Delocalization of the $f$ electron in Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ - the de Haas-van Alphen effect measurement

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Abstract.
We report the first observation of the continuous Fermi surface (FS) variation from La compound (LaRu$_2$Si$_2$) with no $f$ electron to Ce compound (CeRu$_2$Si$_2$) with itinerant $f$ electron via the de Haas-van Alphen (dHvA) effect. The dHvA frequency smoothly varies with Ce concentration and there is no discontinuous change with Ce concentration. It is found that the effective mass and signal amplitude with Ce concentration depends strongly on the Fermi surface sheet, and the effective mass is enhanced toward $x_c$ ($=0.91$) and the signal amplitude reduces around $x_c$ or somewhere between $x_c$ and $x \sim 0.8$.

1. Introduction
Many anomalous physical properties like non Fermi liquid and heavy fermion (HF) have been observed and studied in the highly correlated $f$ electron systems. How the $f$ electron nature changes from impurity state to lattice system is the important issue to resolve the anomalous properties. The $f$ electron nature has two different states, that is the itinerant and localized state. If the $f$ electron is itinerant, the $f$ electron contributes to FS. While, if the $f$ electron is localized, the $f$ electron does not contribute to FS. The dHvA measurement is a powerful tool to measure the FS, so the dHvA measurement have been performed to resolve the issue. When the $f$ electron can be assumed to be localized in the Ce compound like Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ [1], the FS properties can be followed via the dHvA effect measurements from the La compound to the Ce compound. These studies indicate that the $f$ electron is likely to be localized in the impurity state. In the case of $f$ electron is itinerant in the Ce compounds, the dHvA signal can be observed in a small concentration range [2], so how the $f$ electron nature changes from impurity state to lattice system is not obvious.

CeRu$_2$Si$_2$ is a typical paramagnetic heavy fermion compound with the electronic specific heat coefficient $\gamma$ of 350 mJ/mole K$^2$ [3]. The $f$ electron of CeRu$_2$Si$_2$ is itinerant [4]. When a magnetic field is applied parallel to the [001] direction its the tetragonal ThCr$_2$Si$_2$-type structure, a metamagnetic like transition takes place at $B_m$ (= 7.7 T) and Fermi surface (FS) drastically changes from the FS of itinerant $f$ electron to LaRu$_2$Si$_2$ like FS [4]. By replacing Ce with La, Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ changes to antiferromagnetic at $x_c = 0.91$ and finally normal metal.
of LaRu$_2$Si$_2$ with no $f$ electron [6]. When a magnetic field is applied in the (001) plane, the magnetization increases gradually with field and no metamagnetic behavior is observed at least up to 18 T. We have extensively studied Fermi surface properties in dilute Ce concentration samples for magnetic field parallel to the [100] direction [5]. However we have not observed the obvious dHvA signal in high Ce concentration samples. We have observed the continuous Fermi surface variation from $x = 0.0$ (LaRu$_2$Si$_2$) with no $f$ electron to 1.0 (CeRu$_2$Si$_2$) via the dHvA effect for a magnetic field parallel to the [110] direction and we report the variation of dHvA frequency, effective mass and signal amplitude with Ce concentration.

2. Experimentals

Single crystals of Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ were grown by using the Czochralski pulling method in a tetraarc furnace under Ar gas atmosphere and were annealed under vacuum for a week at 900 °C. The lattice parameter was determined by a powder X-ray diffraction analysis. Unit cell volume changes almost linearly with Ce concentration $x$ from $x = 0.0$ to 1.0. Several samples ranging from $x = 0.02$ to 0.5 were analyzed by the inductively coupled plasma method. The measured content $x$ agrees with the nominal content within 10 %. We use the nominal concentration $x$ to label the sample.

The dHvA effect measurements were performed by using the conventional field modulation method in dilution refrigerators under magnetic fields up to 18 T. The signal amplitude is an average obtained from the Fourier analysis between 10 and 16 T at about 0.1 K, and the signal amplitude of LaRu$_2$Si$_2$ is normalized to 1.0 for all of oscillation [6], finally the amplitude make adjustment of the influence of the dHvA frequency and effective mass.

3. Result and discussion

We have successfully observed dHvA oscillation of Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ from $x = 0.0$ to 1.0 for a magnetic field along the [110] direction. Figure 1 shows the dHvA signal and its Fourier spectrum for $x = 0.02$, 0.51, 0.90 and 0.97. The left panels (a1), (b1), (c1) and (d1) of Fig. 1 show the dHvA signals obtained in the field range between 11 and 17.5 T. The right panels (a2), (b2), (c2) and (d2) of Fig. 1 show their corresponding Fourier spectra in the frequency range between 0 and 2000 T. Fermi surfaces of LaRu$_2$Si$_2$ (a) and CeRu$_2$Si$_2$ (b) show in Fig. 2 [7, 8]. The figures indicate the names of the dHvA oscillations and the corresponding orbits when the magnetic field is applied parallel to the [110] direction. We temporally denote the symbol of the dHvA oscillation corresponding to those in LaRu$_2$Si$_2$ from $x = 0.0$ to $x_c$, while the symbol of the dHvA oscillations corresponding to those in CeRu$_2$Si$_2$ from $x_c$ to $x = 1.0$. All the dHvA oscillations corresponding to those in LaRu$_2$Si$_2$ i.e. the $\omega$, $\gamma'$, $\beta'$ and $R$ oscillation can be observed for $x = 0.04$. The $\beta'$ and $\gamma'$ oscillations arise from the ellipsoidal small hole surfaces, the $R$ oscillation arises from the ring electron surface and the $\omega$ oscillation arises from the large hole surface. In antiferromagnetic state of $x = 0.51$, the two clear oscillations are observed, while the $\gamma'$ oscillation is only observed for $x = 0.90$, indicating that there is no maximum in the amplitude reduction at $x = 0.5$. In paramagnetic state of $x = 0.97$, the $\beta$ and $\gamma$ oscillations from small ellipsoidal hole surfaces are observed.

Figure 3 shows the dHvA frequency, effective mass and signal amplitude plotted against Ce concentration for magnetic field parallel to the [110] direction. We plot the dHvA frequencies of $\beta'$-$\beta$, $\gamma'$-$\gamma$ and $R$ oscillations as a function of Ce concentration $x$ in the top panel (a). We have observed the $\gamma'$-$\gamma$ oscillation from $x = 0.0$ to 1.0. The frequency of $\gamma'$-$\gamma$ oscillation continuously varies from $x = 0.0$ to 1.0. Especially there is not discontinuous change at $x_c$.

We also plot the effective masses of observed oscillations as a function of Ce concentration $x$ in the middle panel (b). In low Ce concentration, the effective mass of the $R$ oscillation increases with $x$, while the effective of the $\beta'$ and $\gamma'$ oscillation moderately increase from $x = 0.0$ to about
0.6. The effective mass of $\gamma'$ oscillation increases toward $x_c$, indicating that the effective mass of $\gamma-\gamma'$ oscillation is enhanced around $x_c$.

We also plot the signal amplitude of the $\beta'-\beta$, $\gamma'-\gamma$ and $R$ oscillations as a function of Ce concentration $x$ in the bottom panel (c). The signal amplitudes of the $R$ and $\omega$ oscillation rapidly decreases with increasing $x$, while the amplitude reduction of the $\beta'$ and $\gamma'$ oscillations are much smaller. The signal amplitude of $\gamma-\gamma'$ oscillation largely decreases around $x_c$, and the amplitude is about $10^{-4}$ times as small as that of LaRu$_2$Si$_2$.

We discuss the DHvA effect results. We could observe the DHvA oscillation from the system with no $f$ electron system to the itinerant $f$ electron system for the first time. The frequency of the $\gamma'-\gamma$ oscillation smoothly varies from $x = 0.0$ to 1.0. Our previous study is indicated that the $f$ electron is itinerant in impurity state. If the $f$ electron is itinerant from the impurity state to CeRu$_2$Si$_2$, it is expected that all of the DHvA frequencies smoothly varies from $x = 0.0$ to 1.0. Therefore, the result is consistent with our previous study. However we could observe only the $\gamma'-\gamma$ oscillation arisen from the small Fermi surface with poor $f$ electron content.

The signal amplitude of the $\gamma'-\gamma$ oscillation seems to be minimum around $x_c$ and the effective mass is also enhanced around $x_c$. Since the signal amplitude is a function of effective mass as well as electron scattering, it is not clearwhether or not the minimum of the signal amplitude is at $x_c$. In the sister system CeRu$_2$($Si_{1-x}Ge_x$)$_2$, it is found that the residual resistivity becomes maximum in the antiferromagnetic state near $x_c$ not at $x_c$ [9]. The maximum of the residual resistivity is discussed by Hattori and Miyake in terms of enhanced charge fluctuation due to the competition between the antiferromagnetic interaction and the Kondo effect, and resultant enhancement of the impurity scattering. They predict that the enhancement takes place when $T_N \sim T_K$ [10]. The Ge concentration for the maximum of residual resistivity seems to takes place when $T_N \sim T_K$ in CeRu$_2$($Si_{1-x}Ge_x$)$_2$. On the other hand, $T_N$ is equal to $T_K$ around $x = 0.85$ in Ce$_x$La$_{1-x}$Ru$_2$Si$_2$. Then, considering the correspondence between the two alloy systems, the signal amplitude is likely to be minimum around $x = 0.85$ rather than at $x_c$.

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Figure 2. Fermi surfaces of LaRu$_2$Si$_2$ (a) and CeRu$_2$Si$_2$ (b) [7, 8]. In both the figures, the Fermi surfaces at the top and middle are hole surfaces and those at the bottom are electron surfaces. The figures indicate the names of the dHvA oscillations and the corresponding orbits when the magnetic field is applied parallel to the [110] direction.

Figure 3. DHvA frequencies (a), effective masses (b) and signal amplitude (c) plotted as a function of Ce concentration $x$ in Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ for a magnetic field along the [110] direction. The left axis of (c) is logarithmic scale. The open squares denote the frequencies of the $\gamma'$ and $\gamma$ oscillations. The open triangles denote the frequencies of the $R$ oscillation. The open diamonds denote the frequencies of the $\beta'$ and $\beta$ oscillations.

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