$. An almost spherical FS, which is corresponding to the 47th-band hole FS, was observed. In order to understand the electric and magnetic state further, the measurements under higher magnetic fields including anisotropy are necessary.

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References

[1] Sato H, Sugawara H, Aoki Y and Harima H, 2009 Handbook of Magnetic Materials vol 18, ed K H J Buschow (Amsterdam: Elsevier) p 1.
[2] Takeda N and Ishikawa M 2003 J. Phys. Condens. Matter 15 L229.
[3] Matsuhira M, Hinatsu Y, Sekine C, Togashi T, Maki H, Shirotani I, Kitazawa H, Takamatsu T, Kido G 2002 J. Phys. Soc. Jpn. 71 (Suppl.) 237.
[4] Yoshizawa M, Nakanishi Y, Oikawa M, Sekine C, Shirotani I, Saha S R, Sugawara H, Sato H 2005 J. Phys. Soc. Jpn. 74 2141.
[5] Sanada S, Aoki Y, Aoki H, Tsuiya A, Kikuchi D, Sugawara H, Sato H 2005 Phys. Soc. Jpn. 74 246.
[6] Giri R, Sekine C, Shimaya Y, Shirotani I, Matsuhira K, Doi Y, Hinatsu Y, Yokoyama M, Amitsuka H 2003 Physica B 329-333 458.
[7] Matsuhira K, Doi Y, Wakeshima M, Hinatsu Y, Amitsuka H, Shimaya Y, Giri R, Sekine C, Shirotani I 2005 J. Phys. Soc. Jpn. 74 1030.
[8] Sato H, Kikuchi D, Tanaka K, Aoki H, Kuwahara K, Aoki Y, Kohgi M, Sugawara H, Iwasa K 2007 J. Magn. Magn. Mater. 310 188.
[9] Settai R, Kawai T, Endo T, Muranaka H, Doi Y, Ōmuki Y and Harima H 2008 J. Phys. Soc. Jpn. 77 (Suppl. A) 345.
[10] Kawai T, Muranaka H, Endo T, Dung N D, Doi Y, Ikeda S, Matsuda T D, Haga Y, Harima H, Settai R and Ōmuki Y 2008 J. Phys. Soc. Jpn. 77 064717.
[11] Sugawara H, Iwahashi Y, Magishi K, Saito T, Koyama K, Harima H, Kikuchi D, Sato H, Endo T, Settai R, Ōmuki Y 2009 Phys. Rev. B 79 035104.
[12] Harima H and Takegahara K 2008 Physica B 403 906.
[13] Kikuchi D, Sugawara H, Tanaka K, Aoki H, Kobayashi M, Sanada S, Kuwahara K, Aoki Y, Shishido H, Settai R, Ōmuki Y, Harima and H, Sato H 2008 J. Phys. Soc. Jpn. 77 114705.
[14] Ōmuki Y and Hasegawa A 1995 Hand book on the Physics and Chemistry of Rare Earth, vol 20, chap 135, ed. K A Gschneidner Jr and L Eyring (Amsterdam, Elsevier) p 1.