Anomalous behavior of the dHvA oscillations in Ce$_x$La$_{1-x}$Ru$_2$Si$_2$

Y Matsumoto$^1$, N Kimura$^1$, T Komatsubara$^1$, H Aoki$^1$, M Kimata$^2$, T Terashima$^2$, S Uji$^2$

$^1$Graduate School of Science and Center for Low Temperature Science, Tohoku University, Sendai 980-8578, Japan
$^2$National Institute for Materials Science, Tsukuba, Ibaraki 305-0003, Japan

E-mail: y.mats@mail.clts.tohoku.ac.jp

Abstract.
We report anomalous behaviors of the de Haas - van Alphen (dHvA) oscillations in various Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ samples when magnetic field is applied in the (001) plane. We argue that these behaviors can be explained by assuming that the effective mass of the conduction electron of one spin direction decreases with field while that of the opposite spin direction increases.

1. Introduction
The conduction electrons of highly correlated f electron systems exhibit various anomalous properties under magnetic fields. It is reported in several compounds like CeRu$_2$Si$_2$, Ce$_x$La$_{1-x}$B$_6$, PrPb$_3$, CePd$_2$Si$_2$, CeIn$_3$, and CeCoIn$_5$ [1, 2] that the effective mass ($m^*$) of the conduction electron depends on the direction of spin as well as on magnetic field, while the Fermi surfaces of the up and down spin electrons are nearly the same. It appears in most of the cases that both the $m^*$’s of the up and down spin electrons decrease or increases with magnetic field.

In this paper, we report anomalous behavior of the dHvA oscillations in Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ and argue that it could be explained by assuming that the $m^*$ of one spin direction decreases with magnetic field while that of the opposite spin direction increases.

Before we present the experimental results, we briefly summarize previous observations and our understanding in terms of the spin dependent $m^*$. When the amplitude and frequency of the dHvA oscillation depend on the direction of spin, the fundamental dHvA oscillation $\tilde{M}_{fnd}$ from the up and down spin electrons can be given by

$$\tilde{M}_{fnd} = \tilde{M}_\uparrow(B, T) + \tilde{M}_\downarrow(B, T)$$

$$= A_\uparrow(B, T) \sin \left( \frac{2\pi F_\uparrow(B)}{B} + \xi_\uparrow + \xi_0 \right) + A_\downarrow(B, T) \sin \left( \frac{2\pi F_\downarrow(B)}{B} + \xi_\downarrow + \xi_0 \right) .$$

(1)

Here, $A_\sigma$ and $F_\sigma$ denote spin dependent amplitude and frequency, respectively. In the present case, we may assume that $F_\uparrow = F_\downarrow$. $\xi_0$ is a constant. $\xi_\sigma$ is the spin and magnetic field dependent phase and we assume that it is given by $\mp (\pi g m^*_\sigma(B))/(2m_0 g)[1]$. Here, $m_0$ is the electron rest mass and $g$ is the g factor. $m^*_\sigma(B)$ is the effective mass which depends on spin direction and magnetic field. The $-$ and $+$ signs correspond to the up and down spin electrons, respectively.
When $A_1 \geq A_1$, eq. (1) can be modified as follows.

\[
\tilde{M}_{fnd}(B, T) = \sqrt{A_1^2 + A_1^2 + 2A_1A_1 \cos(\xi_1 - \xi_1)} \times \sin\left(\frac{2\pi F(B)}{B} + \xi_1(B) + \xi_0 + \alpha(B, T)\right).
\]

\[
\alpha(B, T) = \tan^{-1}\left[\frac{A_1}{\xi_1} \sin(\xi_1 - \xi_1) \right] \left[1 + \frac{A_1}{\xi_1} \cos(\xi_1 - \xi_1) \right].
\]

The phase $\alpha(B, T)$ indicates the difference in the phase from that of the dominant oscillation $\tilde{M}_1$. When $A_1 \leq A_1$, an equivalent equation is obtained by exchanging $\uparrow$ and $\downarrow$ [1].

When the $m^*$'s change with field, $\xi_1 - \xi_1$ also changes. As noted from the equations above, the amplitude and phase change with field making anomalous field dependence of the oscillation amplitude. On the other hand, when the amplitudes of the two spin oscillations are comparable but $m^*_1(B) \neq m^*_2(B)$, the ratio $A_1/A_1$ changes with temperature because the variation of the amplitude with temperature depends on $m^*$. Then, the phase $\alpha(B, T)$ changes with temperature. Anomalous changes in the amplitude and phase as a function of field or temperature have been successfully explained in terms of spin dependent oscillations or spin dependent $m^*$ [1, 2].

Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ has the tetragonal ThCr$_2$Si$_2$ - type structure. For $0.02 < x < 0.91$ the ground state is antiferromagnetic and for $x > 0.91$ is a paramagnetic heavy fermion state [4]. When a magnetic field is applied parallel to the [001] direction in the antiferromagnetic state or in the paramagnetic state, it exhibits a metamagnetic transition. However, 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, indicating that Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ has strong Ising character.

2. Experiments

Single crystals of Ce$_x$La$_{1-x}$Ru$_2$Si$_2$ were grown by using the Czochralski pulling method in a tetra-arc furnace and were annealed under vacuum for a week at 900 °C. 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 detected dHvA signal was transferred to a digital filter to study the field dependence of each frequency separately.

3. Result and discussion

Figure 1(a) shows the fundamental frequency oscillation of $\beta_2$ in the $x = 0.04$ sample with fields parallel to the [100] direction at three temperatures. The $\beta_2$ oscillation arises from an ellipsoidal Fermi surface [4]. The oscillation amplitude increases monotonously with increasing field as expected from the standard Lifshitz-Kosevich (LK) formula [3]. However, the amplitude of the oscillation does not increase with decreasing temperature and at fields below about 10 T it decreases with decreasing temperature, while the LK formula tells us that the amplitude should increase monotonously with decreasing temperature. In Figs.1(b) and (c), we show expanded views of the dHvA oscillations at low field side of about 7.07 T and at high field side of about 13.2 T, respectively, to demonstrate how the phase of the oscillation changes with temperature. The phase at the high field side does not change appreciably and that at the low field side slightly changes with temperature. The present observation is in striking contrast to the previous observations where the amplitude does not change monotonously with field and the phase changes considerably with temperature [1, 2]. These previous observations are explained.
Figure 1. (a) $\beta_2$ oscillations plotted against inverse field at 980 mK, 610 mK and 120 mK in $x = 0.04$ sample. Magnetic field is applied parallel to the [100] direction. The small beat structures at the high field side arise from the interference with the second harmonic frequency oscillation of another dHvA oscillation $\beta_3$. It can not be filtered out sufficiently because its frequency is close to that of $\beta_2$. Expanded views of the oscillations at three temperatures around (b) 7.07 T and (c) 13.2 T.

by assuming that both the $m^*$'s of the up and down spin oscillations increase or decrease with magnetic fields.

The anomalous behavior of amplitude as a function of temperature can be explained by assuming that the condition $(\xi_1 - \xi_1) = (2n + 1)\pi$ is nearly satisfied. In this case the amplitude of oscillation in Eq.(2) is given by $| A_\uparrow - A_\downarrow |$. When $m^*_\uparrow(B) = m^*_\downarrow(B)$ and $A_\uparrow = A_\downarrow$, this condition is the same as the so called spin splitting zero condition[3], i.e., for particular values of $m^*$ which satisfy $g(m^*/m_0) = 2n + 1$ ( $n$ : integer), the amplitude of the oscillation vanishes due to dephasing effect of the up and down spin oscillation. If the $m^*$'s of the up and down spin oscillations are different, the amplitude of the oscillation with heavier $m^*$ is smaller than that with lighter $m^*$. On the other hand, that with heavier $m^*$ grows more rapidly with decreasing temperature than that with lower $m^*$ and the growth rate is higher at lower fields [3]. Then, the difference $| A_\uparrow - A_\downarrow |$ will decrease with decreasing temperature particularly at lower fields as observed in the present experiment.

On the other hand, since the $m^*$'s of the up and down spin oscillations should be identical at $B = 0$, they should change differently with field because they are different at high fields. Then the phase will change with field but can remain to be the same only if $(\xi_1 - \xi_1) = (2n + 1)\pi$ and if it does not change with field. Since $(\xi_1 - \xi_1) = \left( m^*_\uparrow(B) + m^*_\downarrow(B) \right) / 2m_0$, this means that the average of the $m^*$'s does not change with field. This also means that if the $m^*$ of one spin direction increases, that of the opposite spin direction decreases with field.

Similar anomalous behaviors are sometimes observed for the other oscillations in samples with $x = 0.02 - 0.10$. We speculate that since the $m^*$ of each oscillation changes with $x$, the condition $(\xi_1 - \xi_1) \sim (2n + 1)\pi$ may be accidentally satisfied for a particular oscillation in a particular sample. In higher concentration samples in the range of $0.1 < x < 0.3$, another anomalous behavior is observed. In this case the amplitude of the dHvA oscillation increases monotonously with decreasing temperature. Therefore, the conventional analysis can be applied to obtain the...
effective mass. Figure 2 shows the $m^*$'s of the fundamental and second harmonic frequency oscillations of $\beta_2$. The $\beta_2$ oscillation arises from another ellipsoidal Fermi surface [4]. The $m^*$ of the fundamental frequency oscillation increases, while that of the second harmonic oscillation exhibits anomalous behavior and decreases with field at high fields. The LK formula predicts that the value of the $m^*$ of the second harmonic frequency oscillation should be just twice as much as that of the fundamental frequency oscillation. We speculate that this observation can be also explained by assuming that the relaxation times as well as $m^*$'s of the up and down spin electrons are different and that $m^*$ of one spin direction increases with field while that of the opposite spin direction decreases.

The field and spin dependences of $m^*$ have been studied theoretically [5, 6, 7, 9, 10] and some theories predict that the $m^*$ of one spin direction increases and that of the opposite spin direction decreases with magnetic field. It should be noted that with fields in the directions near the easy axis [001] the $m^*$ decreases with increasing field and the anomalous behaviors presented above are not observed. This fact could be related with the magnetic properties of the system, that is, the magnetic field along the easy axis may suppress magnetic fluctuation effectively but that perpendicular to the easy axis does not.

Acknowledgments
We thank M. Suzuki and M. Kikuchi for technical support. This work was supported by Grants-in-Aid for Scientific Research (A) 19204034 and for Scientific Research on Innovative Areas “Emergence of Heavy Electrons and Their Ordering” from MEXT Japan. YM acknowledges financial supports from Global COE program “Weaving Science Web beyond Particle-matter Hierarchy” and from Research fellowship for Young Scientists JSPS.

References
[1] Endo M, Kimura N and Aoki H 2005 J. Phys. Soc. Jpn. 74 3295.
[2] Endo M, Nakamura S, Isshiki T, Kimura N, Nojima T, Aoki H, Harima H and Kunii S 2006 J. Phys. Soc. Jpn. 75 114704 and references therein.
[3] D.Shoenberg, Magnetic Oscillations in Metals (Cambridge University Press, Cambridge, England,1984).
[4] Matsumoto Y, Sugi M, Kimura N, Komotsubara T, Aoki H, Satoh I, Terashima T and Uji S 2008 J. Phys. Soc. Jpn. 77 053703 and references therein.
[5] Wasserman A, Springford M and Hewson A C 1989 J. Phys. Condens. Matter. 1 2669.
[6] Edwards D M and Green A C M 1997 Z. Phys. B 103 243.
[7] Spalek J, 2006 Phys. Stat. Sol. (b) 243 78.
[8] Otsuki J, Kusunose H and Kuramoto Y 2007 J. Magn. Magn. Mater. 310 425.
[9] Bauer J and Hewson A C 2007 Phys. Rev. B 76 035118.
[10] Onari S, Kontani H and Tanaka Y 2008 J. Phys. Soc. Jpn. 77 023703.