Enhancement of the cyclotron effective mass in $U_{0.03}Th_{0.97}Ru_2Si_2$

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Abstract. Electronic states of a dilute uranium alloy $U_{0.03}Th_{0.97}Ru_2Si_2$ have been investigated by using de Haas-van Alphen (dHvA) measurements on single crystal samples. Quantum oscillations were successfully observed for the field along the principal axes. The dHvA frequency of the observed branches roughly agrees with those of the reference compound $ThRu_2Si_2$, indicating the change of Fermi surface volume is not significant. On the other hand, the dHvA amplitude is strongly diminished compared to $ThRu_2Si_2$. Furthermore, cyclotron effective masses for corresponding branches are strongly enhanced. The latter effects are indicative of the strong scattering as well as the mass renormalization due to $5f$ moments.

1. Introduction

Electron correlations and their effect on physical properties are one of the central issues in condensed-matter physics. In particular, electrons with large degree of freedom in $d$- or $f$-elements are extensively studied because of their unusual behavior. Among them, $URu_2Si_2$ is known to show heavy fermion superconductivity coexisting with another phase transition whose order parameter is still unidentified.

$ThRu_2Si_2$ crystallizing in the tetragonal $ThCr_2Si_2$-type structure is regarded as a reference compound to the correlated electron system $URu_2Si_2$ [1]. $Th$ usually takes a tetravalent state where the $5f$ shell is empty. The electronic structure of $ThRu_2Si_2$ was previously investigated by the angle resolved photoemission spectroscopy [2], and more recently, by the dHvA technique [3] and the overall Fermi surface topology was well explained by the band calculations. On the other hand, the electronic state of $URu_2Si_2$ is highly different from $ThRu_2Si_2$ and shows enhanced effective masses and a small carrier number due to the symmetry lowering due to the so-called hidden-order transition [4].

The low temperature bulk properties of $ThRu_2Si_2$ doped with dilute uranium have been investigated on single crystals [5]. The uranium atom maintains magnetic moment as deduced...
from the paramagnetic Curie-Weiss behavior at high temperatures. At low temperature, however, a deviation from Curie-Weiss law, enhancement of the electronic specific heat coefficient $\gamma$ and anomalous decrease of the electrical resistivity were observed. They were discussed in terms of multichannel Kondo interaction [5]. The low temperature characteristics are indicative of an enhanced contribution of $5f$ component to conduction bands. It is therefore interesting to observe the change in the Fermi surface properties upon doping $5f$ electrons into nonmagnetic ThRu$_2$Si$_2$.

In the present study, we report the first observation of the quantum oscillation signal in U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$ and the modification of the electronic state of ThRu$_2$Si$_2$ upon replacing Th site with U.

2. Experimental

Single crystal samples of U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$ have been grown using the Czochralski pulling method under argon gas atmosphere in a tetra-arc furnace. The single crystal was then annealed in an evacuated quartz ampoule for a week. The composition and homogeneity are confirmed by an electron probe microanalysis. The lattice parameters and crystallographic parameters were refined by single crystal X-ray diffraction. U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$ has significantly smaller lattice parameters $a = 4.191$ and $c = 9.736$ Å than those of ThRu$_2$Si$_2$ $a = 4.196$ and $c = 9.750$ Å at room temperature, reflecting the smaller atomic radius of uranium.

The paramagnetic susceptibility was measured in a SQUID magnetometer (Quantum Design MPMS). All these analyses show that dilute uranium is incorporated in the host ThRu$_2$Si$_2$. The quality of the sample were also checked by measuring the electrical resistivity using a dc four-probe method. The resistivity ratio between 4 K and 300 K is about 10, in agreement with the previous study. It should be noted that the resistivity steeply decreases below 4 K because of the temperature-dependent scattering by dilute uranium moments. Measurements at very low temperature are required to evaluate the residual resistivity.

Specific heat down to 2 K was measured using the relaxation method (Quantum Design PPMS). The dHvA experiments were performed using a standard field modulation technique in a 17 T superconducting magnet and $^3$He-$^4$He dilution refrigerator.

3. Results and Discussion

Figure 1 shows the temperature dependence of the specific heat divided by temperature $C/T$ of U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$. As a reference without $5f$ electrons, the data for ThRu$_2$Si$_2$ are also plotted. Specific heats of the two compounds agree to each other above 20 K. Below this temperature, however, the data for the dilute alloy shows significantly larger specific heat. This result indicates that the specific heat contains magnetic contribution from $5f$ electrons of uranium only below a characteristic temperature of about 20 K.

The magnetic susceptibility along the [001] direction measured on the same sample (not shown) displays the Curie-Weiss law at high temperature above 20 K, but deviates significantly from the Curie-Weiss law at low temperatures. It should also be noted that the magnetic susceptibility has uniaxial anisotropy along the [001] direction.

These experimental observations are fully consistent with the previous study. [5] Typical dHvA oscillation for U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$ with the magnetic field along the [110] direction and that for the reference compound ThRu$_2$Si$_2$ are shown in Fig. 2. The frequency components of those oscillations are obtained by fast Fourier transform as shown in Fig. 3. The oscillation is clearly seen in both cases. As shown in Fig. 3, the dHvA frequencies for the fundamental branches $A$, $B$ and $C$, as well as their harmonics are very similar each other. This result demonstrates that the Fermi surface volume is unchanged upon doping uranium.

On the other hand, the amplitude of the uranium-doped sample is significantly reduced compared to the measurement on pure ThRu$_2$Si$_2$ under similar experimental conditions. In
particular, the $A$-branch with the largest dHvA frequency and higher harmonics of the $B$-branch are significantly weak in $U_{0.03}Th_{0.97}Ru_2Si_2$. It is also noted that the amplitude of the second harmonic $2B$ in pure $ThRu_2Si_2$ is significantly larger than that of the fundamental $B$-branch. In general, the relative amplitudes between the dHvA branches and their higher harmonics contains information on the spin-splitting factor of the corresponding Fermi surface. Further investigations including the angular dependence is necessary to clarify this point.

There are two possible origins for the reduction of the amplitude of the quantum oscillations. The first one is the electron scattering due to impurity. In the present study, this is most likely a principal origin because 3% of uranium atoms doping should act as the scattering centers. The second one is the enhancement of the cyclotron effective mass. The latter one is due to the small energy splitting $\frac{\hbar eB}{mc^2}$ between Landau levels of heavy conduction carriers. As a result, the dHvA amplitude which reflects the population difference between adjacent Landau levels across Fermi energy smears out quickly with increasing temperature.

We show in Table I the dHvA frequencies as well as cyclotron effective masses of the dHvA branches obtained for $U_{0.03}Th_{0.97}Ru_2Si_2$ and $ThRu_2Si_2$.

The difference of the dHvA frequency, namely the Fermi surface cross section, between $U_{0.03}Th_{0.97}Ru_2Si_2$ and $ThRu_2Si_2$ is extremely small. According to the previous ARPES study...
on URu$_2$Si$_2$, an itinerant 5$f$ character was observed even in the paramagnetic state [7]. It is therefore expected that 5$f$ electrons participate in Fermi surface to modify its volume. If fact, a recent study of dilute Ce$_{0.02}$La$_{0.98}$Ru$_2$Si$_2$ showed a clear change in the Fermi surface volume from the reference compound LaRu$_2$Si$_2$ [6].

The present observation, however, looks disagreeing with this picture, namely, 5$f$ electrons remain localized not participating in the conduction band. Further investigation might be needed to clarify this point, such as detailed analysis of magnetic field dependence on the cyclotron effective mass and dHvA frequency.

Table 1. dHvA frequency $F$ and cyclotron effective mass $m^*_c$ for U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$ and ThRu$_2$Si$_2$.

| branch | ThRu$_2$Si$_2$ | U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$ |
|--------|---------------|---------------------------------|
|        | $F$ (T)       | $m^*_c/m_0$                      | $F$ (T)       | $m^*_c/m_0$ |
| $A$    | 9080          | 1.1                             | 9090          | 2.0         |
| $B$    | 1560          | 0.8                             | 1560          | 2.3         |
| $C$    | 870           | 0.5                             | 880           | 0.9         |

4. Summary
We have grown high quality single crystal samples of U$_{0.03}$Th$_{0.97}$Ru$_2$Si$_2$ and succeeded to detect dHvA oscillation. The dHvA frequency remains the same, indicating that the Fermi surface volume does not change upon uranium doping. On the other hand, the cyclotron effective mass is significantly enhanced, indicating the mass renormalization due to 5$f$ component.

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