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Shubnikov-de Haas Oscillation in $\mathrm{PuIn_3}$

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The Fermi surface of $\mathrm{PuIn_3}$ is investigated using flux-grown single crystals. Shubnikov-de Haas (SdH) oscillations were detected by means of the skin-depth measurement using a proximity-detector-oscillator circuit. Angular dependence of the SdH frequency which corresponds to the extremal cross-sectional area of Fermi surface agrees well with the previous magnetic susceptibility measurement using conventional field-modulation method. The SdH oscillation suddenly vanishes when the magnetic field is tilted from the cubic [111] direction.

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I. INTRODUCTION

Rare-earth and actinide compounds are extensively studied because of their unusual behavior including heavy fermion formation and its superconductivity, various magnetic / multipolar orderings or Kondo-insulating behavior. While the $4f$ electrons in rare-earth elements can be regarded as localized electrons except for Ce and Yb, itinerant character is dominant in light actinide elements, as demonstrated in the Wigner-Seitz radius determined experimentally for light actinide elemental metals. This is most likely due to the larger spatial extent of the wave function of $5f$ electrons compared to that of $4f$ electrons. However, this tendency is no more valid for heavy actinide elements. Namely, Am and heavier actinide metals have almost constant Wigner-Seitz radius similar to that in lanthanide metals.

Plutonium is just located between these two limits. The Wigner-Seitz radius of plutonium at room temperature is slightly larger than neptunium where $5f$ electrons are almost delocalized. It changes discontinuously with increasing temperature and finally approaches to the radius corresponding to the localized $5f$ state.

One of the experimental methods to determine the $5f$ electronic state is the Fermi surface measurement using the de Haas-van Alphen oscillations which can determine the precise cross sectional area of Fermi surface perpendicular to external magnetic field. If the $5f$ electrons participate in the conduction band, they contribute to the Fermi surface volume. The first dHvA measurement on plutonium containing material was conducted using the field modulation technique. The quantum oscillation was clearly observed for the plutonium compound $\mathrm{PuIn_3}$ [1]. $\mathrm{PuIn_3}$ crystallizes in the cubic $\mathrm{AuCu_3}$-type crystal structure. This earlier study reported that this compound shows a heavy fermion behavior.

It was also clarified that the radiation effect due to the alpha decay of plutonium atoms significantly reduces the oscillation amplitude. The observed dHvA signal was compared with the band calculations under the assumption of itinerant $5f$ electron picture.

The aim of the present study is to confirm the quantum oscillation from the Fermi surface of $\mathrm{PuIn_3}$ using the skin-depth measurement using a fresh single crystal to avoid radiation damage.

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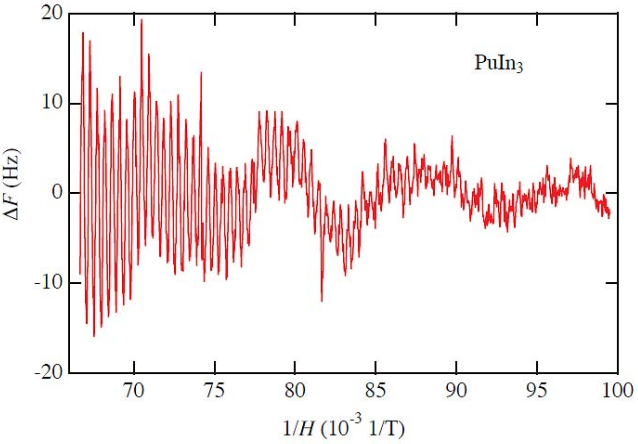

Fig. 1. (Color online) SdH oscillation measured on PuIn$_3$.

II. EXPERIMENTS AND DISCUSSION

Single crystal samples of PuIn$_3$ were prepared using self-flux method at LANL.

For the quantum oscillation measurements, a magnetic torque measurement using a cantilever and an rf skin-depth measurement were performed. The sample was cooled down to about 0.5 K using a ^{3}He cryostat under a static magnetic field up to 15 T. The sample rotator was used to measure the field angle dependence. In order to avoid the accumulation of radiation damage due to alpha decay of ^{239}Pu, the flux grown crystals were transported to the measurement facility in a few hours.

For the magnetic torque measurement, we used a metallic cantilever. The displacement of the cantilever due to the sample's torque was monitored through the change in the capacitance between the cantilever and the reference electrode. In this measurement, however, quantum oscillations were observed only from the indium metal possibly included in the crystal grain boundary during the flux-growth process. It was not possible to extract the signal from PuIn$_3$. On the contrary, the skin-depth measurement described below successfully detected the signal from PuIn$_3$.

In the skin-depth measurement, the sample located in a pick-up coil was put in a metallic container to avoid radioactive contamination. The rf skin-depth was measured using a proximity detector oscillator (PDO) technique to detect quatum oscillations of resistivity as a function of magnetic field [2]. In the skin-depth measurement, only the sample volume corresponding to the skin-depth of the order of 10 μm contributes to the signal. It is therefore expected that the skin-depth measurement is insensitive to the foreign signal coming from the inclusions deep inside the sample. In fact we detected the intrinsic signal from PuIn$_3$ using the skin-depth measurement and the signal from indium can be neglected.

We succeeded in observing a clear SdH oscillation as a function of inverse magnetic field. The raw data has a linear frequency shift as a function of magnetic field. After subtraction of the background, clear oscillation was obtained, as shown in Fig. 1. The SdH frequency is proportional to the cross sectional area of the Fermi surface along the magnetic field direction. Therefore the shape of the Fermi surfaces can be obtained by measuring the angular dependence of the SdH frequency, as shown in Fig. 2. The SdH oscillation was observed when the magnetic field is applied around the [111] axis. The present results (triangle) reproduce well the previous study [2] (solid curve) measured using the conventional field modulation technique. The obtained SdH frequency can be explained by the band calculations assuming the 5f electrons are delocalized. The obtained cyclotron mass which was determined from the temperature dependence of the SdH amplitude is also consistent with the previous study.

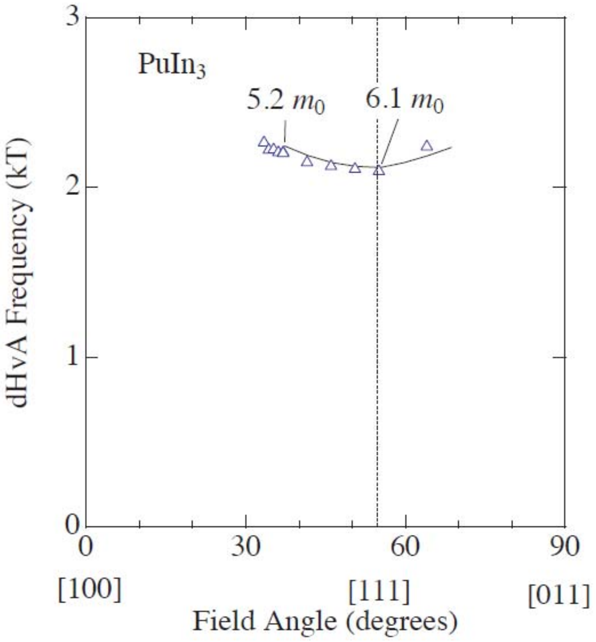

Fig. 2. (Color online) Angular dependence of the SdH frequency in PuIn$_3$. Triangles correspond to the present results, while solid curve is taken from ref. 1.

The SdH oscillation suddenly vanishes at angles around 30 and 65 degrees which cannot be explained from the calculated Fermi surface properties assuming a non-magnetic ground state.

It was recently reported that PuIn$_3$ has a possible antiferromagnetic ordering at 14 K [3]. Although the details of the ordering have not been clarified, one can assume a modification of the Brillouin zone in the ordered state. Among the actinide analogue, UIn$_3$ and NpIn$_3$ are reported to order magnetically. UIn$_3$ is an antiferromagnet with a propagation vector (1/2, 1/2, 1/2) [4]. On the other hand, NpIn$_3$ has two phase transitions. The first one is a ferromagnetic one at 14 K with spontaneous moment along the [111] direction. It changes into an antiferromagnetic ground state with a long-period modulation (3/8, 3/8, 3/8) [5]. In PuIn$_3$, according to the band calculations, a small pocket Fermi surface is located at the Γ point. The observed SdH frequency agrees well with that expected from this Fermi surface. The size of the cross-sectional area is fairly smaller than the Brillouin zone. Assuming a simple antiferromagnetism similar to that of UIn_3, the present Fermi surface is even smaller than the magnetic Brillouin zone. It is likely for the present Fermi surface to survive the magnetic ordering.

For further discussion, detailed understanding of the ordered phase is necessary. Physical properties in both paramagnetic and ordered states are in progress.

III. CONCLUSIONS

We have observed Shubnikov-de Haas oscillations in a plutonium compound $PuIn_3$ using the PDO technique. PDO technique is very powerful in detecting quantum oscillation of flux-grown materials. The angular dependence of the SdH and the cyclotron mass well agree with the previous study. Unusual change in the oscillation amplitude is possibly related to the phase transition at 14 K.

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