DE HAAS–VAN ALPHEN EFFECT IN HEAVY FERMION SUPERCONDUCTOR $PrOs_4Sb_{12}$*

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We have succeeded in observing the de Haas–van Alphen (dHvA) effect in $PrOs_4Sb_{12}$. The Fermi surface topology is similar to the reference compound $LaOs_4Sb_{12}$, indicating the localized character of $4f$-electrons. The cyclotron effective mass, enhanced by about four times compared with $LaOs_4Sb_{12}$, is a direct evidence of the strong electron correlation in this compound.

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1. Introduction

Filled skutterudite compounds RT_4X_{12} (R=rare-earth; T=Fe, Ru and Os; X=P, As and Sb) have attracted much attention because of their interesting anomalous physical properties, such as metal-insulator transition in $PrRu_4P_{12}$ [1] and unusual heavy fermion (HF) behavior in $PrFe_4P_{12}$ [2–4]. In the latter, extraordinarily enhanced effective mass ($m_{\rm c}^* = 81m_0$) and a large difference of the Fermi surface (FS) topology with $LaFe_4P_{12}$ have been confirmed by the de Haas–van Alphen (dHvA) experiments [4]. It is believed that the large c–f hybridization originated from the unique crystal structure of filled skutterudite creates such anomalous properties.

Recently, $PrOs_4Sb_{12}$ was reported to show superconductivity below $T_{\rm C} =$ 1.85 K [5]. The large specific heat jump at $T_{\rm C}$, $\Delta C/T_{\rm C}\sim$ 500 mJ/K$^2\cdot$ mol,

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suggests the strong electronic correlation in this compound, that is the first example of a Pr-based HF-superconductor. To understand the unusual properties, the knowledge of electrical structure is essential. In this paper, we report the first dHvA experiment in $PrOs_4Sb_{12}$, which is the most powerful tool to clarify the FS precisely along with direct evidence of an enhanced effective mass.

2. Experimental

Single crystals of $PrOs_4Sb_{12}$ and reference $LaOs_4Sb_{12}$ were grown by a Sb-self-flux method with excess Sb (ratio R:Os:Sb=1:4:20) [5]. High-purity materials, 4N (99.99% pure)-Pr, 4N-La, 3N-Os and 6N-Sb, were used for the crystal growing. Typical single crystals were of cubic or rectangular shape with a largest dimension of about 3 mm. The residual resistivity ρ_0 and the residual resistivity ratio (RRR) of the present samples are $\rho_0 = 8\mu\Omega\cdot$cm and RRR = 55 for $PrOs_4Sb_{12}$, and $\rho_0 = 2.8\mu\Omega\cdot$cm and RRR = 100 for $LaOs_4Sb_{12}$, indicating high quality of the samples. The dHvA experiments were performed in a top loading dilution refrigerator system with a 17 T superconducting magnet cooled down to 30 mK. The dHvA signals were detected by means of the conventional field modulation method with a low frequency ($\sim$ 10 Hz).

3. Results and discussion

Fig. 1 shows (a) the typical dHvA oscillations and (b) its fast Fourier transformation (FFT) spectra both in $LaOs_4Sb_{12}$ and $PrOs_4Sb_{12}$.

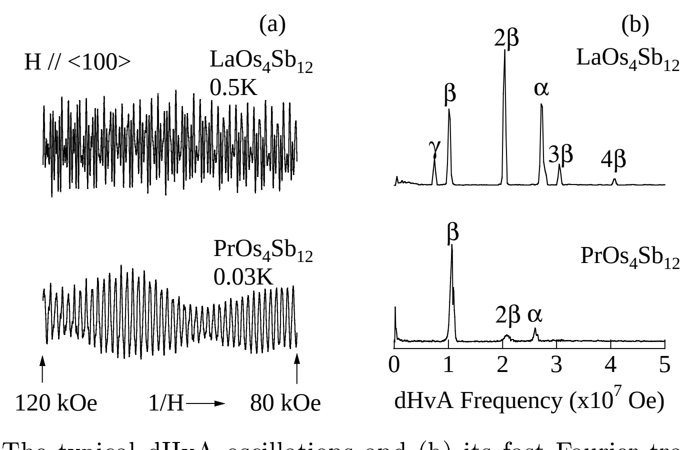

Fig. 1. (a) The typical dHvA oscillations and (b) its fast Fourier transformation (FFT) spectra both in $LaOs_4Sb_{12}$ and $PrOs_4Sb_{12}$.

For $LaOs_4Sb_{12}$, there are at least three dHvA frequency branches denoted as α, β and γ. 2β, 3β and 4β are the β-branch harmonics. The results are in good agreement with the band structure calculation [6]. The frequency branches of $PrOs_4Sb_{12}$ [Fig. 1 (b)] show good agreement with those of $LaOs_4Sb_{12}$, indicating the shapes of FS are close to each other. The result suggests a well localized character of $4f$-electrons in $PrOs_4Sb_{12}$. Note that the small spin-splitting in dHvA frequencies is observed in $PrOs_4Sb_{12}$, which originates from up- and down-spin bands split by the exchange interaction with the induced magnetic moment.

From the temperature dependence of the dHvA amplitude A, we can estimate the cyclotron effective mass m_c^* for β-branch as shown in Fig. 2.

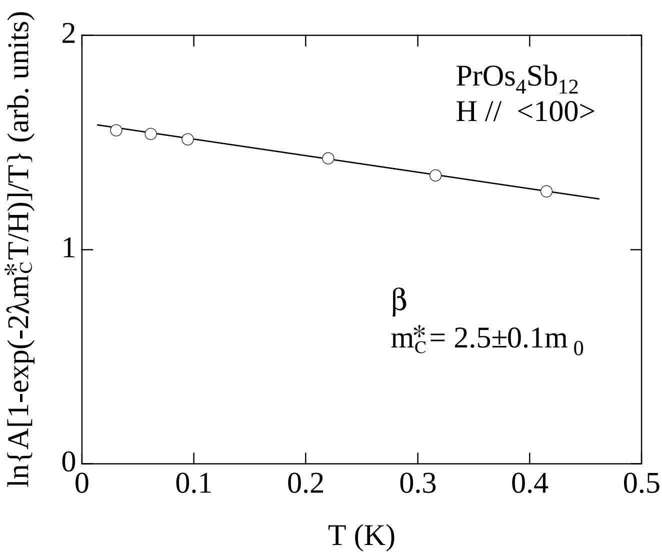

Fig. 2. The semi-logarithmic plot of the reduced dHvA amplitude A *vs* temperature for β-branch in $PrOs_4Sb_{12}$. λ in the vertical-axis label is a constant $\lambda = 2\pi^2 ck_B/e\hbar$. The m_c^* was estimated at around 133 kOe.

The m_c^* is found to be enhanced by about four times compared with $LaOs_4Sb_{12}$. Data of the dHvA frequency and m_c^* for β-branch are listed in Table I for $LaOs_4Sb_{12}$ and $PrOs_4Sb_{12}$. From the comparison of the Sommerfeld coefficient between $LaOs_4Sb_{12}$(39mJ/K^2·mol [7]) and $PrOs_4Sb_{12}$ (500mJ/K^2·mol [5]), the observed m_c^* is too small for $PrOs_4Sb_{12}$. If we simply estimate from the FS volume and m_c^* in the present experiments assuming a spherical FS, the Sommerfeld coefficient should be $\sim$ 20mJ/K^2·mol. This large discrepancy suggests the existence of other FS(s) with heavy mass. The large effective mass $\sim 50m_0$ was also inferred from the slope of the up-

TABLE I

Comparison of the dHvA frequency F and the cyclotron effective mass m_c^* for β-branch between $LaOs_4Sb_{12}$ and $PrOs_4Sb_{12}$ for $H\|\langle 100\rangle$.

	$LaOs_4P_{12}$		$PrOs_4P_{12}$	
Branch	$F(\times 10^7$ Oe)	$m_c^*(m_0)$	$F(\times 10^7$ Oe)	$m_c^*(m_0)$
β	1.02	0.71	1.07	2.5

per critical field near T_C [5]. Under the present experimental conditions, the dHvA signal for such a heavy FS is hardly observable. The localized character of $4f$-electrons in $PrOs_4Sb_{12}$ is the same as for $PrRu_4Sb_{12}$ for which excellent agreement of the dHvA branches with $LaRu_4Sb_{12}$ was clarified [8]. However, the large mass enhancement in $PrOs_4Sb_{12}$ is in sharp contrast to $PrRu_4Sb_{12}$; *i.e.*, $m_c^* = 1.5 \sim 1.8m_0$ and the mass enhancement compared with $LaRu_4Sb_{12}$ is almost negligible. For $PrOs_4Sb_{12}$ and $PrFe_4P_{12}$, the crystal field (CEF) ground state of Pr^{3+} is believed to be the Γ_3 non-magnetic doublet with quadrupole moments, while the Γ_1 singlet is inferred for $PrRu_4Sb_{12}$. Therfore, the quadrupolar interaction is thought to play an important role for the HF behavior and also the HF-superconductivity.

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