DE HAAS–VAN ALPHEN EXPERIMENTS IN THE QUANTUM CRITICAL REGION OF CERIUM AND URANIUM COMPOUNDS*

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When pressure is applied to the cerium and uranium compounds, their magnetic ordering temperatures are suppressed and become zero at a critical pressure P_c. Around P_c, non-Fermi liquid and/or superconductivity are observed. We clarified a change of the electronic state via the de Haas–van Alphen (dHvA) experiment when pressure crosses P_c. The dHvA experiment under pressure was done for antiferromagnets $CeRh_2Si_2$, $CeRhIn_5$ and URu_2Si_2, and a ferromagnet UGe_2. We find an abrupt change of the Fermi surface for $CeRh_2Si_2$ and UGe_2 when crossing P_c, indicating a first-order like phase transition. For $CeRhIn_5$ and URu_2Si_2, a change of the cyclotron mass is clearly observed.

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1. Introduction

In cerium and uranium compounds, the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect compete with each other [1, 2]. Competition between the RKKY interaction and the Kondo effect was discussed by Doniach [3] as a function of $|J_{cf}|D(\varepsilon_{\rm F})$, where $|J_{cf}|$ is the magnitude of the magnetic exchange interaction and $D(\varepsilon_{\rm F})$ is the electronic density of states at the Fermi energy $\varepsilon_{\rm F}$. Most cerium compounds order magnetically, when the RKKY interaction overcomes the Kondo effect at low temperatures. The magnetic order is formed by localized-$4f$ moments. The topology of the Fermi surface for the conduction electrons is therefore quite similar to that of the corresponding non-$4f$ lanthanum compound, although the cyclotron mass of the cerium compound is one to two orders of magnitude larger than that of the lanthanum compound.

On the other hand, some cerium compounds such as $CeCu_6$ and $CeRu_2Si_2$ show no long-range magnetic ordering, because the Kondo effect overcomes the RKKY interaction. These compounds are called heavy fermion compounds since they have a large electronic specific heat coefficient γ : $\gamma \simeq 10^4/T_{\rm K}$ (mJ/K^2 mol) [1, 2]. In fact, a large cyclotron effective mass of $120m_0$ was detected in the de Haas–van Alphen (dHvA) experiment for $CeRu_2Si_2$ [4]. Moreover, the topology of the Fermi surface is well explained by the $4f$-itinerant band model, although the cyclotron effective mass is much larger than the corresponding band mass.

Some uranium compounds such as UPd_2Al_3 and UPt_3 are very similar to the heavy fermion compounds. In many aspects, however, uranium compounds are different from the cerium compounds. They appear to possess the dual nature, both itinerant and localized.

Recently a new aspect of cerium and uranium compounds with magnetic ordering has been discovered. When pressure P is applied to the compounds with antiferromagnetic ordering such as $CeIn_3$ and $CePd_2Si_2$ [5], the Néel temperature $T_{\rm N}$ decreases, and a quantum critical point corresponding to the extrapolation $T_{\rm N} \to 0$ is reached at $P = P_{\rm c}$. Here, $|J_{cf}|D(\varepsilon_{\rm F})$ in the Doniach model can be replaced by pressure. Surprisingly, superconductivity appears around $P_{\rm c}$. Moreover, the heavy fermion state is also formed around $P_{\rm c}$, where the non-Fermi liquid nature is found in some compounds. Similar pressure-induced superconductivity was reported for other compounds such as $CeRh_2Si_2$ [6], $CeRhIn_5$ [7] and UGe_2 [8].

The crossover from the magnetically ordered state to the non-magnetic state under pressure, crossing the quantum critical point, is the most interesting issue in strongly correlated f-electron systems. The purpose of the present work is to clarify the nature of f electrons by using a microscopic probe — the dHvA method — on $CeRh_2Si_2$, UGe_2, $CeCoIn_5$ and

URu_2Si_2. Previous dHvA works were carried out for magnetic and non-magnetic
f-electron compounds and were compared to the results of the corresponding non-$4f$ lanthanum compounds and/or energy band calculations. The present work was done for a single compound across the quantum critical point, due to the extreme technical challenge method.

2. Experimental results and discussion

2.1. $CeRh_2Si_2$ and UGe_2 — a drastic change of the Fermi surface

$CeRh_2Si_2$ with the tetragonal structure orders antiferromagnetically below the Néel temperature $T_{N1} = 36$ K. Below $T_{N2} = 25$ K, there occurs a change of the magnetic structure, from the antiferromagnetic state with the propagation vector $\boldsymbol{q}_1 = \left(\frac{1}{2}\,\frac{1}{2}\,0\right)$ to the $4\boldsymbol{q}$-structure [9]. $CeRh_2Si_2$ is considered to be a usual $4f$-localized system. When pressure is applied to $CeRh_2Si_2$, $T_{N2} = 25$ K and $T_{N1} = 36$ K are suppressed to 0 K at $P_c' \simeq 0.6$ GPa and $P_c = 1.06$ GPa, respectively, and superconductivity appears at $T_{sc} \simeq 0.4$ K around P_c [6, 10].

Fig. 1 shows the pressure dependence of T_{N1}, T_{N2} and T_{sc}, which was obtained by the electrical resistivity measurement [11]. Superconductivity appears around P_c, as shown in Fig. 2. An indication of superconductivity appears in the pressure region from 0.97 to 1.20 GPa, which is shown in

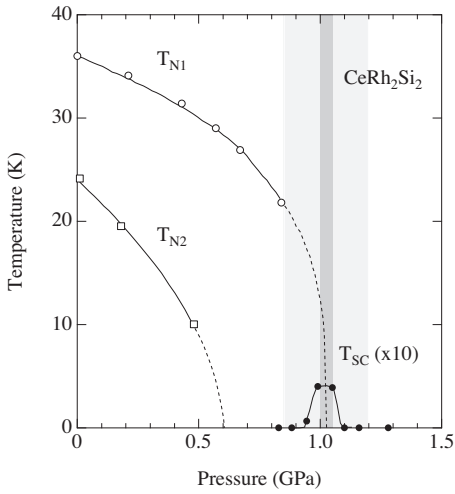

Fig. 1. Pressure dependence of T_{N1}, T_{N2} and T_{SC} in $CeRh_2Si_2$. The temperature of T_{SC} is ten times enlarged compared to those of T_{N1} and T_{N2}. The data on T_{N2} are cited from Ref. [9].

Fig. 1 as a gray region. The resistivity zero is, however, observed in an extremely narrow pressure region around P_c, which is shown in Fig. 1 as a dense-gray region. Namely, the resistivity at $P = 1.05$ and 1.06 GPa in Fig. 2 decreases below 0.5 K and becomes zero at 0.4 K. This implies that homogeneous bulk-superconductivity is realized in an extremely narrow pressure region around P_c.

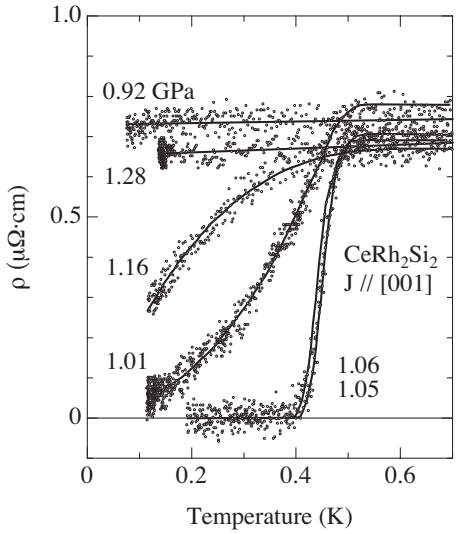

Fig. 2. Low-temperature resistivity under pressure in $CeRh_2Si_2$.

The pressure dependence of the dHvA frequency up to 1.54 GPa is plotted in Fig. 3. Here, the dHvA frequency $F(= \hbar c S_F/2\pi e)$ is proportional to the extremal cross-sectional area S_F of the Fermi surface, and is expressed as a unit of magnetic field [1,2]. The dHvA branches are slightly changed at $P_c' \simeq 0.6$ GPa. Branches o, κ and ζ disappear completely and new branches named p, q and r appear above $P_c' \simeq 0.6$ GPa. This is because the magnetic structure changes from the $4\boldsymbol{q}$-structure to the $\boldsymbol{q}_1$-structure at $P_c' \simeq 0.6$ GPa. Branches d and π are unchanged against pressure up to P_c. Note that branches d and π are found at 1.03 GPa, while at 1.08 GPa they disappear completely and a new branch named A appears. With further increasing pressure, two branches named B and C are also detected. These branches are well identified by the $4f$-itinerant band model [12].

The cyclotron effective masses m_c^* were determined from the temperature dependence of the dHvA amplitude [1, 2]. The cyclotron masses increases with increasing pressure, reaching $28\,m_0$ at 1.03 GPa for branch d, which is four to five times larger than that at ambient pressure. Above P_c, the cyclotron masses of branches A, B and C are large, ranging from 24 to 34 m_0.

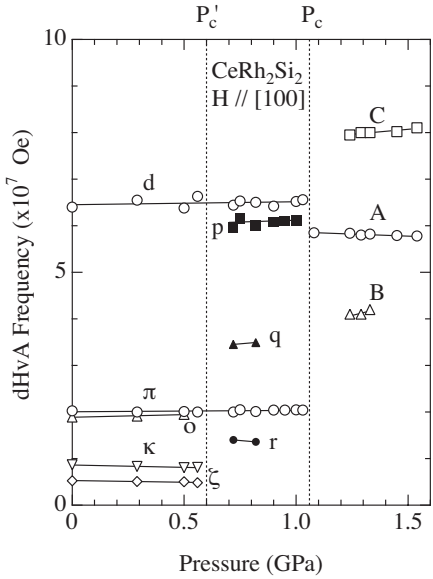

Fig. 3. Pressure dependence of the dHvA frequency in $CeRh_2Si_2$.

It is thus cocluded that the detected dHvA frequencies clearly change at $P_c \simeq 1.06$ GPa, implying a first-order like plase transition. This indicates a discontinuous change of the Fermi surface.

Next we note a novel electronic state of UGe_2. UGe_2 with the orthorhombic crystal stacture is a new pressure-induced superconductor in the ferromagnetic state [8,13]. In the pressure experiment, it was clarified that with increasing pressure P, the Curie temperature T_C (= 52 K) becomes zero roughly at $P_c^* \simeq 1.5$ GPa [14]. The second phase transition was later found below T_C, at $T^* \simeq 30$ K at ambient pressure [15]. T^* also becomes zero at $P_c^* \simeq 1.2$ GPa. Around P_c^*, namely in the pressure region from 1.0 to 1.5 GPa, still in the ferromagnetic state, superconductivity was observed below $T_{SC} = 0.7$ K [8, 13]. The temperature region from T_C to T^* and/or the pressure region from P_c^* to P_c were named the weakly polarized phase, while the lower temperature region ($T < T_C$) and/or the pressure region below P_c^* were named the strongly polarized phase [16]. When the magnetic field is applied along the a-axis, the weakly polarized phase is changed into the strongly polarized phase at $H = H^*$ even in the pressure region of $P > P_c^*$ [13, 17]. This phenomenon corresponds to the metamagnetic transition, indicating a step like increase of the magnetization curve. The P–H phase diagram for the field along the a-axis is shown in Fig. 4. The phase boundary from the paramagnetic phase to the weakly polarized phase is also indicated at H_c. These phase boundaries were obtained from the ac-susceptibility measurement [18].

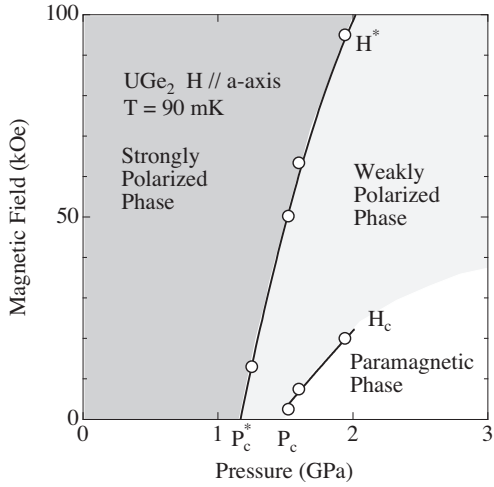

Fig. 4. P–H phase diagram for $H \parallel a$-axis in UGe_2.

Recent spin-polarized band calculations clarified the existence of cylindrical Fermi surfaces along the b-axis [19,20]. The dHvA experiments under pressure for the field along the b-axis indicated that corresponding dHvA branches are clearly observed up to P_c^* (in the strongly polarized phase) but are scarcely seen in the pressure region form P_c^* to P_c (in the weakly polarized phase) [19,21]. This is mainly due to an extremely large cyclotron mass of conduction electrons in the weakly polarized phase. It is, however, remarkable that new dHvA branches with large cyclotron masses $m_c^* \simeq 60$ m_0 appear clearly in the paramagnetic region, $P > P_c$. A drastic change of the Fermi surface occurs at P_c. It is thus suggested from a view point of the Fermi surface property that the phase boundary at P_c is of the first-order like phase transition.

2.2. $CeRhIn_5$ — a Fermi surface unchanged under pressure

$CeRhIn_5$ as well as related compounds $CeCoIn_5$ and $CeIrIn_5$ crystallize in the tetragonal structure with alternating layers of $CeIn_3$ and $RhIn_2$, stacked sequentially along the [001] direction (c-axis). Hegger *et al.* found that $CeRhIn_5$ orders antiferromagnetically below $T_N = 3.8\,K$, but reveals an antiferromagnetic to superconducting transition at a relatively low critical pressure $P^* = 1.63\,GPa$ [7]. The superconducting transition temperature $T_c = 2.2\,K$ at $2.5\,GPa$ is the highest value in the pressure-induced superconductors, as shown in Fig. 5 [22,23]. On the other hand, $CeCoIn_5$ and $CeIrIn_5$ are superconductors at ambient pressure, with $T_c = 2.3$ and $0.4\,K$, respectively [24].

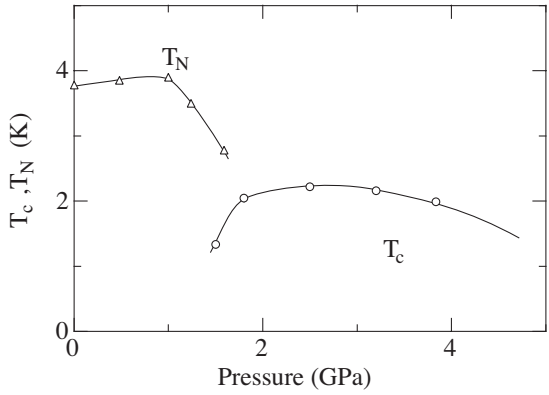

Fig. 5. Pressure dependence of the Néel temperature T_N and the superconducting transition temperature T_c.

The topology of main Fermi surfaces in the antiferromagnet $CeRhIn_5$ is nearly cylindrical, and is found to be in good agreement with that of a reference non-$4f$ compound $LaRhIn_5$, indicating that the $4f$ electrons in $CeRhIn_5$ are localized and do not contribute to the volume of the Fermi surface [25,26].

Fig. 6 shows the fast Fourier transformation (FFT) spectra of the dHvA oscillations under $P = 1.3$, 1.8 and 2.0 GPa. Main three branches named α_1, $\alpha_{2,3}$ and β_2 in Fig. 6 are due to a band 15-electron Fermi surface (α_i) and a band 14-electron Fermi surface (β_2), respectively. With increasing pressure, the dHvA amplitude is strongly reduced, but the dHvA frequencies for main cylindrical Fermi surfaces are unchanged up to 2.1 GPa, which is above $P^* = 1.63$ GPa.

The cyclotron effective mass m_c^* was determined by the temperature dependence of the dHvA amplitude [1,2]. Fig. 7 shows the pressure dependence of the cyclotron mass, which was determined at $H = 120$ kOe. The cyclotron mass increases steeply above 1.6 GPa, where superconductivity sets in. For example, the cyclotron mass of branch β_2 is $5.5\,m_0$ at ambient pressure, $20\,m_0$ at 1.6 GPa and $45\,m_0$ at 2.1 GPa. Futhermore, we found that the cyclotron mass is strongly field-dependent with increasing pressure: at 2.0 GPa the cyclotron mass of $40\,m_0$ at 110 kOe is reduced to $33\,m_0$ at 158 kOe. We point out that a considerable mass reduction is observed above $P^* = 1.63$ GPa. A much steeper mass reduction due to magnetic fields is observed in $CeCoIn_5$ at ambient pressure [27].

From these experimental results, it is concluded that the topology of the Fermi surface is unchanged up to 2.1 GPa, but the heavy fermion state is formed above 1.6 GPa where superconductivity sets in. It is thus suggested

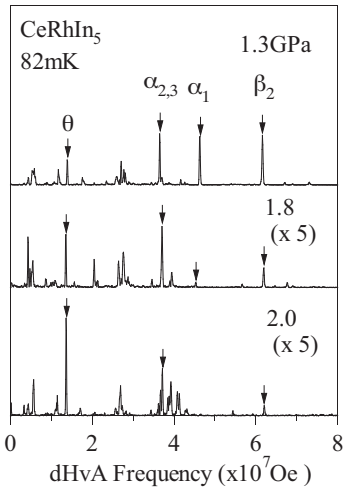

Fig. 6. FFT spectra under $P = 1.3$, 1.8 and 2.0 GPa in $CeRhIn_5$.

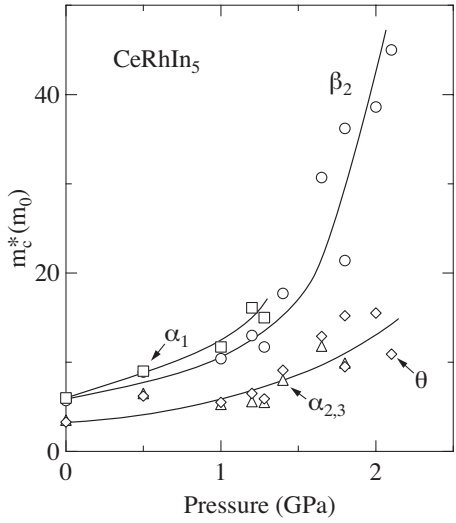

Fig. 7. Pressure dependence of the cyclotron mass for four dHvA branches in $CeRhIn_5$.

that the antiferromagnetic state remains in the superconducting phase becuase the topology of the Fermi surface in antiferromagnets is quite similar to that of the corresponding non-$4f$ reference lanthanum compounds [1,2]. The present result is consistent with the NMR result showing the $\sqrt{T}$-dependence of the nuclear spin-lattice relaxation rate, indicating that the superconducting phase still remains in a nearly antiferromagnetic state [28].

2.3. URu_2Si_2 — magnetic moment vs cyclotron mass

The heavy-fermion compound URu_2Si_2 exhibits two successive transitions at $T_c = 1.4$ and $T_o = 17.5\,K$. The former is the superconducting transition temperature. On the other hand, the latter has still remained unidentified, although characteristic features are associated with this phase transition. The neutron diffraction study indicated the development of a simple type-I antiferromagnetic order with a tiny $5f$-magnetic moment of $0.03\ \mu_B$ along the tetragonal [001] direction below T_o [29]. Recent neutron scattering and NMR experiments under pressure shed a new insight to this phase transition [30, 31]. It was clarified from the neutron scattering experiment that the magnetic moment increases linearly up to 1 GPa, reaching 0.25 GPa, but has a jump from 0.23 to $0.4\,\mu_B$ at $P_c = 1.5$ GPa [30]. The lattice constant (a-value) is also associated with a step-like decrease at P_c. The result of NMR experiment furthermore indicated that there exist distinct antiferromagnetic and paramagnetic regions, and with increasing pressure, the antiferromagnetic region increases in space, reaching 100% of the antiferromagnetic volume fraction at 1.0 GPa [31].

Previously we studied the dHvA effect of URu_2Si_2 [32]. If the NMR proposal is right, the detected dHvA branches were mainly due to the paramagnetic region because volume fraction of the paramagnetic and antiferromagnetic regions are about 99 and 1%, respectively, from the tiny moment of $0.03\,\mu_B$. The topology of the Fermi surface is generally influenced by the antiferromagnetic ordering. At 0.5 GPa we expect two kinds of dHvA branches based on the paramagnetic and antiferromagnetic regions. To clarify it we carried out dHvA experiment under pressure in magnetic fields up to 170 kOe and low temperatures down to 120 mK.

A detected branch is named α, which is observed at ambient pressure and is known to be nearly spherical in shape. There is no beat pattern in the dHvA oscillation at 0.5 GPa, meaning that there are no two kinds of branches. The dHvA frequency named α increases very slightly without a change at $P_c = 1.5$ GPa, as shown in Fig. 8(a). The increase of the dHvA frequency corresponds to an increase of the volume of the Fermi surface, which is consistent with the decrease of the lattice constant mentioned above. Fig. 8(b) shows the pressure dependence of the cyclotron mass m_c^*. The cyclotron mass decreases considerably with increasing pressure. In the heavy fermion system, the magnetic specific heat of $5f$ electrons is partially changed into an electronic specific heat. The present result is consistent with the pressure dependence of the magnetic moment. The larger the magnetic moment, the smaller the electronic specific heat coefficient or the cyclotron mass. It is, however, noted that a change of the cyclotron mass at P_c is not observed, which is very different from the magnetic moment.

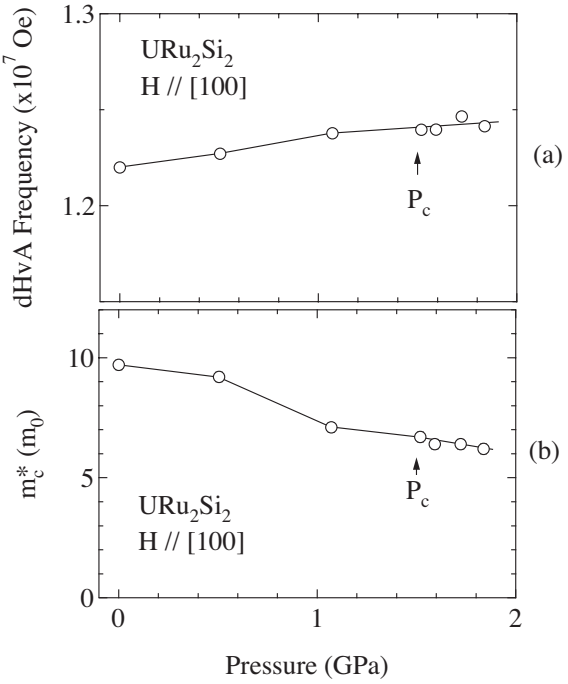

Fig. 8. Pressure dependence of (a) the dHvA frequency and (b) the cyclotron mass for branch α in URu_2Si_2.

We also determined the Dingle temperature and estimated the mean free path for branch α. The mean free path is 1100 Å ($\pm$ 50 Å), approximately independent on the pressure.

It is thus concluded that the present dHvA experiment under pressure is inconsistent with the phase-separated proposal based on the NMR experiment. The cyclotron mass under pressure changes considerably, reflecting a change of the magnetic moment.

3. Conclusion

We have clarified a drastic change of the Fermi surface for an antiferromagnet $CeRh_2Si_2$ and a ferromagnet UGe_2 when pressure crosses the critical pressure P_c. Superconductivity in $CeRh_2Si_2$ is observed in an extremely narrow pressure region around P_c.

In an antiferromagnet $CeRhIn_5$, the Fermi surface is unchanged up to 2.1 GPa, which is larger than the critical pressure $P^* = 1.63$ GPa where superconductivity sets in. Interesting is that the topology of the Fermi surface is different between $CeRhIn_5$ with pressures up to 2.1 GPa and $CeCoIn_5$ at

ambient pressure. It is, however, expected that the topology of the Fermi surface in $CeRhIn_5$ is changed into that in $CeCoIn_5$ either at pressures higher than 2.5 GPa at which the superconducting transition temperature T_{sc} becomes a maximum, or at pressures higher than about 5 GPa at which T_{sc} becomes zero. A higher-pressure technique is needed to clarify it. It is left to the future study.

Finally no distinct change of the Fermi surface is observed at $P_c = 1.5$ GPa in URu_2Si_2, although the cyclotron mass decreases considerably with increasing pressure, reflecting an increase of magnetic moment.

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