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Thermal expansion and magnetostriction of heavy fermion $CeRu_2Si_2$ at millikelvin temperatures

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Abstract

We have measured linear thermal expansion and magnetostriction of single crystal $CeRu_2Si_2$ that is well known as a heavy fermion metamagnetic compound. Thermal expansion and magnetostriction along the a-axis (B $\parallel a$) and the c-axis (B $\parallel c$) were measured by the capacitive dilatometer at temperatures down to 12 mK and in magnetic fields up to 9 T. We observed a strong anisotropy between a and c axis. In addition, negative deviations from Landau-Fermi liquid behavior for thermal expansion and magnetostriction coefficients were found below 50 mK and 0.4 T indicating non Fermi liquid behavior.

Keywords: Heavy fermion, $CeRu_2Si_2$, Thermal expansion, Magnetostriction, Capacitive method

1 Introduction

Pressure and magnetic field may alter the ground state on heavy fermion systems near a quantum critical point which is a continuous order-disorder transition point at zero temperature. Ce heavy fermion systems have interesting properties taken up as a subject for investigating quantum phase transitions.

We focus on $CeRu_2Si_2$ which has a $ThCr_2Si_2$-type crystal structure and is well known as a heavy fermion compound. This system generally exhibits a Landau-Fermi liquid (LFL) state with an electronic specific heat coefficient $\gamma = 350$ mJ/K^{2}mol below the Kondo temperature $T_K = 14$ K. In the LFL state, thermal expansion and magnetostriction coefficients are proportional to the temperature and magnetic field, respectively [1, 2]. Applying a magnetic field on $CeRu_2Si_2$ induces a pseudo-metamagnetic transition from a Pauli paramagnetic to a highly polarized paramagnetic state at $B_m \sim 7.8$ T [3]. La or Rh substitution on the Ce or Ru sites in $Ce_{1-x}La_xRu_2Si_2$ and $Ce(Ru_{1-x}Rh_x)_2Si_2$ respectively, changes the volume via a negative chemical pressure effect of a few kbar, which leads to a quantum critical transition (QPT) from a Pauli paramagnetic to an antiferromagnetic (AFM) state. With decreasing x, the transition temperature decreases to absolute zero at the critical concentration $x_c = 0.075$ in $Ce_{1-x}La_xRu_2Si_2$ [4],

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and at $x_c = 0.03$ in $Ce(Ru_{1-x}Rh_x)_2Si_2$ [5]. Therefore, $CeRu_2Si_2$ is regarded as an example of a pressure-driven QPT material with the critical pressure $p_c \sim -0.3$ GPa [6] and several critical phenomena down to 0.1 K have been explained by the self-consistent renormalization (SCR) spin fluctuation theory [5, 7, 8]. In contrast to this nature, our previous experimental results of the magnetic susceptibility for undoped $CeRu_2Si_2$ along the c-axis at ultralow temperatures and in small magnetic fields have shown the non-Fermi liquid (NFL) behavior against the temperature independent Pauli paramagnetic susceptibility [9]. The measured NFL behavior could not be explained as the quantum criticality due to the AFM quantum critical point (QCP) and showed a resemblance to the magnetic-field-turned QCP.

Thermal expansion and magnetostriction are the strain responses of temperature and external or internal magnetic field, respectively. Their temperature and magnetic field derivatives are thermal expansion and magnetostriction coefficients which are relating to the partial derivative of specific heat and magnetization with respect to pressure. Therefore, these properties, which can be observed sensitively around the phase boundary [10], have been used for the study of phase transitions, quantum criticality, and other interesting phenomena [11].

Accordingly, we have developed a compact capacitive dilatometer and a capacitance bridge, and have reported results of the heavy fermion compound $CeRu_2Si_2$ along the *c*-axis at temperatures down to 1 mK in magnetic fields up to 52.6 mT [12, 13]. In addition, we have developed an experimental system using a ^{3}He-^{4}He dilution refrigerator in a high magnetic field up to 9 T [14]. In this report, we present thermal expansion and magnetostriction of the $CeRu_2Si_2$ along the *a*-axis at temperatures down to 12 mK and in magnetic fields up to 9 T.

2 Experimental Procedures

2.1 Dilatometer using the capacitive method

Among the various methods for measuring length changes induced by temperature and magnetic field, a capacitive method has high sensitivity and low power dissipation suitable for the low millikelvin temperature measurements. Our dilatometer has a three-terminal capacitive structure with a diameter of 12 mm and length of 30 mm as shown in Fig. 1. Its size is compact enough for being used in the limited space at millikelvin temperatures [13]. It consists of three parts made of oxygen-free high-conductivity copper for high thermal conductivity. Capacitor plates with a diameter of 5 mm and thickness of 2 mm are glued to the upper, middle holder and sample using Stycast 2850FT. A sample is cut into a cylinder or a cube with 3∼5 mm in diameter and 4∼5 mm in length and is glued to the lower holder using Arzerite VL-10 silver paste for thermal conduction. Each plate is polished by the sandpaper in order to have the

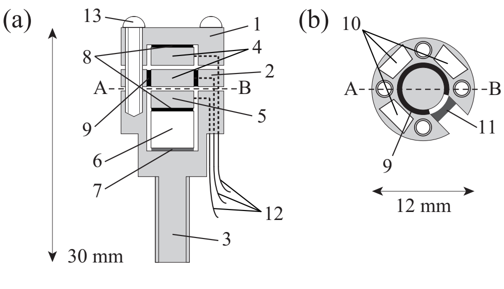

Figure 1: Schematic drawing of the capacitive dilatometer. (a) Three-terminal capacitive dilatometer. (b) cross-sectional view of the capacitor: (1) Upper holder; (2) middle holder; (3) lower holder; (4) fixed capacitor plates; (5) movable capacitor plate; (6) sample; (7) silver paste; (8) Stycast FT and cigarette paper; (9) Stycast FT; (10) copper foil spacers; (11) silver epoxy; (12) silver-coated copper coaxial cables; (13) M2 brass screws (four in total) [14].

same plane with the holder. Copper foil spacers are inserted to obtain a close spacing between capacitance plates. The measured capacitance and loss of capacitor are ∼11 pF and ∼0.01 nS at 1 K in a vacuum, respectively.

The dilation ΔL of sample length L is related with spacing d between a pair of capacitor plates as $\Delta L = -\Delta d$, and measured capacitance is given as $C = \varepsilon_0 S/d$, where ε_0 is the permittivity of vacuum, S is the area of the capacitor plate. In actual measurements, the dilation of holders, capacitor plates and Stycast by temperature and field change affect the measured capacitance change, which is known as the cell effect. The change rate of C in the sample and the reference capacitance are given by

$$\frac{\Delta C_x}{C_x} = \frac{L}{d_x}\left\{\left(\frac{\Delta L}{L}\right)_{\mathrm{sample}} - \left(\frac{\Delta l}{l}\right)_{\mathrm{Cu}}\right\} + \frac{l_{\mathrm{sty}}}{d_x}\left\{\left(\frac{\Delta l}{l}\right)_{sty} - \left(\frac{\Delta l}{l}\right)_{Cu}\right\} - \frac{S_p}{d_x}\left(\frac{\Delta l}{l}\right)_{\mathrm{Cu}}, \quad (1)$$

$$\frac{\Delta C_{ref}}{C_{ref}} = \frac{l_{\mathrm{sty}}}{d_{ref}}\left\{\left(\frac{\Delta l}{l}\right)_{\mathrm{sty}} - \left(\frac{\Delta l}{l}\right)_{\mathrm{Cu}}\right\} - \frac{S_p}{d_{ref}}\left(\frac{\Delta l}{l}\right)_{\mathrm{Cu}}, \quad (2)$$

where C_x and C_{ref} are the sample and the reference capacitance, d_x and d_{ref} are distance between capacitor plates of C_x and C_{ref}, and l_{sty}, l_{Cu}, S_p are the thickness of Stycast, capacitor plate, copper foil spacer, respectively. In Eq. (1), the second and last terms is the cell effect, which are usually obtained by measurements for the copper standard sample. From Eqs. (1) and (2), the cell effect is equivalent to the change of reference capacitor in the case of $d_x = d_{ref}$ and $\Delta d \ll d$. We measured that $\Delta C_{ref}/C_{ref}$ was proportional to T^2 and less than 2% for $\Delta C_x/C_x$ below 1 K [14].

2.2 Capacitance bridge

We have used a capacitance bridge based on ratio-transformer, as shown in Fig. 2. A seven-decade ratio transformer with output impedance R_{r} of 5 Ω is used to compare a sample capacitance with a reference capacitance. A six-decade resistor R_{b} with resolution of 1 mΩ is used to balance the resistive parts R_1 and R_2. A high-sensitivity lock-in amplifier is used as an off balance voltage detector. The input voltage of the circuit V_i is less than 5 V_{rms} at frequency about 1.5 kHz. A balance ratio r is obtained by an off balance voltage V_0, a ratio sensitivity of V_0 and a setting of the ratio transformer. When the in-phase component of V_0 is balanced, the ratio gives as $C_x/C_{ref} = r/(1-r)$. A linear expansion is given by $\Delta L/L = (d_x/L)\Delta C/C$ in an ideal case. Assuming that the cell effect is equal to the change rate of reference capacitance, the $\Delta L/L$ is given by

$$\left(\frac{\Delta L}{L}\right)_{sample} - \left(\frac{\Delta l}{l}\right)_{Cu} = \frac{d_x}{L}\left(\frac{\Delta C_x}{C_x} - \frac{\Delta C_{ref}}{C_{ref}}\right) = \frac{d_x}{L}\frac{\Delta r}{r(1-r)}. \quad (3)$$

Here, thermal expansion of copper, which is less than $(\Delta l/l)_{Cu} \sim 10^{-10}$ below 1 K [15], is negligibly small compared with that of $CeRu_2Si_2$, $\Delta L/L \sim 10^{-6}$ at 1 K. Thus, the linear expansion of sample can be measured within the accuracy of $10^{-9} \sim 10^{-10}$ [13,14].

2.3 High magnetic field measurements

We used a ^{3}He-^{4}He dilution refrigerator and a 9 T superconducting magnet for thermal expansion and magnetostriction measurements [14]. In high magnetic field measurements, we have to take into account the nuclear specific heat of the copper and the eddy current by applying magnetic fields because our dilatometer and a thermal link are made of OFC. At millikelvin

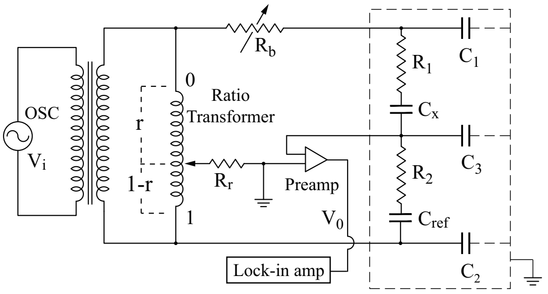

Figure 2: Diagram of the ratio-transformer-based capacitance bridge, where C_x and C_{ref} are respectively the sample and the reference capacitance, and C_1, C_2 and C_3 are the cable capacitances. R_b is adjusting resistive part of the bridge. The dashed line indicates parts being located in the cryostat [14].

temperatures, the eddy current heating caused by applying magnetic fields affects isothermal magnetostriction measurements, therefore the thermal link has slits for reducing the eddy current heating. The eddy current heating is given by $\dot{Q}_{eddy}(t) \propto (\partial B/\partial t)^2$ which is estimated to be ~ 0.5 μW when a rate of applied magnetic field is $\dot{B}(t) = 1.44$ T/hour, and temperature stability ΔT is less than 1 mK at 98 mK at this field sweep rate [14].

Temperature was measured using a ^{3}He melting curve thermometer (MCT) at $T < 250$ mK and a RuO_2 resistance thermometer at $T > 250$ mK, respectively. The field influence for thermometry is negligible in our measurements because the stray field around thermometers is compensated to be less than 16 mT by the compensating coil when a magnetic field of 9 T is applied. Furthermore, the field dependence of a MCT is known to be $\Delta T/T = 2$ % at 25 mK and 5 T [16], and the accuracy of temperature measurement for the MCT is ensured in high magnetic fields.

3 Results

Figure 3 shows the temperature dependence of the linear thermal expansion of $CeRu_2Si_2$ measured along the a-axis in magnetic fields up to 5 T. As shown in the inset of Fig. 3, the linear thermal expansion shows T^2 dependence up to 1 K in the zero field, expressed as $\Delta L(T)/L = aT^2/2$. The coefficient a is obtained to be 2.68×10^{-6} K^{-2}. From this result along the a-axis and our previous result along the c-axis [12], the anisotropy ratio of the thermal expansion coefficient turns out to be $c/a \sim 2$. As shown in Fig. 3, the linear thermal expansion shows T^2 dependence above 50 mK. This temperature dependence indicates that the system is in the LFL state. However, with decreasing temperature, the pronounced negative deviations from the LFL behavior are observed in all measured fields. Figure 4 shows the temperature dependence of the linear thermal expansion coefficient $\alpha = d(\Delta L(T)/L)/dT$. The deviation temperature of about 50 mK is in agreement with our previous report based on linear thermal expansion and magnetostriction measurements as well as studies of the magnetic susceptibility along the c-axis [9, 12]. Thus, $CeRu_2Si_2$ shows the crossover from the LFL to the NFL state at about 50 mK.

Figure 5 shows the magnetic field dependence of linear magnetostrictions, $\Delta L(B) \parallel c$ $(B \parallel c)$ and $\Delta L(B) \parallel a$ $(B \parallel a)$ at 2 K. In the low field region, the linear magnetostriction expressed as $\Delta L(B)/L = bB^2/2$ indicates the LFL state. The coefficients b in the LFL state are obtained to be $b_c = 8.8 \times 10^{-6}$ T^{-2} and $b_a = 1.33 \times 10^{-8}$ T^{-2}, respectively. Thus, the anisotropy ratio of magnetostriction coefficients turns out to be $c/a \sim 660$. A pseudo-metamagnetic transition is observed along the c-axis at $B_m \sim 8$ T, but not observed along the a-axis up to 9 T. Figure 6 shows the field dependence of the linear magnetostriction coefficient $\lambda = d(\Delta L(B)/L)/dB$ along the a-axis in the low field region. The magnetostriction coefficient is proportional to B above

0.4 T in all measured temperatures. However, the negative deviation from the B dependence is observed below 0.4 T at low temperatures. This negative deviation becomes large with decreasing temperature, and shows a maximum at around 0.1 T.

In the temperature vs pressure phase diagram with an antiferromagnetic(AFM) ordered phase extended to a finite temperature from a quantum critical point(QCP), the isentropes have a minimum on the phase boundary and at the QCP [17]. Thus, in the high pressure side of QCP, the entropy decreases with increasing pressure. It means that the sign of a quantum critical contribution to the thermal expansion coefficient should be positive from the Maxwell relation $(\partial V/\partial T)_p = -(\partial S/\partial p)_T$. In the T–p diagram of $CeRu_2Si_2$, the AFM transition boundary ends to the AFM QCP at -0.3 GPa [6]. Thus, the critical contribution to the thermal expansion coefficient due to the AFM QCP is expected to have a positive value. One possible explanation for our observed negative deviation in thermal expansion is that a hidden QCP may exist in the region of higher-than-ambient pressure. Assuming that $CeRu_2Si_2$ at ambient pressure is located closer to this hidden QCP than the AFM QCP, the entropy increases with increasing pressure owning to the hidden QCP, thus the negative deviation appears as a quantum critical contribution. From the Maxwell relation $(\partial M/\partial p)_B = -(\partial V/\partial B)_p$, the negative deviation of the magnetostriction coefficient implies that magnetization increases with pressure, which is expected from the quasiparticle mass enhancement in the approach to the QCP. Thus, the negative deviation of the magnetostriction coefficient is consistent with the existence of a hidden QCP at higher-than-ambient pressure.

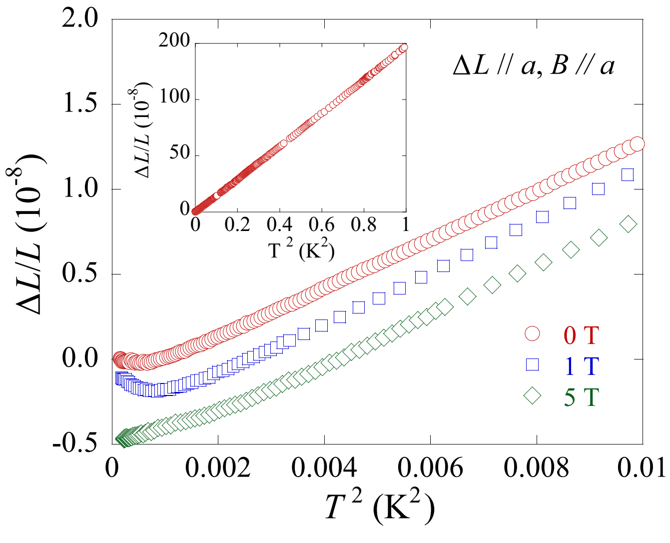

Figure 3: Linear thermal expansion of $CeRu_2Si_2$ along the a-axis at different applied fields, plotted as a function of T^2. The data are subsequently shifted. The inset shows T^2 dependence of $\Delta L(T)/L$ below 1K.

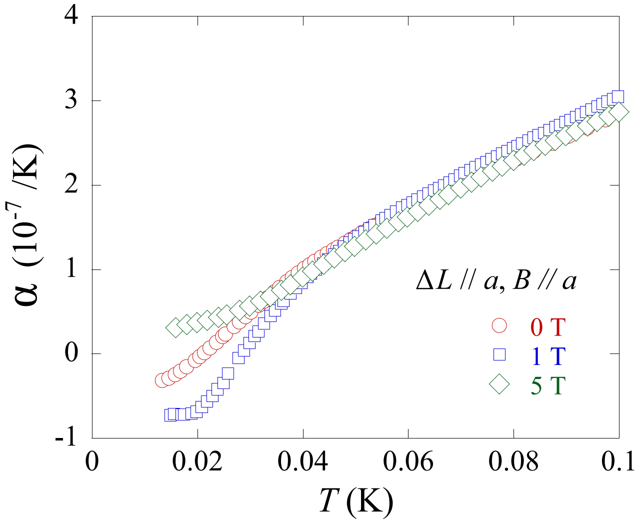

Figure 4: Linear thermal expansion coefficient α of $CeRu_2Si_2$ along the a-axis, obtained from the data in Fig. 3.

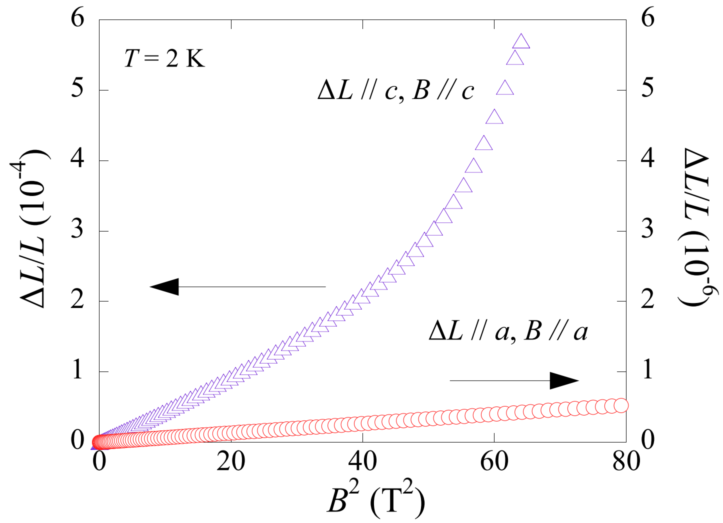

Figure 5: Linear magnetostriction of $CeRu_2Si_2$ along the c-axis and the a-axis at 2 K, plotted as functions of B^2.

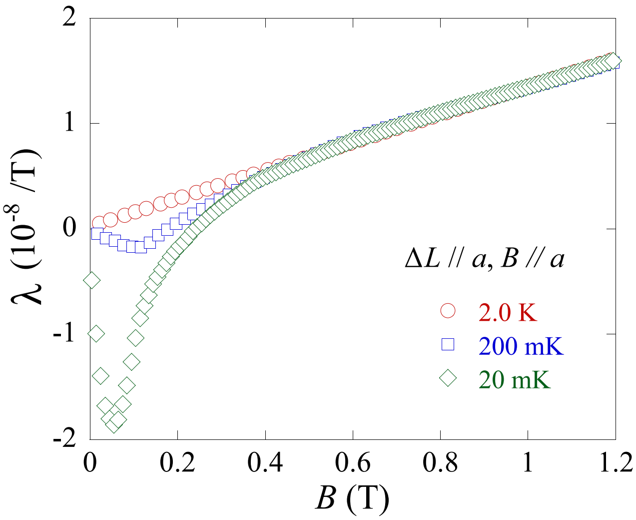

Figure 6: Linear magnetostriction coefficient λ of $CeRu_2Si_2$ along the *a*-axis at millikelvin temperatures as a function of magnetic field.

4 Summary

We report the linear thermal expansion and magnetostriction measurements of the heavy fermion compound $CeRu_2Si_2$ along the *a*-axis in the magnetic fields up to 9 T. The large anisotropy ratio of magnetostriction coefficients was found and the pseudo-metamagnetic transition was not observed along the *a*-axis up to 9 T. This compound showed the pronounced NFL behavior in linear thermal expansion and magnetostriction below 50 mK and below 0.4 T. The negative deviations from the LFL behavior suggest increases in entropy and magnetization with pressure, and we discuss the possibility of a hidden QCP.

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