

Thermal expansion and magnetostriction measurements using a high sensitive capacitive dilatometer at millikelvin temperatures

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Abstract. We have developed a dilatometric measuring system for thermal expansion and magnetostriction, those are more singular than specific heat in approaching to a quantum critical point. With decreasing temperature, thermal expansion becomes small in proportional to the square of temperature, thus, high sensitivity and reproducibility are necessary for the dilatometric measurements in millikelvin temperatures. Our dilatometer composed of the sample and the reference capacitor provides the extremely high resolution of $\Delta L/L \sim 10^{-10}$ using the ratio-transformer-based capacitance bridge. The dilatometer was installed on the ^3He - ^4He dilution refrigerator with the 9 T superconducting magnet, and temperature was measured by the ^3He melting curve thermometer. We have measured thermal expansion and magnetostriction of the typical heavy fermion compound CeRu_2Si_2 along a -axis at temperature down to 10 mK in magnetic fields up to 9 T.

1. Introduction

Dilatometry has been a powerful technique for investigating the thermodynamic properties of materials. Thermal expansion and magnetostriction, those are the strain responses of a material to temperature and an external or internal magnetic field, have a close relationship with the partial derivative of specific heat and magnetization with respect to pressure. Thus, these properties have been good measures of phase transitions, quantum criticality, and other interesting phenomena [1]. Among the various methods for measuring length changes induced by temperature and magnetic field, capacitive dilatometry excels in its sensitivity reaching values of $\Delta L/L \sim 10^{-10}$ at low temperatures in magnetic fields. Many types of capacitive dilatometers have been developed for various experiments, such as absolute expansion dilatometer [1], dilatometer with an open architecture sample mounting arrangement [2], and small dilatometers for magnet systems [3]. In the low millikelvin temperature range, the capacitive dilatometer has another advantage in low power dissipation. However, sufficient thermal contact for a sample cooling without any heat exchange gases and liquids is important in this temperature range. The limited space in magnet systems for magnetostriction measurement also requires a small size of dilatometer. Therefore, the capacitive dilatometer has been used in only a few investigations down to low millikelvin temperatures [4–6]. We developed the compact capacitive dilatometer and the capacitance bridge, and have reported the results of the heavy fermion compound CeRu_2Si_2 along c -axis at temperature down to 1 mK in magnetic fields up to 52.6 mT [7,8]. In



the present report, we have developed our dilatometric measurements extending the magnetic field range up to 9 T at 10 mK. The thermal expansion and magnetostriction of CeRu_2Si_2 along a -axis have been measured where the accuracy of $\Delta L/L$ is better than 1×10^{-10} and the temperature stability $\Delta T/T$ is $\sim 10^{-2}$ in isothermal magnetostriction measurements.

2. Experiment

2.1. Dilatometer

Figure 1 shows our three-terminal capacitive dilatometer with a diameter of 12 mm and length of 30 mm, which was small enough to be used in the limited space [8]. Most of the parts were made of oxygen-free high-conductivity copper for high thermal conductivity. Capacitor plates with a diameter of 5 mm and thickness of 2 mm were glued to the upper, middle holder and sample using Stycast 2850FT. The thickness of Stycast on the upper and the movable capacitor plate were typically 0.1 mm. Cigarette papers were inserted in the Stycast for ensuring an electrical insulation. The sample was cut into a cylinder or a cube with 3~5 mm in diameter and 4~5 mm in length, and was glued to the lower holder using Arzerite VL-10 silver paste for thermal contact. The copper spacers were inserted among holders to obtain a close spacing which was less than 20 μm , and three holders and plates composed two capacitors, sample and reference. Thermal expansion and magnetostriction as a linear strain is given by $\Delta L/L = d_x/L \times \Delta C/C$ in an ideal parallel plate model, where L , C , and d_x are the sample length, capacitance, and distance between plates, respectively. However, the dilation of holder, capacitor plates and Stycast affect the measured capacitance values [9]. Hence, change rates of $\Delta C/C$ in the sample capacitance C_x and the reference capacitance C_{ref} are given by

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

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

where d_x and d_{ref} are distance between capacitor plates of C_x and C_{ref} , and $l_{\text{sty}} \sim 0.1$ mm, $l_{\text{Cu}} = 2$ mm, $S_p = 15$ μm are the thickness of Stycast, capacitor plate, copper foil spacer, respectively. The second and last terms in Eq. (1), known as the cell effect, are usually obtained by measurements for the copper standard sample using the same dilatometer. In our dilatometer design where the sample is glued to the holder and capacitor plate, it is difficult to replace the sample so that the cell effect has completely the same value. From Eqs. (1) and (2), the cell effect of the sample capacitor is expected to be same as that of the reference capacitor, if $d_x = d_{\text{ref}}$. Figure 2 shows the temperature dependence of the sample capacitance C_x and the reference capacitance C_{ref} of the dilatometer for the heavy fermion compound CeRu_2Si_2 along the a -axis measured by the automatic capacitance meter AH 2700A [10]. Both capacitance changes are proportional to T^2 , but $\Delta C_{\text{ref}}/C_{\text{ref}}$ is less than only 2% for $\Delta C_x/C_x$ at 1 K.

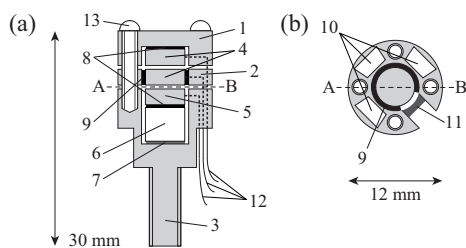


Figure 1. Schematic drawing of the capacitive dilatometer. (a) Three-terminal capacitive dilatometer. (b) Cross-sectional view of the capacitor: (1) Upper holder; (2) middle holder; (3) lower holder; (4) fixed capacitor plate; (5) movable capacitor plate; (6) sample; (7) silver paste; (8) Stycast FT 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).

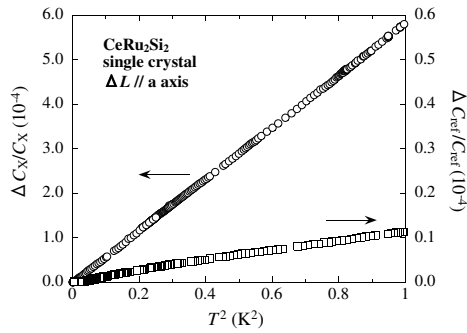


Figure 2. Capacitance change rate of the sample and the reference capacitor for CeRu₂Si₂ along the *a*-axis as a function of T^2 in a zero magnetic field.

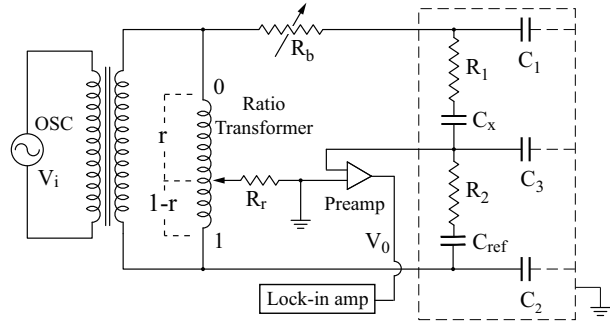


Figure 3. Diagram of the ratio-transformer-based capacitance bridge, where C_x and C_{ref} are respectively the sample and the reference capacitor, and C_1 , C_2 and C_3 are the cable capacitances. R_b is adjusting resistor. The region among the dashed line is installed in the refrigerator.

2.2. Capacitance bridge

Our ratio-transformer-based capacitance bridge is shown in Fig. 3. A seven-decade ratio transformer with output impedance R_r of 5 Ω was used to compare a sample capacitance to be determined to a reference capacitance. A six-decade resistor R_b with resolution of 1 m Ω and stability of approximately 20 ppm/K was used to balance the resistive parts R_1 and R_2 . A high-sensitivity lock-in amplifier with a 40 dB preamplifier was used as a null detector. The input voltage of the circuit V_i was 5 V_{rms} at frequencies between 1 and 3 kHz. If the resistive part of the bridge is adjusted by R_b , the output off-balance voltage V_0 can be given as [11]

$$\frac{V_0}{V_i} = \frac{(r-1)C_x + rC_{ref}}{C_x + C_{ref} + C_3} - j\omega R_r \frac{C_x + C_{ref}}{C_x + C_{ref} + C_3} ((r-1)C_1 + rC_2), \quad (3)$$

where r is the ratio-transformer setting. When the in-phase component is balanced, that gives $C_x/C_{ref} = r/(1-r)$ equating to the Wheatstone bridge. Practically, the capacitance change resulting from thermal expansion and magnetostriction induces the off-balance V_0 . Thus, balanced r is calculated from the off-balance voltage of the in-phase component, the ratio setting, and the ratio sensitivity. Accordingly, thermal expansion and magnetostriction as $\Delta L/L$ using the capacitance bridge are given by

$$\left(\frac{\Delta L}{L}\right)_{\text{sample}} - \left(\frac{\Delta l}{l}\right)_{\text{Cu}} = \frac{d_x}{L} \left(\frac{\Delta C_x}{C_x} - \frac{\Delta C_{ref}}{C_{ref}}\right) = \frac{d_x}{L} \frac{\Delta r}{r(1-r)}. \quad (4)$$

Thermal expansion of CeRu₂Si₂ is $(\Delta L/L)_{\text{sample}} \sim 1 \times 10^{-6}$ at 1K, therefore, $(\Delta l/l)_{\text{Cu}}$ is negligibly small because this is $\sim 10^{-10}$ below 1 K [12].

2.3. High magnetic field system

We developed an experimental setup in the ³He-⁴He dilution refrigerator with the 9 T superconducting magnet for measuring thermal expansion and magnetostriction in high magnetic fields. Figure 4 shows our experimental set up and magnetic field profile in the field of 9 T at $Z = 0$ mm. Thermally-annealed oxygen-free high-conductivity copper thermal link intervening ³He melting curve thermometer (MCT), the dilatometer, and the experiment stage had the particular structure that was the “U” shaped with a cross-section of 12×12 mm² and height of 256 mm. The dilatometer placed in the center of the magnetic field can be settled in either direction, parallel and perpendicular to the magnetic field. One extremity of the thermal link was connected to the experiment stage where there were a carbon resistance thermometer and the heater for

temperature control. In contrast, the other extremity was connected to the MCT but thermally insulated to the experiment stage by Vespel. The field dependence of the MCT is known to be $\Delta T/T = 2\%$ at 25 mK and 5 T [13], hence the location of the MCT at $Z = 240$ mm where $B \sim -16$ mT at the maximum field of 9 T ensured the accuracy of temperature measurement. Thermal link has six slits in order to reduce the eddy current heating $\dot{Q}_{\text{eddy}}(t) \sim (\partial B/\partial t)^2$ which is estimated to be $\sim 0.5 \mu\text{W}$ when a rate of applied magnetic field is $\dot{B}(t) = 2 \times 10^{-4}$ T/sec. Figure 5 shows the typical temperature stability in field sweeps at 98 mK. It indicates that the temperature change by the field sweep is less than about 1 mK even at the rate of 4×10^{-4} T/sec. In fact, we applied the magnetic fields in the rate of $\sim 10^{-5}$ T/sec for measuring the magnetostriction, thus, the typical temperature stability was $\Delta T/T = 5 \times 10^{-3}$ at 98 mK, and less than $\sim 10^{-2}$ down to 14 mK.

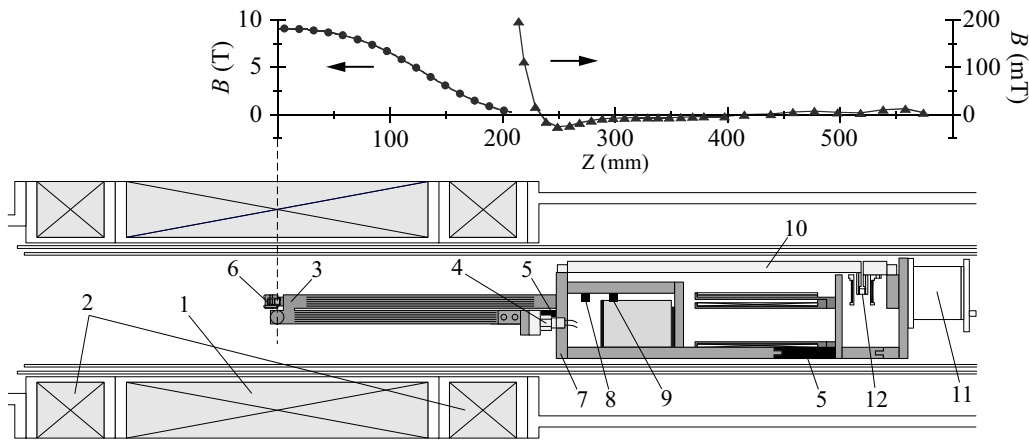


Figure 4. Schematic drawing of our experimental set up and magnetic field profile: (1) main magnet; (2) compensation coil; (3) copper thermal link; (4) ^3He melting curve thermometer; (5) Vespel; (6) dilatometer; (7) experiment stage; (8) carbon resistance thermometer; (9) heater; (10) silver thermal link; (11) mixing chamber of the ^3He - ^4He dilution refrigerator; (12) indium heat switch. The field profile is represented by the circle in the left scale and the triangle in the right magnified scale.

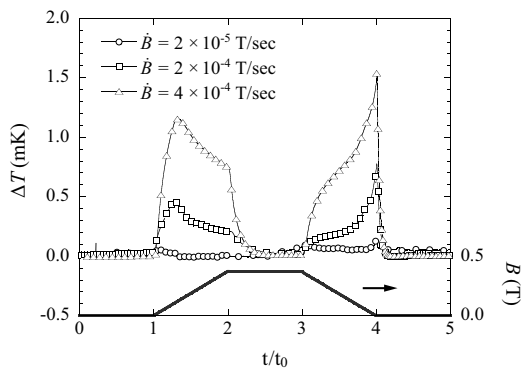


Figure 5. Temperature stability in field sweeps at 98 mK as a function of normalized time t/t_0 . t_0 are 9.0×10^2 , 1.8×10^3 , and 1.8×10^4 s in the rate of 4×10^{-4} , 2×10^{-4} , and 2×10^{-5} T/s, respectively. Solid line represents magnetic field strength.

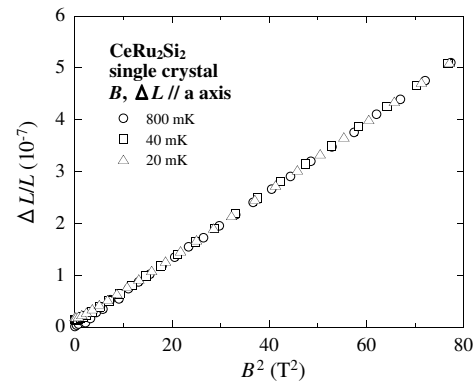


Figure 6. Isothermal magnetostriction of CeRu_2Si_2 along the a -axis as a function of B^2 .

3. Results

We have measured magnetostriction of a typical heavy fermion compound CeRu_2Si_2 at temperature down to 10 mK for the magnetic field direction along the a -axis up to 9 T. Figure 6 shows measured magnetostrictions up to 9 T at 20, 40, 800 mK, where the field sweep rate were $\dot{B}(t) = 2.4 \times 10^{-5}$, 4×10^{-5} , and 2×10^{-4} T/sec, respectively. Below 200 mK, the accuracy of magnetostriction measurements being better than 1×10^{-10} were achieved by the ratio-transformer-based capacitance bridge. In contrast, the accuracy was 2×10^{-8} at 800 mK using the capacitance meter AH 2700A in the averaging time of 40 sec. The isothermal linear magnetostriction is proportional to B^2 and the metamagnetic behavior cannot be observed up to 9 T. The magnetostriction coefficient along a -axis, $\lambda_a = \partial(\Delta L/L)/\partial B$, is independent of temperature as $\lambda_a/B = 1.3 \times 10^{-8} \text{ T}^{-1}$, which is 800 times smaller than that along c -axis [14]. These differences among axes in the high field region agree with the anisotropic nature of this compound observed by other magnetic properties [15]. On the other hand, the deviation from B^2 dependence became significant around zero magnetic field with decreasing temperature. The similar behavior was observed by the previous measurements along c -axis [7]. Thus, it indicates that this compound shows an axis-independent quantum criticality below 40 mK around zero field.

4. Summary

We reported the new design of a capacitive dilatometer, a capacitance bridge system and high magnetic field system for accurate measurement of thermal expansion and magnetostriction at millikelvin temperatures in high magnetic fields. The capacitance bridge system ensured the high accuracy and high stability of $\Delta L/L$ being less than $\sim 2 \times 10^{-10}$ in the long-term experiment. The high magnetic field system enabled the isothermal magnetic measurement up to 9 T in the rate of $\dot{B}(t) \sim 10^{-5} \text{ T/sec}$. Thus, our experimental device provides sufficient performance under multi-extreme conditions.

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