

# An Ultrasensitive Differential Capacitive Dilatometer

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In this paper, we describe an ultrasensitive differential capacitive dilatometer ( $\Delta L/L \sim 10^{-9}$ ) and novel ratiometric measurement method in the temperature range of  $1.9 \text{ K} < T < 400 \text{ K}$  for the investigation of thermal expansion and magnetostriction of single crystal as well as polycrystalline samples. The dilatometer provides a capacitive measurement based on a differential capacitance or the capacitance ratio measured between two parallel plate capacitors of the dilatometer. In this regard, an absolute capacitance bridge is not required, and even more advantageously, the effect of adsorbed gas on the capacitor plates is greatly reduced compared to conventional dilatometers. The differential capacitive dilatometer provides a symmetrical configuration which reduces the effect of temperature gradients. The capabilities of the dilatometer are demonstrated by the measurement of magnetic ordering of a  $\text{URu}_2\text{Si}_2$  sample, and of de Haas–van Alphen oscillations in an aluminum sample.

**Index Terms**—Capacitor, differential, dilatometer, magnetostriction, thermal expansion.

## I. INTRODUCTION

A DILATOMETER is an ultrasensitive instrument for measuring the dilation of a material, often measured along a specific crystallographic axis, which is brought about by changes in quantized vibrations of the lattice (phonons), and to a smaller degree of conduction electrons or cooperative ordering of spins when either is present. Thermal expansion is a fundamental property of all materials; yet, the capability of measuring the coefficient of thermal expansion is not readily available in most laboratories, especially in an external applied magnetic field and at cryogenic temperatures in the order of a few degrees of Kelvin [1], [2]. Various applications for dilatometers may include: locating phase transitions in materials, predicting pressure effects in superconductors, characterizing cryogenic construction materials, magnetostriction studies, or providing information that is complementary to heat capacity data<sup>1</sup> [3].

In this paper, we introduce a miniature dilatometer constructed entirely from fused silica (FuSi) that benefits from low background versus oxygen-free high thermal conductivity (OFHC) copper as shown in Fig. 1, and is specifically configured for measuring a capacitance ratio rather than an absolute value. Thanks to the ratiometric nature of the measurement, the effect of adsorbed gas on the capacitance plates is greatly reduced. This is of particular importance for cryogenic applications where helium exchange gas is used to cool the dilatometer probe. In previous designs, changes in helium

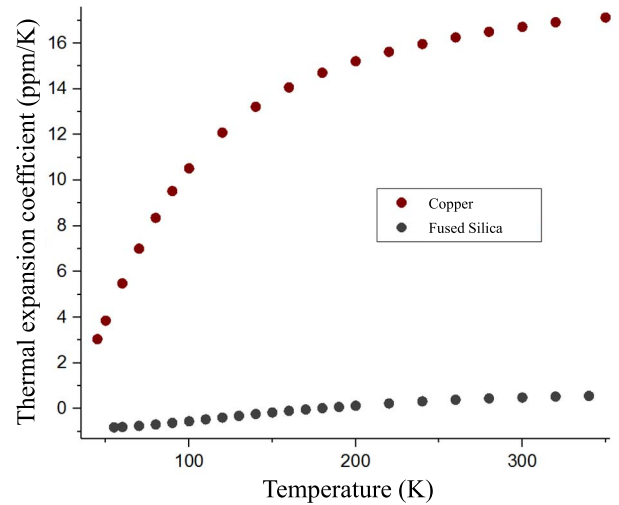


Fig. 1. Reported thermal expansion coefficients of copper, a common dilatometer construction material, and FuSi, the material comprising the QD dilatometer [6].

density over the temperature range of an experiment would require that these effects be subtracted out manually from the data. The small size of the dilatometer cell and symmetry of the capacitor plates reduce the effect of temperature gradients. By mounting the sample on the cell centerline, detrimental torque effects on the cell can also be avoided. Finally, multiple electrodes attached at various points of the cell provide for balancing of forces, which eliminates spurious signals that would otherwise need to be subtracted from the dilation measurement of the sample. This system has been proven to be compatible with the cryogen-free physical property measurement system (PPMS) platforms, which employ either a Gifford McMahon or a pulse tube cryorefrigerator.

## II. APPARATUS DESCRIPTION

The dilatometer consists of the following hardware: capsule, probe, and Controller Area Network bus module.

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<sup>1</sup>Grüneisen parameter:

- $\gamma = \left( \frac{dP}{d(E/V)} \right)_V = \frac{\partial V}{C_V \chi T}$
- $C_V$  = heat capacity,  $\chi T$  = compressibility
- $\gamma$  describes how volume changes effect vibrational properties
- For one mode:  $\gamma_i = -\frac{V}{\omega_i} \frac{\partial \omega_i}{\partial V}$
- Ehrenfest Relation:  $\frac{dT_C}{dP} = VT \frac{\Delta\beta}{\Delta C_P}$

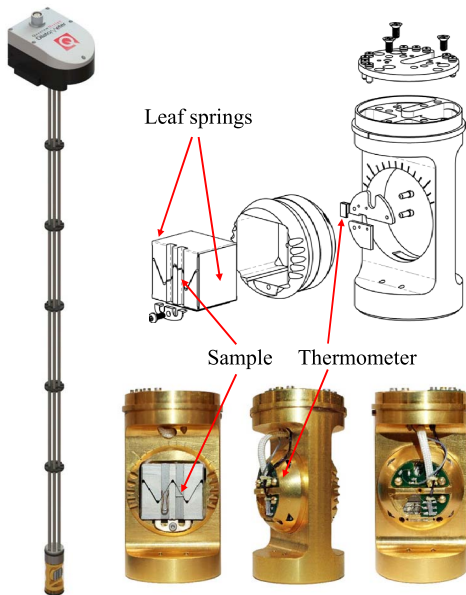


Fig. 2. Dilatometer probe assembly with exploded view of the capsule showing the leaf springs, the sample location, and the location of the thermometer behind the sample sphere.

### A. Cell Design and Construction

The dilatometer cell cube is made up of platinum-coated FuSi parts with isolated electrodes that comprise multiple capacitor plates. The gap between the plates is set to 0.12 mm by soldering two FuSi leaf springs in Fig. 2 to the sides of the cube. The springs provide a counterpoise force in a direction opposite to sample expansion. In general, it is preferred that the sample to be measured be 2 mm in length,  $\pm 50 \mu\text{m}$ . The smaller sample lengths can be accommodated by using FuSi shims, which can then be sandwiched together with the sample to make up the required 2 mm sample length.

Fig. 2 illustrates an exploded view of the capsule assembly. Within the cell cube, the electrical configuration consists of electrodes ablated from the dilatometer cell. Two sets of electrically connected positive capacitor “receiver” plates form the moveable top half of the cell. Two sets of fixed negative capacitor “transmitter” plates form the bottom half of the cell (i.e., rigid with respect to the dilatometer probe reference frame). The top portion of the cell will be displaced either to the right or left with respect to the bottom half of the cell according to the sample dilation. The displacement of the transmitter plates will, in the case of sample expansion, increase the gap and thus decrease the capacitance of the first set of capacitors, and decrease the gap and thus increase the capacitance of the second set of capacitors. Assuming an effective area  $A$  of the capacitor plates and gaps  $d_1$  and  $d_2$ , the capacitance of each capacitor is given by:  $C_1 = \epsilon(T)A/d_1$  and  $C_2 = \epsilon(T)A/d_2$ , where:  $\epsilon(T)$  is the dielectric constant of the gas between the capacitor plates. At equilibrium  $d_1 = d_2 = d$  is dictated by the length of the sample at room temperature and in zero applied magnetic field, and is nominally 2 mm. When the sample expands by an infinitesimal amount  $x$  due to a change in temperature and/or applied magnetic field, then the gaps in the capacitors will change to  $d_1 = d + x$  and  $d_2 = d - x$ .

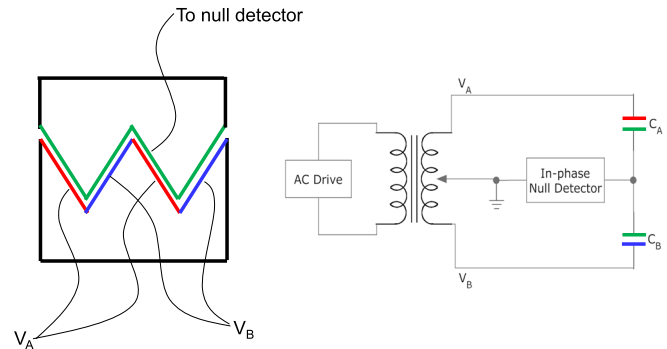


Fig. 3. Cell schematic illustrating drive voltages  $V_a$  and  $V_b$ , and parallel plate capacitors  $C_a$  and  $C_b$ . Capacitors  $C_a$  and  $C_b$  share common receiver plates which are connected to the null detector.

### B. Dilatometer Capsule and Probe

The dilatometer cell is mounted on a specially designed capsule at the end of the dilatometer probe. The probe is inserted into an isothermal chamber within the commonly owned and commercially available PPMS [4]. The PPMS is a versatile, low temperature cryostat capable of providing an environment with temperatures between 1.9 and 400 K, with fields up to 16 T. To accurately measure the sample temperature, a thermometer is mounted near the sample in the capsule, as shown in Fig. 2. The capsule is configured to allow the dilatometer cell to be rotated and locked in place via a set screw in order to accomplish measurements at various angles, over a  $110^\circ$  range with respect to an applied magnetic field. Shielded cables electrically connect the individual capacitance plates of the dilatometer with a preamplifier mounted on the top of the cryogenic probe.

### C. Ratiometric Capacitance Electronics

The dilatometer module contains electronics that simultaneously control the voltage applied to the capacitors, measure the imbalance signal, and measure the cell temperature. Fig. 3 shows an alternating current used to create differential drive voltages  $V_a$  and  $V_b$  for the two parallel plate capacitors [5]. The module operates at a null point which is important to minimize the effect of nonlinearities and stray capacitance. In particular, if the capacitance ratio measurement is seen as a transformer ratio arm bridge, then terms related to bridge excitation, detector impedance, and capacitance to ground fall out when the bridge is kept in balance. Closed-loop control is used to maintain a null signal on the phase detector by adjusting a switched ratio transformer and multiplying digital-to-analog converters. This is essential for adjusting the drive circuit to remain in balance and calculate sample expansion. This combination provides for an adjustment resolution of one part in  $10^7$ . Before closing the control loop, an initial balance is obtained by a binary search. In addition, the module reads temperature with high accuracy, which is synchronized at uniform time intervals with the imbalance.

In practice, the proportional–integral–derivative control loop will adjust the voltage amplitude across the ratio transformer windings by an amount  $\Delta U$ , to compensate for the change in gap across each of the two capacitors. The voltage across each

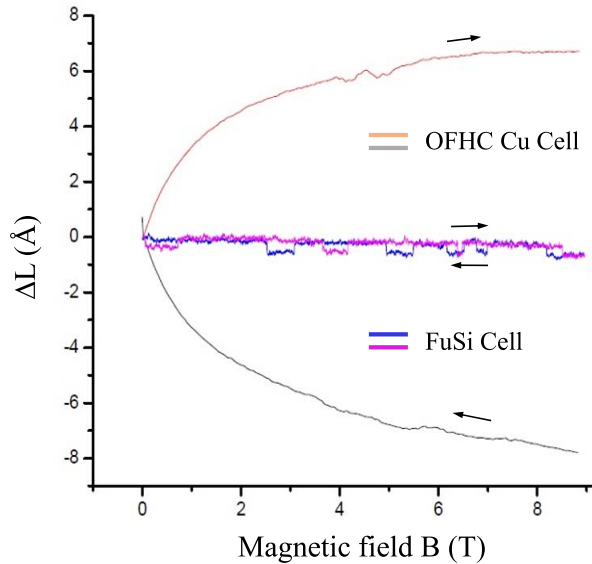


Fig. 4. Absolute change in the length scale of copper and FuSi dilatometer cells, as a function of applied magnetic field [11].

capacitor will then be

$$U_0 - \Delta U = \frac{I}{j\omega C_1}, U_0 + \Delta U = \frac{I}{j\omega C_2} \quad (1)$$

where  $j$  is the square root of  $-1$ , and  $\omega$  is the excitation angular frequency. Eliminating current  $I$  from (1) and solving for the gap expansion  $x$  we obtain the total imbalance

$$\frac{\Delta U}{U} = \frac{x}{d} \quad (2)$$

which thus provides a direct measurement of changes in the gap size. Note that in (2),  $x$  refers to the change in the gap of the cell. The change in the sample length (dilation) is  $\Delta L = x/\cos 30^\circ$  due to the geometry of the cell.

The simple result in (2) shows the important and experimentally observed fact that the measurement of the sample expansion is independent of the temperature-dependent permittivity  $\varepsilon(T)$  of the gas medium between the capacitor plates, thus greatly reducing the contribution of gas adsorbed on the capacitor plates of the cell to the raw data of the dilatometer. In practice, this will mean that when the first pumping out the dilatometer sample chamber from ambient pressure, permittivity changes by approximately  $\Delta\varepsilon \approx 590$  ppm while imbalance changes only by  $\Delta I \approx 26$  ppm, a dielectric medium rejection of over  $20\times$  [10].

### III. MEASUREMENT DATA

An important advantage of the current dilatometer design when compared to metallic dilatometers is the extremely low dilation background in an applied magnetic field [11], [12]. Eddy currents in metallic parts will induce heating in the sample and torque, which often translates into a large magnetic field-dependent background. Fig. 4 shows the comparison of the data taken at 10 K during up and down field sweeps with the Quantum Design (QD) dilatometer, and with a copper-based dilatometer. The OFHC copper body of the metallic dilatometer makes it susceptible to eddy currents while the

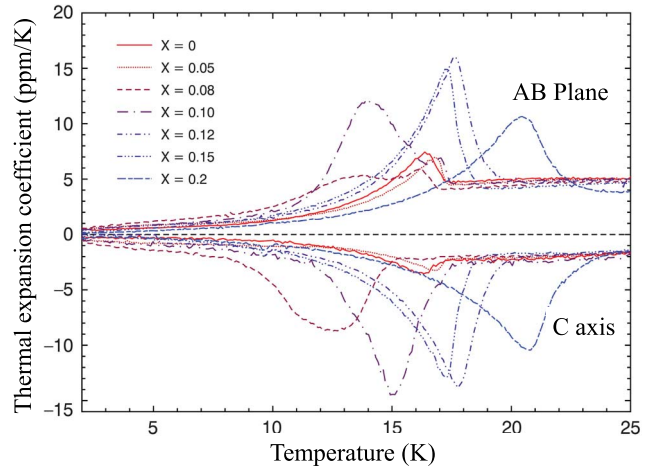


Fig. 5. Thermal expansion data along two unique crystallographic axes in  $URu_{2-x}Fe_xSi_2$ . The evolution of the material's phase transitions as Fe is substituted on the Ru site can be seen.

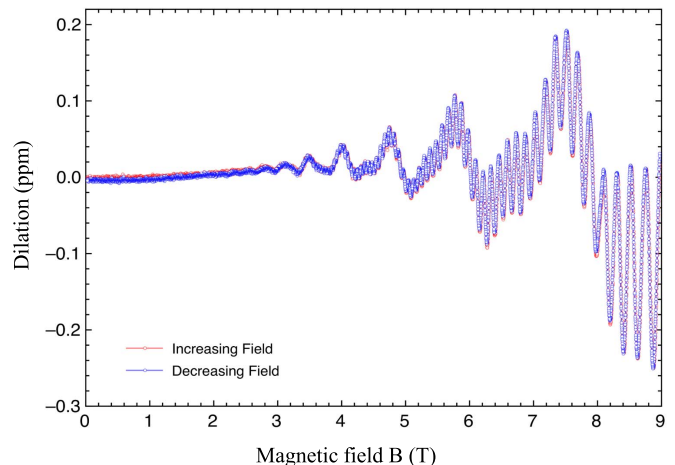


Fig. 6. Dilation data of an Al sample subjected to successive field sweeps up to a maximum field of 9 T and back down to 0 T at 10 K.

magnetic field is changing, leading to a much larger magnetic hysteresis. The QD dilatometer shows that negligible eddy current contributions are present in the background signal of the cell while sweeping the magnetic field. This makes the measurement less susceptible to artifacts that might be induced by torque and heating effects.

#### A. Thermal Expansion Measurements

Analysis of thermal expansion coefficient data was used to identify the ground state in  $URu_{2-x}Fe_xSi_2$ . Signatures of a large moment antiferromagnetic phase, as well as a hidden order phase, are shown in Fig. 5 [9]. Measurements along two distinct crystallographic axes are shown.

#### B. Magnetic Field Measurements

The transverse magnetostriction of a 2 mm aluminum polycrystalline sample (99.9995% pure) was measured at 10 K. The result, shown in Fig. 6, clearly depicts quantum oscillations due to the de Haas-van Alphen effect [7], [8], [11]. Furthermore, the agreement of the data collected on increasing and decreasing field sweeps demonstrates the superior repeatability of the measurement.

#### IV. CONCLUSION

In this paper, we described a versatile dilatometer constructed entirely from FuSi that employs a unique ratiometric capacitance measurement method. These novelties provide a new measurement capability for the PPMS system which presents several advantages over existing dilatometer designs. In the QD FuSi capacitive dilatometer, vanishingly small thermal and magnetic backgrounds of the empty cell allow for a simpler determination of sample effect when compared with competing designs. Ratiometric capacitance measurement allows for a dielectric medium rejection ratio of over  $20\times$ , important for instruments such as the PPMS in which the sample relies on a background partial pressure of helium for temperature control. While changes in helium density affect other cells to the first order with pressure-dependent background, the QD cell is insensitive to these changes.

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#### REFERENCES

- [1] T. H. K. Barron and G. K. White, *Heat Capacity and Thermal Expansion at Low Temperatures*. New York, NY, USA: Kluwer, 1999.
- [2] B. Yates, *Thermal Expansion*. New York, NY, USA: Plenum, 1972.
- [3] K. Andres, "The measurement of thermal expansion of metals at low temperatures," *Cryogenics*, vol. 2, no. 2, pp. 93–97, 1961.
- [4] N. Dilley, "PPMS family options compatibility," Quantum Design Inc., Nov. 2015. [Online]. Available: [https://www.qdusa.com/sitedocs/productBrochures/PPMS\\_Family\\_Options\\_Compatibility.pdf](https://www.qdusa.com/sitedocs/productBrochures/PPMS_Family_Options_Compatibility.pdf)
- [5] M. B. Simmonds, "Ultrasensitive ratiometric capacitance dilatometer and related methods," U.S. Patent 8992077, Mar. 31, 2015.
- [6] F. R. Kroeger and C. A. Swenson, "Absolute linear thermal-expansion measurements on copper and aluminum from 5 to 320 K," *J. Appl. Phys.*, vol. 48, no. 3, pp. 853–864, 1977.
- [7] W. J. de Haas and P. M. van Alphen, "The dependence of the susceptibility of diamagnetic metals upon the field," *Proc. Acad. Sci. Amsterdam*, vol. 33, pp. 1106–1118, Dec. 1930.
- [8] R. Griessen and J. L. Olsen, "The oscillatory magnetostriction in aluminum," *Solid State Commun.*, vol. 9, pp. 1655–1658, Oct. 1971.
- [9] S. Ran *et al.*, "Phase diagram and thermal expansion measurements on the system  $\text{URu}_{2-x}\text{Fe}_x\text{Si}_2$ ," *Proc. Nat. Acad. Sci. USA*, vol. 113, no. 47, pp. 13348–13353, 2016.
- [10] L. G. Hector and H. L. Schultz, "The dielectric constant of air at radiofrequencies," *Physics*, vol. 7, no. 4, pp. 133–136, 1936.
- [11] G. M. Schmiedeshoff *et al.*, "Versatile and compact capacitive dilatometer," *Rev. Sci. Instrum.*, vol. 77, p. 123907, Dec. 2006.
- [12] J. J. Neumeier, R. K. Bollinger, G. E. Timmins, C. R. Lane, R. D. Krogstad, and J. Macaluso, "Capacitive-based dilatometer cell constructed of fused quartz for measuring the thermal expansion of solids," *Rev. Sci. Instrum.*, vol. 79, p. 033903, Mar. 2008.