De Haas - van Alphen effect and Fermi surface properties in $U_{1-x}Th_xPd_3$ alloys

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Abstract. We report the first observation of the de Haas-van Alphen (dHvA) oscillations in U-based alloys $U_{1-x}Th_xPd_3$ up to $x = 0.5$. In $U_{0.5}Th_{0.5}Pd_3$, the dHvA oscillations $\beta_2$ can be observed in the (1100) plane and the frequencies of the oscillations as well as their angular dependences are found to be nearly the same as those in UPd$_3$. On the other hand, the effective mass of the $\beta_2$ oscillation is found to decrease with $x$ from 1.78$m_0$ of UPd$_3$ to 1.32$m_0$ of $U_{0.5}Th_{0.5}Pd_3$. From these observations we argue that the f electrons in UPd$_3$ are nearly localized and the interactions between the f electrons and the conduction electrons are weak.

1. Introduction

There are a number of rare earth and actinide compounds where higher order multipoles (quadrupolar, octupolar, etc) play a key role in the physical properties. Although a great number of studies have been performed experimentally as well as theoretically, the mechanism how the multipolar interactions are mediated has not been clarified yet. To investigate the mechanism of multipolar order, it is useful to replace the magnetic element by other elements and see the effect of the substitution on the properties. On the other hand, some previous experiments such as that on PrPb$_3$ [1] suggest that the quadrupolar interaction is mediated via the conduction electrons. Then it is very useful to reveal the change in the electronic structure with the substitution together with the change in the properties. In this paper, we report the first successful observation of the de Haas-van Alphen (dHvA) effect in $U_{1-x}Th_xPd_3$ to investigate the mechanism of the quadrupolar order in UPd$_3$ and $U_{1-x}Th_xPd_3$.

UPd$_3$ crystallizes in the double-hexagonal close-packed structure. Previous studies on UPd$_3$ indicate that four phase transitions occur below 8K including antiferroquadrupolar ordering [2]. On the other hand, ThPd$_3$ has no 5f electron and crystallizes in the same crystal structure as UPd$_3$. The unit cell volume of ThPd$_3$ is larger than that of UPd$_3$ by about 5.3%.

The dHvA effect measurement is a powerful method to measure the properties of conduction electrons. It is very difficult, in general, to observe the dHvA signal in alloys because of the scattering of the conduction electrons by the doped elements. However, there are already several successful attempts to observe the dHvA signal over the whole concentration range in alloys, such as Ce$_x$La$_{1-x}$B$_6$ [3, 4, 5], Ce$_x$La$_{1-x}$Sb [6], Ce$_x$La$_{1-x}$RhIn$_5$ [7], Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ [8], Pr$_x$La$_{1-x}$B$_6$ [4]. In these compounds, the 4f electrons are well localized and the substitution of Ce or Pr by La does not seem to cause serious scattering of the conduction electrons. Moreover, the Fermi
The dHvA effect measurements were performed in several alloys with different Th concentrations. Results and Discussion

Field modulation method in a dilution refrigerator under magnetic fields up to 16T. To label the sample, the dHvA effect measurements were performed by using the conventional technique. The dHvA effect will also give us deeper insights in the nature of the 5f electrons in the actinide compounds.

2. Experimental

Single crystals of $U_{1-x}Th_xPd_3$ were grown by using the Czochralski pulling method in a tetra-arc furnace. The X-ray diffraction and Electron Probe Micro Analysis confirmed that there were no other phases and Th were distributed uniformly. We used the nominal concentration of Th to label the sample. The dHvA effect measurements were performed by using the conventional field modulation method in a dilution refrigerator under magnetic fields up to 16T.

3. Results and Discussion

The dHvA effect measurements were performed in several alloys with different Th concentrations from UPd$_3$ to ThPd$_3$. The results of UPd$_3$ are consistent with those of previous reports [9, 10, 11]. Figures 1 (a) and (b) show the dHvA signal in $U_{0.5}Th_{0.5}Pd_3$ at 100mK and its Fourier spectrum, respectively. The field direction is tilted by 30 degree from [1120] to [0001] in the (1100) plane. The $\beta_2$ oscillation is clearly observed. This is the first observation of the dHvA oscillation in U-based alloys. Any other oscillations than the $\beta_2$ oscillation are not detected in this plane in $U_{0.5}Th_{0.5}Pd_3$. Although a peak around 2100T, whose frequency is close to that of $\gamma$ oscillation in UPd$_3$, is observed in Fig.1 (b), it does not arise from a dHvA oscillation because its magnitude is comparable to the noise and we could not detect any clear peaks around this frequency for other orientations. In Fig.1 (c), we show the FFT spectrum of the dHvA signal in UPd$_3$ detected with a sample whose volume is almost the same as that of $U_{0.5}Th_{0.5}Pd_3$. The panels (b) and (c) indicate that the amplitude of the $\beta_2$ oscillation in $U_{0.5}Th_{0.5}Pd_3$ is about 50 times smaller than that in UPd$_3$. However it is noted that the suppression of signal amplitude is extremely smaller than those observed in normal metal alloys: Addition of 1% alloy element decreases the signal amplitude by several orders of magnitude and makes the dHvA signal unobservable.

Figure 2 shows the angular dependences of $\beta$ frequencies in $U_{0.5}Th_{0.5}Pd_3$ in the (100) plane together with those in UPd$_3$ for comparison. The field direction is rotated from [1120] to [0001] in the (1100) plane. According to the band structure calculation by Tokiwa et al. [10], $\beta$ branch is one of the main Fermi surfaces and originates from the closed hole sheet centered at $\Gamma$ point. For the $\beta$ branches, the shape and frequency in $U_{0.5}Th_{0.5}Pd_3$ are very similar to those in UPd$_3$. On the other hand, the effective mass of the $\beta_2$ frequency decreases from 1.78$m_0$ of UPd$_3$ to 1.32$m_0$ of $U_{0.5}Th_{0.5}Pd_3$ for the field direction tilted by 30 degree from [1120] to [0001].

The present results for $U_{0.5}Th_{0.5}Pd_3$ together with those of $U_{1-x}Th_xPd_3$ with $x < 0.5$ suggest that the Fermi surface topology of $U_{1-x}Th_xPd_3$ with $x < 0.5$ is almost the same as that in UPd$_3$. The successful observations of the dHvA signal even in the high concentration alloy of $U_{0.5}Th_{0.5}Pd_3$ also confirm that the 5f electrons are well localized and the Fermi surface may not change significantly with the Th concentration up to at least $x = 0.5$. On the other hand, the value of the effective mass and its decrease with Th content imply that the interactions between the 5f electrons and the conduction electrons are appreciable but may not be strong.

Our recent DC magnetization measurements indicate that the quadrupolar order is rapidly destroyed with the Th substitution and there is no trace of the quadrupolar transition above 1.8...
K for $x > 0.1$. Since the Fermi surface topology and the interactions between the 5f electrons and the conduction electrons are not likely to change significantly for $x > 0.1$, we conjecture that the RKKY like mechanism may not be responsible for the quadrupolar order in $\text{U}_{1-x}\text{Th}_x\text{Pd}_3$.

In summary, we have grown high-quality single crystals of $\text{U}_{1-x}\text{Th}_x\text{Pd}_3$. We are successful for the first time in observing the dHvA effect in U-based alloys. The present results indicate that the f electrons are well localized and the interactions between the f electrons and the conduction electrons are weak in $\text{U}_{1-x}\text{Th}_x\text{Pd}_3$ alloys.

![dHvA Signal](image1)

**Figure 1.** DHvA signal (a) and its Fourier spectrum (b) in $\text{U}_{0.5}\text{Th}_{0.5}\text{Pd}_3$ at 100mK and (c) Fourier spectrum of the dHvA signal in UPd$_3$ at 100mK. The field direction is tilted by 30 degree from [1120] to [0001] in the (1100) plane.

![dHvA Frequency](image2)

**Figure 2.** Angular dependences of the dHvA frequencies of the $\beta_1$ and $\beta_2$ oscillations in UPd$_3$ (○) and $\text{U}_{0.5}\text{Th}_{0.5}\text{Pd}_3$ (■) in the (1100) plane. The field direction is rotated from [1120] to [0001].
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